

Zeitschrift: Helvetica Physica Acta
Band: 51 (1978)
Heft: 1

Vereinsnachrichten: Rapport de la session de la Société Suisse de Physique à Berne les
7 et 8 octobre 1977

Autor: [s.n.]

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Rapport de la Session d'automne de la Société Suisse de Physique à Berne les 7 et 8 octobre 1977

Président: Prof. Ph. CHOQUARD, ETH Lausanne
Vice-président: Mme Prof. IRIS ZSCHOKKE-GRÄNACHER, Université de
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Secrétaire: Prof. PAUL ERDÖS, Université de Lausanne
Membres du bureau: Prof. PETER F. MEIER, Université de Zürich
Prof. ALEXANDRE HEYM, EPF Lausanne

Partie Administrative

1. Membres

Les membres ordinaires suivants ont été reçus au sein de notre Société:
A. BARATOFF (Gattikon), J.-L. BERTCHIER (Genève), D. BICHSEL (Genève), P. COLLET
(Genève), F. FUCHS (Zürich), J. GASS (Zürich), H. GRUNDER (Cal., USA), W. JOSS
(Zürich), B. JOYE (Fribourg), A. A. MANUEL (Genève), D. MEIER (Berne), U. MOSER
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(Lausanne), P. STREIT (Urdorf), F. THIEBAUD (Zürich), B. T. ULRICH (Genève),
B. VAUCHER (Fribourg), G. VIERTEL (Berne), H. P. WALKER (Berne), L. WEISS-
PARMEGGIANI (Genève), K. YVON (Genève).

La Société comprend actuellement 1179 membres individuels, 6 membres
collectifs et 13 bibliothèques.

2. Séance administrative

Ouverture de la séance: vendredi 7 octobre à 17 h 15.—Présents: 35 membres.

1. Procès-verbal de l'Assemblée Générale du 14.4.1977:

Approuvé à l'unanimité. Pas de commentaires.

2. Communications:

Concernant l'enquête sur la situation professionnelle des physiciens en Suisse,
deux envois ont été effectués, aux physiciens non membres et aux membres. Un
troisième envoi sera fait aux physiciens licenciés ou diplômés cette année et dont
la liste nous a été fournie par les universités suisses. Cette catégorie sera invitée à se
joindre à notre Société. Le Prof. J. P. Amiet (Neuchâtel) est chargé de l'évaluation
des résultats de l'enquête qui se fera probablement à Genève.

Un groupe des membres de la Société Suisse de Physique de Genève a émis le
voeu de créer des bourses destinées à des physiciens membres de la Société



Européenne de Physique pour passer 1 ou 2 ans dans un des instituts de physiciens suisses. Cette proposition est bien accueillie et le Président fera une enquête pour voir dans quelle mesure cela est possible.

3. Prochaine session

La session de printemps 1978 se tiendra à Neuchâtel les 30 et 31 mars. L'échéance pour l'envoi des communications est fixée au 10 février 1978. Il est à noter que les universités de Bâle, Zürich et Fribourg ont peu accueilli la Société jusqu'à présent et il serait donc souhaitable que ces villes soient mises à contribution ces prochaines années.

Dans le cadre de cette session de printemps, la maison Addison and Wesley a fait part de son désir d'exposer des livres. La suggestion est acceptée mais à condition que d'autres maisons puissent aussi participer. Une séance de posters est également à envisager et le Prof. Ch. Enz (Genève) est chargé de l'organiser.

4. Autres groupes

Du 25 au 28 septembre prochain une réunion commune est prévue avec la Société Autrichienne de Physique à Innsbruck. Le Comité suggère de donner des bourses à une vingtaine de jeunes pour qu'ils puissent s'y rendre. La proposition est adoptée.

D'autre part la Société est invitée par la Société Allemande de Physique à participer à une réunion commune avec la Société Autrichienne de Physique à Ulm lors de la réunion d'automne du 24 au 28 septembre 1979. La Société ne peut accepter cette invitation car, d'une part, elle a déjà une réunion commune avec la Société Autrichienne en 1978 et, d'autre part, le groupe suisse de physique nucléaire envisage de prendre une initiative en 1980 pour réunir les spécialistes de cette branche des pays limitrophes en Suisse. Enfin, il faut noter que l'Université de Berne a décidé d'organiser un symposium à l'occasion du 100ème anniversaire de la naissance d'Einstein en 1979.

5. Commissions

La Commission de Recherche présidée par le Prof. S. Steinemann (Lausanne) va s'achever par un rapport sur la Recherche et un autre sur la 'Relation entre Recherche et Enseignement dans les Universités Suisses'.

IUPAP: jusqu'à l'an dernier la représentation suisse pour l'IUPAP était nommée par la Société Helvétique des Sciences Naturelles, indépendamment de la Société Suisse de Physique. Il apparaît souhaitable d'intégrer le représentant suisse de l'IUPAP à la Société en invitant leur président à devenir membre du Comité de la Société qui serait ainsi au courant des activités et des projets de l'IUPAP. Le Prof. H. Staub (Zürich), président actuel, démissionne et le Prof. Mercier de Berne est proposé pour le remplacer. Il se choisira un adjoint. Ces propositions sont acceptées.

6. Nomination

Le Prof. I. Sick (Bâle) propose de nommer le Prof. H. Staub membre honoraire de la Société et donne un aperçu de son oeuvre pour la physique en Suisse. L'Assemblée approuve par applaudissements.

Fin de la séance à 18 h 00.

P.S.: Le Comité a réalisé que les dates du 25 au 29 septembre 1978 prévues pour la réunion commune avec la Société Autrichienne de Physique coïncideraient avec la réunion de la Société Européenne de Physique à York.

Comme la réunion de la Société Autrichienne de Physique ne peut être déplacée, la Société a renoncé à la réunion commune. La réunion d'automne 1978 se tiendra donc dans le cadre de la réunion annuelle de la Société Helvétique des Sciences Naturelles à Brigue les 3 et 4 octobre 1978. En conséquence, la Société accepte l'invitation de la Société Allemande de Physique de participer à une réunion commune avec la Société Autrichienne de Physique à Ulm du 24 au 28 septembre 1979.

Physique de la Matière Condensée

Wasserstoff-Speicherung in LaNi_5 und La_7Ni_3

P. FISCHER und W. HÄLG

(Institut für Reaktortechnik ETHZ EIR, 5303 Würenlingen)

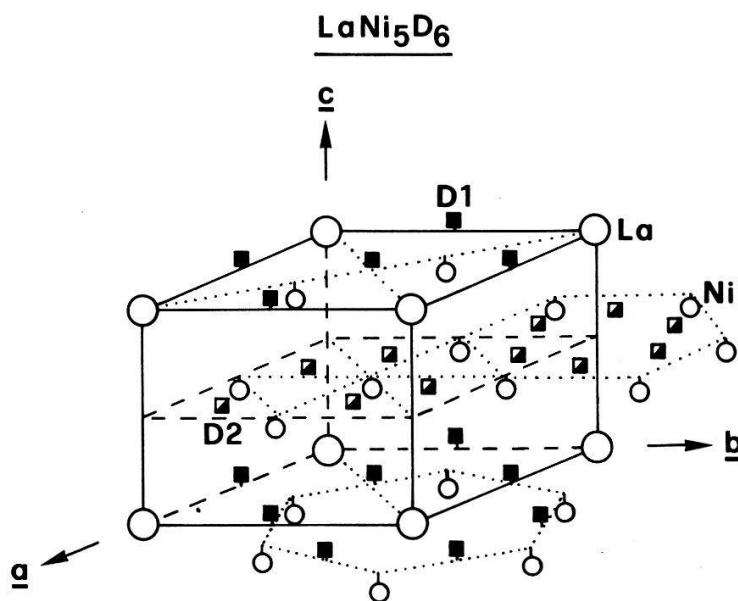
L. SCHLAPBACH und TH. VON WALDKIRCH

(Laboratorium für Festkörperphysik ETHZ, 8093 Zürich)

La-Ni-Systeme eignen sich für technische Anwendungen, insbesondere als Wasserstoffspeicher [1]. Durch Neutronenbeugungsuntersuchungen wurde Deuterium in LaNi_5D_x ($x = 0 \div 6$) und $\text{La}_7\text{Ni}_3\text{D}_{20}$ lokalisiert. Im Gegensatz zu ternären LaNi_5D_x -Verbindungen [2] wird La_7Ni_3 durch Deuterierung in LaD_3 und LaNi_5 zersetzt [3]. Die Struktur von LaNi_5D_6 ist in Figur 1 dargestellt. Als Ergänzung zu [2] sind in Tabelle 1 aus gemessenen Neutronenintensitäten verfeinerte Strukturparameter zusammengestellt ($R_{x=0.4} = 5.6\%$, $R_{x=6} = 6.4\%$). Für $x = 6$ wurde im Gegensatz zu [2] auch die Inäquivalenz der Reflexe $111/1\bar{1}\bar{1}$, $112/11\bar{2}$ und $221/22\bar{1}$ berücksichtigt.

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Figur 1

Kristallstruktur von LaNi_5D_6 (Raumgruppe P31m). a , b , c = hexagonale Basistranslationen. Die Positionen D1 und D2 sind 100 resp. ca. 50% von Deuterium besetzt.

Tabelle 1

Strukturparameter von LaNi_5D_x . * x -Werte entsprechend Desorptions-Messungen. Standardabweichungen sind in runden Klammern angegeben.

x	0.4*, 0.36(2)	6*, 5.8(3)
Gitterkonstanten a_N (Å)	5.027(5)	5.411(5)
c_N	3.989(5)	4.293(5)
Raumgruppe	P6/mmm	P31m
Positionsparameter:		
z_{Ni1}	0	0.916(9)
x_{Ni2}	0.5	0.482(3)
z_{Ni2}	0.5	0.444(6)
D-Positionen	3f	3c(D1) 6d(D2)
x_{D1}	0.5	0.474(4)
z_{D1}	0	0.046(7)
x_{D2}		0.181(6)
y_{D2}		0.831(5)
z_{D2}		0.490(9)
D-Verteilung: D1	0.36(2)	3.0(1)
D2		2.8(2)
Debye-Waller-Parameter B (Å ²)	1.4(1)	1.3(2)

Wasserstoffspeicherung in Metallen: Magnetische Ausscheidungen in LaNi_5

G. BUSCH und L. SCHLAPBACH

(Lab. für Festkörperphysik der ETHZ, 8093 Zürich)

Die intermetallische Verbindung LaNi_5 absorbiert schnell und reversibel grosse Mengen Wasserstoff [1]. Gründe für die ausgezeichneten kinetischen Eigenschaften sind bis jetzt keine bekannt. Ebenso sind die magnetischen Eigenschaften des LaNi_5 -Hydrides noch nicht untersucht worden. Nach Wallace [2] ist LaNi_5 ein Pauliparamagnet. Für Pauliparamagnete kann aus der magnetischen Suszeptibilität oder aus dem Koeffizienten γ des Elektronenanteils der spezifischen Wärme die Zustandsdichte an der Fermienergie bestimmt werden.

Wir berichten über Messungen der magnetischen Suszeptibilität nach der Faradaymethode an LaNi_5 und an LaNi_5 -Hydrid. Als Folge mehrerer Hydrierungszyklen werden magnetische Ausscheidungen festgestellt.

Wir haben aus 99.9% reinem La und 99.998% reinem Ni Verbindungen der eingewogenen Zusammensetzung $\text{LaNi}_{5.00}$ hergestellt [3]. Ihre Suszeptibilität beträgt $25 \cdot 10^{-6}$ emu/g bei 20°C und 4 kOe und ist feld- und temperaturabhängig. Offenbar ist die Probe nicht stöchiometrisch, sondern enthält Ni- oder Ni-reiche Ausscheidungen. Proben der eingewogenen Zusammensetzung $\text{LaNi}_{4.97}$ und $\text{LaNi}_{4.95}$, sowie LaNi_5 -Einkristalle, weisen übereinstimmend die Suszeptibilität $\chi = (4.5 \pm 0.3)10^{-6}$ emu/g auf, temperaturunabhängig von 100 bis 300 K und feldunabhängig von 0 bis 100 kOe. Unterhalb 100 K steigt die Suszeptibilität an, so dass wir geringe Abweichungen von der Stöchiometrie nicht ausschliessen können.

Wir haben die LaNi_5 -Proben bei Zimmertemperatur in einem Autoklaven bei 50 bar Wasserstoffdruck hydriert. Dabei zerfällt LaNi_5 in ein Pulver der mittleren Korngrösse 10 bis 20 μm . Die Suszeptibilität des LaNi_5 und des LaNi_5 -Hydrides wurde nach mehreren Absorptions- und Desorptionszyklen gemessen. Dazu wurden die pulverförmigen Proben in Quarzröhrchen bei 20°C und 6 bar hydriert. Nach Abkühlen der Proben auf die Temperatur des flüssigen Stickstoffes wurden die Quarzröhrchen zugeschmolzen und die Suszeptibilität der hydrierten Probe gemessen. Die dehydrierten Proben wurden zur Messung in evakuierte Quarzröhrchen eingeschmolzen. Die Resultate sind in der Tabelle 1 zusammengestellt.

Table 1
Magnetische Suszeptibilität von LaNi_5 und
 LaNi_5 -Hydrid nach mehreren Hydrierzyklen.
(20°C).

Probe	Anzahl Hydrierzyklen	Suszeptibilität 10^{-6} emu/g
LaNi_5	0	4.5 ± 0.3
LaNi_5	1	5.4
LaNi_5H_6	2	3
LaNi_5	2	8
⋮		
LaNi_5	9	32
LaNi_5H_6	10	23
LaNi_5	10	37

Zwei Ergebnisse sind ersichtlich: (1) Die Suszeptibilität des LaNi_5 -Hydrides ist kleiner als diejenige von LaNi_5 . Somit wird durch das Hydrieren die Zustandsdichte kleiner. (2) Die Suszeptibilität nimmt im Verlauf der Hydrierungszyklen stark zu. Diese Zunahme führen wir wegen ihrer Feld- und Temperaturabhängigkeit auf magnetische Ausscheidungen zurück. Als Ausscheidung kommt nur Ni oder eine Ni-reiche Phase in Frage.

Kürzlich durchgeführte Oberflächenuntersuchungen mit Hilfe der Photoelektronenspektroskopie im Röntgengebiet bestätigen das Vorhandensein von metallischem Ni an der Oberfläche von LaNi_5 [4].

Feinverteiltes metallisches Ni ist ein Katalysator für Wasserstoffreaktionen, besonders für die Dissoziation des Wasserstoffmoleküls an der Oberfläche. Wir vermuten deshalb, dass die hier festgestellten Ni-Ausscheidungen für die ausgezeichneten kinetischen Eigenschaften des LaNi_5 beim Hydrieren und Dehydrieren verantwortlich sind.

Weitere Untersuchungen über die Art und Grösse der Ausscheidungen, wie auch über ihre Folgen auf die Lebensdauer des LaNi_5 beim zyklischen Hydrieren und Dehydrieren, sind im Gang.

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Oszillatorische Magnetostriktion und die Druckabhängigkeit der Fermi-Fläche von Palladium

W. JOSS und W. VAN DER MARK

(Laboratorium für Festkörperphysik, ETHZ, 8093 Zürich)

Die Amplituden des de Haas-van Alphen Drehmoments und der oszillatorischen Magnetostriktion wurden bei 1.3°K in Magnetfeldern bis zu 10.5 T simultan gemessen. Aus diesen Daten wurde die uniaxiale Druckabhängigkeit mehrerer extremaler Bahnen auf der Fermi-Fläche von Palladium bestimmt.

Die Messungen wurden an einem hochreinen Einkristall mit Restwiderstandsverhältnis 5000 durchgeführt, und digitale Filtrierung erlaubte uns, die Amplituden der verschiedenen Frequenzkomponenten (z.B. Γ -zentrierter Elektronenteil) genau zu bestimmen. Die Summe der uniaxialen Druckabhängigkeiten in Richtung der kristallographischen Hauptachsen des X-zentrierten Löcher-Ellipsoids und des Γ -zentrierten Elektronenteils stimmt gut mit den experimentell gemessenen hydrostatischen Druckabhängigkeiten überein.

Elektronen Streuung an Germanium Verunreinigungen in Aluminium

W. VAN DER MARK und W. WEJGAARD

(Laboratorium für Festkörperphysik, ETHZ, 8093 Zürich)

Es werden die ersten genauen Messungen von de Haas-van Alphen Streuraten in einer Reihe von Aluminiumproben mit unterschiedlicher Zulegierung von Germanium vorgestellt. Die Dingle-Temperaturen, die proportional zu den Streuraten sind, wurden an drei verschiedenen repräsentativen Bahnen der dritten Zone der Fermi-Fläche von Aluminium gemessen. Einige Aspekte, die im Zusammenhang mit der Charakterisierung der Proben stehen, und weitere Resultate an anderen Legierungen werden behandelt.

On the work function of simple metals

R. MONNIER

(Nordita, Copenhagen)

J. P. PERDEW and D. C. LANGRETH

(Rutgers University)

J. W. WILKINS

(Cornell University)

[présentation orale seulement.]

Über das Bindungsproblem und die Zusammenhänge zwischen Struktur und physikalischen Eigenschaften in 'Chevrel-Phasen' $M_x Mo_6 X_{8-y}$ ($M = \text{Kation}$, $X = \text{Chalkogen}$)

K. YVON

(Laboratoire de Cristallographie aux Rayons X, Université de Genève, 24, quai Ernest Ansermet, 1211 - Genève 4)

Infolge der besonderen Tieftemperatureigenschaften sind die Chevrel-Phasen zur Zeit Gegenstand zahlreicher Untersuchungen [1]. Röntgenbeugungsuntersuchungen und strukturelle Überlegungen führen zu einem Bindungsmodell welches sich auf das Auffüllen eines Leitungsbandes von Mo d-Zuständen durch Ladungstransfer von M zu Mo stützt [2]. Die für die elektronischen Eigenschaften massgebenden strukturellen Parameter sind die Mo-Mo Abstände innerhalb und zwischen den Mo_6 -Oktaedergruppen [2]. Wichtig für das Verständnis der Gitterdynamik dieser Phasen ist die Tatsache, dass die M-Atome anharmonisch um ein Inversionszentrum schwingen ('delokalisiert' sind), das Schwingungsverhalten der Mo_6 -Gruppen vermutlich stark beeinflussen und zu Gitterinstabilitäten führen [3].

LITERATUR

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Physikalische Eigenschaften von TmSe mit veränderlicher chemischer Zusammensetzung

B. BATLOGG, E. KALDIS, H. R. OTT und P. WACHTER

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Die physikalischen Eigenschaften von TmSe sind entscheidend von der chemischen Zusammensetzung beeinflusst. Wir haben deshalb an einphasigen $Tm_x Se$ -Kristallen ($0.87 < x < 1.05$) mit wohlbestimmten x eine Reihe von thermischen, elektrischen, magnetischen und mechanischen Untersuchungen durchgeführt. Der Tm Gehalt spiegelt sich in einer systematischen Veränderung aller Messgrößen wider. Die mittlere Wertigkeit des Tm scheint sich mit wachsendem x von 3^+ in Richtung 2^+ zu verschieben, hydrostatischer Druck hingegen induziert mehr Dreiwertigkeit. $Tm_x Se$ ($x > 0.97$) ordnet antiferromagnetisch ($T_N = 3-4$ K) und zeigt ein ungewöhnliches magnetisches Phasendiagramm. Der elektrische Widerstand wächst für $T < T_N$ und für $x > 1$ auf Werte an, die an der Grenze metallischer Leitung liegen.

Facteurs gyromagnétiques g' de métaux de transition paramagnétiques

GUY MAURER et R. HUGUENIN

(Institut de Physique Expérimentale, 1015 Lausanne)

Nous présentons un appareillage permettant de mesurer, à température ambiante, le facteur gyromagnétique g' de métaux paramagnétiques selon la méthode Einstein-

De Haas. Les valeurs que nous avons obtenues pour V et Pd sont en accord avec celles qu'ont trouvées R. Huguenin, G. P. Pells et D. Baldock [1]. Nous avons également effectué des mesures de g' dans le domaine paramagnétique du système Ni-Cu; nos valeurs se situent dans le prolongement de la courbe de g' en fonction de la concentration de Cu obtenue par Scott et Reck [2] dans le domaine ferromagnétique.

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Electronic structure and electrical conductivity of graphite acceptor intercalation compounds

L. PIETRONERO, S. STRÄSSLER, H. R. ZELLER

(Brown Boveri Research Center, CH-5405 Baden, Switzerland)

M. J. RICE

(Xerox Webster Research Center, Webster, N.Y. 14580, U.S.A.)

It is shown that the change in the graphite electronic structure in dilute ($n > 2$) intercalation compounds cannot be described in terms of a uniform shift of the overall Fermi level. The transferred charge resides predominantly on the graphite layers adjacent to the intercalate. A calculation of the conductivity and plasma frequency for the acceptors compounds based on simple approximations is in good agreement with the experimental data of some of these compounds. Since this model also provides an upper limit on σ_{dc} (of about 1/3 that of copper) we conclude that the high conductivity reported for the compound $C_{8n}AsF_5$ cannot be described by simple models.

Über die Anzahl Leitungselektronen bei den dreiwertigen Seltenen Erden

B. DELLEY, H. BECK, H.-U. KÜNZI, H.-J. GÜNTHERODT

(Institut für Physik, Universität Basel, 4056 Basel)

Der elektrische Widerstand ρ der dreiwertigen flüssigen Seltenen Erden zeigt folgendes Verhalten: der Wert von ρ oberhalb des Schmelzpunktes steigt von La bis zu Lu mehr oder weniger monoton an, während sich der Temperaturkoeffizient von positiv bei La zu leicht negativ bei Lu ändert. Diese Tatsachen sind verträglich mit kürzlich veröffentlichten selbstkonsistenten Bandstrukturrechnungen von Duthie und Pettifor [1]. Letztere zeigen, dass der Bruchteil der drei Valenzelektronen, der s-artige Zustände besetzt (n_s), stark von der relativen Grösse des Ionenrumpfes abhängt, und liefern $n_s \approx 0.5$ für La und $n_s \approx 1.5$ für Lu. Somit wächst der Durchmesser der Fermikugel der Leitungselektronen, $2k_F$, von La zu Lu, was im Rahmen der Einionenstreunäherung für den Widerstand ungeordneter Systeme die Zunahme von

ρ erklärt. Für flüssiges Lu liegt überdies $2k_F$ in der Nähe von k_p , dem Wellenvektor des ersten Maximum des Strukturfaktors, was zu einem negativen Temperaturkoeffizienten führt.

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Elektronische Struktur des antiferromagnetischen Halbleiters UO_2

J. SCHOENES und A. SCHLEGEL

(Laboratorium für Festkörperphysik, ETH Zürich, 8093 Zürich)

Das Reflexionsvermögen von UO_2 Einkristallen wurde bei Zimmertemperatur zwischen 0.03 eV und 13 eV gemessen. Mittels einer Kramers–Kronig Analyse wurde daraus die komplexe dielektrische Funktion, der Energieverlust und die Zahl der effektiven Elektronen berechnet. Zusätzlich wurde an Dünnschliffen aus Transmissionsmessungen im schwach absorbierenden Spektralgebiet direkt der Absorptionsindex bestimmt. Die Absorptionskante wird bei 2 eV gefunden und einem $5f^2 \rightarrow 5f^1 6d^1 e_g$ -Uebergang zugeordnet. Ein Energieniveau Schema wird vorgeschlagen, das eine selbstkonsistente Zuordnung von optischen Uebergängen zwischen Maxima der gewichteten Zustandsdichte und der Struktur von $\varepsilon_2(\hbar\omega)$ erlaubt. Das 6d-Band wird dabei, als durch das Kristallfeld in zwei Teilbänder aufgespalten, betrachtet. Die Aufspaltung beträgt ca. 2.8 eV. Im Infraroten werden die LO und TO Phononen Moden bestimmt.

Gewichtete Bandstruktur ferromagnetischer Halbleiter

W. NOLTING

(ETH Zürich, Institut für Theoretische Physik)

Das sf-Modell für ferromagnetische Halbleiter wird ausgehend von der exakt lösbaren atomaren Grenze für Leitungsbänder endlicher Breite diskutiert. Das ursprüngliche s -Band spaltet infolge der sf -Wechselwirkung in eine Reihe von Quasiteilchenbändern auf, deren Zahl, Breite und spektrale Gewichte durch die Bandbesetzung, die Temperatur und die Grösse der sf -Kopplungskonstanten bestimmt sind. Die übliche Annahme, dass das Leitungsband in zwei spinpolarisierte Teilbänder aufspaltet, trifft höchstens für sehr schwach gekoppelte Systeme zu.

Propriétés électroniques des composés III–VI semiconducteurs à structure en couches

Y. DEPEURSINGE

(Laboratoire de Physique Appliquée, EPF-Lausanne, Switzerland)

Nous avons effectué une étude comparative des critaux de InSe, GaSe et GaS par l'intermédiaire de leurs structures de bandes et densités de charge respectives. Les structures de bandes ont été calculées grâce à la méthode du pseudopotentiel empirique

alliée à la méthode de perturbation $k \cdot p$. Comme hypothèses de travail, nous avons pris le même potentiel du Sélénium dans les cristaux de InSe et de GaSe, ainsi que le même potentiel du Gallium, dans les cristaux de GaSe et de GaS. La compatibilité requise entre les résultats obtenus pour l'ensemble des 3 cristaux et les données expérimentales fait que cette méthode présente un intérêt théorique particulier, puisqu'elle lève l'arbitraire lié à la redondance des paramètres apparaissant dans la méthode du pseudopotentiel empirique. Cette technique de 'consistance sur les 3 composés' nous a permis de donner un schéma cohérent explicatif des propriétés optiques et de photoémission de la famille considérée. De plus, les résultats obtenus sont en bon accord avec le calcul de la ionicité par la méthode de Phillips.

Electronic properties of cubic spinel CdIn_2S_4

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The electronic energies of the normal spinel CdIn_2S_4 are calculated using recently developed atomic model potentials [1]. The results show that CdIn_2S_4 is indirect with the conduction band minimum at Γ and the valence band maximum along the Σ direction. Keeping the atomic form factors unchanged, we have studied the effects produced on the electronic energies by varying the internal distortion parameter u or the distribution of the cations between tetrahedral and octahedral sites. The results are used to derive information on the chemical bonds of this compound.

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$k \cdot p$ method and nonlocality of crystal potentials

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The use of $k \cdot p$ method in non-local band structure calculations is improper and leads to errors of the order of 1 eV for semiconductors like Si and as high as 5 eV for d -band metals like Nb. Extending the currently used $k \cdot p$ expressions to include the momentum dependence of the non-local crystal potential, good results are obtained at the expense of an increased numerical effort. The consequences of these results on the most frequent applications of the $k \cdot p$ method are discussed.

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Renormalized orbital radii

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The orbital radii of Simons and Bloch, based on non-integral angular momenta and determined from spectroscopic valence term values, are refined by fitting to valence pseudo-wave function maxima as well. Two renormalization schemes are proposed; the first based on non-integral angular momenta *and* core charges, and the second by damping the additional centrifugal potential of Simons model. Results are presented for all atoms of the first period.

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Effet du champ cristallin et écrantage des ions d'uranium

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La séparation des neuf niveaux les plus bas d'énergie de l'ion U^{4+} dans un champ octaédrique [1] et cubique a été calculée tenant compte du mélange de huit termes spectroscopiques. Les résultats sont portés en Figures 1 et 2 en fonction du paramètre $V_4 = A_4 \langle r^4 \rangle$ du champ cristallin et le quotient V_6/V_4 des paramètres des composants des sixième et quatrième ordres du champ. Ici $\langle r^4 \rangle$ est la moyenne de la quatrième puissance du rayon de l'électron $5f$. Les valeurs négatives de ce quotient correspondent au champ cubique. En contraste avec les terres rares [1] où les diagrammes correspondants sont des lignes droites, le mélange des termes spectroscopiques produit ici une courbure. On remarque aussi que l'état singulet Γ_1 peut être l'état fondamental dans le cas cubique au lieu de l'état magnétique Γ_5 [2].

Les facteurs d'écrantage de Sternheimer ont été calculés pour U^{4+} et pour U^{3+} , et sont donnés dans la Table 1.

Une discussion plus détaillée paraîtra prochainement dans le 'Journal of Chemical Physics'.

Table 1

Facteurs d'écrantage de Sternheimer du champ cristallin selon la formule $B_l^m = (1 - \sigma_l)A_l^m$. Les A_l^m et B_l^m sont les coefficients du champ d'ordre l respectivement sans et avec écrantage.

l	2	4	6
U^{3+}	0.83	0.026	-0.039
U^{4+}	0.88	0.012	-0.046

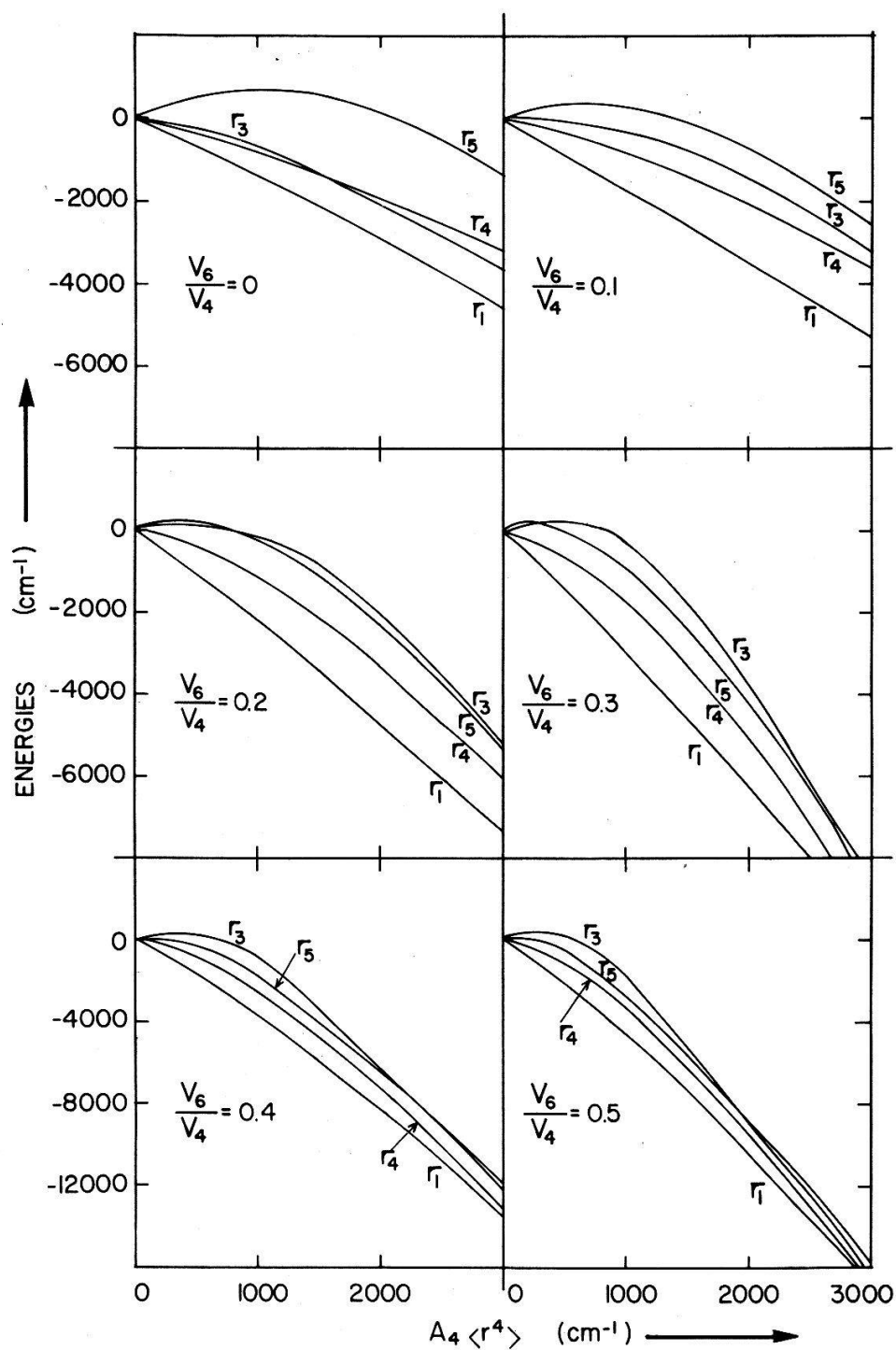


Figure 1

Energies des niveaux de provenance 3H_4 de l'ion U^{4+} dans un champ cristallin octaédrique. $V_4 = A_4 \langle r^4 \rangle$ est le coefficient du champ de quatrième ordre, V_6 celui du sixième ordre. Les symboles Γ_i dénotent les représentations irréductibles du groupe de symétrie ponctuel du champ.

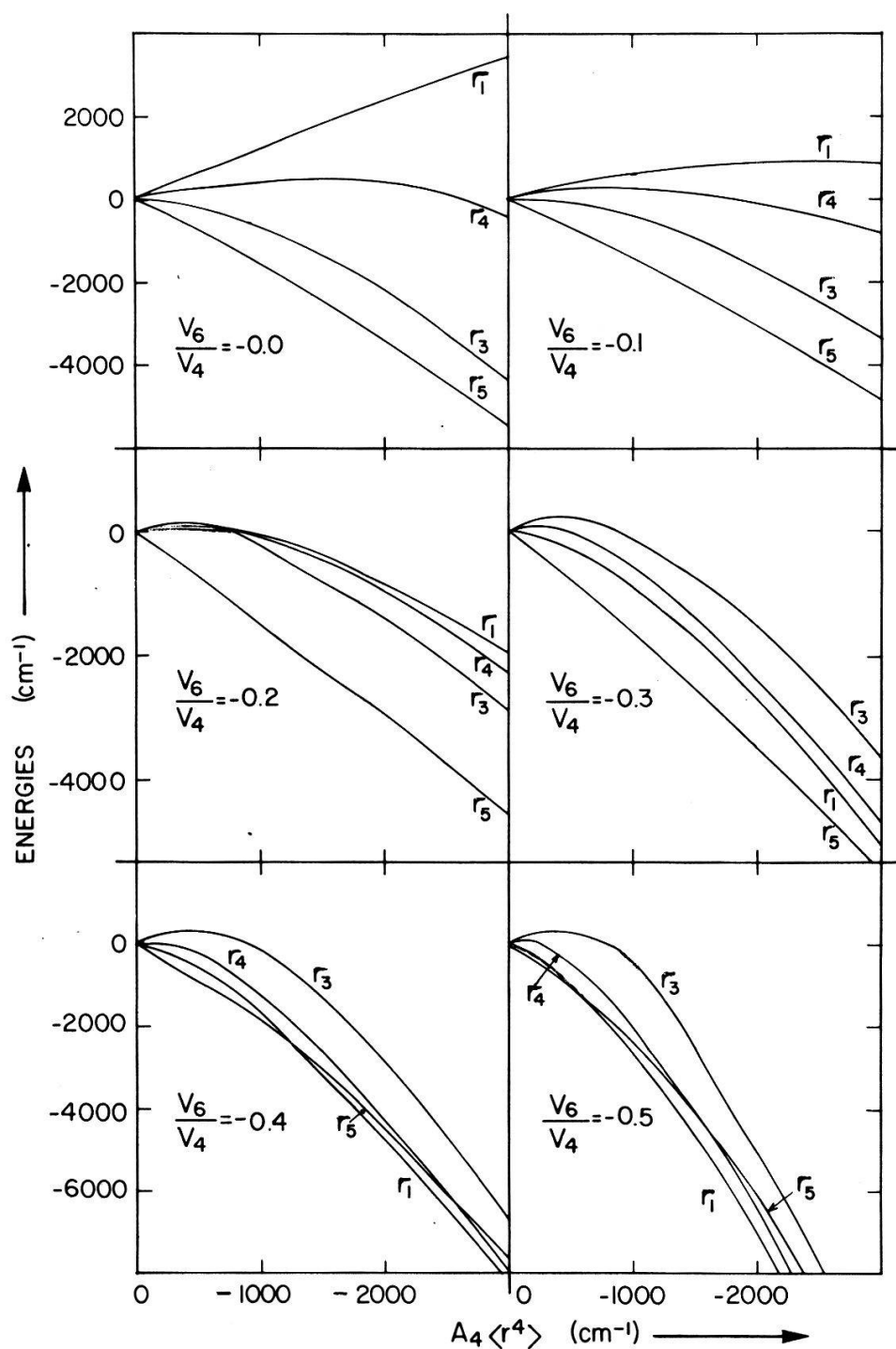


Figure 2
Energies des niveaux de provenance 4H_4 de l'ion U^{4+} dans un champ cristallin cubique. Les symboles sont les mêmes que dans Figure 1.

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Die Struktur der Tieftemperaturphasen von Kaliumhyperoxid KO_2

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Die Phasenumwandlungen bei 12 und 7 K wurden mit Röntgenstreuung an KO_2 -Einkristallen untersucht. In der Phase IV, oberhalb 12 K, ist KO_2 monoklin. Es treten 4 kristallographische Domänenorientierungen auf, herrührend von der Phasenumwandlung $\text{III} \rightarrow \text{IV}$ bei 196 K. In der Phase V, zwischen 12 K und 7 K, besteht der Kristall aus einem Gemisch von zwei verschiedenen Strukturen, die röntgenkristallographisch den Strukturen der Phase IV und der Phase VI entsprechen. Der Volumenanteil der Struktur der Phase IV nimmt stetig ab mit sinkender Temperatur und verschwindet schliesslich, wenn man sich dem Néel-Punkt bei 7 K nähert. Die Struktur der Phase VI ist triklin. Die Elementarzelle konnte bestimmt werden.

Temperaturabhängigkeit des $(110) T_1$ Phononenzweiges in Silber – ein Uebergang erster Schall $\rightarrow$ nullter Schall?

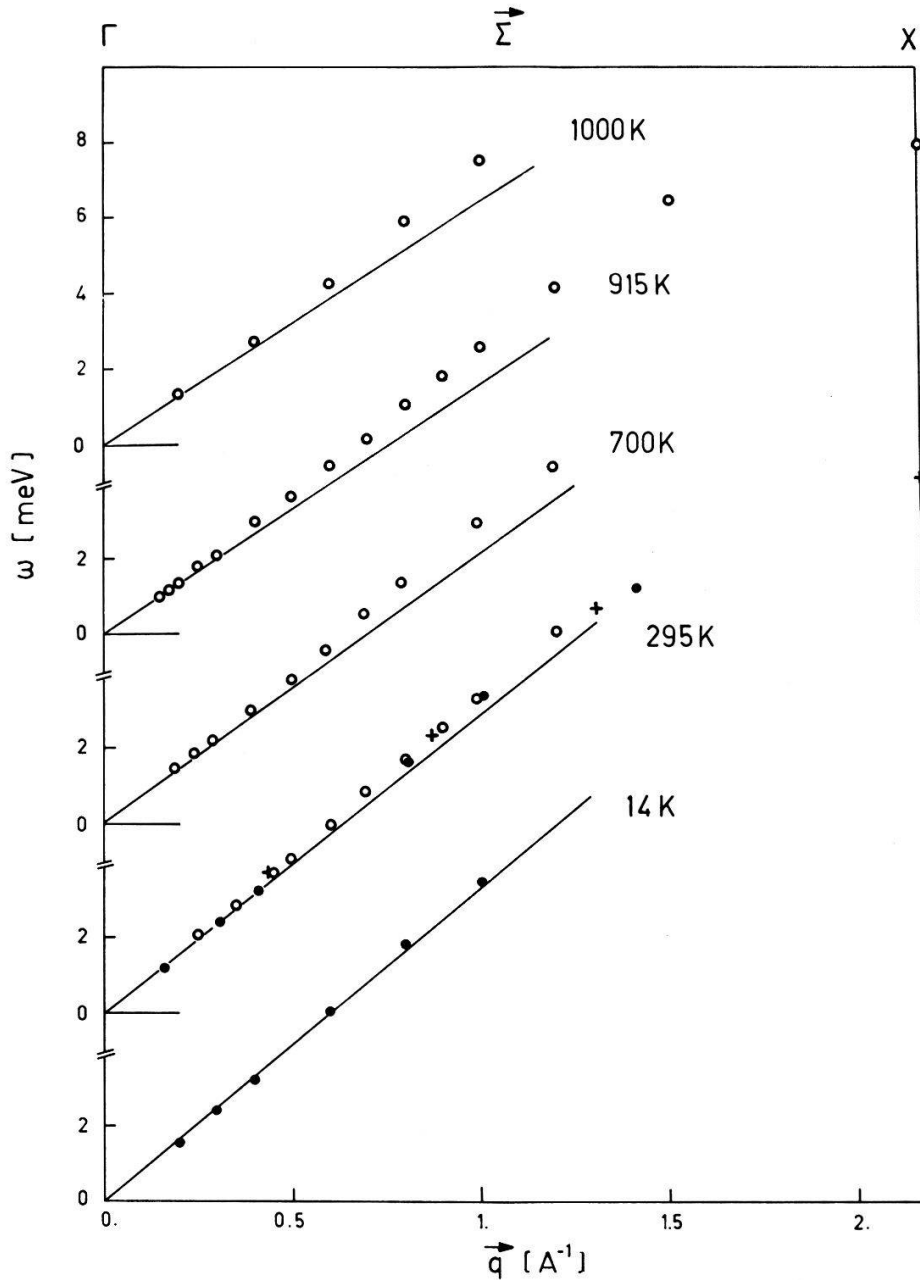
W. BÜHRER

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In einem anharmonischen Kristall sind die elastischen Konstanten von der Frequenz abhängig. Bei niedrigen Frequenzen misst man die adiabatischen oder ersten Schallgeschwindigkeiten, während bei hohen Frequenzen die elastischen Konstanten des nullten Schalls beobachtet werden [1]. Der Uebergang vom ersten Schall zum nullten Schall erfolgt bei einer Frequenz $\Omega = \bar{\Gamma}$, wobei $\bar{\Gamma}$ die mittlere reziproke Lebensdauer der thermischen Phononen im Kristall ist. Die Differenz 0. Schall – 1. Schall, welche bei 0 K verschwindet, nimmt als Funktion der Temperatur linear zu, und da die mittlere reziproke Lebensdauer $\bar{\Gamma}$ selbst von der Temperatur abhängig ist, wird der Uebergangsbereich mit steigender Temperatur zu höheren Frequenzen geschoben. Experimentell sind im allgemeinen beide Bereiche zugänglich: Ultraschalltechnik im Gebiet des 1. Schalls und unelastische Neutronenstreuung im Gebiet des 0. Schalls.

In Silber zeigt der transversale akustische Phononenzweig entlang $(\zeta\zeta 0)$ (Polarisation parallel $(1\bar{1}0)$, elastische Konstante $c'' = (c_{11} - c_{12})/2$) bei Raumtemperatur eine positiv gekrümmte Dispersion $\omega(\mathbf{q})$ [2, 3]. Im vorliegenden Experiment haben wir durch unelastische Neutronenstreuung das ungewöhnliche Verhalten dieses Phononenzweiges als Funktion der Temperatur untersucht; Experimente im Bereich des 1. Schalls sind von Chang und Himmel [4] veröffentlicht worden.

Die Resultate der mit einem Dreiachs-Spektrometer am Reaktor Diorit durchgeführten Messungen sind in Figur 1 dargestellt. Im Bereich 14 K – 1000 K zeigt sich für Wellenvektoren $q < 0.3 \text{ \AA}^{-1}$ eine stärkere Frequenz-Erniedrigung als für $0.3 < q < 1.2 \text{ \AA}^{-1}$, d.h. die Frequenzen bei kleinen Wellenvektoren folgen, als Funktion der Temperatur, den ersten Schallgeschwindigkeiten (ausgezogene Linien). Bei mittleren Wellenvektoren nimmt die Differenz 0. Schall – 1. Schall mit steigender Temperatur zu, die Gruppengeschwindigkeit bleibt jedoch praktisch konstant. Ein



Figur 1

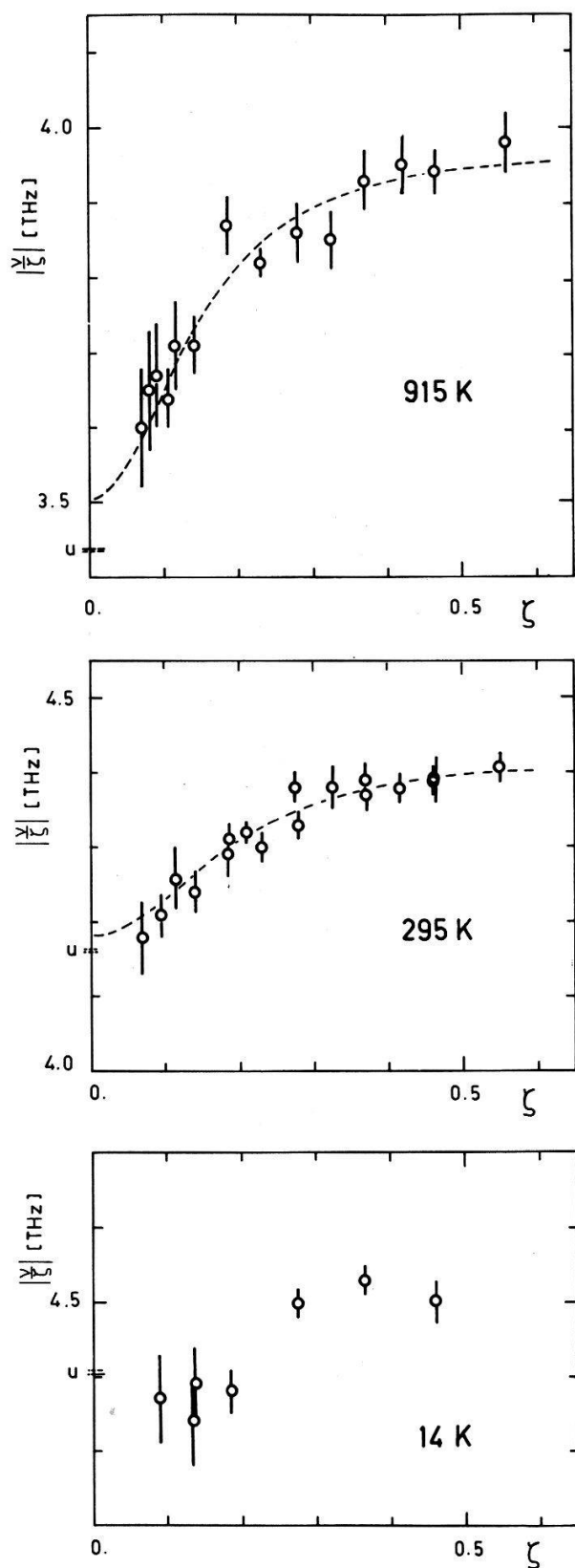
Temperatur-Abhängigkeit der Phononendispersion des $(\zeta\zeta 0) T_1$ Zweiges in Silber. $\circ \bullet$ Dieses Experiment (Vernichtung, Erzeugung); $+$ Kamitakahara und Brockhouse [3]; — 1. Schall, Chang und Himmel [4], extrapoliert zu 1000 K.

ähnliches Verhalten ist in Kupfer beobachtet [5] und als Uebergang 1. $\rightarrow$ 0. Schall interpretiert worden [6].

Die Temperatur- und Frequenzabhängigkeit der elastischen Konstanten $c'' = (c_{11} - c_{12})/2$ ist bei hohen Temperaturen in erster Näherung gegeben durch [6]:

$$c''(T, \Omega) = c''_{is}(T) + k_B T \delta^2 \frac{1}{v_a} \frac{\Omega}{\Omega + 2i\Gamma},$$

dabei ist c''_{is} die isotherme elastische Konstante und δ eine mittlere anharmonische Kopplungskonstante (proportional zum Grüneisen-Parameter). Die Anpassung



Figur 2

Phasengeschwindigkeit $|v/\zeta|$ in Silber entlang $(\zeta\zeta 0)$. ζ = reduzierter Wellenvektor (in Einheiten von $2\pi/a$); $u = 1$. Schall [4]; ---- Theorie 1. Schall $\rightarrow$ O. Schall.

dieses theoretischen Ausdrucks an die experimentellen Phasengeschwindigkeiten ergibt bei hohen Temperaturen eine gute Uebereinstimmung (Figur 2, 295 K, 915 K), und die angepassten Parameter c''_{is} und δ nehmen bei allen Temperaturen physikalisch sinnvolle Werte an. Die Grösse $\bar{\Gamma}$ jedoch, charakteristisch für den Uebergang $1. \rightarrow 0$. Schall, erweist sich als von der Temperatur unabhängig ($295K \leq T \leq 1000K$); dieses Verhalten ist mit einer Interpretation als reziproker Lebensdauer nicht verträglich, im weiteren zeigt bereits die Messung bei tiefer Temperatur (Figur 2, 14 K) eine wellenvektorabhängige Phasengeschwindigkeit welche im Rahmen der Theorie $1. \rightarrow 0$. Schall nicht erklärt werden kann.

Die in Silber bei Raumtemperatur gemessenen Dispersionskurven sind mit Born-von Karman-Modellen analysiert worden [2, 3]. Obwohl bei diesen Anpassungen von interatomaren Kraftkonstanten nur wenige Phononen aus dem fraglichen Bereich verwendet worden sind, zeigen die gerechneten Kurven entlang ($\zeta\zeta 0$) erstaunlicherweise eine positive Krümmung. Dies legt den Schluss nahe, dass die Information 'positiv gekrümmte Dispersion für ($\zeta\zeta 0$) T'_1 ' auch in den übrigen Phononen mit höherer Energie und aus anderen Richtungen enthalten ist. Die Bindungskräfte in Silber (Elektron-Phonon Wechselwirkung) müssen dergestalt sein, dass bereits bei 0 K eine positive Krümmung entsteht; ein Verhalten, wie es zum Beispiel in Niob [7] sehr ausgeprägt in Erscheinung tritt.

Um die gemessene Temperatur- und Wellenvektor-Abhängigkeit der Phasengeschwindigkeit in Silber erklären zu können, müssen die beiden Effekte überlagert werden: bei tiefen Temperaturen ist der Einfluss der Bindungskräfte dominant, bei höheren Temperaturen bringen die anharmonischen Wechselwirkungen eine weitere Zunahme der Phasengeschwindigkeit. Eine Trennung der beiden Beiträge ist nur mit Hilfe detaillierter Modellrechnungen möglich, welche ihrerseits aber auf temperaturabhängigen Phononen-Messungen in der ganzen Brillouinzone basieren müssen.

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Exciton annihilation in molecular crystals at high densities

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Suna's kinematic equations [1] describing exciton annihilation in aromatic molecular crystals are solved for high exciton densities [2]. It is found that in case of diffusion limited annihilation the annihilation rate γ is a monotonically increasing function of exciton density n even for $n \ll$ lattice site density. These density effects depend on the dimensionality of the exciton motion. For both triplet and singlet excitons γ is a function of n in one-dimensional and two-dimensional systems. For singlet excitons γ depends on n even in three-dimensional systems, if reabsorption is a dominant mechanism of exciton motion.

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Magnetic resonance of vitamin B₁₂ single crystals

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It has been found, that crystalline vitamin B₁₂ (cyanocobalamin) and B_{12b} (hydroxocobalamin) may be reduced to the cobaltous B_{12r} by hydrogen gas at high temperature, or (and) high pressure. At temperatures below 273 K an oxygen uptake was observed resulting in a superoxocobalamin compound. The new reduction method was used to get single crystal ESR spectra for the paramagnetic vitamin B_{12r} diluted in B_{12b} or B₁₂, respectively. For the first time, single crystal ESR spectra of B_{12r} and the superoxocomplex are shown and the influence of the motion of water molecules and the superoxide ion (O₂⁻) on the spectra will be discussed.

Étude de la surface de la glace près de 0°C par la canalisation de protons

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La perfection cristalline de la face basale de la glace est étudiée entre -130°C et -2°C par la canalisation de protons de 100 keV. Le cristal de glace est en équilibre thermodynamique avec sa vapeur pendant ces mesures. Pour éviter la divergence du faisceau protonique dans la vapeur (par exemple $p = 3.9$ Torr à -2°C), celui-ci est injecté dans la chambre à diffusion à travers une buse conique de 6° d'ouverture, ayant un trou de sortie de 0.1 mm de diamètre. L'existence d'une couche désordonnée à la surface de la glace est ainsi mise en évidence par la première méthode directe. L'épaisseur de cette couche varie de quelques nm à -35°C à 90 nm à -2°C, suivant une loi en $-C \cdot \log(273 - T)$. Ces résultats, qui sont en accord avec des mesures de propriétés mécaniques de la glace, pourraient être expliqués par des vibrations anharmoniques de surface.

Flussquantisierung am einzelnen Oberflächenatom eines Neutronensterns?

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Kürzlich wurde in der Pulsarstrahlung von Herkules X-1 eine 53 keV Röntgenlinie entdeckt und hieraus auf das Sternfeld $4.6 \cdot 10^{12}$ G geschlossen [1]. Ueberträgt man heuristisch das Modell [2] auf den Sternfestkörper (kosmischer Funke!), so folgt: (i) die Emissionslinie lässt sich dem Gap-Uebergang $2\varepsilon(0)$ zuordnen; (ii) dies entspricht gleichzeitig einer atomlokalen Flussänderung $\phi_L = h/e$ des ele-

mentaren Quanta mit der mittleren Induktion $B = 4,6 \cdot 10^{12}$ G; (iii) die zugehörige Teilchendichte beträgt $n = 3,6 \cdot 10^{28} \text{ cm}^{-3}$ (bzw. $\rho_{\text{Fe}} = 3,4 \cdot 10^6 \text{ g cm}^{-3}$); für die (athermische) Quantentemperatur folgt $T_c = 160$ Mio. Grad, für die höchstens zuzulassende thermische Temperatur $T_{\text{th}} \approx 5$ Mio. Grad. (iv) Kein ausgedehntes Gasplasma erforderlich, feldfreie Oberfläche wahrscheinlich B - E -kondensiert, statistisches 'Krustenkrachen' wahrscheinlich mit induzierten Phasenübergängen verknüpft.

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Energy selective measurement of electron spin polarization (ESP) in field emission from W-EuS junction emitters

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We have measured energy selectively the ESP of field emitted electrons from W-EuS junction field emitters with an energy resolution of about 0.2 eV. The energy distribution depends on the annealing history of the emitter. It is similar to that of a cold metal emitter if the emitter has been annealed at 840 K, and the spin polarization is about 85% (perfect junction). Annealing at a higher temperature generates a double peak energy distribution. The separation between both peaks can be several eV and depends on film thickness and electric field strength. Energy selective measurements of the ESP show that the high energy electrons are weakly polarized ($P < 0.2$), while the electrons with low kinetic energies are highly polarized ($P \sim 0.7$). With the perfect emitter the high P is generated by the W-EuS interface acting as a spinfilter. Its impact is lowered by traps after higher annealing. Thus tunnelling from W into EuS results in a moderate P only, whereas the higher P of the low energy electrons is determined by scattering at traps forming a bound magnetic polaron.

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Änderung der Elektronenzustandsdichte des Galliums beim Phasenübergang fest-flüssig

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Wir haben Photoemissionsmessungen an Gallium bis zu einer Photonenenergie von 6.5 eV über einen Temperaturbereich von 220 K bis 350 K durchgeführt und damit erstmals die Energieverteilung der Photoelektronen aus der flüssigen Phase direkt mit derjenigen der festen Phase vergleichen können.

Während die Spektren des flüssigen Galliums im untersuchten Energiebereich mit dem Modell des freien Elektrons erklärt werden können, lassen die Messungen an den polykristallinen Proben auf eine gegen die Fermienergie zu stark abnehmende Elektronenzustandsdichte schliessen.

Photoemissionsuntersuchung zum Dichteverlauf an der Oberfläche von flüssigem Quecksilber

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Der Dichteverlauf in der Uebergangszone zwischen Volumenbereich und Dampfphase ist bei Oberflächen flüssiger Metalle weitgehend unbekannt und wird sowohl durch örtlich oszillierende [1] wie auch monoton abfallende [2] Modell-Dichten beschrieben. Wir haben die Photoemission zur Untersuchung der Uebergangszone an flüssigem Quecksilber, des einen dichteabhängigen Metall-Halbleiter-Uebergang zeigt, angewendet. Unsere Messungen der Temperaturabhängigkeit der Energieverteilung der Photoelektronen (EDC) bei verschiedenen Lichteinfallswinkeln deuten darauf hin, dass die Uebergangszone klein ist verglichen mit der Austrittstiefe der Photoelektronen ($\sim 30 \text{ \AA}$). Die starke Temperaturabhängigkeit der EDC's kann quantitativ durch Streuung der Elektronen am Quecksilberdampf erklärt werden, so dass bis 40 Grad oberhalb des Schmelzpunktes kein mit der Temperatur sich verbreiterndes Dichteprofil anhand der EDC's beobachtet wird.

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Spin polarisierte Photoelektronen aus Wolfram

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Der Polarisationsgrad der Photoelektronen, emittiert durch zirkular polarisiertes Licht aus der (100)-Oberfläche eines W Einkristalles, wurde für Lichtenergien von 1.94–5.7 eV gemessen. Durch Aufdampfen von Cs wurde die Photoschwelle von 4.3 eV auf 1.8–2.4 eV gesenkt. Die maximale gemessene Polarisation beträgt 10%. Das Polarisationspektrum $P(h\nu)$ lässt sich qualitativ mit Hilfe der Dipol-Auswahlregeln für die optische Anregung von Elektronen innerhalb des Leitungsbandes erklären. Entscheidend für das Auftreten einer von Null verschiedenen Polarisation ist die Spin-Bahn Wechselwirkung.

Winkelverteilung von Roentgen-Photoelektronen aus Au-, Ag- und Cu-Einkristallen

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Abstract. The angular dependence of X-ray photoelectrons from single-crystal gold, silver and copper was measured. The samples were prepared by vacuum deposition on KCl single crystals. The distributions exhibit a strong modulation related to diffraction effects. Differences are observed in the distributions of electrons of near equal energy (Au_{4f} , Ag_{3d} and Cu_{3p}) or emitted from different shells in the same crystal (Cu_{3d} , Cu_{3p} , Cu_{2p} and Cu LMM-Auger). Some features of these distributions can hardly be explained by the crystal structure alone.

Einleitung

Die Bewegung eines Elektrons in einem Einkristall gibt Anlass zu Beugungserscheinungen. Diese Tatsache ist eine Folge der Gitterperiodizität und tritt dann auf, wenn die de Broglie-Wellenlänge kleiner als die Gitterkonstante ist. Die Intensität der Photoelektronen wird in Funktion der Kristallorientierung gemessen. Durch die Energieanalyse ist es möglich, nur solche Elektronen zu erfassen, die elastische oder quasielastische Streuungen erlitten haben. Die so beobachteten Beugungserscheinungen sind sehr ausgeprägt. Diese Anomalie der Transmission tritt in denjenigen Richtungen auf, für welche die Laue-Bedingungen annähernd erfüllt sind. Wegen der Prägnanz des Vorgangs sind die Bedingungen, unter welchen eine kinematische Theorie anwendbar ist, nicht mehr erfüllt. Infolgedessen sind Aussagen nur im Rahmen einer dynamischen Theorie zu suchen.

Die Beugungserscheinungen von Röntgen-Photoelektronen wurden bisher verhältnismässig wenig erforscht. Intensitätsverteilungen von Rumpf und Valenzelektronen wurden nur in NaCl [1–2], Au [3–6], KCl [7], BaTiO₃ [8], Si [5, 9–10] und Graphit [11–12] gemessen. In dieser Arbeit wird über eine systematische Untersuchung an Au-, Ag- und Cu-Einkristallen berichtet.

Experimentelles

Die vorliegenden Messungen wurden mit einem speziell dafür eingerichteten magnetischen Spektrometer durchgeführt. Zur Anregung der Photoelektronen wurde die AlK _{α} -Strahlung (1486.6 eV) verwendet. Das Vakuum betrug ca. $2 \cdot 10^{-7}$ Torr. Um eine Adsorption von Fremdatomen zu vermindern, wurden die Proben auf 300°C geheizt. Die trotzdem vorhandene Bedeckung durch Kohlenstoff und Sauerstoff (siehe Tabelle I) hat auf die vorliegenden Ergebnisse keinen Einfluss.

Die Probe wird um eine senkrecht zur Spektrometerebene stehende Achse gedreht. Der Winkel zwischen einfallender Röntgenstrahlung und analysierten Photoelektronen bleibt dabei konstant und beträgt 68°. Der polare Winkel θ ist durch die Flächennormale der Probe und die Richtung der analysierten Photoelektronen definiert. Die Winkelauflösung ist durch eine Blende bestimmt und beträgt $\pm 1.3^\circ$ in der Spektrometerebene und $\pm 3.5^\circ$ senkrecht dazu.

Alle Au-, Ag- und Cu-Proben wurden in einer separaten Aufdampfanlage durch epitaxiales Wachstum auf KCl-Einkristallen (Spaltebene (100)) hergestellt. Die verwendeten Mo-Schiffchen, sowie das Ausgangsmaterial (99.999%) wurden wiederholt in einem Vakuum von ca. $5 \cdot 10^{-8}$ Torr ausgeheizt. Der Kristallhalter wurde bis 500°C ausgeheizt, wobei ein Abdampfen von KCl beobachtet wurde. Weitere Daten sind aus Tabelle I zu entnehmen.

Die geschätzte Aufdampftrate betrug jeweils ca. 500 Å/s, die Aufdampfdauer 2–4 s. Aus dem Substrat konnten keine Photolinien nachgewiesen werden; die Bildung von diskontinuierlichen Aggregaten kann daher ausgeschlossen werden.

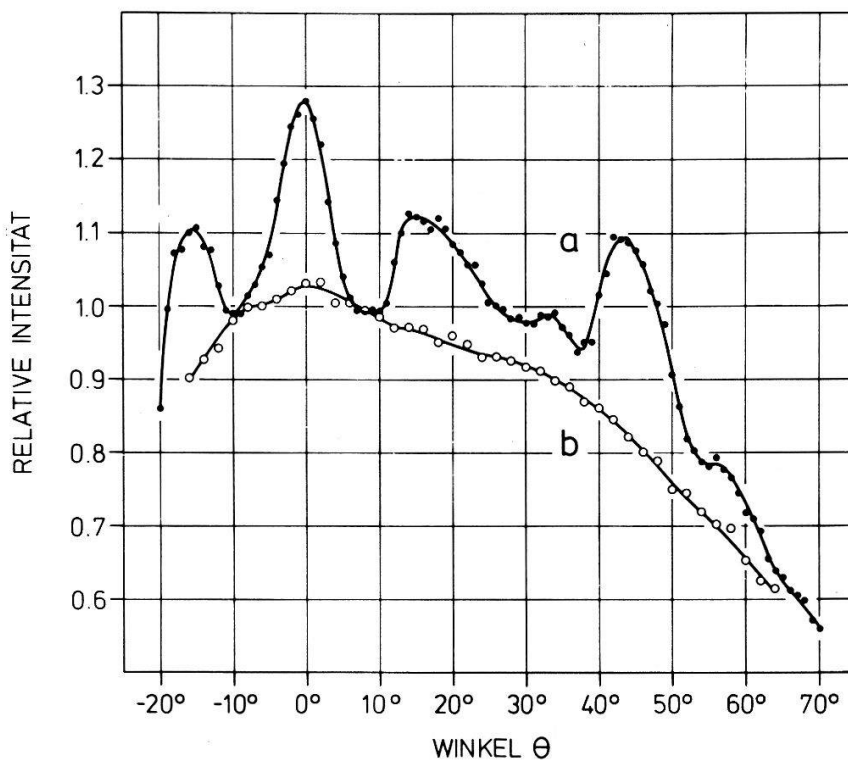
Tabelle I

Au (100)	C _{1s} /Au _{4f} = 0.02	O _{1s} /Au _{4f} = 0.02	T _s = 320°C
Ag (100)	C _{1s} /Ag _{3d} = 0.02	O _{1s} /Ag _{3d} = 0.01	T _s = 480°C
Cu (100)	C _{1s} /Cu _{2p} = 0.04	O _{1s} /Cu _{2p} = 0.04	T _s = 480°C

Bekanntlich ordnen sich Metallschichten bei geeigneten Bedingungen epitaxial [13]. Die vorliegenden Messungen zeigen zudem, dass der Ordnungszustand bis an die Kristalloberfläche erhalten bleibt.

Messungen

Die Winkelverteilung der $4f_{7/2}$ -Photoelektronen aus einer epitaxial geordneten Goldprobe weist eine ausgeprägte Struktur auf (Figur 1, Kurve a). Bei einer Probe, die bei einer Temperatur hergestellt wurde, bei der keine Epitaxie zu erwarten ist, ist diese Struktur nicht vorhanden (Figur 1, Kurve b). In beiden Fällen war die Rotationssachse parallel zur (001)-Richtung des Substrats. Die Messpunkte sind Mittelwerte von mindestens zwei unabhängigen Messreihen und werden keinerlei Korrektur unterworfen.

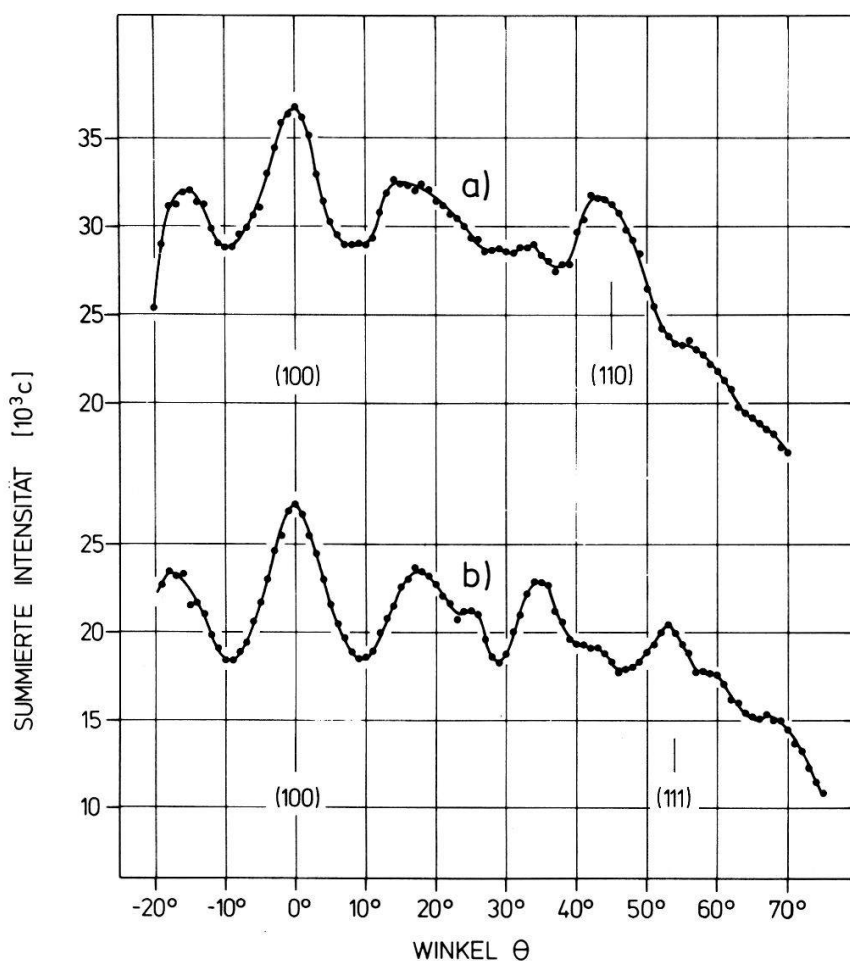


Figur 1.

Einfluss der Substrattemperatur bei der Aufdampfung auf die Winkelverteilung der Photoelektronen aus dem Gold $4f_{7/2}$ Niveau. Die Punktdurchmesser entsprechen ungefähr dem statistischen Fehler. Die Winkelauflösung von θ beträgt $\pm 1.3^\circ$. Die Intensität ist auf die Werte von $6-8^\circ$ normiert. (a) $T_s = 320^\circ\text{C}$; (b) $T_s = 70^\circ\text{C}$.

Der Zusammenhang zwischen Kristallorientierung und Winkelverteilung der Photoelektronen ist aus Figur 2 erkenntlich. Die Proben wurden um eine (001)-Achse, Kurve (a) und um eine (011)-Achse, Kurve (b) gedreht. In beiden Fällen treten Intensitätsmaxima bei Rotationswinkeln auf, die mit den eingetragenen Kristallrichtungen übereinstimmen.

Die Ähnlichkeit mit den an NaCl-Einkristallen gemessenen Verteilungen ist bemerkenswert [2]. Die Intensitätsverteilungen der Photoelektronen sind somit im wesentlichen durch die Kristallstruktur bestimmt. Bekanntlich ist aber die erreichbare Probentiefe durch die inelastische freie Weglänge der Photoelektronen begrenzt;



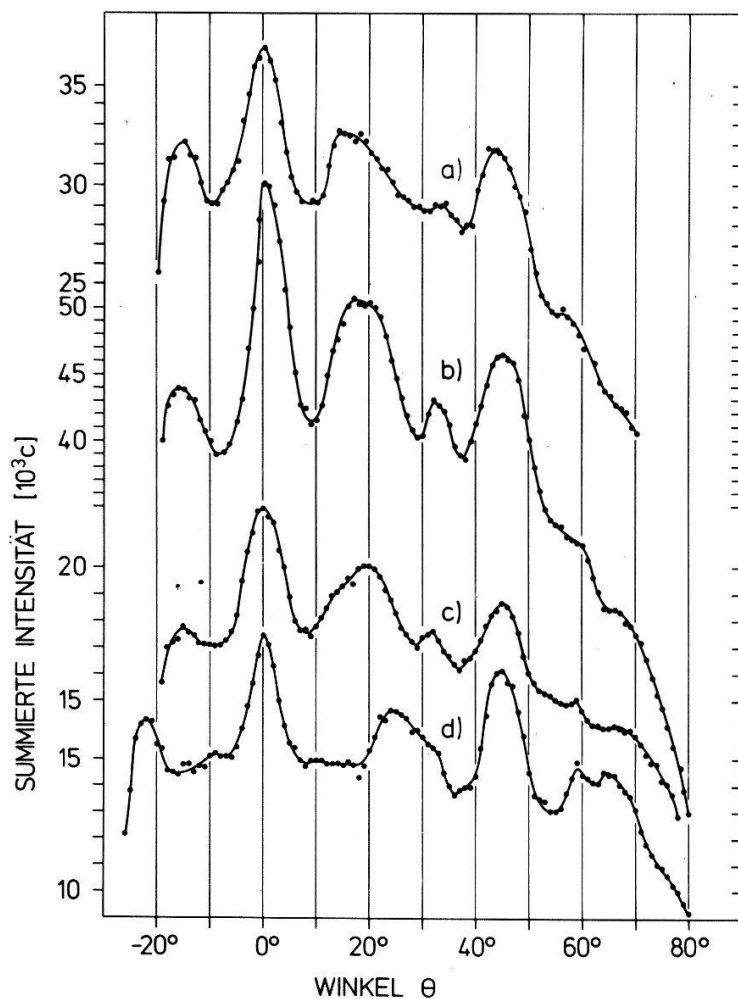
Figur 2

Winkelverteilung der Photoelektronen aus dem $4f_{7/2}$ Niveau von epitaxial gewachsenen Goldkristallen um zwei verschiedene Rotationsachsen. (a) Rotationsachse parallel zur (001)-Achse; (b) Rotationsachse parallel zur (011)-Achse.

diese beträgt für die $4f$ -Linie in Gold ca. $10 \text{ \AA}$. Die Messung zeigt also, dass die epitaxiale Ordnung bis in unmittelbare Nähe der Kristalloberfläche erhalten bleibt.

Beim Vergleich mit der einzigen bisher veröffentlichten Messung an einem Au-Einkristall [6] ist Vorsicht am Platz. Die Winkelauflösung bei dieser Messung betrug nur $\pm 3.5^\circ$. Ferner war in [6] die Flächennormale um 8° gegen die (100)-Richtung verdreht. Gewisse Unterschiede bei der Rotation um die (001)-Achse könnten daher durch diese Fehljustierung bedingt sein. Wir haben versuchsweise eine Probe um 6° geneigt und dabei wesentliche Veränderungen der Winkelverteilung festgestellt. Hingegen sind die Resultate in [6] mit unseren Messungen bei (011)-Drehung vergleichbar.

In Figur 3 sind die Winkelverteilungen von Photoelektronen aus den drei Edelmetallen mit einer entsprechenden Verteilung aus einem KCl-Einkristall verglichen. Die Rotation erfolgte jeweils um eine (001)-Achse. Die gewählten Photolinien haben vergleichbare kinetische Energien: Au $4f_{7/2}$: 1403.8 eV, Ag $3d_{5/2}$: 1119.9 eV, Cu $3p_{3/2}$: 1413 eV und K $2p_{3/2}$: 1193.0 eV. Bekanntlich besitzen diese Kristalle f.c.c. - Struktur. Bei allen Verteilungen treten bei den Winkeln 0° (100) bzw. 45° (110) Intensitätsmaxima auf. Hingegen sind die dazwischen gelegenen Strukturen voneinander verschieden. Die Winkelverteilungen von Photo- und Auger-Elektronen verschiedener Schalen aus einem Cu-Einkristall sind aus Figur 4 zu ersehen.

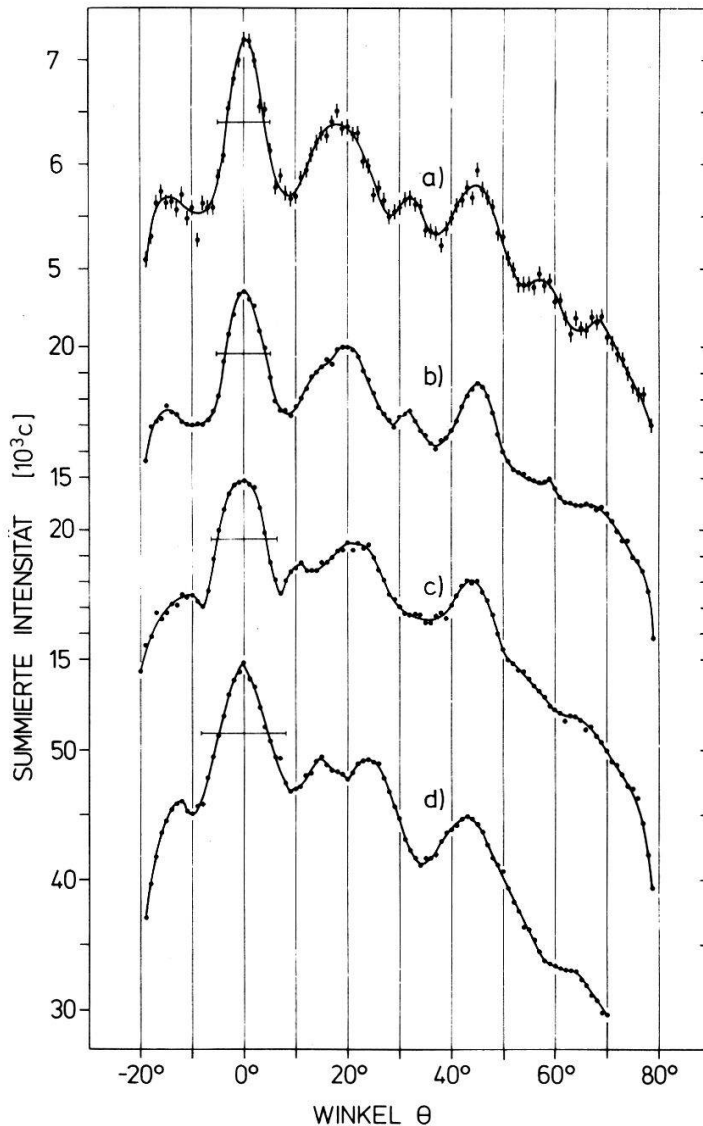


Figur 3

Winkelverteilung der Photoelektronen von epitaxial gewachsenen Edelmetallkristallen aus Schalen mit vergleichbaren Bindungsenergien. (a) Gold $4f_{7/2}$; (b) Silber $3d_{5/2}$; (c) Kupfer $3p$; (d) gleich behandeltes KCl Substrat; $K2p_{3/2}$.

Diskussion

Alle gemessenen Intensitätsverteilungen von Photoelektronen aus f.c.c.-Einkristallen weisen in der (100)-, (110)- und in der (111)-Richtung ausgeprägte Maxima auf. Eine qualitative Erklärung dafür gibt die dynamische Theorie der Elektronenbeugung [14]. Die Bewegung des Photoelektrons im periodischen Kristallpotential wird als Überlagerung von Bloch-Wellen dargestellt. Die Amplitude dieser Wellen wird in Funktion der Kristallrichtung berechnet. Bei Richtungen, für welche die Laue-Bedingungen annähernd erfüllt sind, werden die Wellen verschieden stark angeregt. Die Intensitätsverteilung des austretenden Elektrons wird durch Interferenz zwischen diesen Wellen bestimmt. In der Zweistrahl-näherung wird angenommen, dass nur eine starke Reflexion auftritt. Die so berechnete Verteilung ist bezüglich des Bragg-Winkels θ_B asymmetrisch: Das Maximum liegt bei einem kleineren Winkel und ist somit in Richtung der reflektierenden Ebene verschoben. Den Beitrag eines Photoelektrons, das auf der anderen Seite der Ebene reflektiert wird, erhält man durch Spiegelung [15]. Die Gesamtintensität weist in Richtung der Kristallebene ein Maximum auf. Seine Breite ist im wesentlichen durch die Energie, die Kristallrichtung



Figur 4

Winkelverteilung von Photo- und Auger-Elektronen aus einem epitaxial gewachsenen Kupferkristall. Die Kurven sind nach der kinetischen Energie der Elektronen geordnet. (a) Cu 3d; $E_K = 1485$ eV; (b) Cu 3p; $E_K = 1413$ eV; (c) Cu Auger $L_3 M_{4.5} M_{4.5}$; $E_K = 920$ eV; (d) Cu $2p_{3/2}$; $E_K = 555$ eV.

und durch das Kristallpotential bestimmt und beträgt ungefähr $2\theta_B$. Der Einfluss der Absorption wird durch die Einführung eines Imaginärteils im Potential beschrieben. Dadurch wird die Form der Linie zwar geändert, ihre Breite jedoch erhalten. Bemerkenswert ist das Auftreten von Flügeln, die sich in ein breites Winkelgebiet erstrecken.

Die Existenz von Intensitätsmaxima in Richtung von niederindizierten Kristallebenen wird durch das Experiment bestätigt. Ihre Breite ist jedoch selbst ohne Berücksichtigung des endlichen Auflösungsvermögens kleiner als erwartet. In Figur 4 sind beim Winkel Null die Breiten $2\theta_B$ eingetragen. Die zwischen 10° und 40° beobachteten Strukturen zeigen die Unzulänglichkeit der oben skizzierten Theorie. Dabei sind drei Punkte hervorzuheben: (A) Obwohl die drei Edelmetalle sowie KCl f.c.c.-Struktur besitzen, ist der Verlauf der Verteilung von Photoelektronen mit annähernd gleichen kinetischen Energien zwischen 10° und 40° verschieden (vgl. Figur 3). (B) Das Nebenmaximum bei 32° kann keiner Kristallrichtung zugeordnet werden. (C) Die Struktur bei 20° (Figur 4) spaltet sich mit abnehmender kinetischer Energie

auf, was bei den (100)- und (110)-Maxima nicht beobachtet wird. Es ist daher anzunehmen, dass diese Strukturen der Intensitätsverteilung nicht durch die Kristallstruktur allein bestimmt sind.

Zusammenfassung

Die vorliegenden Messungen zeigen, dass epitaxial gewachsene Metallkristalle bis in die äussersten Atomlagen geordnet sind. Die beobachteten Interferenzerscheinungen der Photoelektronen weisen Strukturen auf, die noch ausgeprägter sind als in Falle der Alkalihalogenide. Die Prägnanz dieser Effekte ermöglicht es, Strukturänderungen in dünnen Oberflächenschichten nachzuweisen [12]. Die Interpretation der Beugungserscheinungen im Rahmen einer einfachen dynamischen Theorie ist nur zum Teil möglich. Eine quantitative Beschreibung verlangt eine Vertiefung dieser Theorie.

Dem Schweizerischen Nationalfonds zur Förderung der wissenschaftlichen Forschung danken wir für die finanzielle Unterstützung dieser Arbeit.

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Electronic structure of Ce-pnictides

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The core levels and the valence band region of the Ce-pnictides (CeN, CeP, CeAs and Ce Sb) have been studied by high resolution X-ray photoemission spectro-

scopy. By comparison of the core level spectra clear evidence is found for the intermediate valence character of CeN. Here the f level is found to be pinned at the Fermi level and distinctly separated in energy from the N -derived p valence band. It overlaps the $5d$ conduction band, which is not well resolved because of the small photocross-section at high photon energies. In contrast, in the other pnictides the stable f^1 level overlaps in energy the anion derived p -valence band. It therefore cannot be unambiguously resolved. Its position in energy is well below the Fermi level and because of this fact a large lifetime broadening is expected, as it is indicated by the data.

Splitting of 2p levels in transition metal oxides

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The analysis of far-ultraviolet photoelectron spectra ($90 > h\nu > 20$ eV) of the transition metal (TM) oxides, SrTiO_3 , V_2O_3 , Cr_2O_3 , MnO , FeO , CoO , NiO , and ZnO , yields a consistent picture of the $2p$ oxygen bands. These form bonding and nonbonding subbands with an energy separation of about 5 eV. Their energy with respect to the valence band edge depends on the $3d$ occupancy of the TM ions. This novel interpretation is consistent with the electrochemical and structural properties of these compounds and allows exact determination of the $3d$ excitation spectrum.

Messung der 4f-Elektronen Coulomb Korrelation in EuO, GdTe und GdSb mittels inelastischer Elektronenstreuung (ELS) und Photoemission mit Röntgenstrahlen (XPS)

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Die Anregungsenergie für die Streuung eines $4d$ Elektrons in einem unbesetzten $4f$ -Zustand ist $A = U + E_B$ mit $U = U_{ff} - U_{df}$ und $E_B = E_B^{4d} - E_B^{4f}$. U_{ff} und U_{df} sind bzw. die effektive intra-atomare Coulomb-Energie zwischen einem Paar von $4f$ Elektronen und einem $4f$ und einem $4d$ Elektron. E_B sind XPS Bindungsenergien von $4f$ bzw. $4d$ Elektronen bezogen auf das Fermi-Niveau. Wir finden $U = 5 \pm 1$ eV in EuO und $U = 6 \pm 1$ in GdSb. Der Fehler wird durch die Unsicherheit in der Bestimmung der Schwerpunkte der $4d$ -Multipletts verursacht. Die Resultate von GdTe sind mit denjenigen von GdSb identisch.

Critical currents of a one-dimensional vortex array in a periodic pinning potential

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A theoretical study of the static interaction of a one-dimensional vortex chain with a periodic pinning potential is presented. The critical current, I_c , of the vortex array is obtained by determining the metastable positions of the vortices from a set of coupled non-linear equations describing their equilibrium under the influence of the Lorentz force, the harmonic pinning force and the pinning induced restoring force of the chain. The calculation of I_c is elementary when the (undistorted) vortex row matches the periodic pinning structure. Under non-matching conditions I_c , with the exception of a set of highly symmetric chain configurations, vanishes for a harmonically distorted chain. A significant pinning effect occurs only when a fairly resolved periodic array of defects develops along the vortex row. Results of numerical calculations of I_c based on a vortex-defect superlattice model are presented and discussed in terms of a parameter measuring the relative strength of the pinning force with respect to that of the repulsive interaction force between the vortices.

Dynamic interaction of a lattice of superconducting vortices with a periodic pinning potential

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The flux-flow régime in superconducting films with harmonically modulated thickness is theoretically investigated when steady state vortex motion results from the equilibrium of the Lorentz driving force, the viscous damping force, the harmonic pinning force due to the thickness modulation and the pinning induced lattice restoring force. A highly coherent flux-flow régime with rf properties similar to those of series arrays of resistively shunted Josephson junctions acting in phase and frequency coherence is found when the vortex lattice matches the periodic pinning structure. For non-matching configurations damped lattice modes of deformation dynamically excited by vortex motion in the periodic pinning potential modify the current-voltage (I-V) characteristic and attenuate the interference effect between the natural oscillation of the moving lattice and an applied rf field. The width of the interference transitions appearing in the I-V curves is found to be inversely proportional to the relaxation time for transverse deformation modes of the vortex lattice.

Anomalous temperature dependence of the critical currents of thickness modulated superconducting films

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An unusual temperature dependence of the critical currents, $I_c(T)$, required to unpin the vortex lattice has been observed in thin superconducting granular Al-films where a controlled pinning structure arises from a one-dimensional periodic thickness modulation. In contrast to the monotonic behaviour usually observed in type-II superconductors, $I_c(T)$ shows a characteristic minimum for a temperature T^* , where the critical currents of a modulated layer are essentially equal to those of a flat reference film. This indicates that the pinning mechanism associated with the thickness modulation is ineffective for $T = T^*$, an interpretation consistent with the absence of macroscopic quantum phenomena (rf interference effect, quantum oscillation of the moving vortex lattice) in which the periodic pinning potential plays an essential role. A switching of the pinning sites from the minima to the maxima of the thickness modulation is believed to be responsible for the anomalous behaviour of $I_c(T)$. Possible models providing a switching mechanism are discussed.

Far-infrared absorptivity of very thin Nb/Cu sandwiches with and without perpendicular applied magnetic field

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The far-infrared absorptivity of two Nb(300 Å)/Cu(200, resp. 360 Å) sandwiches and of a simultaneously evaporated Nb(300 Å) reference film was measured at 1.7 K. The measurements were performed both with and without applying a magnetic field perpendicular to the films. We interpret the absorption spectra using a *local* electrodynamic model based on the Abrikosov–Gorkov theory of the pair-breaking effects [1], since we are concerned with very thin films in the dirty limit. The measured absorptivity is expressed as the sum of up to 5 contributions arising from different parts of the film. The concentration c_i and the depairing parameter Γ_i of each part are determined by fitting the calculated absorptivity to the experimental one and by geometrical and magnetic flux considerations. The strong Γ -dependence of the absorption edge of a homogeneous film and the sharp absorption onset allow the unambiguous determination of c_i and Γ_i ; the corresponding order parameter values Δ_i are then calculated.

The most striking result of the analysis is obtained without applied magnetic field. The absorption spectrum of the thinner sandwich is fitted by taking $\Delta = 0.92 \Delta_0$ in the Nb film, and $0.83 \Delta_0$ in the Cu one, where Δ_0 is the order parameter without pair-breaking effect. For the thicker sandwich we get $\Delta = 0.91 \Delta_0$ in the Nb film, while the Cu film has to be divided into three parts to fit the experimental curve:

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230 Å with $\Delta = 0.76 \Delta_0$, 65 Å with $\Delta = 0.53 \Delta_0$, and 65 Å of normal metal. Thus, a rather sharp decrease of the order parameter takes place in the Cu film, approximately 250 Å from the interface. This effect is not observed in the thinner sandwich where the Cu film thickness is less than 250 Å.

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Destruction de la supraconductivité dans un fil creux

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Lorsqu'un fil creux FC supraconducteur de type I est parcouru par un courant statique I_0 , il apparaît un état mixte à 2 dimensions dit de Landau sur la face intérieure du métal. Si l'on fait passer un autre courant I_b à l'intérieur de la cavité, cet état mixte peut se déplacer à l'intérieur de FC [1]. Nous avons calculé les caractéristiques tension courant de FC pour toutes les valeurs possibles de I_b et I_0 . Ces caractéristiques peuvent présenter des paliers horizontaux aussi bien que des discontinuités (sauts) qui accompagnent l'apparition ou la disparition de domaines normaux, supraconducteurs, intermédiaires ou mixtes.

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Two-dimensional current and field distribution in superconductors of rectangular cross-section

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The problem of current and field distribution in a superconducting cylinder carrying a steady current I smaller than the critical current is treated by solving London's and Laplace's equations inside and outside the conductor, respectively.

In this note, we restrict ourselves to the case of an infinitely long slab with square cross-section for which no solution exists in the literature. For this two-dimensional problem, a system of reduced Cartesian coordinates x, y is introduced so that the center of the square is at (0, 0) and the edge of the first quadrant at (1, 1). If the current density $J_z(x, y)$ is normalized,

$$j_z(x, y) = \frac{J_z(x, y)}{\bar{J}},$$

where $\bar{J} = I/F$ = average current density,

$F = 4a^2$ = area of cross-section, and

$\beta = 1/\lambda$ = reciprocal London penetration depth λ ,

then London's equation can be written

$$\frac{\partial^2 j_z}{\partial x^2} + \frac{\partial^2 j_z}{\partial y^2} = (\beta a)^2_{jz}. \quad (1)$$

Equation (1) is solved by a vector potential in London's gauge

$$\nabla \cdot \mathbf{v} = 0, \quad v_{\perp} = 0:$$

$$v_z(x, y) = -\frac{j_z(x, y)}{(\beta a)^2} = -a_0 \left(\frac{\cosh \beta a x}{\cosh \beta a} + \frac{\cosh \beta a y}{\cosh \beta a} \right) - \sum_{k=1}^n a_k \left\{ \frac{\cosh \Gamma_k a x \cos \gamma_k a y}{\cosh \Gamma_k a \cos \gamma_k a} + \frac{\cosh \Gamma_k a y \cos \gamma_k a x}{\cosh \Gamma_k a \cos \gamma_k a} \right\}, \quad (2)$$

where

$$\Gamma_k a = \sqrt{(\beta a)^2 + (\gamma_k a)^2}. \quad (3)$$

With the separation constant

$$\gamma_k a = k\pi \quad (4)$$

the sum $\sum$ does not contribute to the total current I and the coefficient of the first term is simply

$$a_0 = 1/2\beta a \tanh \beta a. \quad (5)$$

In terms of the tangential component $h_{\parallel}$ of the magnetic field, a scalar potential

$$u_s = - \int h_{\parallel} dx + \text{const} \quad (6)$$

can be defined in the surface of the superconductor.

Outside the superconductor Laplace's equation

$$\frac{\partial^2 v_z}{\partial x^2} + \frac{\partial^2 v_z}{\partial y^2} = 0, \quad \frac{\partial^2 u}{\partial x^2} + \frac{\partial^2 u}{\partial y^2} = 0 \quad (7)$$

is solved according to the four-fold symmetry by a vector potential

$$v_z(x, y) = -\frac{2}{\pi} \ln r - \sum_{k=1}^n c_k \frac{\cos 4k\varphi}{r^{4k}} \quad (8)$$

and by a scalar potential

$$u(x, y) = -\frac{2}{\pi} \varphi + \sum_{k=1}^n c_k \frac{\sin 4k\varphi}{r^{4k}}, \quad (9)$$

where

$$r^2 = x^2 + y^2 \quad (10)$$

and

$$\varphi = \arctg \frac{y}{x}. \quad (11)$$

The coefficients a_k and c_k are determined by the boundary conditions, namely, that the vector and scalar potentials are continuous in the surface. This is done here using Gauss' method of least squares. The solutions are regular and of good convergence. As an example, Figure 1 shows the current density in the surface and in the x (and y) axis calculated with $n = 5$ terms.

The procedure described can easily be extended to cases of different cross-sections as well as to systems of conductors. This is the subject of forthcoming detailed publications.

It is a pleasure to thank Mr. Heinrich Mueller of this laboratory for most useful advice and support in numerical calculations.

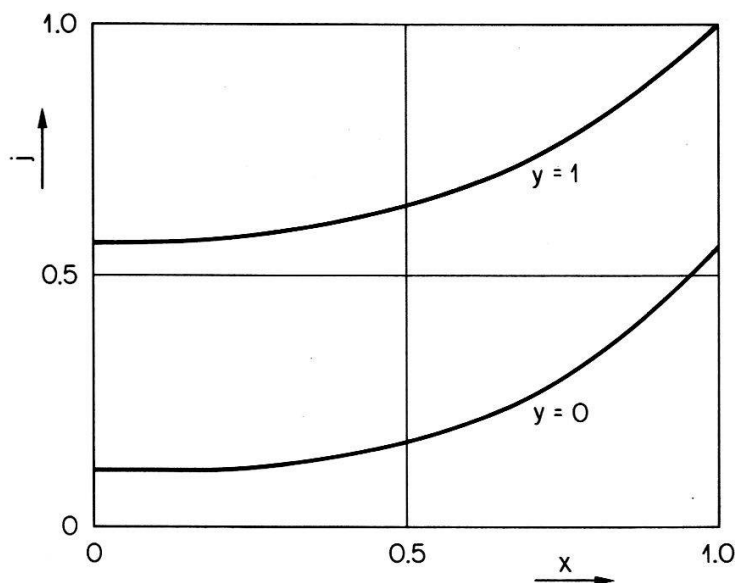


Figure 1
Current density $j = j_z(x, y)/j_z(1, 1)$ in the surface ($y = 1$) and in the x -axis ($y = 0$) of a superconductor of square cross-section, $\beta a = 3$, $n = 5$.

Quasiequivalence of annihilative e^-e^+ and ionizative e^-e^- interactions on dielectric solids and their relationship with valence-orbital flux quantization

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Abstract. The discovery of a far-reaching correlation between the general intensity structure of the spark mass plasma and the average positron lifetime of real AgBr crystals has brought forth the existence of a general relationship between the average charge center shift δ of the atomic ions in the spark stereoplasma and the average interaction rate λ of e^+ annihilation as a function of the chemical binding in dielectric solids. The true reason for this relationship lies in the bond-dependent electron spin coupling. We show in an approximate way that the empirical laws can be cast into a fully symmetric form if the differential interaction rates of both processes are suitably normalized with regard to valence and the respective differential changes of the (heteropolar) chemical bond. The interaction rates themselves are found to be essentially identical in both phenomena. It is shown that the deeper physical cause for this relationship must be sought in the phenomenon of flux quantization on the extended valence-orbitals mediated by inductive action of impacting positrons or electrons. The e^-e^+ ortho- and para-positronium states find an almost perfect pendant in the electron-hole states of the stereoplasma.

Introduction

The purpose of this study is to show the quasiequivalence of annihilative and ionizative elementary interactions of impacting positrons and electrons when acting on the chemical bond in dielectric solids. The experimental results were obtained by positron annihilation lifetime measurements on the one hand and spark source mass spectrographic analyses on the other hand. The e^+ lifetime investigations on real AgBr crystals were performed at this laboratory by H. Surbeck [1, 2]. For the elaboration of the λ -shift laws we used the λ -results of Bertolaccini et al. [3] on alkali and alkaline earth halides and chalcogenides. The definite advantage of using these results is the fact that they were performed by only one laboratory applying always the same evaluation procedure and the same source correction [4]. The mass spectrographic results on AgBr and the rare earth compounds were obtained by the present author. The charge center shift (δ -shift) due to ionizative electron interactions in the stereoplasma was obtained on doped and undoped rare earth compounds crystallizing in the rocksalt structure (B_1). The stereoplasma which precedes the atomic disintegration of the solid is defined here as the primary collective plasma state which has a lifetime of about 200 ps (1 ps = 10^{-12} s) and involves electronic Bose-Einstein condensation. The observation of the δ -shift dates back to 1973 but could not be explained at that time.

This work is divided into two parts: In the first exclusively empirical part we present a few hitherto unpublished results derived from primary data obtained by the methods mentioned above. For brevity primary data as well as the description of the experimental techniques are omitted. As a consequence of the universal nature of the stereoplasma, mass spectra may be examined by an almost infinite number of aspects. We therefore confine our analysis to the most important ones. In the case of real AgBr we are mainly interested in the matrix molecular ions and the impurities, while in the case of rare earth compounds we are especially interested in the distributions of mono- and multi-positive matrix cations and anions. Since AgBr is monovalent, it does not allow a reliable δ -shift analysis. The case of real AgBr touches on the difficult problem of stereoplasma molecule formation from an ideally non-molecular solid. This problem, in its idealized version, has been overcome by a partial solution [5]. The complicated role of the imperfect or 'real' nature of a solid in the process of molecule formation was partly elucidated by plasma valence analysis based on the study of complex ions. This kind of study clearly indicates that many-body effects are important both in positron annihilation and in plasma molecule formation. Fajans' early concept of electron quanticules, when suitably extended to non-integral electron clusters, forms an excellent means to describe such effects. The study of the formation of binary molecules (more strictly speaking: molecular ions) has unique microcosmological aspects, but a better understanding of short-range coherence superconductivity is awaited. The asymmetrizing effect of Schottky and Frenkel defects as well as of static and dynamic Jahn-Teller effects has become evident by the valence phenomenon [6], however, at present there is no quantitative model. The molecule problem of real AgBr is similar but we shall mention it only in a few marginal notes though the plasma/ e^+ lifetime correlation originally was discovered by the anomalous behaviour of AgBr matrix molecules [7]. The extremely complex and subtle nature of interrelationships generally associated with real nature phenomena and principally inherent to the stereoplasma will be discussed briefly in a table. The main emphasis of this work is not on the molecules but rather on the matrix information obtained

from the atomic ions of simple dielectric compounds. In order to do full justice to this difficulty we shall bear in mind (1) that both methods have only used real solids (ideal solids are fictions); (2) that all primary data must, for experimental reasons and those of data processing, be considered as approximate; (3) that the results derived from both λ -shift and δ -shift are at best semiquantitative; an 'exact' determination of the δ -shift is principally impossible; negative ions as a rule are missing, and neutral species are completely unknown; (4) that for the theoretical treatment of the equivalence we use the approximation of the ideal gas; this simplification of the problem is permissible only for small δ -values.

Taking all these facts into account, it may easily be recognized that the quantitative side of the equivalence can at best be proved to a factor of 2. Any better agreement is merely chance. We are primarily interested in the fact of the equivalence as such, while we are glad to obtain the order of magnitude for the strength of interaction.

In the second part the example of the δ -shift is used to demonstrate how the extremely complicated problem of non-thermal atomic disintegration of solids can be simplified to a tractable case. This is only possible if, on the one hand, we neglect the detailed contributions of the actually unknown distributions of the oxidation states of the chemical bond for either kind of atoms and if, on the other hand, we content ourselves with the average contribution of anions and cations to the δ -shift. The δ -shift thus can be regarded as an average value for the electron spin coupling of the integral valence shell quanticule. The purpose is to describe only simple binary solids of formula MX . In view of these circumstances it appears fully justified to use Pauling's (atomic) electronegatives as the parameters describing the degree of heteropolarity of MX . The use of more adequate (solid state) electronegativities has no basic advantage. In the ideal gas approach we recognize that the empirical δ -shift relationship is, in fact, a manifestation of the Eggert-Saha equation. But the description of the process as some kind of quantum thermionic emission from the plasma atom, based on the Richardson-Dushman equation would lead to an essentially identical result. This point of view provides the average ionization energies which operate in the δ -shift. In this way we are prepared to define the corresponding plasma interaction rate. Finally the equivalence permits a few basic statements as to the symmetry of e^-e^+ and e^-e^- interactions, the number of quanticule electrons in both cases and the strength of the interactions. However, only the deeper understanding of the plasma dynamics, which reveals the stereoplasma atom to be a true harmonic quantum oscillator, allows us to realize that both the λ -shift as well as the δ -shift are connected with the phenomenon of flux quantization on the atomic scale. The flux quantization takes place in the case of the λ - and δ -shift on the slightly extended valence orbitals of the matrix ions. From the study of this process the general meaning of the λ -shift and the δ -shift within the framework of the stereoplasma becomes obvious. With this in mind the conclusion becomes obvious that the electron-positron interaction has an adequate image in the form of the electron-hole interaction of the plasma. This latter aspect becomes of vital importance if we try to guess the true superconducting energy gap associated with the electronic Bose-Einstein condensation.

I. Empirical situation

Molecular ions

In 1975 we reported about a correlation between the spark mass plasma intensities of molecular as well as atomic ion species (matrix and impurity) and the average e^+ lifetime for the example of real AgBr crystals grown by different techniques and laboratories [7]. Both the present understanding of molecular formation in the plasma and the discussion of the analytical results regarding the role of the chemical impurities left little doubt that the longer lifetimes had to be attributed to the more ideal crystals ($\tau_{\text{ideal}} \simeq 340$ ps), characterized by the arbitrary reality parameter value $x = 0$, and the shorter ones to the more real crystals. Despite the fact that the AgBr^+ concentration, when extrapolated to 100%, $c_{\text{AgBr}} \rightarrow 1$ (i.e. perfect electronic non-ideality), characterized by $x = 1$, agreed with the shortest measured lifetime, $\tau_{\text{real}} \simeq 120$ ps, neither the physical nature of the disturbance which raised the interaction rate, nor the true physical significance of the components of the time spectra was then evident. Further analysis of the results led to an ever-increasing complexity of the correlation in the case of AgBr and to a general relation between the phenomena of e^+ annihilation and spark disintegration of dielectric solids.

Figure 1 shows the mass spectrographic concentrations of four typical elemental impurities as a function of the matrix molecule concentration $[\text{AgBr}^+]$ in the case of three identical crystals from the same batch, without etching ($c_1^{(1)}$ and $c_1^{(3)}$) and after etching in ultrapure 1-molar KCN ($c_1^{(2)}$). The impurities are completely unaffected

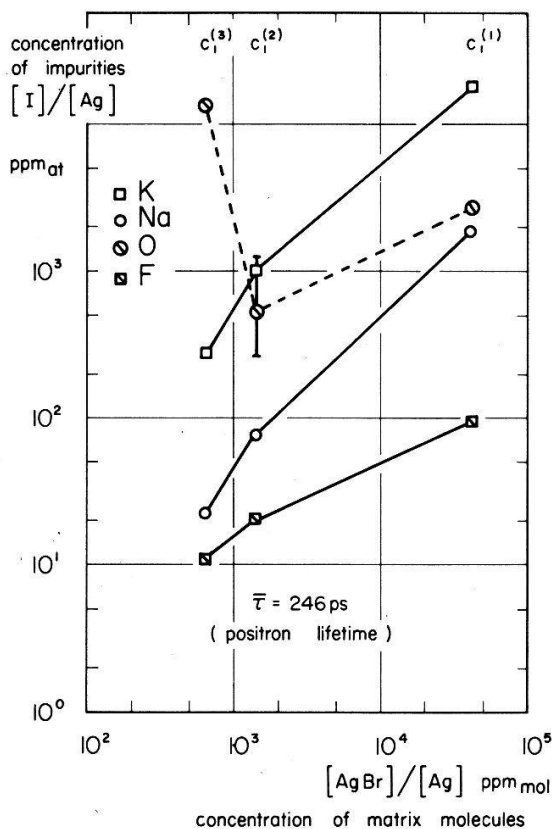


Figure 1
Concentration of impurities as a function of $[\text{AgBr}]$ for 3 identical AgBr crystals (same batch), showing identical positron lifetime. Strong correlation of all impurities with the molecular ion intensity ($C_1^{(2)}$ etched in 1m-KCN).

by the etching procedure. They may therefore be considered as true bulk impurities, where surface contributions appear to be completely negligible. For both cationic (K, Na) and anionic (O, F) impurities the correlation is obvious and similarly strong. On the other hand, all three samples have, within a few ps, the same average lifetime $\bar{\tau} = 246$ ps. This unequivocally demonstrates that the positrons do not see the strongly localized asymmetry potential which is due to the individual impurity atom and which is responsible for the 'real nature' fraction of the molecule formation. This situation may easily be understood in terms of 'thermalized' positrons – whatever this means! – having a De Broglie wavelength of 50–100 Å, which therefore see much larger crystal regions than the plasma electrons having a coherence length of only a few Å.

Figures 2 and 3 show the same situation for cationic and anionic impurities, respectively, but for crystals classified according to a common average ideal or real lifetime which are otherwise different in origin and crystal growth technique. Again, the longer lifetimes are on the average associated with smaller impurity concentrations than the shorter ones. A further conspicuous aspect is the smaller statistical scatter for the more ideal samples. This fact supplies a further independent method to estimate the ideal lifetimes. We again find $\tau_{\text{ideal}} \simeq 320\text{--}340$ ps. On the other hand, there are crystals which have a short lifetime – i.e. a high degree of reality – and therefore

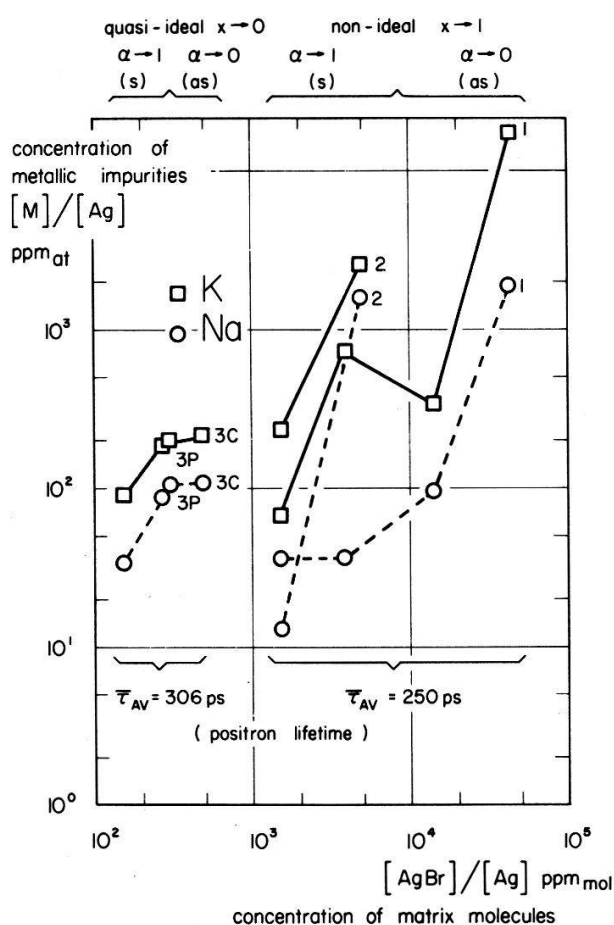


Figure 2
Concentration of cationic impurities as a function of $[\text{AgBr}]/[\text{Ag}]$ for non-ideal and quasi-ideal AgBr crystals, and for low and high lateral symmetry.

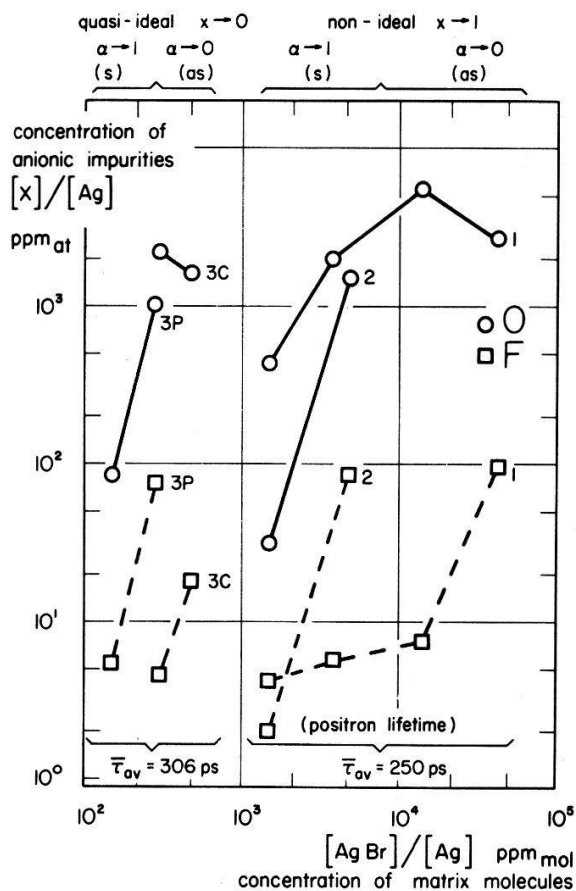


Figure 3
Concentration of anionic impurities as a function of $[\text{AgBr}]/[\text{Ag}]$ for non-ideal ($x \rightarrow 1$, $\bar{\tau} = 250$ ps) and quasi-ideal ($x \rightarrow 0$, $\bar{\tau} = 306$ ps) AgBr crystals, and for low ($\alpha \rightarrow 0$) and high ($\alpha \rightarrow 1$) lateral symmetry.

appear to have a higher degree of chemical purity while not having an equally low molecule rate. This puzzling situation is of particular interest with regard to the physical nature of the local asymmetry. It is true that on the average the impurities indicate the degree of reality. But there must exist other physical factors which determine the extent of local asymmetry in the presence of an impurity. What are these factors? We are confronted with extremely subtle effects, which probably can only be observed by a very limited number of physical techniques, and maybe by the present method only. We imagine that local phase perturbations of binding electrons form the true cause of the local asymmetry. Therefore we are confronted with some kind of electronic amorphicity. Such a state could easily be created by small changes of thermal gradients during the crystal growth. We presume therefore a close connexion with the crystal growth technique itself, which in fact appears to be the case. This provides a useful hint as to the physical nature of such frozen-in paraelectronic perturbations. This kind of imperfection should not be confused with the more usual mechanical strains, even if these imperfections would also create important local strains – depending on the actual size and chemical nature of the impurity. For instance, if the thermally created asymmetry potential is of the order of a few eV, there is no hope to get rid of it by annealing the crystal if the chemical bond acts in the wrong direction. This fact stresses the importance of local catalytic action. It is not excluded that the photographic quality of AgBr is connected with paraelectronic perturbations rather than with the chemical purity or the presence of a particular impurity. This possibility could explain the well-known difficulty to parametrize the photographic sensitivity. This leads to the concept of local ‘left-to-right’ symmetry characterized by a factor α . If at the site of the atom the symmetry in a particular direction is low, $\alpha \rightarrow 0$, neighbouring nuclei will inevitably flow together and create a molecule (Jahn–Teller effect). If, on the other hand – even in the presence of non-negligible amounts of impurities and essentially reduced potential barriers – the degree of symmetry is high, $\alpha \rightarrow 1$, there is still little stimulus for neighbouring atoms to form molecules. This peculiar situation is well demonstrated in Figure 4. The upper part (a) shows molecule rates as a function of $\bar{\tau}$ for undoped AgBr crystals which were prepared by different laboratories: Ciba–Geigy (C), Max Planck–Institut (P) and Merck–Lab (M). For a discussion of the techniques we refer to Surbeck’s thesis [2]. Note that C_2 and C_3 crystals were in fact produced by the (P) technique.

The lower figure (b) shows the same data as in (a), however curves for different degrees of local symmetry have been added; these are given by the simple expression

$$\frac{c_\alpha(x)}{c_{\text{ideal}}} = \left(\frac{c_{\text{real}}}{c_{\text{ideal}}} \right)^{x - \alpha x^2} \quad (1)$$

The physical idea of expression (1) is as follows: In the extreme case of full local asymmetry ($\alpha = 0$) the molecule rate will be uniquely determined by the arbitrary degree of x of reality assumed to be proportional to the actual lifetime reduction $\tau_{\text{ideal}} - \bar{\tau}$. Increasing symmetry ($\alpha > 0$) corresponds to a more balanced asymmetry potential on either side of the atom. This negative trend is proportional to x^2 . The square is due to the law of independent probabilities. The exponential growth (epic) is indicative of the extended ‘collective’ nature of paraelectronic perturbations in the presence of impurity catalysts. By this kind of chemical overlapping the efficiency of molecule formation becomes much stronger than in the case of a simple linear action. The dotted line (laic) shows such a linear active-impurity catalyst behaviour in the

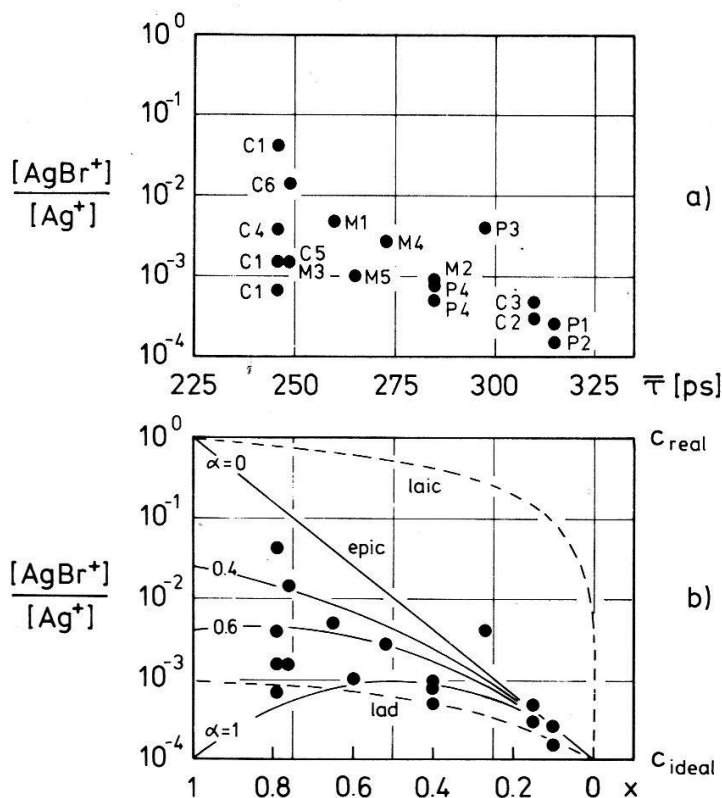


Figure 4

(a) Concentration of $[AgBr^+]$ for real AgBr crystals as a function of the e^+ lifetime. Samples with different growth techniques (see text) (b) Experimental and calculated $[AgBr^+]$ as a function of the degree x of electronic disturbance for exponential passive-impurity-catalyst asymmetry (epic) and lateral symmetry α , together with linear active-impurity-catalyst (laic) and -disturbance (lad) behaviour.

case of magnesium metal, where the rate of Mg_2^+ formation is normalized to the amount of oxygen. In this case the asymmetry potential appears to be perfectly local. There is no contradiction to the linear and exponential growth rates in Figure 4b. In fact we did not yet mention the strength of the average impurity interaction in AgBr which is weaker than that of oxygen in magnesium by a factor of 10^4 . Thus the respective linear active-disturbance behaviour would result in a very small increase of the molecule rate (lad). This fact explains why most methods fail to observe electronic impurities unless they are associated with considerable amounts of elemental impurities. We may now try to get a better estimate for τ_{real} by extrapolating from small degrees of reality as well as pure exponential growth ($\alpha = 0$) and obtain roughly $\tau_{real} \simeq 200$ ps. This value compares well with the value $1/\lambda_1 = 230$ ps of Surbeck's model [1, 2]. Finally, a rough estimate can be made of the range of interaction of paraelectronic perturbations. From Figure 4b we see that for $x < 0,1$ a distinction between linear (lad) and exponential (laic) growth rates becomes meaningless. Thus the range of interaction must at least enclose the twelve equi-distant neighbours of the 'impurity' if AgBr is assumed to crystallize in B_1 -structure. This extent essentially coincides with that of the valence orbitals and also makes transparent in the case of molecule formation, the role of flux quantization. If, as a result of overlapping orbitals, the atoms become 'multiply'-connected, the flux-trapping occurs much more easily. This is equivalent to saying that the effective molecular ionization energy decreases as a result of the decreased spin coupling, thus favouring both the observable amount of molecular ions and the effective positron interaction. But this is the essence

of the interaction equivalence applied to the situation of the real nature (imperfection) of a solid, viewed by the complicated molecule formation on the one hand, and the anomalous increase of the e^+ interaction on the other hand.

Atomic ions: λ -shift

We shall now turn to the question whether and in which way the average positron interaction rate λ is connected with the chemical bond. (In the present work we disregard the role of the components.) We envisage only simple dielectric solids, in particular those of the type MX . For this purpose, we shall only use the results of the Milano positron group [3]. There is no need to emphasize the great merits of that laboratory for investigating entire crystal series. In a future study it shall be attempted to do justice to all presently existing dielectric data, which, in a most meritorious way, have recently been made available [8]. The Italian authors find that λ , in the case of alkali and alkaline earth halides as well as chalcogenides may well be represented by a linear function

$$\lambda = \lambda_0 + \frac{1}{4}c_s\xi'n^* = \lambda_0 + c_D\xi'n^*. \quad (2)$$

From a least squares fit they obtain $\lambda_0 = 0.195 \cdot 10^{10} \text{ s}^{-1}$. This value very nearly coincides with the calculated annihilation rate of the negative positronium ion in the vacuum, $\lambda_{ps^-} = 0.199 \cdot 10^{10} \text{ s}^{-1}$ (Ferrante). $c_D = c_s/4 = \pi r_e^2 c = 7.48 \cdot 10^{15} \text{ cm}^3 \text{ s}^{-1}$ is the Dirac fundamental interaction cross section, where r_e is the 'electromagnetic' radius of the electron ($2.8 \cdot 10^{-13} \text{ cm}$) and c is the velocity of light. $\xi' = 6.09$ is tentatively identified with the number of electrons in the anion valence shell. For the anionic particle density the authors choose

$$n^* = nv_- / \left(1 - \frac{4\pi}{3} R_+^3 nv_+ \right) \quad (3)$$

where $n = N\rho/M$; N = Avogadro's number, ρ = mass density and M = molecular weight of MX . R_+ is the radius of the positive ion, and v_- and v_+ respectively, denote the number of negative and positive ions in the molecule: In the case of MX $v_{\pm} = 1$.

Results obtained by the recently discovered method of flux quantization on the atomic scale [9] provide for the first time a direct proof that electrons in the stereoplasma occupy the cationic space. By virtue of symmetry, this justifies the assumption that positrons prefer the anionic regions of the solid. Thus n^* is the best possible choice, but we ought to bear in mind the approximate nature of this choice in view of the fundamentally unsolved radius problem.

Figure 5 shows the λ -results for dielectric solids as a function of n^* as given by [3]. For clarity, we have slightly modified the representation. One may argue that we are looking for physics where there is merely statistics. This is certainly not true, but we must postpone the answer to the end. Without doubt, the following analysis is, for a number of reasons, at best semi-quantitative. Nevertheless, Figure 5 reveals a few important and interesting aspects: (Ia) For a homologous series MX , where the anion X is fixed, the slope $d\lambda/dn^*$ is obviously correlated with the degree of heteropolarity of MX , i.e. also with the electronegativity of X ; the further down X is located in the periodic table the greater is the slope. (Ib) The slopes are furthermore connected with the valence v_2 of $M^{v_2} X^{-v_2}$. If only the degree of polarity was important we should expect the oxides to lie between the monochlorides and the monofluorides and the

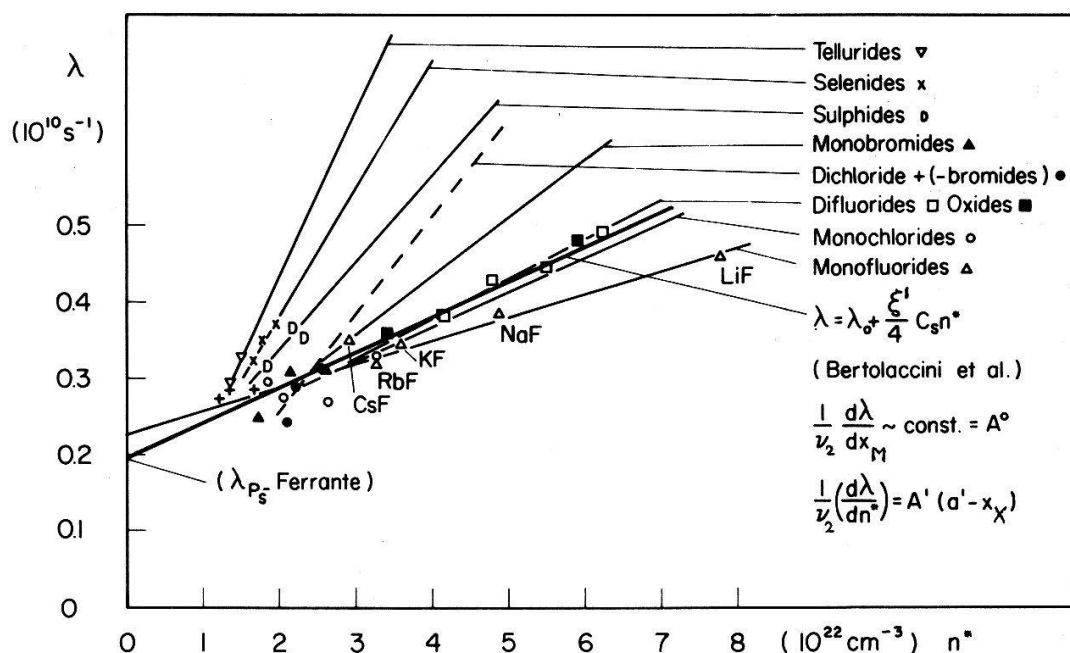


Figure 5

Positron annihilation rate as a function of the anion electron density for various solids (compiled by M. Bertolaccini, A. Bisi, G. Gambarini and L. Zappa); $\Delta\lambda$ proportional to cation EN change Δx_M , valence v_2 , and change Δn^* , resp. (pres. author).

monochlorides between the oxides and sulphides, respectively. This is not at all the case. On the contrary the oxides rise even more strongly than the chlorides; thus we conclude that the slope is directly proportional to the valence. (IIa) Somewhat less obvious but still clearly recognizable, we observe that, within a series MX (where X is fixed) the interaction rate is larger when M becomes more electronegative.

The rise in λ is approximately proportional to the change of the cationic electronegativity. (IIb) Again the change in λ , when normalized to a unit change of the cationic electronegativity, is approximately proportional to the valence.

For the sake of completeness we represent the derived values in Table 1 with v_2 being the presumable valence of the anion. We are also including dihalides which viewed from the positron act as monovalent. x_X denotes the Pauling electronegativity, whereby for dihalides x_X is somewhat smaller than the respective value of the monohalide on account of the repulsive action among the anions. The very small difference Δx_M , as determined by the boundary cations, explains the large mean error of 30%. The true solid-state value is, of course, unknown and may easily differ by 100%. The result is still very good.

The results of the second and third groups in Table 1 are represented in Figure 6 and may well be described by a linear relationship. The regularities as obtained from IA and IIA halides and IIA chalcogenides can be expressed in the following way:

$$\frac{1}{v_2} \frac{d\lambda}{dn^*} = A'(a' - x_X) \quad (4)$$

$$\frac{1}{v^2} \frac{d\lambda}{dx_M} \sim \text{const} = A^0 \quad (5)$$

where $A' = 5.10^{-14} \text{ cm}^3 \text{ s}^{-1}$, $a' = 4.2$ and $A^0 = (3.0 \pm 0.9) 10^9 \text{ s}^{-1}$.

Table 1

Slopes per unit valence of the average e^+ interaction rate λ as a function of the anionic density n^* together with the Pauling anionic EN x_X ; the cationic EN difference Δx_M belonging to the boundary cations of the investigated series MX (X fixed), and the interaction rate change per unit valence and unit cationic EN change are given for a variety of series of alkali and alkaline earth halides and chalcogenides, as derived from the experimental data [3].

(1a, 1b)				(11a, 11b)		
MX	$\frac{1}{v_2} \frac{d\lambda}{dn^*}$	$(10^{-13} \text{ cm}^3 \text{ s}^{-1})$	x_X	M_1X-M_2X	Δx_M	$\frac{1}{v_2} \frac{d\lambda}{dx_M} (\cdot 10^9 \text{ s}^{-1})$
Monofluorides	1	0.33	4.0	CsF-LiF	0.3	4.9
Monochlorides	1	0.45	3.0	RbCl-LiCl	0.2	2.8
Monobromides	1	0.77	2.8	KBr-LiBr	0.2	3.4
Monoiodides	1	0.33	2.5	RbI-LiI	0.2	
Difluorides	1	0.52	3.3	BaF ₂ -MgF ₂	0.3	3.6
Dichlorides	1	0.80	2.9	BaCl ₂ -BaBr ₂	0.1	2.8
Monoxides	2	0.25	3.5	SrO-MgO	0.2	3.2
Monosulphides	2	0.57	2.5	BaS-CaS	0.1	2.5
Monoselenides	2	0.85	2.4	BaSe-CaSe	0.1	2.4
Monotellurides	2	1.1	2.1	BaTe-SrTe	0.1	1.6

$A^0 = (3.0 \pm 0.9) 10^9 \text{ s}^{-1}$

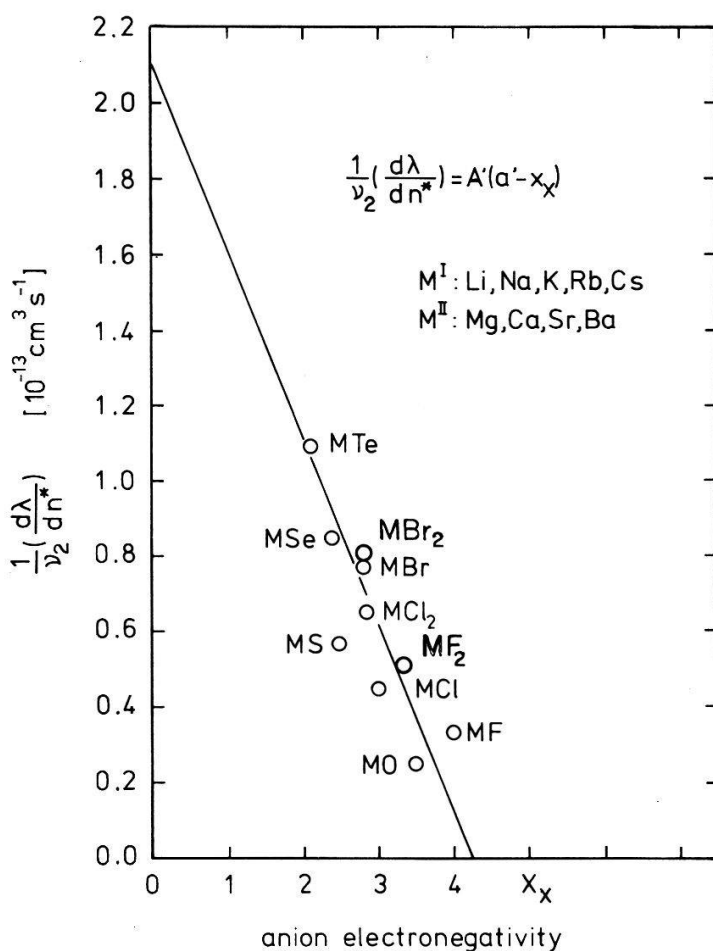


Figure 6

Change of e^+ annihilation rate per unit change of anion electron density and valence unit as a function of the anion electronegativity for various solids.

Preliminary interpretation

Equations (4) and (5) may be understood as electron spin pair decoupling effects. decreasing heteropolar bond draws, according to (5), the cation to a more neutral state A ($\Delta x_M > 0$), therefore increasing the interaction with e^+ . The rate increase $\Delta\lambda$, normalized per unit valence, appears reasonably independent of the individual cation. The role of the cation is qualitatively described by the electronegativity increase Δx_M . Of course, no information is obtained in this way about the absolute value of λ . In fact Δx_M stands for the average cationic 'field' action. Similarly, according to (4), the rate change per unit electron density change Δn^* and unit valence is greater for less heteropolar bonds (x_X decreasing). The accompanying electron spin pair decoupling may be interpreted as an effective single-electron component. This fact corresponds to the quasifree electron state of the 'real' component of AgBr (i.e. λ_1 in [1. 2]). In a broader sense (4) and (5) appear to suggest that any rate change $\Delta\lambda$ automatically entails $\Delta x_M \neq 0$ (even when M is fixed!) and $\Delta n^* \neq 0$ (neglecting Δx_X), although these facts are not reflected by the formulae. For a strongly heteropolar bond $\Delta\lambda/\Delta n^*$ becomes small, thus n^* must vary considerably to achieve an actual $\Delta\lambda$. This means that the anion valence shell behaves like a compressible gas suggesting strong pair coupling, or in other words: weak inter-pair interaction (each pair acts as a good boson). The reverse appears to hold for more covalent compounds where in virtue of pronounced inter-pair interaction the anion shell rather behaves like an incompressible fluid owing to the increasing importance of the Fermi-hole aspect.

Atomic ions: δ -shift

The interesting phenomenon of the δ -shift has to be interpreted as a demonstration of the fact that the quasiparticles of the stereoplasma which effectively carry the Bose-Einstein condensation (i) are of composed nature, and (ii) have only a limited bond-dependent stability. If now the heteropolar bond becomes weaker, the composed particles will be more easily decomposed or ionized, as the effective ionization energy must have been diminished. From this finding we may draw an important conclusion: even in the case of electrically insulating (or semiconducting) compounds the intensity distributions of the atomic ions, as derived from the mass spectrum, are not automatically identical with the distributions of the crystalline oxidation states of the matrix and impurity ions, because even the majority ground state – which by definition of the harmonic charge oscillator has to be assumed as strictly non-ionizative! – has become partly ionized. This is almost obvious as no chemical bond is infinitely strong. Does this mean that the non-ionizative oscillator ground state does not really exist? We do not think so. Recent investigations leave no longer any doubt about the true existence of the oscillator. One must, therefore, conclude that the actually observed spectra represent a linear superposition of a system of quantum oscillators – in principle one per oxidation state – and of an additional quantum thermionic ionization attributable to the finite bond strength of all quasi-particles (excited as well as non-excited ones). In the ideal case this additional rate of ionization must not depend on the degree of (dynamic) ionization. We therefore expect, in its most idealized version, a common charge center shift of all species toward more positive values. This is the δ -shift. But only since very recently we know that also the

δ -shift, and not only the excited oscillator states, must be associated with atomic flux quantization [10].

In practice the δ -shift is rather difficultly obtained. One of the reasons for this fact is the outstanding sensitivity of flux quanta on local perturbations. As a rule real solids are very far from ideality. This holds even for elemental solids such as simple metals if the single crystals were not obtained by applying extreme care during the crystal growth and profiting of outstanding environmental conditions. This is unfortunately very rarely the case. Thus, only long experience and great patience can provide us with reliable (average) δ -results. But even then the uncertainty is relatively large.

There is another equally important reason why δ -values are difficultly obtained. δ is by definition the arithmetic mean of the anionic and cationic shifts, i.e. the shift of the respective charge centroid. In practice these shifts are obtained by extrapolation of the oscillator intensity curves to an apparent 100% concentration (in the semilogarithmic plot multipositive ion intensities are linear insofar as electron shell influences may be neglected). Now, one of the most important pillars in the proof of atomic flux quantization in the case of dielectric solids is the fact that anionic radius changes manifest themselves, under certain conditions, as K_X -changes (K_X is the slope of the anion curve), while on the other hand the cation slope is universal, neglecting small structure dependent effects. By the varying anion slope, yet, the extrapolation becomes problematic if the shift is small. For this reason we must content ourselves with an approximate average cation-anion δ_{MX} -shift.

Again, we shall, for the sake of completeness, indicate a few of the δ_{MX} -data in Table 2. All values were obtained on simple non-monovalent rare earth chalcogenides and pnictides. We shall not bother about the fact that the chemical bond in these cases

Table 2

Approximate spark valence ν_1 as derived from the valence effect [6], approximate average cation-anion δ_{MX} -shift as determined from the charge centroid shift of multipositive ions in the spark plasma, and Pauling EN difference Δx for a variety of doped and undoped rare earth compounds and in the case of GdP grown by a variety of crystal growth conditions (1)–(7).

Subst.	ν_1	δ_{MX}	Δx	Subst.	ν_1	δ_{MX}	Δx
EuO	2	0.04	2.4	GdS	2.5	0.56	1.4
EuS	2	0.64	1.4	GdSe	2.5	0.75	1.3
EuSe	2	1.0	1.3	GdTe	2.5	0.80	1.0
EuTe	2	0.62	1.0	GdN	3	0.23	1.9
		0.66		GdP(1)	3	1.0	1.0
		0.85		GdP(2)	3	0.82	
	Av.	0.71		GdP(3)	3	0.61	
EuO:Gd(ppm _{at})				GdP(4)	3	0.88	
EuO: 25	2	0.05	2.4	GdP(5)	3	0.91	
EuO: 150	2	0.13		GdP(6)	3	1.0	
EuO: 520	2	0.18		GdP(7)	3	0.64	
					Av.	0.85	
EuO: 1600	2	0.04		GdAs	3	0.97	0.9
EuO: 3300	2	0.20		GdAs	3	1.2	
EuO: F (1%)	2	0.24					
	Av.	0.13		DyP	3	0.65	1.0

also involves contributions from the incomplete f -shell. The δ -shift cannot allow for small effects. v_1 denotes this time the (formal) spark valence as derived by the valence effect [6]. In contrast to λ -results we shall use now the electronegativity difference $\Delta x = x_X - x_M$ as the bond parameter, which appears more adequate to the fact that the δ -shift is the arithmetic mean of the individual shifts: $\delta_{MX} = (n_M + n_X)/2$.

All data in Table 2 belong to crystal systems which have been investigated under quite different aspects of the stereoplasma and have already been published [11, 6, 12, 13, 5, 9] or shall be published. Thus, the indication of the precise numbers is unavoidable – even if, to a certain extent, they belong to the real nature of the solid – if the total character of the interrelationship shall become manifest (some time in the future).

Figure 7 shows δ_{MX}/v_1 from Table 2 as a function of Δx . Although a true linear behaviour may only be expected for small δ -values we approximate $(\delta_{MX}/v_1)/\Delta x$, just as for the λ case, by a linear function:

$$\frac{\delta_{MX}}{v_1} = A''(a'' - \Delta x) \quad (6)$$

where $A'' = 0.2$, $a'' = 2.6$, and $\Delta x = x_X - x_M$.

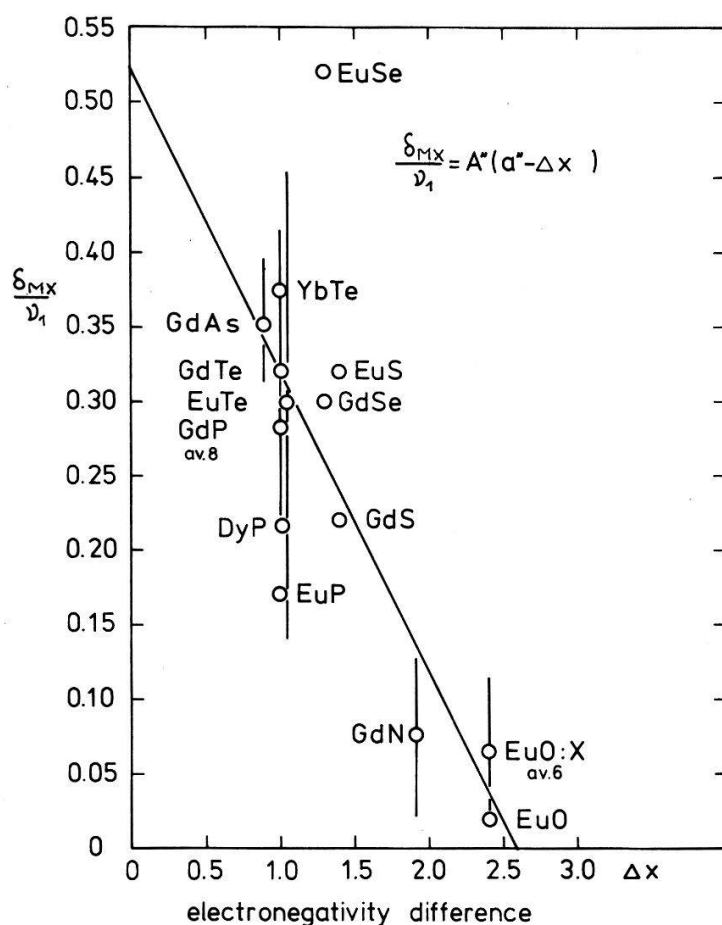


Figure 7

Average plasma atomic ion charge center shift per valence unit as a function of the Pauling anion-cation electronegativity difference for binary solids.

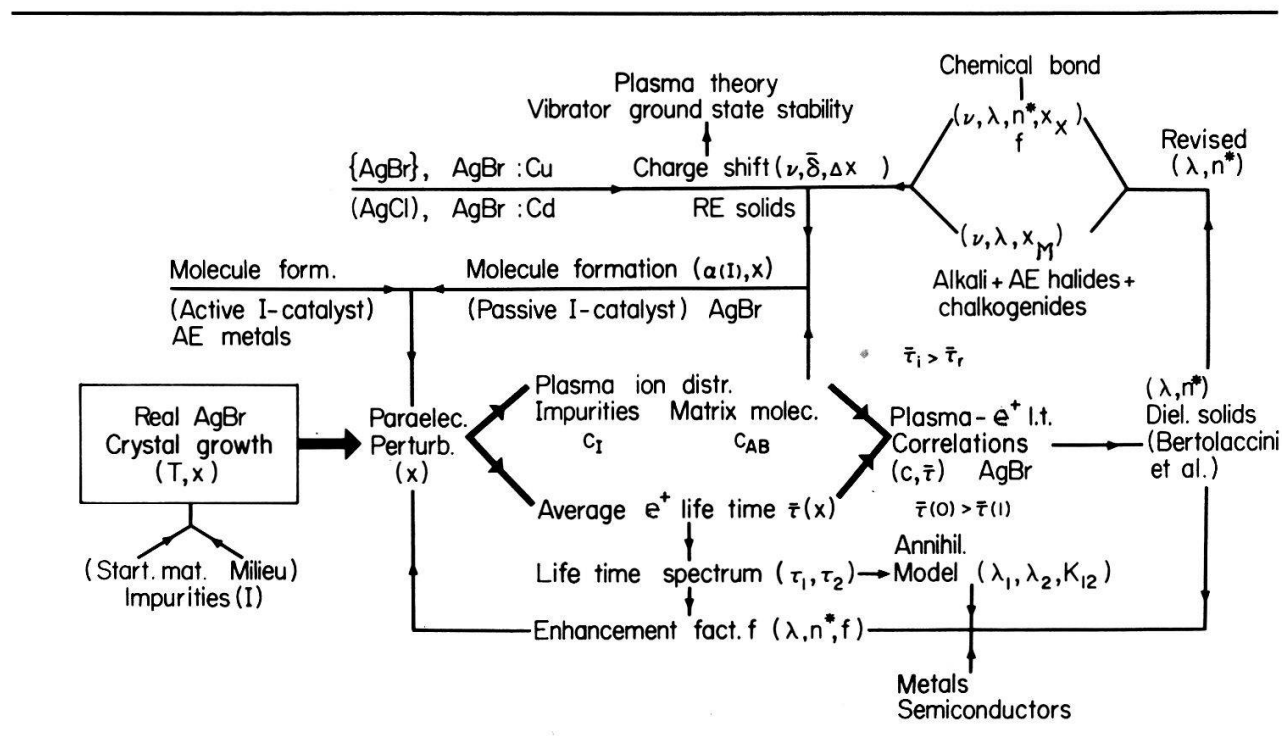
Preliminary interpretation

Equation (6) is the plasma analogue of (4) and (5). δ_{MX} if normalized to the valence unit is a measure of the tunnelling probability for a paired electron to escape from the plasma oscillator. According to the harmonic oscillator picture of plasma atoms, ground states ideally should not become ionized. Thus, (6) is taken as a proof that the relevant quasiparticle of the ground state is of composed, dissociable (i.e. ionizable) nature – in fact, a more or less strongly coupled electron pair according to the actual chemical binding. δ_{MX} corresponds effectively to $\Delta\lambda$. The plasma pendant of the moving vacuum hole is represented by the effective Fermi-hole (at rest) of the atom, seen by an incoming hot electron that is going to ionize the atom during the extremely short 'last moment' of isotropic disintegration – a time interval of at most the period of the cyclotron resonance in the local flux quantum field, $\tau_c \simeq 5.10^{-16}\text{s}$ [10].

General frame of the correlation: résumé

Table 3 shows in the upper part plasma associated aspects, in the lower part those of the positron. Starting from the left, real AgBr crystals as a rule differ not only by their growth techniques, but also by starting purities, milieu influences and thermal history. In the case of AgBr this gives rise to the occurrence of paraelectronic perturbations of concentration x , which apparently influence directly the lifetime of the thermalized positrons (abscissa). Their influence on the molecular formation (ordinate) – in particular of AgBr^+ – on the other hand is of rather complicated indirect nature, insofar as impurities play a vital role in the dynamic formation of local asymmetries. Microscopic asymmetry is an absolute necessity for the formation

Table 3
Spark plasma structure/Positron annihilation life time correlations on real AgBr, and associated relationships.



of molecules. In a highly simplified and approximate way molecular formation, showing an exponential law for full asymmetry, may be ascribed to (i) the asymmetrizing action of fortuitous passive impurity catalysts and (ii) the collective strengthening of dynamic asymmetry due to the overlapping of individual disturbances. From the linearization of the exponential curve we estimate the spatial extent of the individual disturbances, comprising roughly the space of 10–100 atoms (2–5 Å linearly). This makes plausible the strong interaction with the e^+ having an even longer De Broglie wavelength.

There are fair grounds to believe that $1/\tau_1$ may be regarded as the sum of the interaction rate λ_1 corresponding to the annihilation of the positrons with the 'quasifree' electron gas, and k_{12} corresponding to the transition of positrons into the intrinsic state of the ideal solid with the interaction rate λ_2 ($1/\tau_1 = \lambda_1 + k_{12}$, $1/\tau_2 = \lambda_2$) [1, 2]. In this way the real nature behaviour of AgBr may be reconciled with both typical interaction rates of metals and semiconductors and of dielectric solids. Bertolaccini et al. [3] proposed an attractive model which allows them to relate their interacting rate ' λ_1 ' (corresponding to our λ_2 of AgBr: in Figure 5 ' λ_1 ' = λ) of dielectrics linearly with the anion electron density n^* . Beyond any doubt the empirical ansatz of the Italians has very attractive features. Indeed when looking more closely into the compiled data two independent, qualitative relationships may be derived which involve not only the valence of the solid but also the (atomic) anion and cation electronegativities x_X , x_M . Moreover they mediate new aspects of the nature of the heteropolar bond in general, and of real AgBr in particular. These findings prove furthermore of great interest for the spark plasma theory in that they deliver an explanation for the charge centre shift δ_{MX} of atomic ions hitherto not understood. This may be taken as a proof that the oscillator ground state quasiparticle in the collective plasma phase is of composed nature, becoming decomposed by some kind of tunnelling effect. Finally the present image provides a qualitative understanding of the multipositive ion spectra of Cu- and Cd-doped AgBr when compared with lifetime results.

II. Phenomenological model of the interaction equivalence

We must now discuss the circumstances under which the δ -shift occurs. One of the most conspicuous aspects of the stereoplasma is its detailed solid-state 'memory'. In other words: the detailed chemical bond forms the very complicated *randbedingung* of the stereoplasma dynamics. Even in the case of quasiideal solids this bond is strictly speaking unknown. Real nature imperfectness, of course, rather adds to the problem than diminishes it. Whatever the true situation of the electron distributions in the real solid, it will result in an apparent over-all charge distribution of the detected ionic species, and represent a complex picture of the solid viewed with the 'glasses' of the dynamics. Luckily, the stereoplasma has lost much of the horror it had a few years ago. Even if we can hardly hope to determine reliably the quantum-chemical weight of the individual oxidation states, we get, at least, a reasonable average insight into most bond properties. In rather rare instances, yet, the experiment delivers outstanding information about the relative behaviour of individual oxidation states. Such detailed information may become of great importance for the understanding of structural transitions and critical phenomena in general.

Definitions

In order to improve the clearness in what follows it appears indispensable to define and comment briefly the most important quantities used. We shall only investigate dielectric solids of the simple type MX crystallizing in the sodium chloride structure B_1 . M = cation, X = anion. $MX = M^{v_1}X^{-v_1}$. v_1 is the formal 'spark valence'. Formal means, that v_1 is taken as an integral or half-integral number. v_1 may be determined roughly by the plasma valence effect based upon matrix or impurity complex ions [6, 12]. The spark valence is identical with the usual formal valence if v_1 is integral. For the sake of simplicity we assume equal and opposite charges on the cation and the anion.

v is the detected (apparent) degree of ionization of an ion. v may either be a true crystalline oxidation state or such a state altered by (dynamic) ionization or electron capture. If the δ -shift is small, or may even be neglected, the detected major ion charge represents ideally the plasma oscillator ground state which in this case coincides with the corresponding major crystalline oxidation state. If v is the smallest oxidation state realized by the chemical bond, ions which carry the charge $v - 1$ are said to be due to plasma electron capture (some kind of a resonance process in the solid). If on the other hand v is the highest oxidation state, ions carrying the charge $v + 1$ are said to have become singly ionized insofar they belong to v , and doubly ionized insofar they belong to $v - 1$. In the majority of crystals, as a rule, one oxidation state is prevailing. In many rare earth chalcogenides, yet, second and third oxidation states may be comparably strong, while the fourth is almost negligible. We then attribute to these compounds the formal spark valence $3/2$. Eu- and Yb-chalcogenides are clearly bivalent ($v_1 = 2$), and the rare earth pnictitides are clearly trivalent ($v_1 = 3$). But v_1 may not only differ from an integral value but also vary in a complicated way when suitably doped [12].

ξ is the average apparent degree of ionization per valence unit ($v_1 \xi$ may be any non-integral number $>$ average crystalline oxidation state) = sum of the average crystalline oxidation state and the average dynamic ionization, the sum being divided by v_1 . As only positive ions are obtained ξ can be determined reliably only (i) for the cation, and (ii) for solids having a formal valence $v_1 > 1$. $c^v = c_{M^v}^{(v_1)}$ is the concentration of the cation of charge v from a solid of valence v_1 .

$\delta_{MX}^{(1)} = \delta_{MX}^{(v_1)}/v_1$ is the δ -shift per valence unit, i.e. the single-bond shift of an atom having valence v_1 ; $\delta^{(1)}$ represents a normalized measure of the (heteropolar) bond strength.

$I(\Delta x)$ is the δ -shift active single-bond ionization energy. $\Delta x = \Delta x_{MX} = x_X - x_M$ is the Pauling (anion-cation) electronegativity difference.

$K_{M,X}$ is the (negative) slope of the normalized (multipositive) M or X ion intensities as determined from the natural logarithmic intensities c^v as a function of v ; numerically it holds $K > 0$. Very often the first degree of ionization is not usable for the determination of K for reasons of the chemical bond (in metals one electron is almost fully delocalized, and in heteropolar compounds one electron is often removed from the cation to the anion). On the other hand the first degree of ionization must, of course, be used for normalizing $\{c^v\}$.

T_c is the critical temperature for Bose-Einstein condensation as it follows from the gas degeneration of (indistinguishable) particles obeying Bose statistics. T_c holds both for true electron pair bosons and exciton like electron-hole bosons in the approximation $2m_e^* = m_e^* + m_h^* = 2m_e$ (m_e = electron rest mass). By 'condensation'

we mean in fact that more than only one particle may occupy the same space. It is almost certain that the multiply excited oscillator states represent such a condensation. This is not too strange as the excitations in this case have a strong exciton aspect. It is not certain but probable that also the electron pair ground state bosons represent a condensed state in the short period which precedes the establishment of the oscillator equilibrium. In the equilibrium, of course, one atom has at most one particle in the ground state. I , E mean the atomic (first) ionization potential and electron affinity, respectively.

M is the Mulliken electronegativity: $M = \frac{1}{2}(I + E)$; (try not to confuse with $M = \text{cation}$). sW is the specific (single pair) probability frequency for successful dissociation, i.e. the probability that, by one step, a $\delta_{MX}^{(1)}$ shift ionization occurs; if there occur in succession ν_1 steps the process results in a total shift $\delta_{MX}^{(\nu_1)}$.

$\lambda_p = 1/\tau_p$ represents the (order of magnitude) collective stereoplasma interaction rate. τ_p means the average lifetime of the stereoplasma, i.e. the period between the onset of the electron impact and the end of the short-range coherence interaction. In the plasma molecule formation model [5] we used $\tau_p = \tau_1$; after the discovery of the plasma $/e^+$ lifetime correlation we must be more careful as τ_1 in the e^+ lifetime language means the first, shortest lifetime component. p is intended to remind of 'plasma').

A^0 , A'' , a'' stand for empirical constants (or are at least weak functions).

Distinguishable ionization mechanisms

For merely heuristic reasons it is advisable to distinguish between two important ionization mechanisms: (i) the δ -shift ionization, and (ii) the oscillator-bound ionization. It is now becoming more and more obvious that in both mechanisms atomic flux quantization must be involved. While in the δ -shift the quantization of electric currents occurs on the slightly extended valence orbitals, the second mechanism involves current quantization on strictly localized atomic orbitals. Insofar as any shell structure details may be neglected the radii of the flux trapping atomic orbitals are nearly identical. The radius which belongs to the circular atomic orbital coincides in the case of metallic elements essentially with the X-ray effective crystalline radius r_x . In the case of dielectric solids things are more complicated on account of the heteropolar charge distributions [9, 10]. Furthermore we have to attribute the same dimensions to oscillators being in different states of excitation. This implies that the n -fold excitation effectively means n simple excitations in complete accordance with the corresponding situation of the black body radiation where the quantity n of the photon energy $n\hbar\omega$ has to be interpreted as the number of light quanta with energy $\hbar\omega$ [14]. This means, on the one hand, that the spatial description of the simple harmonic oscillator for multiple excitation does no longer hold, and, on the other hand, that several particles may coexist in the same space. The plasma atom viewed as a harmonic oscillator is the statistical picture of the excited atomic entity having well defined dimensions and being in collective (chemical) contact with all the rest of the crystal during the lifetime τ_1 . In this picture the majority of the atoms are in the ground state, characterized by a certain zero-point energy.

As the ground state wave function has no node this state is, according to the oscillator picture, strictly non-ionizative, while the excited states are strictly ionizative. In other words: the ground state quasiparticle appears as ideally non-dissociable. On

the other hand there are the slightly extended δ -active valence orbitals which have an ionization energy given by the average strength of the heteropolar bond. The ionization energy associated with the δ -shift is, of course, smaller than that one of the oscillator excitations, as the valence orbitals are larger than the atomic orbitals. It is important to note that in the ionization picture the role of the spin does not explicitly come under discussion for both mechanisms (at least we may do so as if it did not exist). For the discussion of the chemical bond strength, as reflected by the δ -shift, we prefer the electron-electron aspect. The quasiparticle is then describable as a (covalent) pair of electrons with their spins opposed (total spin $S = 0$), i.e. a quasiparticle in the singlet state. If the spins are perfectly antiparallel (i.e. to the extent allowed by the uncertainty principle) the single-electron component of the quasiparticle is ideally zero. Such a particle is assumed to represent the oscillator ground state of the plasma atom in the case of an ideally perfect solid. In the case of the valence orbitals the spins are still opposed although not perfectly. The difference is a result of the finite bond strength. In this case the single-electron component is not zero. The less heteropolar the bond the larger the component. It is just this component which by virtue of its 'metallic' character makes the Fermi-hole become of increasing importance. Thus, an increased δ -shift is equivalent to saying that the electron pair presents to an incoming electron a larger ionization cross section on the one hand, and to an incoming positron an (equivalent) annihilative cross section on the other hand. It is in this way also we understand the real nature influence of any electron perturbations when describing them as a trend toward a more 'metallic' state. It is clear that the effective coupling is a result of the many-body interactions of all electrons which belong to the valence shell quanticle given by the magnitude of the plasma active flux quantum, which, just as in ordinary superconductivity, is found to be half the classical (London) flux quantum $\phi_p = \frac{1}{2}(h/e)$. For δ -shift ionization, as well as for the oscillator ionization, we fortunately need not make any precise explicit statement as to the role of the spin in both electron-electron and electron-hole pictures. Of course, the spin is taken into account implicitly in both the Eggert-Saha equation and the Bloch theorem which warrants the coexistence of Bose and Fermi systems [15].

Empirical δ -shift relationships

The investigation of dielectric solids leads to the simple (approximate) result that the δ -shift per valence unit (5) may be described by the single-bond electronegativity difference Δx in the following way:

$$\delta_{MX}^{(1)} = A''(a'' - \Delta x). \quad (7)$$

Furthermore, as a result of the finite coupling strength of the valence electron pairs the charge pattern simply appears to undergo a parallel shift toward higher degrees of ionization by the total amount $\delta_{MX}^{(v1)}$. That we have in fact to do with a parallel shift is convincingly proved by the (average) constancy of the cation slope K_M for all solids of a series MX (M fixed!) while at the same time δ varies considerably. Owing to the validity of the Pauling electroneutrality principle, which looks fairly safe in the case of MX compounds [16], the δ -shift makes itself felt primarily in a change of c^{+2} (the electroneutrality principle states that even for the strongest heteropolar bond the effective cation charge is at most one electron charge, which fact appears well confirmed by the plasma polarity analysis [12]. Therefore it holds approximately

$$\Delta \ln \left(c^{+2} / \sum_v c^v \right) = K_M \delta_{MX}^{(v_1)} \quad (8)$$

where by definition $-K_M = \ln (c^{v+1}/c^v)$ for degrees of ionization $v \gtrsim v_1 + 1$. By definition we have

$$\delta_{MX}^{(1)} = \xi - \xi_1$$

where δ varies in a range $0 \lesssim \delta_{MX}^{(1)} < 1$. In fact, for all investigated valences $v_1 = 2; 3/2; 3$; $v_1 \xi < 2$ has been found. ξ_1 corresponds to the absolute shift of the most heteropolar compound of the series. As a rule ξ_1 is small and only difficultly determined. We repeat once more that the empirical relative-shift quantity δ has to be considered as semiquantitative, one reason being the lack of information about c^0 and c^{-1} (multiply negative species have never been observed in spark mass spectra). From the fact, that in the weakly heteropolar bond δ is no longer a small quantity we conclude that locally beginning 'metallic' degeneracy will become important. Of course, this trend toward a δ -shift active degeneracy must not be confused with the Fermi gas degeneracy of the metal. In fact, the action of the truly metallic electrons manifests itself in the plasma process by an increased recombination tendency. It is therefore difficult to observe a δ -shift in the case of metals.

Physical approach to δ

A large δ means a large 'free electron' concentration n_e . We confine our analysis to the case of a classical non-degenerate electron gas, that is for $\delta^{(1)} \ll 1$ where the Saha equation is a meaningful approximation. By introducing the specific concentration per valence unit we use the Saha equation in the simple form:

$$\frac{sC^{+2}}{sC^{+1}} = \mathcal{H} \exp(-I(\Delta x)B). \quad (9)$$

Here we have assumed that the first oxidation state is prevailing. Furthermore we neglect the role of second and third dynamic ionizations. This shall still be true for decreasing Δx . c^{+1} is, therefore, assumed dominant, although the experiment is strictly not able to show it. $B = 1/k_B T_c$, where k_B is the Boltzmann constant. The modern version of the Eggert-Saha equation gives for $\mathcal{H}$,

$$\mathcal{H} = \frac{1}{n_e} \frac{g_{2,0}}{g_{1,0}} g_e \frac{(2\pi m_e k_B T_c)^{3/2}}{h^3} \quad (10)$$

where the ordinary lattice temperature T has been substituted by the quantum temperature T_c . It goes without saying that in (10) we are using the electron mass, as the formula deals with fermions. $g_{2,0}, g_{1,0}$ are the statistical weights of the M^{+2}, M^{+1} ions in the ground state. We shall not bother about optically excited states, $s \neq 0$. $g_e = 2$ corresponds to the two spin orientations of the free electron. We shall now assume that the δ -shift free electron (fermion) gas and the quasiparticle (boson) oscillator gas are in quantum thermal equilibrium. Such an equilibrium – if it ideally exists – is warranted by a theorem of Bloch [15] which provides the coexistence of ideal gases belonging to the two different kinds of quantum statistics: Fermi-Dirac statistics and Bose-Einstein statistics. The phonon system appears at the same time to be rather

little affected by the impact associated phenomena. There is nothing such as a thermal equilibrium between the system of the 'nuclei' and the electronic system. This follows conclusively from the phenomena of plasma induced phase transitions [12, 17]. The critical temperature T_c is obtained from the theory of gas degeneration and has in the approximation of free particles the value

$$T_c = \frac{h^2}{2\pi(2m_e)k_B} \left(\frac{n_M}{s\zeta_{3/2}(1)} \right)^{2/3} \quad (11)$$

where we approximate the boson mass by $2m_e$, and n_M signifies the cationic particle density; n_M must be used in consequence of the fact that the electrons prefer to go to the positive cations. n_M corresponds to n^* in the e^+ case (2,3). $s = 1$ is the multiplicity corresponding to the spin $S = 0$ singlet state. T_c as given by (11) is a typical boson particle result. Its use stands in logical agreement with the pair nature of the quasi-particles which constitute the harmonic oscillator. Furthermore it would be difficult to interpret the results of the improved polarity analysis by another form of T_c than that one given by (11). The form (11) is also independently suggested by a variety of plasma effects such as the lattice constant correlation [13], the molecule formation [5] and so on. The fact of beginning degeneracy means that the degeneracy parameter of the δ -active Fermi gas comes to lie somewhere near 1, thus n_e becomes of the order of $n_e \sim 2(2\pi m_e k_B T_c)^{3/2} h^{-3}$. By comparison of n_e with (10) follows immediately $\mathcal{H} \approx 1$. This is a rather late justification of the early finding [12] that multipositive atomic ions may simply be described by a Boltzman distribution. It also confirms – though from a quite unexpected side – that δ -shift ionization and oscillator-bound ionization must finally rest on the same physical cause, i.e. atomic flux quantization. T_c is of the order of 10–30 000 K while the lattice temperature remains somewhere above room temperature. We approximate now (9) by

$$\frac{{}^s C^{+2}}{{}^s C^{+1}} = \exp(-I(\Delta x)B) \quad (12)$$

(12) may be interpreted as the probability ${}^s W = W^{(1)}$ for successful δ -shift active ionization of a single valence electron pair. According to the law of independent probabilities the integral δ -shift of a v_1 valent atom has a probability

$$W^{(v_1)} = \prod_{v_1} W^{(1)} = {}^s W^{v_1} = \exp(-v_1 I(\Delta x)B). \quad (13)$$

$W^{(v_1)}$ corresponds to $c^{+2}/\sum_v c^v$ in (8). This result leads immediately to the magnitude of the ionization energy $I(\Delta x)$; by using (13) and (7) in (8) we obtain:

$$\Delta \ln W^{(v_1)} = \Delta(-v_1 I(\Delta x)B) = K_M v_1 A''(a'' - \Delta x). \quad (14)$$

Now, as n_M , and thus also T_c and B , for a series MX (M fixed) remain constant, we may write (14) in the simple form

$$-v_1 B(I(\Delta x) - I(a'')) = K_M v_1 A''(a'' - \Delta x) \quad (15)$$

where $I(a'')$ corresponds to the case of perfect coupling, $\Delta x_{\max} = a''$. Thus it follows from (15) that in the general case $I(\Delta x)$ has the form

$$I(\Delta x) = \frac{K_M A''}{B} \Delta x = \frac{K_M A''}{B} (x_X - x_M). \quad (16)$$

But $K_M/B = K_M k_B T_c = \hbar \omega_M = I_{\varepsilon^*}$ is just the atomic orbital ionization energy [10].

Therefore it follows, because for all physical cases holds $A''(x_X - x_M) < 1$,

$$I(\Delta x) < I_{\varepsilon*}. \quad (17)$$

The important result (17) implies, as $I_{\varepsilon*} = \hbar^2/m_e r^2$, that the δ -shift is due to the ionization of orbitals which are slightly larger than the atomic orbitals; such orbitals are the valence orbitals of radius $r_v > r \simeq r_x$ [9].

The Pauling electronegativity is connected with that one of the Mulliken scale by $x = 1/2.78 M = 1/2.78 \cdot \frac{1}{2}(I + E)$. One easily recognizes that x is in fact proportional to the free atom ionization energy. Therefore $I(\Delta x)$ has the meaning of a reduced average ionization energy. $I(\Delta x)$ is considerably smaller than the atomic value itself; this can be taken as another manifestation of the fact that also the δ -shift is a true solid state event. It again points to the fact that the ionization must precede the disintegration of the solid. The limiting case $\Delta x \rightarrow 0$ is physically not meaningful, as $\Delta x \rightarrow 0$ in our approximation simply means a vanishing heteropolarity. But for Δx small we are in the covalent regime which has not been taken into account.

We may now define the differential ionizative δ -shift active interaction rate for the v_1 valent case in the obvious form

$$d\lambda^{(v_1)} = v_1 d\lambda^{(1)}$$

$$v_1 d\lambda^{(1)} = \frac{1}{\tau_p} \frac{dW^{(v_1)}}{W^{(v_1)}} = \frac{1}{\tau_p} d \ln W^{(v_1)}$$

$$d \ln W^{(v_1)} = -v_1 K_M A'' d(x_X - x_M) \quad (18)$$

or, as for M fixed, follows $dx_M \simeq 0$ we obtain

$$d\lambda^{(v_1)} = - \frac{v_1 K_M A''}{\tau_p} dx_X \quad (19)$$

which results, slightly arranged, gives

$$\frac{1}{v_1} \frac{d\lambda^{(v_1)}}{d(-x_X)} = \frac{K_M A''}{\tau_p} = \text{const.} \quad (20)$$

Equation (20) is the perfect counterpart of the differential positron interaction rate change (5)

$$\frac{1}{v_2} \frac{d\lambda^{(v_2)}}{dx_M} \simeq A^0 = \text{const.} \quad (21)$$

The comparison of (21) and (20) shows that the interaction rate increase for e^+ annihilation in a system $M^{v_2} X^{-v_2}$ (where X is fixed) comes about by an increase of the cationic electronegativity, while in a system $M^{v_1} X^{-v_1}$ (where M is fixed) the interaction rate increase for δ -shift Fermi hole ionization comes about by a decrease of the anionic electronegativity. In both cases means $d\lambda > 0$ a differentially reduced heteropolar bond. It is interesting to note that the rate change is governed by the respective bond change of that crystal ion which carries the same charge as the impacting particle. This indicates once more the importance of the integral cation-anion valence shell quanticle [1, 2, 5].

Comparison of the interaction rates

Both for body-centered cubic (A_2) and face-centered cubic (cubic close packing) (A_1) structures K is of the order of 2: in fact $^{A_2}K = 2,028$, $^{A_1}K = 1,917$, and for sodium chloride structure (B_1): $^{B_1}K = 2,415$ [9]. $A'' \simeq 0,2$ and $\tau_p = \tau_1 \simeq 2 \cdot 10^{-10}$ s [5]. Thus it follows

$$\frac{K_M A''}{\tau_p} \simeq (2,4 \pm 2) 10^9 \text{ s}^{-1}.$$

For A^0 we found previously $A^0 \simeq (3 \pm 1) 10^9 \text{ s}^{-1}$ (Table 1).

Therefore, we obtain the interesting result that annihilative e^-e^+ and ionizative e^-e^- interactions do not only depend in a symmetric way on the heteropolar bond but also have nearly equal strength. We may, therefore, call them quasiequivalent and express this result by the approximate relationship

$$\frac{1}{v_1} \frac{d\lambda_{e^-e^-}^{(v_1)}}{d(-x_X)} \simeq \frac{1}{v_2} \frac{d\lambda_{e^-e^+}^{(v_2)}}{dx_M} \simeq \text{const} \quad (22)$$

obtained from corresponding series of compounds

$$M_{\text{fixed}}^{v_1} X^{-v_1} \quad \therefore \quad M^{v_2} X_{\text{fixed}}^{-v_2}$$

where the constant has the appropriate value $(2-3) \cdot 10^9 \text{ s}^{-1}$.

Interpretation of the λ -shift laws

We will now treat the relationship between the two e^+ -shift laws. It is clear that (4) and (5) only represent different possible aspects of one and the same physical quantity. Equation (5) reflects a purely phenomenological point of view, while (4) partly takes into account the spin pair coupling and partly the electron density. (4) integrated gives simply

$$\lambda(x) = \lambda_0(x) + A' v_2 (a' - x_X) n^* \quad (23)$$

where $\lambda_0(x) \simeq \text{const.}$ cannot be further discussed, since our analysis is of a relative nature only. By comparison of (23) with (2) we recognize that $v_2(a' - x_X)$ adopts the valence- and coupling-dependent role of the enhancement factor more commonly used in the case of metals [1, 2]. The empirical quantity A' compares with $\xi' c_D$. We obtain $\xi' = 6.7-7.3$, which agrees excellently with the value $\xi' = 6.09$ of Reference [3] and emphasizes once more the importance of the valence shell quanticle. Increasing valence effectively diminishes the spin coupling, thus increasing the interaction rate.

If we differentiate (23) with respect to x_X , it follows

$$\frac{1}{v_2} \frac{d\lambda}{d(-x_X)} = A' n^* \simeq A^0. \quad (24)$$

We see that (24) amalgamates effectively with (5) because, if we put $dx_M = -dx_X$, the same $d\lambda$ results. From $\lambda_0(x) = 1/\tau(x) \simeq 0.195 \cdot 10^{10} \text{ s}^{-1}$ follows that with $\tau \simeq 500$ ps the absolute lifetime of annihilative and ionizative processes are comparable.

Finally, (24) has also a trivial interpretation which becomes evident when it is compared with the δ -shift result (20): a spark experiment effected on antimatter would lead to a negative δ -shift, while K_M would then be $K_{\bar{M}}$. The impact of an electron, finally would lead by annihilation to a negative 'hole' in the antimatter.

Conclusion

On the basis of the analysis of the (positron) λ -shifts and the (electron) δ -shift we could show that annihilative e^-e^+ and ionizative e^-e^- interactions are fundamentally related with regard both to symmetry and to strength. The associated relationships are of very complex nature: it is found that the elementary flux quantum acts as a universal connecting link. The basic physical reason for this fact is given by the simultaneous facts of the quantization of action in units h on the one hand and the charge quantization in units e on the other hand. Both for e^+ annihilation and spark plasma ionization, charge processes play an important part. The interaction equivalence – as demonstrated on the example of the λ -shifts and the δ -shift – is able to reveal a basic relation, which finds further confirmation in the real nature-bound e^+ interaction rate increase and molecule formation increase in the case of AgBr. On the other hand the equivalence is in fact connected with the phenomenon of the plasma oscillator. In all cases it is flux quantization on the atomic scale which acts as the mediating process. Through the magnitude of the flux quantum itself the importance of boson-like quasiparticles becomes manifest. But only by the deeper insight into the mechanism of the harmonic plasma oscillator we may realize (i) that the parapositronium has a perfect analogue in the short-lived excited oscillator state describable by an electron-hole exciton, and (ii) that the ortho-positronium finds an analogue in the long-lived oscillator ground state. Finally we perhaps ought to mention that part of the results in this work have been presented on positron conferences [18].

It is a pleasure to thank Professor G. Busch for his continuous support, Dr. H. Surbeck for an intensive and fruitful collaboration, Mr. R. Hauger, Drs. F. Hulliger and E. Kaldis from this laboratory, Dr. P. Junod from Ciba-Geigy, Photochemie, Fribourg, and Dr. E. Schönherr from the MPI, Stuttgart, for supplying many of their crystals.

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Electron-electron contributions to the scattering frequency in cadmium as measured in radio frequency size effect experiments

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Lawrence and Wilkins [1, 2] have calculated that Cd should have one of the largest electron-electron scattering coefficients of the polyvalent metals considered: $\rho_{ee}/T^2 \sim 5 \times 10^{-14} \Omega \text{cm K}^{-2}$. We have made sensitive, tilted field RFSE measurements of the limiting point scattering frequency $\bar{\nu}_k(T)$ for $\mathbf{k}$ near $\langle 0001 \rangle$, on the nearly free electron-like region of the 3rd band lenticular Fermi surface of Cd. Below ~ 2 K the logarithm of the RFSE amplitude, $\ln A(T) \propto \bar{\nu}(T)$, is found to vary as T^2 . From ~ 2 –4 K, $\ln A$ is approximately proportional to T^3 , reflecting the normal (intra-band) electron-phonon contribution. At higher temperatures $\ln A$ varies more rapidly than T^3 , indicative of inter-sheet scattering [3] resulting from enhanced coupling to transversely polarized phonons. The (large angle) electron-electron scattering is completely effective and presumably the electron-phonon scattering is too, since if it were ineffective, a T^5 followed by a T^3 temperature dependence would have been observed. Depending on the explicitly assumed temperature dependence for the electron-phonon scattering contribution used to extrapolate to $T = 0$ [4], the coefficient of the T^2 contribution ($\propto T^{-2} \ln(A(0)/A(T))$) is:

$$\nu_{ee}/T^2 = (6\text{--}22) \times 10^6 \text{ s}^{-1} \text{ K}^{-2} \pm 50\% \quad (\text{RFSE})$$

where we have used the free electron Fermi velocity $1.6 \times 10^6 \text{ m s}^{-1}$.

The theoretically expected value for Cd is enhanced by a factor $(1 + \Delta)/\Delta \sim 3$ (see reference 1, 2) over the value for the bulk resistivity since the RFSE is sensitive to both normal and Umklapp processes and is:

$$\nu_{ee}/T^2 = 4 \times 10^6 \text{ s}^{-1} \text{ K}^{-2} \quad (\text{Lawrence, Wilkins}).$$

In view of the assumption of nearly free electrons throughout (except for the calculation of Δ), the agreement between experiment and theory is reasonable. This constitutes the first observation of electron-electron scattering in a non-transition metal using the FRSE [5].

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Positronenannihilation in stromdurchflossenen Halbleitern

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Die Positronenannihilation in den Halbleitern Germanium und Silizium wurde mit den verschiedensten Methoden recht detailliert untersucht. Die stark anisotropen Winkelverteilungen bei der 2γ -Annihilation ([1], [2]) können mit dem independent-particle-Modell nach Stroud und Ehrenreich [3] gut interpretiert werden. Shulman et al [4] konnten zeigen, dass bei 2γ -Winkelkorrelationsexperimenten zwischen hochdotierten und intrinsischen Proben kein Unterschied festgestellt werden kann. Die Beweglichkeit der Ladungsträger in Ge und Si wurde von vielen Autoren sehr gründlich untersucht. Auch über die Beweglichkeit von Positronen in diesen Halbleitern liegen Informationen vor. (Für Silizium: Brandt und Paulin [5], für Germanium: Mills und Pfeiffer [6]); die letzteren sind in der Regel um einen Faktor 5–10 kleiner als die der Ladungsträger.

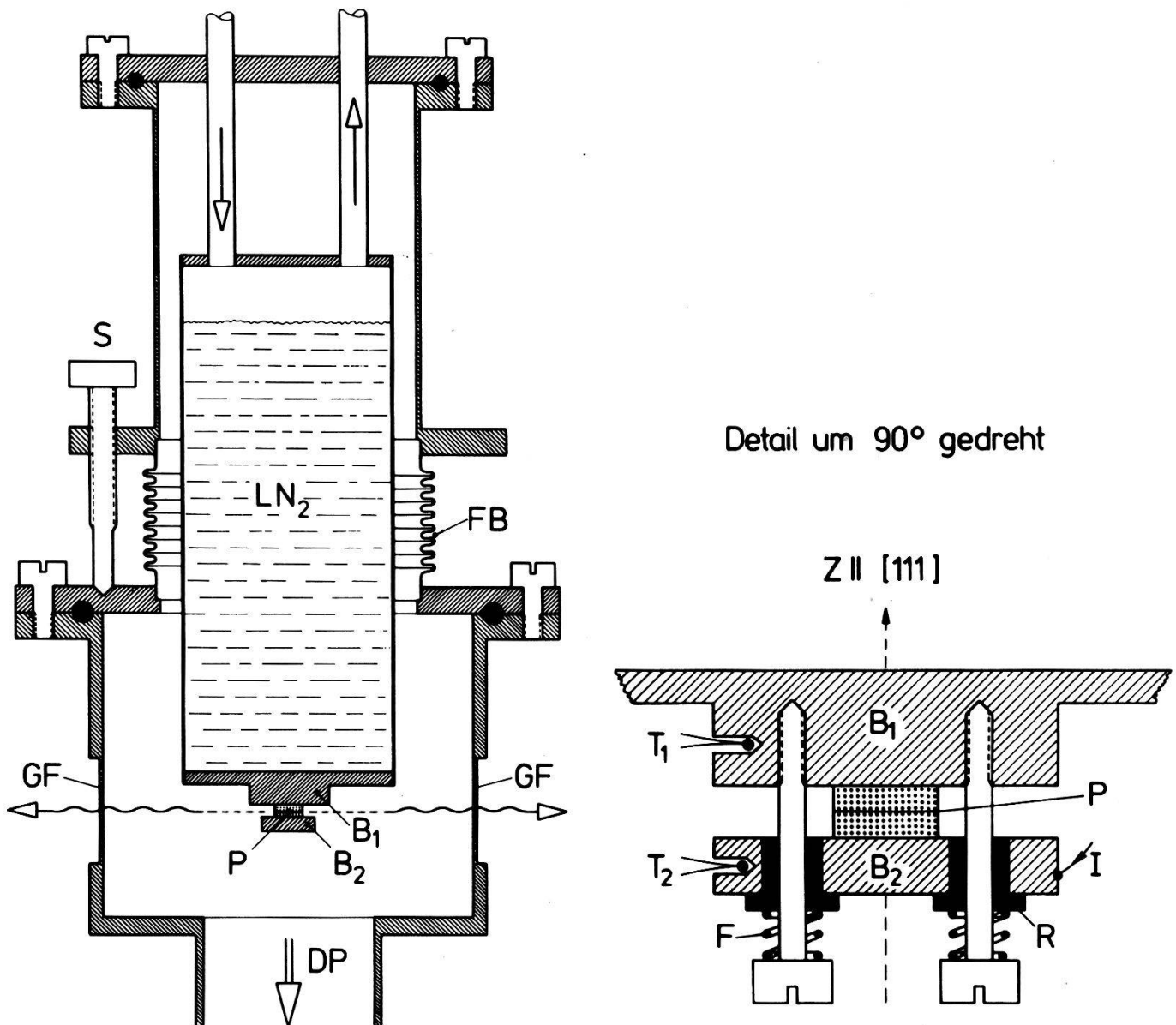
Umso mehr überraschte eine kürzlich erschienene Publikation von Rozenfeld et al [7], in der berichtet wird, dass die 2γ -Winkelkorrelationsfunktion drastisch asymmetrisch wird, wenn an p-Siliziumproben ein elektrisches Feld senkrecht zur Quantenemissionsrichtung angelegt wird. Die Deformation ist am ausgeprägtesten bei einem Korrelationswinkel $\theta = p_z/mc \simeq 3\text{--}4$ mrad. Die Zentroidverlagerung ist so gerichtet, als wäre der Schwerpunkt des annihilierenden Elektron-Positronpaares negativ geladen. Selbst bei extremsten Annahmen über die applizierten Feldstärken und über die Beweglichkeiten wäre eine Asymmetrie zu erwarten, die um einige Grössenordnung kleiner ist, als die hier berichtete. Eine plausible Erklärung für diesen Effekt wird nicht geboten.

Wir haben versucht, diesen merkwürdigen Effekt unter günstigeren experimentellen Bedingungen zu reproduzieren. Als Proben wurden sehr reines Germanium (ca. $40\ \Omega\cdot\text{cm}$) und n-dotiertes Silizium (ca. $120\ \Omega\cdot\text{cm}$) verwendet. Die Probenanordnung im Innern eines N_2 -Kryostaten ist in Figur 1 dargestellt.

Die Proben ($8 \times 8 \times 2.5$ mm) sind so orientiert, dass die [111]-Achse senkrecht zur γ -Emissionsrichtung steht. Bei der Präparation ohmscher Kontaktflächen hat uns Herr Dr. R. Jaggi (IBM-Zürich Research Laboratory, Rüschlikon) in dankenswerter Weise beraten. Zwischen den Probenstücken P befindet sich eine ca. $10\ \mu\text{m}$ dicke ^{64}Cu -Positronenquelle. Durch diese Sandwichgeometrie wird eine optimale Positronenausbeute erzielt. Der Beitrag von Annihilationsprozessen im Quellenmaterial beträgt

$$P_Q = \alpha \frac{1 + r}{1 - r(1 - 2\alpha)} \simeq \begin{cases} 23\% \text{ (Ge)} \\ 19\% \text{ (Si)}, \end{cases}$$

wobei α die Positronenintensitätsschwächung in der Cu-Folie und r der Rückstreuoeffizient der Positronen am Probenmaterial [8] ist. Für jeden Korrelationswinkel wurde in rasch aufeinanderfolgenden Messungen (um Drifts weitgehend auszu-

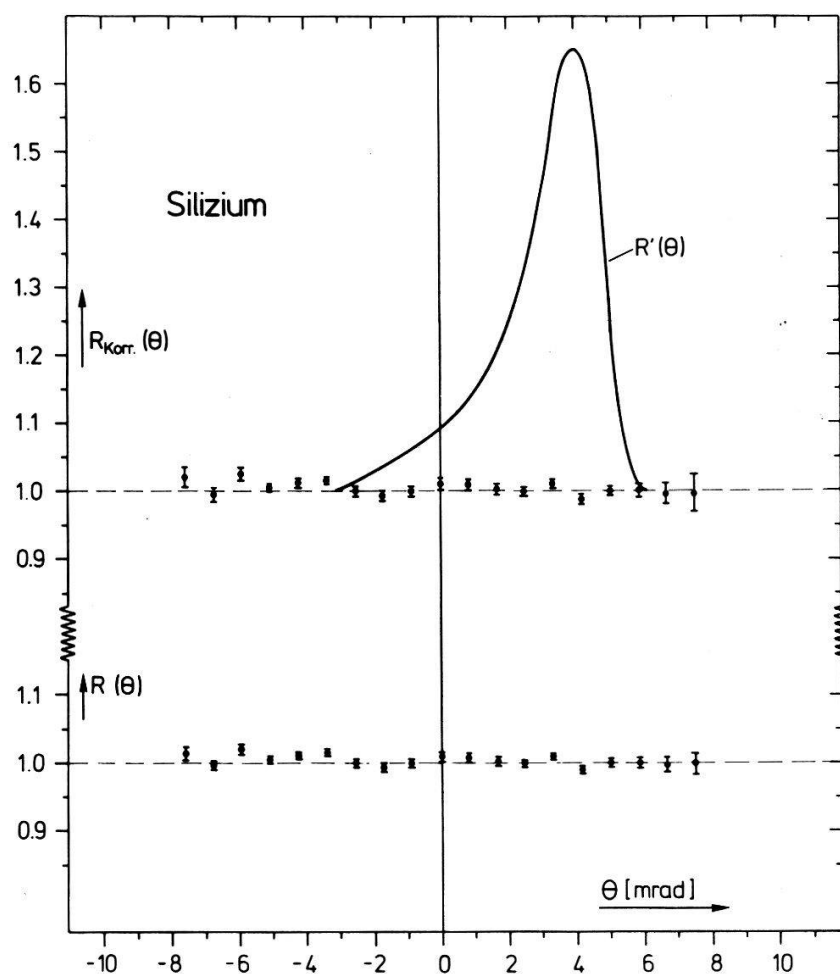


Figur 1

Probenanordnung: LN_2 = flüssiger Stickstoff, P = Sandwich Probe-Quelle-Probe, B = Kupferbacken, FB = Federbalg, S = 3 Justierschrauben, GF = Gammafenster, DP = Diffusionspumpe, T = Thermoelemente, R = Isolatoren, I = Stromzuführung, F = Spannfedern.

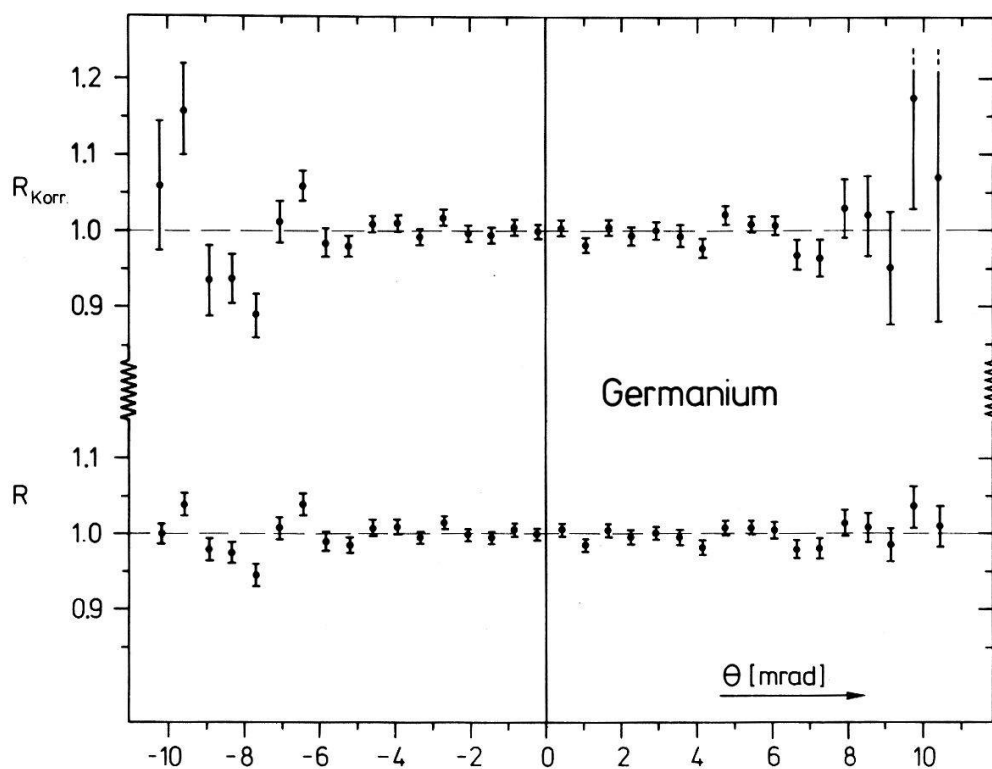
schliessen und um die Korrektur bezüglich der Halbwertszeit von ^{64}Cu [9] klein zu halten) das Verhältnis $R(\theta)$ der Koinzidenzraten mit bzw. ohne elektrisches Feld bestimmt. In den Figuren 2 und 3 sind die Messresultate für Silizium und Germanium dargestellt; die Grösse $R_{\text{Korr}}(\theta)$ beinhaltet die Korrekturen für Annihilationen im Quellenmaterial, die vor allem bei Germanium zu beträchtlichen Unsicherheiten bei grossen Winkeln führt.

Eine Abweichung des Verhältnisses $R_{\text{Korr}}(\theta)$ vom Wert 1 kann innerhalb der insbesondere für Silizium sehr engen Fehlergrenzen nicht festgestellt werden. Die Symmetrie der Korrelationsfunktionen wird durch die vorliegenden Messresultate innerhalb von 0.3% bestätigt. Die in Figur 2 eingetragene Kurve $R'(\theta)$ nach Rozenfeld et al [7] ist auf gleiche Stromdichten bezogen und zwar aus folgenden Gründen. Die Angaben dieser Autoren über die elektrischen Eigenschaften ihrer Proben und über die applizierten Feldstärken sind widersprüchlich. So ist z.B. ein Resistivitätswert



Figur 2

$R(\theta)$ und $R_{\text{Korr.}}(\theta)$ für Si in Orientierung [111] bei 79 K und $j = 47 \text{ mA/cm}^2$. Die eingezeichnete Kurve $R'(\theta)$ repräsentiert die Messresultate nach [7] bezogen auf gleiche Stromdichte.



Figur 3

Messresultate für Ge bei $j = 300 \text{ mA/cm}^2$.

($1.44 \cdot 10^6 \Omega \cdot \text{cm}$) angegeben, der weit über dem Wert für intrinsisches Si liegt und überdies im Widerspruch zur angegebenen Löcherkonzentration steht. Ferner wird angegeben, dass die Kontaktierungsflächen ohmisch sind und zwar unabhängig von der Art des aufgedampften Kontaktierungsmetalls; das dürfte – insbesondere bei hochohmigem Material – kaum möglich sein. Daher muss angenommen werden, dass der angegebene Wert für die Feldstärke (10^4 V/cm !) unrealistisch hoch ist. Darum haben wir uns auf den Wert für die Stromdichte (ca. 10 mA/cm^2 , private Mitteilung) bezogen.

Es ist zu vermuten, dass bei den Untersuchungen nach [7] eine starke Wärmeentwicklung bei den Kontakten auftrat, die zu einem Eindiffundieren des Kontaktmaterials während der Messung und dadurch zu einer tiefgreifenden Veränderung des Probenmaterials führte.

Herrn. R. Spring danken wir für technische Hilfe bei diesen Messungen und dem Schweizerischen Nationalfonds zur Förderung der wissenschaftlichen Forschung für die finanzielle Unterstützung dieser Arbeit.

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Bestimmung der Bildungsenergie für Monovakanzen in Blei mit Hilfe der Positronenannihilation

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Aus der Temperaturabhängigkeit charakteristischer Observabler bei Positronenannihilationsexperimenten kann die Bildungsenergie Q_v von Vakanzen in Metallen bestimmt werden. (Zusammenfassende Darstellung vgl. [1]). Geeignete Observable sind z.B. die mittlere Lebensdauer der Positronen, die Intensität der langlebigen Komponente im Lebensdauerspektrum, die Dopplerbreite der 511 KeV-Annihilationsstrahlung und die sogenannte 'Peakhöhe' $\hat{N}(T)$, d.h. die Koinzidenzrate bei 2γ -Winkelkorrelationsexperimenten beim Korrelationswinkel $\theta = p_z/mc = 0$ [2]. Die Methodik der Positronenannihilation zur Bestimmung von Defektbildungsenergien bietet eine Reihe von Vorteilen: Die Messungen erfolgen im thermodynamischen Gleichgewicht der Defektkonzentration, d.h. die Vorgeschichte der Probe spielt eine weniger wichtige Rolle als bei Nicht-Gleichgewichtsexperimenten (z.B. Einfrieren der Defekte durch Abschrecken); die Defektkonzentration kann über einen sehr grossen Temperaturbereich verfolgt werden, und die maximale Empfindlichkeit der Methode liegt in der Regel relativ weit unterhalb des Schmelzpunktes; dadurch kann

in günstigen Fällen zwischen Monovakanzen und Vakanzagglomeraten unterschieden werden.

Im Rahmen einer Studie über die Bildung von Vakanzen im Legierungssystem Tl-Pb haben wir eine Reihe von Messungen der Bildungsenergie für Monovakanzen in Pb nach der Peakhöhenmethode vorgenommen. Eine relativ starke Zunahme von $\hat{N}$ mit der Temperatur verbunden mit einer Abnahme des Annihilationsbeitrages der Rumpfelektronen (Core annihilations) wurde schon in früheren Messungen von Kusmiss und Stewart [3] sowie Badoux und Heinrich [4] festgestellt. Den Zusammenhang zwischen $\hat{N}$ und den Parametern der Monovakanzkonzentration $c_v(T) = e^{S_v/k} \cdot e^{-Q_v/kT}$ (S_v = Entropie für Monovakanzbildung) vermittelt das sog. Trapping Modell [5] [6]. Ist λ_f die Rate der Annihilationen aus dem ungestörten Kristall und $\mu \cdot c_v(T)$ die Rate, mit der Positronen in Vakanzen eingefangen werden, so ist

$$\hat{N}(T) = \hat{N}_f \frac{\lambda_f}{\mu c_v + \lambda_f} + \hat{N}_v \frac{\mu c_v}{\mu c_v + \lambda_f}. \quad (1)$$

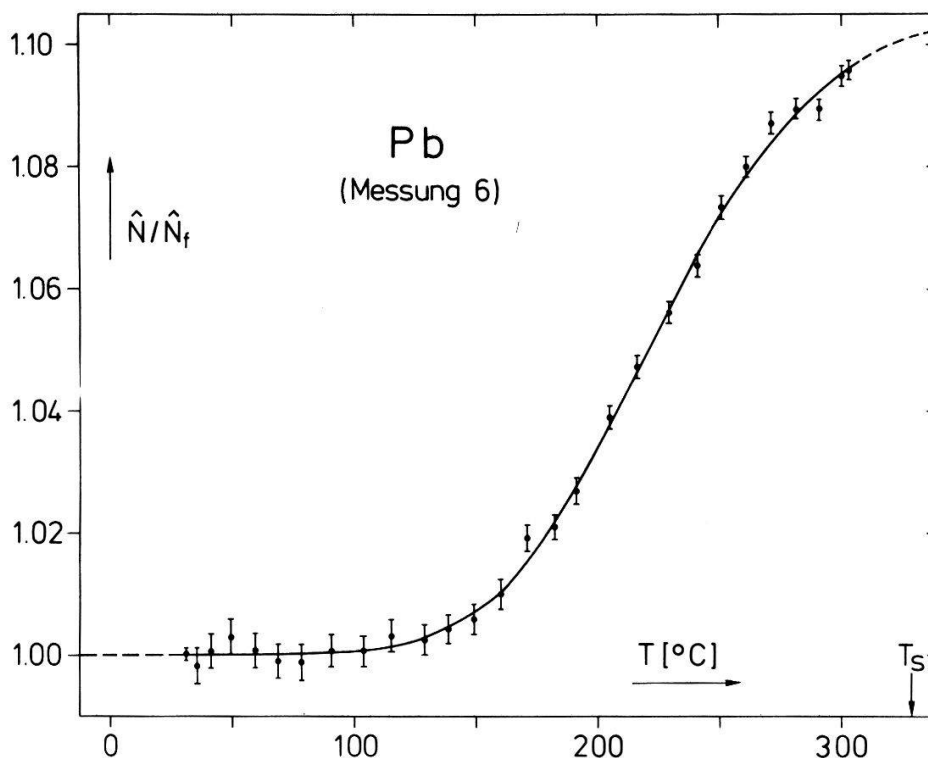
$\hat{N}_f$ und $\hat{N}_v$ sind die Peakhöhen für den Fall, dass keine Positronen bzw. alle Positronen eingefangen sind (Grenzfälle tiefer bzw. hoher Temperaturen). Durch Einsetzen von $c_v(T)$ in (1) folgt

$$\phi(T) = \frac{\hat{N}(T) - \hat{N}_f}{\hat{N}_v - \hat{N}(T)} = \frac{\mu}{\lambda_f} e^{S_v/k} \cdot e^{-Q_v/kT}. \quad (2)$$

$\phi(T)$ ergibt in Arrheniusdarstellung eine Gerade, aus deren Steigung die gesuchte Monovakansbildungsenergie Q_v folgt. Der Temperaturgang von $\hat{N}(T)$ enthält zudem Effekte, die von der thermischen Expansion des Gitters herrühren und die sich hauptsächlich auf $\hat{N}_f$ auswirken. Sharma et al [7] haben allerdings bei Positronenlebensdauer-messungen an Pb festgestellt, dass die thermische Expansion zudem einen (wenn auch kleineren) Temperaturgang von $\hat{N}_v$ verursacht. Bei der Auswertung der vorliegenden Messungen wurde lediglich der in guter Näherung lineare Temperaturanstieg in $\hat{N}_f$ berücksichtigt; wir konnten jedoch feststellen, dass der Wert von Q_v nur geringfügig verändert wird, wenn auch für $\hat{N}_v$ ein Temperaturkoeffizient eingesetzt wird, der ohnehin vorhanden ist, weil nach Malmfors [8] besonders bei Proben mit starker γ -Absorption eine durch die thermische Expansion bedingte Aenderung der γ -Emissionsrate mit der Temperatur auftritt. Dieser Effekt ist offenbar bei Annihilationsexperimenten mit variabler Probentemperatur bisher übersehen worden; er rührt daher, dass sich beim Erhitzen der Probe um ΔT die Dichte wie $-3\alpha\Delta T$, der Quantenabsorptionsweg aber wie $+\alpha\Delta T$ ändert (α = linearer thermischer Expansionskoeffizient).

Für die Messungen wurde eine mechanisch sehr stabile Korrelationsapparatur mit starrem Korrelationswinkel $\theta = 0$ verwendet. Die relativ bescheidene Winkelauflösung von 3 mrad (FWHM) ist ein Kompromiss zwischen Zähleffizienz und Empfindlichkeit für Peakhöhenänderungen. Als Positronenquelle wurde ^{64}Cu verwendet (ca. 0.5 Ci). Die Proben wurden aus hoch-reinem, vakuumgeschmolzenem, sorgfältig entgastem Blei hergestellt. Die Probenoberfläche ist dachförmig, um die γ -Absorption in den Proben zu reduzieren. Die Koinzidenzraten wurden jeweils auf die zugehörige Einzelrate eines der Detektoren bezogen, wodurch temperaturbedingte Aenderungen der Raumwinkelgeometrie zwischen Probe und Quelle herausfallen. Für die Halbwertszeitkorrektur wurde der Wert $T_{1/2} = (12.701 \pm 0.001)\text{h}$ [9] benützt. Es wurden 7 voneinander unabhängige Messungen unter

verschiedenen experimentellen Bedingungen vorgenommen: Variationen bei der Probenpräparation und bei der Geometrie Quelle – Probe, Messungen nach steigender bzw. fallender Temperatur, Änderungen der Temperaturraten (11–22°C/h) usw.



Figur 1

In Figur 1 ist $\hat{N}(T)/\hat{N}_f$ für eine dieser Messungen im Temperaturbereich 20–310°C aufgetragen. Die Messpunkte sind bereits bezüglich der ^{64}Cu -Halbwertszeit, des Untergrundes und der thermischen Expansion der Probe korrigiert. Die eingetragene Kurve ist ein Bestfit an (1). Für Q_v ergeben sich 7 statistisch konsistente Resultate mit dem gewichteten Mittelwert

$$Q_v = (0.631 \pm 0.012) \text{ eV},$$

der sehr gut mit dem Resultat von Shulman [10] übereinstimmt, aber um einiges grösser ist als die Messungen anderer Autoren wie

$Q_v = 0.49 \pm 0.1 \text{ eV}$	Feder et al [11]
0.49	Köhler et al [12]
0.50 ± 0.03	McKee et al [2]
0.49 ± 0.03	McKee [13]
0.54 ± 0.02	Triftshäuser [14].

Sehr sorgfältige Untersuchungen von Sharma, Berko und Warburton [7] an Blei mit Hilfe der Positronen-Lebensdaueremethode zeigen, dass bei dieser Methode die Resultate für die Vakanzbildungsenergie Q_v in hohem Masse von der Analyse der Lebensdauerspektren abhängen.

Den Herren R. Spring und R. Meier sind wir für die Mitwirkung beim Bau der Apparaturen und wertvolle technische Assistenz dankbar. Dem Schweizerischen

Nationalfonds zur Förderung der wissenschaftlichen Forschung danken wir für die finanzielle Unterstützung dieser Arbeit.

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Énergie de liaison de la molécule excitonique

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L'état lié de deux excitons est bien compris dans la limite de la molécule d'hydrogène où $m_{\text{électron}}/m_{\text{trou}} = \sigma \ll 1$. Dans la limite $\sigma = 1$ (molécule de positronium), un groupe d'auteurs a obtenu, à l'aide de méthodes variationnelles, de faibles énergies de liaisons. D'autres auteurs prédisent de grandes énergies de liaison, mais utilisent des méthodes controversées. Il est important de trancher entre ces résultats discordants, car la valeur de l'énergie à $\sigma = 1$ est cruciale pour l'estimation de l'énergie de liaison pour $0 < \sigma < 1$. Nos calculs préliminaires, obtenus à l'aide de fonction variationnelles très élaborées, mais qui négligent la corrélation entre particules égales, donnent des énergies de liaison plus grandes que le groupe des calculs non controversés, dont certains tiennent compte de la corrélation trou-trou. Il n'est pas encore possible d'estimer si l'adjonction de corrélations supplémentaires à notre fonction d'onde permettra de confirmer les valeurs les plus élevées proposées jusqu'ici pour l'énergie de liaison.

Optically observed second sound resonances

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Although second sound in Helium II is ideally a temperature wave at constant liquid density, there are in fact pressure gradients in second order associated with the

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motion of the normal and the superfluid components [1, 2]. We have shown that this makes it possible to observe standing waves of second sound optically, and have reported on the use of this method to study the resonance frequencies of a cavity.

The configuration studied has been a vertical cylinder of radius 1 cm with a free helium surface excited by alternating current in a vertical axial wire. The high Kapitza resistance and the low thermal conductivity of the cylinder led to the assumption of $\nabla_n T = 0$ normal to the cylinder wall as a boundary condition. The resonance frequencies for this case are equivalent to those given by Rayleigh [3] for radial vibrations of a cylinder and our observations showed fair agreement with the Rayleigh solution [4].

The boundary condition given above and the Rayleigh solution ignore two important aspects of the real experimental situation, namely that heat is transferred at both boundaries and that the cavity is an annular region and not a cylinder. The second sound is produced by a heater in the cylinder axis, and the heat produced is allowed to escape through holes in the cylinder wall (these were bored to remove d.c. heat from the helium). To take account of this heat transfer, a more general boundary condition may be used:

$$\nabla_n T + \xi(T - T_0) = 0 \quad (1)$$

where T_0 is the average temperature and ξ is a constant for each boundary surface. The general solution is then

$$T - T_0 = [aJ_n(kr) + bN_n(kr)] \cos n\theta \sin \omega t \quad (2)$$

with $b \neq 0$ because the cavity is an annular region rather than a cylinder. $J_n(kr)$ and $N_n(kr)$ are the Bessel and Neumann functions of order n , a and b are constants, and $k = \omega/c$ where c and ω are the velocity and angular frequency of second sound. For $\xi = 0$ and $b = 0$, the boundary conditions lead to the resonance condition:

$$J'_n(Z) = 0 \quad (3)$$

where $Z = kR$ and R is the cylinder radius. Let $Z_R(n, s)$ be the solutions of (3) and v_{ns} be the resonant frequencies of the a.c. heater. Then, if (3) holds,

$$v_{ns} = Z_R(n, s)c/4\pi R$$

$$\text{and } \frac{v_{ns}}{Z_R(n, s)} = \frac{c}{4\pi R} = \text{constant.}$$

For $\xi \neq 0$, $b \neq 0$, the resonances can be enumerated using the same n and s as for the Rayleigh solution. $v_{ns}/Z_R(n, s)$ is not, however, a constant any longer.

We have now made more exact measurements of some 80 resonance frequencies than was possible in our earlier experiments. Great care was taken to obtain all results keeping the temperature very constant, because the variations in frequency due to temperature are larger than those due to differing boundary conditions. The data are plotted as v/Z_R against v in Figure 1.

Values of ξ at the heater and outer surface of the cavity were determined by a least square fit of the observed frequencies to the theoretical expression. The curve in Figure 1 is a line drawn through the theoretical points $v_{ns}/Z_R(n, s)$ calculated using the least square values of ξ . Although there are several such curves for different n values, they have been omitted for the sake of clarity.

It is clear that while the Rayleigh boundary condition yielding $v/Z_R = \text{constant}$

is in very good agreement with the observations at the higher frequencies it fails at the lowest frequencies. On the other hand the modified boundary condition reproduces the observed data well within the experimental error. It is easy to show mathematically that the value of the high frequency resonances is not sensitive to the boundary condition assumed.

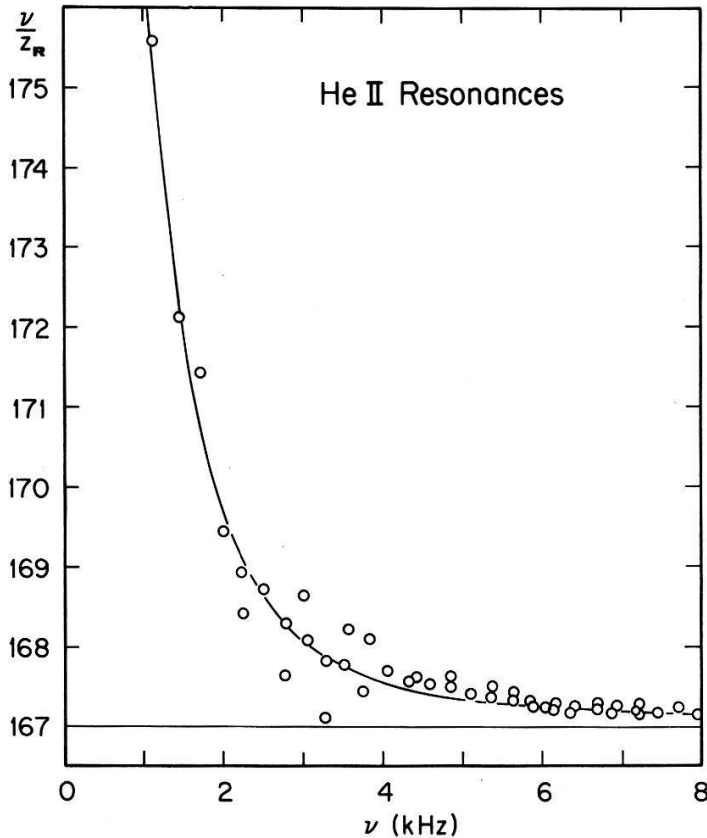


Figure 1

The ratio ν/Z_R of the observed resonance heater frequency to the Rayleigh solutions Z_R of $J'_n(Z) = 0$, as a function of ν .

Having determined optimum values of ξ at the boundary surfaces, it is now possible to relate ∇T at the surface of the heater wire to the power delivered, and hence to make calculation of the expected height of the waves. This is done using the expressions given by Sorbello in a recent paper [5].

The conclusion from the present set of experiments is that for our case Sorbello's theoretical height estimates yield wave heights of the order of 0.5 mm. Our experimental technique does not at present allow a very reliable measurement of the wave height but suggests maximum wave heights of approximately 0.1 mm. It is at present not possible to determine whether this discrepancy is due to the uncertainty of the experimental data or if it indicates some weakness in the theory.

A more detailed account of this work will be published elsewhere.

This work was made possible by financial support from the Swiss National Science Foundation.

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Dynamische Prozesse an der Phasengrenze Eis-Wasser

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Mit Rayleigh-Lichtstreuung wird die Dynamik des Gefrierprozesses an der Phasengrenze eines in c -Richtung wachsenden Kristalls untersucht. Die Intensität des Streulichtes hängt von der Wachstumsgeschwindigkeit ab und zeigt eine Hysterese, sodass man das Einsetzen der Fluktuationen bei einer bestimmten Wachstumsgeschwindigkeit beobachtet. Am schmelzenden Kristall wird keine Streuung beobachtet. Man misst eine Linienbreite des gestreuten Lichtes, die proportional zum Quadrat des Streuvektors ist und bei einem Streuwinkel von 90° etwa 2 kHz beträgt. Die Linienbreite hängt nicht von der Wachstumsgeschwindigkeit ab. Das Licht wird isotrop gestreut. Dies deutet darauf hin, dass die Korrelationslänge im Vergleich zur Wellenlänge klein ist.

Thermische Rayleigh-Streuung in SrTiO_3

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In einer 0.03% Fe-dotierten Probe wurde mittels zwei interferierender und zeitlich modulierter Argonlaserstrahlen ein thermisches Gitter erzeugt. Die Streuung eines dritten Lichtstrahls von einem He-Ne Laser an diesem Gitter ist proportional zu Brechungsindexänderung und damit zur Temperaturvariation. Die Frequenzabhängigkeit entspricht dem klassischen Rayleighspektrum.

Wir haben das Rayleighspektrum in SrTiO_3 auf diese Weise mit hoher Frequenzauflösung bestimmt. Es wurde für Streuwinkel von $\theta = 10$ bis 20 mrad im Gebiet 90 bis 300 K gemessen. Die Halbwertsbreite ist proportional zu θ^2 ; die nimmt bei Abkühlung stark zu im Einklang mit der abnehmenden spezifischen Wärme und zunehmender Wärmeleitung. Am 105 K Phasenübergang ist keine Anomalie in der Halbwertsbreite zu beobachten. Die Intensität der Streuung variiert stark, bei 105 K, jedoch wurde eine quantitative Auswertung durch Domainenbildung verhindert. Zusätzlich zum thermischen Rayleighspektrum wurden in diesem Kristall photochromische Effekte sowie ein noch nicht identifizierter Hochfrequenzanteil beobachtet.

Inelastische Lichtstreuung an Metallen mit NaCl-Struktur: Ein-Phonon-Zustandsdichte in GdTe , GdSe , GdS und LaTe

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Wir untersuchten die kubisch-flächenzentrierten Metalle GdS , GdSe , GdTe und LaTe mit Ramanstreuung. In den Gadoliniumchalkogeniden ist die inelastische

Lichtstreuung wegen der hohen Neutronenabsorption und metallischen IR-Reflexion die bisher einzige Methode zur Bestimmung des Phononspektrums, Dabei wird in den untersuchten Materialien das Symmetrieverbot für 1. Ordnung-Ramanstreuung durch den Einfluss von Fehlstellen aufgehoben, und man findet im Frequenzbereich von $10\text{--}300\text{ cm}^{-1}$ defektinduzierte Ramanspektren erster Ordnung, die einer gewichteten Ein-Phonon-Zustandsdichte entsprechen. Mit zunehmender Masse der Anionen zeigen sich starke Veränderungen im Phononspektrum, wobei die Frequenzlücke zwischen den akustischen und optischen Phononen verschwindet. Es ergeben sich Analogien zum Phononspektrum der Europiumchalkogenide, allerdings wurde kein Einfluss der magnetischen Ordnung auf die Ramanstreuung gefunden.

Magneto-optische Messungen an $(\text{CH}_2)_2(\text{ND}_3)_2\text{MnCl}_4$

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Die lineare Doppelbrechung (LD) von $(\text{CH}_2)_2(\text{ND}_3)_2\text{MnCl}_4$ ($^2\text{C}_2\text{Mn}$) wurde bei $\lambda = 632.8\text{ nm}$ zwischen 4 K und 300 K und in Magnetfeldern bis zu 100 kOe gemessen. In $^2\text{C}_2\text{Mn}$ bilden die magnetischen Mn^{2+} Ionen zusammen mit den Cl^- Anionen schichtartige Netzwerke, die durch kovalente Bindungen miteinander verknüpft sind. Durch Vergleich mit dem isomorphen Diamagneten $(\text{CH}_2)_2(\text{NH}_3)_2\text{CdCl}_4$ konnte der magnetische Beitrag zur LD abgetrennt werden. Die Temperaturabhängigkeit der linearen magnetischen Doppelbrechung (LMD), gemessen senkrecht zu den Schichten, hat die Form der magnetischen Energie. Sie hat einen sehr ausgeprägten Hochtemperaturausläufer bis auf 200 K. Die 3-dim. Ordnung äussert sich nicht in eindeutiger Weise in einer Anomalie am Néelpunkt ($T_N \sim 48\text{ K}$). Verglichen mit der Temperaturabhängigkeit ist die Feldabhängigkeit schwach. Der Spinflop Übergang konnte aber eindeutig bestimmt werden bei $H_{SF} = 33.5 \pm 2\text{ kOe}$ ($T = 4\text{ K}$). H_{SF} steigt mit der Temperatur an in Übereinstimmung mit Magnetisierungsmessungen. Die LMD nimmt bei H_{SF} um $\sim 1\%$ ab ($T = 4\text{ K}$). Aus der LMD folgt, dass sich $^2\text{C}_2\text{Mn}$ in guter Näherung wie ein 2-dimensionaler Heisenberg Antiferromagnet verhält.

Untersuchung von sensibilisierter Phosphoreszenz mit Hilfe der site selection Spektroskopie

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Wir haben die Phosphoreszenzspektren von Naphthalinjodid (Donor) und perfluoriertem Naphthalin (Akzeptor) in EPA-Lösungen bei 4.2° K gemessen. Dabei wurde nur der Triplettzustand des Donors mit einer geeigneten Argonlaserlinie angeregt. Die Phosphoreszenz des Donors, aber auch des Akzeptors, zeigt scharfe ($1\text{--}2\text{ cm}^{-1}$) O-Phononenlinien. Für diese Linien muss also gelten, dass eine Energieübertragung Donor-Akzeptor nur an denjenigen 'sites' stattfindet, wo die Donor Emissionsenergie genau mit einer Akzeptor Absorptionsenergie zusammenfällt. Der Energietransport selbst muss dabei auch ein O-Phononenprozess sein.

Partial circular polarization of the spectral thermal emission from ferromagnetic iron

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Abstract. The spectral thermal emission from a single crystal of ferromagnetic iron is found to be partially circular polarized. The circular polarization changes sign near a photon energy of 1.3 eV.

We have measured the thermal spectral emission from the (100) surface of a single crystal of ferromagnetic iron. The electropolished sample was heated in an uhv-chamber and magnetically saturated in an external field of $B \simeq 1.5$ T. The emitted radiation was detected in the photon energy range $0.4 \text{ eV} < h\nu < 1.8 \text{ eV}$. The right circularly polarized (RCP) and the left circularly polarized (LCP) component of the radiation were separated with a Soleil-Babinet compensator and detected individually.

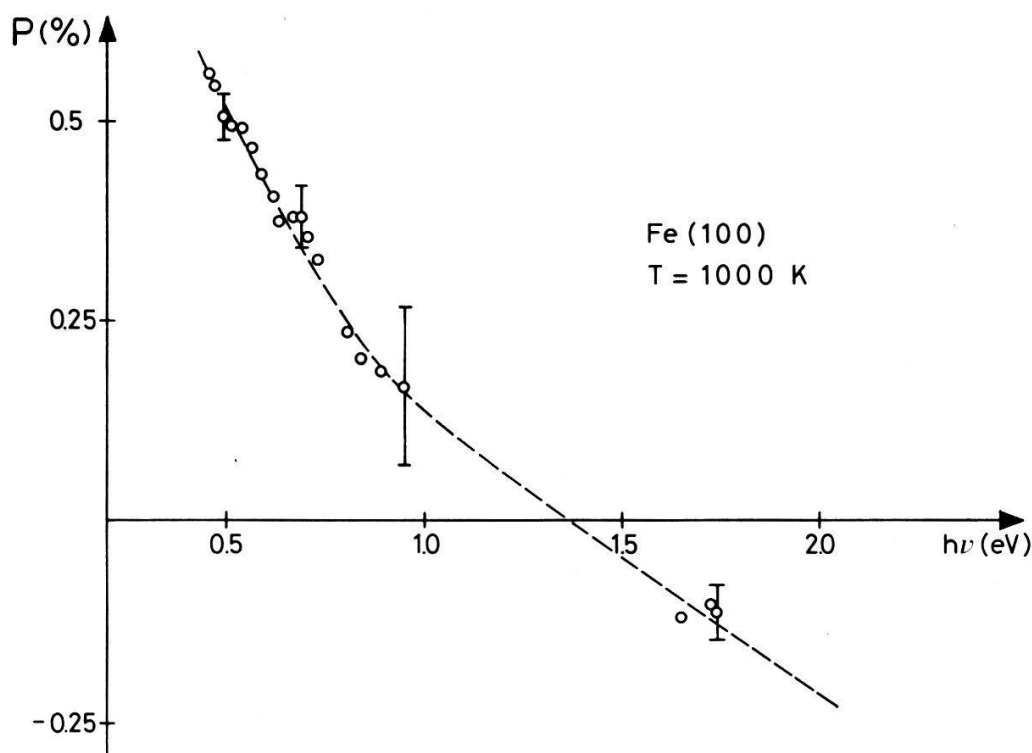


Figure 1

Degree P of the circular polarization of the thermal spectral emission from a single crystal of iron as a function of the photon energy.

In Figure 1 the degree P of the circular polarization of the spectral thermal emission from the iron at a temperature of 1000 K is plotted versus the photon energy. P is represented by the intensity ratio

$$P = 2 \frac{I_{\text{LCP}}(\mathbf{B}) - I_{\text{RCP}}(\mathbf{B})}{I_{\text{LCP}}(\mathbf{B}) + I_{\text{RCP}}(\mathbf{B})}$$

LCP and RCP radiation are defined by the optics convention. The direction of the external magnetic field $\mathbf{B}$ perpendicular to the surface of the sample equals the direction opposite to the emission.

The most striking feature of Figure 1 is the change of the sign of the circular polarization near a photon energy of 1.3 eV.

The described magneto-optic effect is due to spin-orbit coupling in the ferromagnetic iron [1]. It is related to the Kerr effect, which was measured in Fe, Co, Ni [2] and calculated for Fe by Singh et al. [1] and for Ni by Erskine and Stern [3]. Singh et al. [1] computed the off-diagonal term of the interband conductivity for Fe, which represents the difference [3] between the absorption of the LCP and the RCP radiation. They found this term to be positive for photon energies up to 5 eV. The corresponding intraband conductivity should be positive also, as a consequence of the calculated positive spin polarization in the vicinity of the Fermi level [1]. According to the principle of detailed balance (Kirchhoff's law) the difference of the absorptions of the LCP and the RCP radiation corresponds to the reported partial circular polarization of the spectral emission. Therefore, the observed partial circular polarization of the spectral thermal emission from the (100) surface of a single crystal of Fe seems to contradict the magneto-optic interband conductivity calculated by Singh et al. [1].

The authors wish to thank Prof. Dr. W. Baltensperger, Prof. Dr. H. C. Siegmann, Dr. W. Eib, ETH Zurich and Dr. J. Helman, Mexico, for valuable discussions, and G. von Holzen, EIR, Würenlingen for technical assistance.

This work is supported by the Swiss National Science Foundation and by E.I.L. Zurich.

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Phase locking in coupled Josephson weak links

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Abstract. It has been experimentally demonstrated that two closely coupled Josephson weak links can synchronize their phase motion over a certain range of biasing currents. We show from the equations of motion that the oscillations in two Josephson junctions can phase lock when the junctions are coupled by a mutual capacitance. When one junction is biased at finite voltage by a constant current i_2 , the junctions phase lock over an appreciable range of bias currents i_1 through the other junction. The apparent critical current in the (i_1-v_1) current-voltage characteristic for constant i_2 is asymmetric with respect to the relative sign of i_1 and i_2 with the calculated asymmetry in the same direction as that observed experimentally. The phase locked periodic solutions found numerically are shown to be asymptotically stable through use of the Poincaré-Floquet mapping technique.

In a superconducting weak link which exhibits the Josephson effect, the current varies like $\sin \phi(t)$ where

$$\frac{d\phi}{dt} = \frac{2 \text{ eV}}{\hbar} \quad (1)$$

is proportional to the voltage V across the weak link. External electromagnetic radiation can synchronize the internal oscillation frequency to the external frequency,

resulting in a change in the voltage across the link so equation (1) remains valid. There is increasing experimental evidence [1] that the oscillating currents in closely coupled superconducting weak links can phase lock, to oscillate synchronously, even in the absence of externally applied electromagnetic radiation. In the experiments of D. W. Jillie et al. [2], two superconducting microbridges, separated by a $2\ \mu\text{m}$ wide strip of superconductor were fed by two currents of opposite sign: both directed toward, or both away from the central strip. When one of the microbridges was biased to a finite voltage V_1 by a current I_1 , the microbridges interacted so this same voltage, V_1 , appeared also across the second microbridge for an appreciable range of currents I_2 through the second microbridge. Voltage locking was not observed for currents of the same sign. Recently, P. E. Lindelof and J. Bindslev Hansen [3] observed coherent microwave radiation from two closely spaced microbridges when the voltages locked to the same value, showing that voltage locking implies phase locking in the internal oscillations of the microbridges. Locking occurred for both relative signs of the bias currents. Similar results had been reported earlier by D. W. Palmer and J. E. Mercereau [4] in linear arrays of up to 2000 junctions. These arrays can phase lock when fed in series if the center to center spacing is less than $2\ \mu\text{m}$.

The purpose of this note is to report that mutual capacitance between two superconducting weak links can induce phase locking. Within the resistively shunted junction model [5], the current flowing through a single weak link, or Josephson junction, is

$$I = I_0 \sin \phi + \frac{V}{R} + C \frac{dV}{dt} \quad (2)$$

where I is a constant bias current, I_0 the critical current, R the shunt resistance, C the shunt capacitance of the junction, V the voltage and ϕ the quantum phase difference across the junction. After substituting V in terms of ϕ from (1) into (2), the resulting equation for ϕ can be integrated numerically to find $\phi(t)$ and the time average voltage $\bar{V}$ observed in low frequency current-voltage (I-V) characteristics [6].

Two junctions coupled by mutual capacitance are described by two analogous equations which are now coupled. The interaction via the electric field for junction electrode sizes small compared to the wavelength of the emitted radiation can be described by capacitance coefficients C_{ij} which linearly relate the potential V_i of electrode i to the charge q_j on electrode j according to

$$V_i = \sum_{j=1}^3 C_{ij}^{-1} q_j \quad (3)$$

where the index 2 corresponds to the electrode region between the two junctions, and 1 and 3 to the outer ends. With this interaction, two junctions with identical critical currents I_0 and shunt resistances R are described by

$$\begin{aligned} C \left(\frac{d^2 \phi_1}{dt^2} - \alpha \frac{d^2 \phi_2}{dt^2} \right) &= \frac{2e}{\hbar} I_1 - \frac{2e}{\hbar} I_0 \sin \phi_1 - \frac{1}{R} \frac{d\phi_1}{dt} \\ C \left(\frac{d^2 \phi_2}{dt^2} - \alpha \frac{d^2 \phi_1}{dt^2} \right) &= \frac{2e}{\hbar} I_2 - \frac{2e}{\hbar} I_0 \sin \phi_2 - \frac{1}{R} \frac{d\phi_2}{dt} \end{aligned} \quad (4)$$

with

$$C = \frac{-C_{13}^2 - C_{21}^2 - 2C_{13}C_{21} + 2C_{11}C_{23} + C_{11}C_{22} + C_{11}C_{33}}{\sum_{i=1}^3 \sum_{j=1}^3 C_{ij}} \quad (5)$$

and

$$\alpha = \frac{-C_{13}^2 - C_{13}C_{23} - C_{13}C_{12} - C_{13}C_{22} + C_{11}C_{33} + C_{11}C_{23} + C_{12}C_{23} + C_{12}C_{33}}{-C_{13}^2 - C_{21}^2 - 2C_{13}C_{21} + 2C_{11}C_{23} + C_{11}C_{22} + C_{11}C_{33}} \quad (6)$$

In terms of the natural units of current, time, and capacitance, (4) can be rewritten as

$$\begin{aligned} c(\ddot{\phi}_1 - \alpha\ddot{\phi}_2) &= i_1 - \sin \phi_1 - \dot{\phi}_1 \\ c(\ddot{\phi}_2 - \alpha\ddot{\phi}_1) &= i_2 - \sin \phi_2 - \dot{\phi}_2 \end{aligned} \quad (7)$$

where $i_1 = I_1/I_0$, $i_2 = I_2/I_0$, $\tau = 2eI_0Rt/\hbar$, $c = 2eI_0R^2C/\hbar$.

The dot denotes differentiation with respect to normalized time τ . Expressed in units of voltage $V_c = I_0R$, the voltage across each junction is $v = \dot{\phi}$.

It can be seen that the equations (7) depend on four relevant parameters, i_1 , i_2 , α and c (the normalized capacitance c is the same as the Mc Cumber and Stewart parameter β). The equations were integrated numerically with an order 17 10 Runge Kutta method [7], and the voltage across each junction, $\dot{\phi}_i$, was time averaged over a large interval of ϕ_i after the disappearance of a transient due to initial conditions.

The results of the calculations are illustrated in a manner similar to that for the data which exhibit voltage locking [2]. For fixed bias currents i_2 through junction 2, the current i_1 through junction 1 was varied, and the voltage across each junction calculated. Figure 1 illustrates the calculated voltages for $\alpha = 0.5$ and $c = 2$. It can be seen that the average voltage across the junctions becomes identical over a range of currents i_1 , both when i_1 has the same, as well as when it has a sign opposite to that of i_2 . Note that in our model, the mutual capacitive coupling leads to an asymmetry in the apparent critical currents of junction 1: the apparent critical current of junction 1 is lower when i_1 and i_2 are of opposite sign, than when they are of the same sign (series biased junctions). The direction of this calculated asymmetry is the same as that observed experimentally [2, 3].

In the region of voltage locking, the numerical results indicate that $\phi_1(t)$ and $\phi_2(t)$ converge to phase locked solutions in which $\dot{\theta} = \dot{\phi}_1 - \dot{\phi}_2$ is a periodic function of time, with exactly zero time average. Such a converged solution is shown in Figure 2. The technique of 'period-advance mapping' (Poincaré-Floquet mapping [8]) has been used to demonstrate rigorously that the phase locking is exact.

The solutions of (7) can be viewed most conveniently as trajectories in a phase space defined by the coordinates $(\dot{\phi}, \phi, \dot{\theta}, \theta)$, where $\phi = \phi_1 + \phi_2$ is the total phase drop across the junctions and $\theta = \phi_1 - \phi_2$ is the difference in phases mentioned above. When (7) is rewritten in terms of ϕ and θ , it can be seen that the coefficients are periodic with period 4π in ϕ and in θ . Thus, the structure of the phase space is that of a four dimensional manifold $M = R^1 \times S^1 \times R^1 \times S^1$, with the phases (ϕ, θ) defining a point on a two dimensional torus $S_1 \times S_1$, and the voltages $(\dot{\phi}, \dot{\theta})$ defining a point in a plane R_2 . A phase locked solution corresponds to a closed trajectory in the phase space M , such that ϕ advances by 4π in time period T , and the other variables

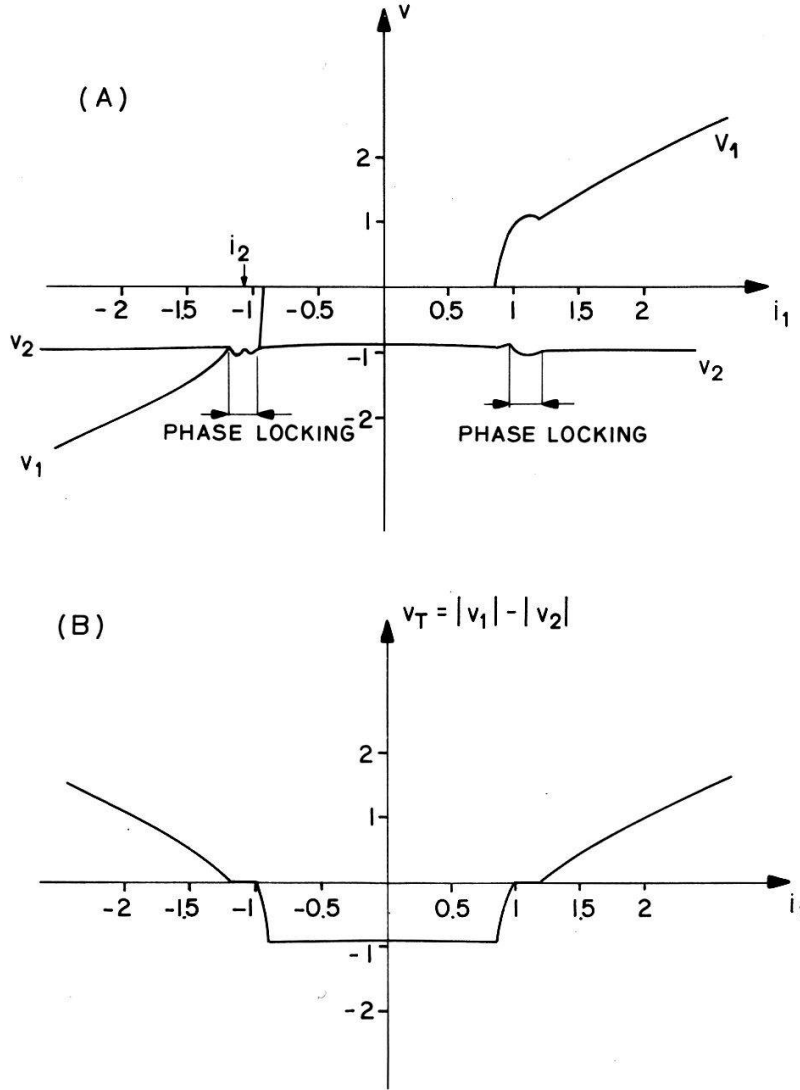


Figure 1

(A) Average voltages v_1 and v_2 , across each junction, expressed in units of RI_0 , when the current i_2 is held constant as i_1 is varied, for identical junctions with $c = 2$, and coupling parameter $\alpha = 0.5$. The ranges of i_1 over which phase locking occurs are indicated. Note that in the presence of coupling, the critical current for i_1 is lower when i_1 and i_2 have opposite signs, than when they have the same sign. (B) To illustrate the regions of phase locking, the difference in the absolute value of the voltages, $v_T = |v_1| - |v_2|$ is plotted versus i_1 for the same parameters as in (A).

are periodic. Starting at $\phi = 0$, the other coordinates of the periodic trajectory are $x_{0p} = (\phi_{0p}, \theta_{0p}, \theta_{0p})$. After period T , the periodic trajectory again crosses the surface defined by $\phi = 0 \pmod{4\pi}$, at the point $x_{1p} = x_{0p}$. Starting points x_0 near x_{0p} define trajectories that also cross the surface $\phi = 0 \pmod{4\pi}$ for the first time again near x_{0p} at $x_1 = g(x_0)$, defining the Poincaré map g as illustrated in Figure 3. The periodic trajectory corresponds to a fixed point x_{0p} in this map. Near the fixed point, the linear part of the map g determines the stability properties of the fixed point, and also the stability properties of the periodic trajectory. In particular, if the absolute value of the eigenvalues of the Jacobian $Dg = (\partial x_1 / \partial x_0)$ are less than 1, i.e., if $|\lambda| < 1$, then g is a contraction near x_{0p} , and the periodic orbit is asymptotically stable.

This technique was applied to demonstrate that the entire regions of i_1 exhibiting the voltage locking of Figure 1 correspond to exact phase locking: the trajectory in phase space is periodic and asymptotically stable. First, the equations (7) were

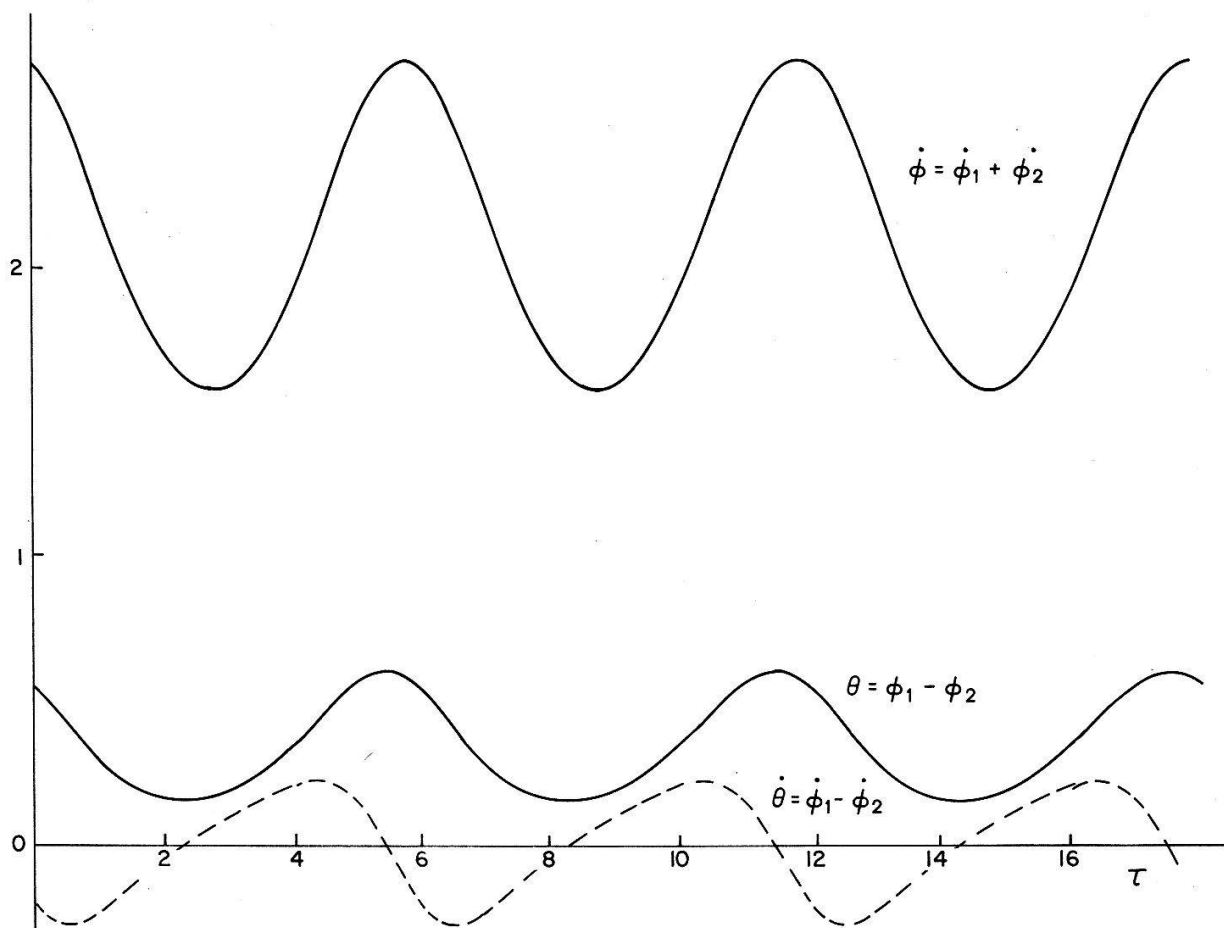


Figure 2

Total voltage drop across both junctions $\dot{\phi} = \dot{\phi}_1 - \dot{\phi}_2$, relative phases $\theta = \phi_1 - \phi_2$, and voltage difference $\dot{\theta} = \dot{\phi}_1 - \dot{\phi}_2$ for a phase locked solution as a function of normalized time for $c = 2.$, $\alpha = 0.5$, $i_1 = 1.11$, and $i_2 = 1.08$. The solution is periodic and asymptotically stable, as shown by the Poincaré-Floquet mapping technique.

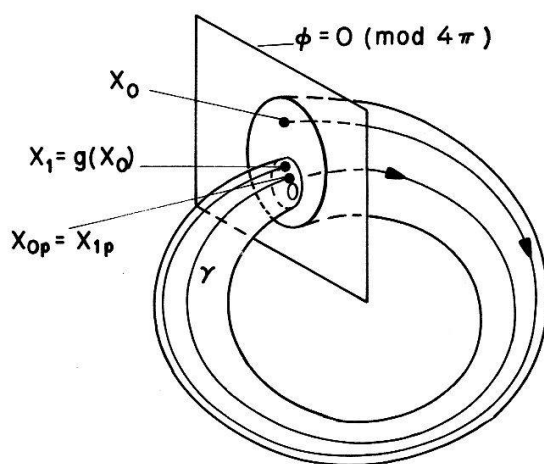


Figure 3

The Poincaré-Floquet map g illustrated for an asymptotically stable periodic orbit γ (an attractor). The map contracts a neighborhood of initial conditions x_0 for trajectories passing through the surface $\phi = 0 \pmod{4\pi}$ near the orbit γ .

integrated numerically to find the approximate location of a periodic trajectory. Then Newton's method was used to find the fixed point $g(x_{0p}) - x_{0p} = 0$, and the Jacobian matrix calculated [9]. In all cases, the absolute value of each of the eigenvalues was less than 1, demonstrating that the periodic trajectory was asymptotically stable. The question of whether all initial conditions x_0 converge to this periodic trajectory for the parameters of Figure 1, as well as the range of junction parameters which permit phase locking, will be discussed separately.

The values of i_2 , and i_1 chosen for Figure 1 are typical for the experiments reported so far [2, 3]. The normalized capacitance c is not known very accurately. Richards et al. [10] estimate values ranging from 3×10^{-6} to 3 for weak links. Their estimate for C ($C \simeq 10^{-12}$ F) seems, however, to be quite large. Rough calculation of C gives a value 10 to 100 times smaller for the present experimental geometries for weak links. Thus, the value of $c = 2$, used in the calculations for Figure 1, is probably an overestimate of the capacitance for the present experiments. Within our model, numerical calculations also indicate phase locking for $c = 0.5$, but require larger bias currents i_1 and $i_2 \sim 2$. We have not yet been able to determine whether smaller values of c lead to phase locking. The coupling parameter, α , also is not easy to estimate accurately. For parameters of Figure 1, phase locking occurs for $0.2 < \alpha < 0.6$, and estimates of α for simple geometries give $0.2 < \alpha < 0.5$.

The coupled equations (4) are certainly not the only ones leading to phase locking. Recently, Frank et al. [11] proposed a coupling energy of the form $\cos \phi_1 \cos \phi_2$, which leads to terms of the form $\cos \phi_1 \sin \phi_2$ in the coupled differential equations. These equations seem also to have the phase locking property.

We thank Prof. G. Wanner for valuable discussions on numerical techniques, and for the use of his computer programs.

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Relaxation des Myonspins in Myonium in Festkörpern

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Das Verhalten der Myonspinpolarisation eines Myoniumatoms in einem Festkörper wird in einem Modell untersucht, das die Spin-Wechselwirkungen des gebundenen Elektrons und des μ^+ mit der Umgebung durch zwei Parameter beschreibt. Es enthält die Gleichungen von Ivanter und Smilga [1] als Spezialfall. Numerische Lösungen für den Uebergang von langsamer zu rascher Elektronenspinrelaxation werden diskutiert, zusammen mit den analytischen Lösungen in den beiden Grenzfällen.

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Oscillations nonlinéaires d'une jonction Josephson avec échange d'énergie entre la jonction, le champ magnétique, et le champ électrique

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On a étudié numériquement et qualitativement les oscillations d'une jonction Josephson dans laquelle le stockage d'énergie de la jonction dans le champ magnétique et dans le champ électrique a été représenté par un circuit avec une inductance et une capacitance. Quand les énergies magnétique et électrique sont comparables à l'énergie intrinsèque de la jonction, la jonction peut osciller à une fréquence subharmonique $f_n = 2\text{eV}/h\nu$ de la fréquence Josephson. Les bifurcations entre les ordres n de l'oscillation subharmonique, et les propriétés qualitatives de l'équation nonlinéaire de troisième ordre seront discutées.

Physique Appliquée

Der photoelektrische Effekt in Aerosolen

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Die Photoemission eines e^- aus feinsten, in Gasen schwebenden Partikeln führt zur elektrischen Leitung σ , die zur Vermeidung von Raumladungserscheinungen mit einer Wechselstrombrücke gemessen wird. Der zeitliche Anstieg von σ bei Einschalten des Lichts sowie sein Gleichgewichtswert sind mit dem Durchmesser, der Konzentration und dem Photoquerschnitt der Teilchen verknüpft. Das Ziel des

Experiments besteht darin, zusammen mit weiteren bekannten Methoden Aerosole zu charakterisieren und insbesondere die Oberflächeneigenschaften feinsten Teilchen zu studieren.

Résonances paramétriques optiques

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Un système atomique à deux niveaux en interaction avec une onde électromagnétique résonnante, intense et modulée en amplitude présente des variations de populations lorsque la fréquence de Rabi est un multiple de la fréquence de modulation. Ce phénomène, bien connu en résonance magnétique (radiofréquences), a été calculé et observé dans le domaine optique sur la transition $|3S_{1/2}, F = 2, m_F = 2\rangle \leftrightarrow |3P_{3/2}, F = 3, m_F = 3\rangle$ du sodium (jet atomique). La largeur des résonances optiques observées reflète l'origine différente de la relaxation (émission spontanée).

Isotopenanomalien in Edelgasen aus terrestrischem Josephinit

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Josephinit ist eine silikathaltige Eisennickelverbindung die in Form von kleinen Knollen von einigen cm Grösse in Josephine County in Oregon gefunden wird. Der Ursprung dieses seltenen Gesteins ist unklar, doch es gibt eine Hypothese, wonach Josephinit von der Grenze Erdkern-Erdmantel herkommen könnte. Wir haben mittels Extraktion bei verschiedenen Temperaturstufen eine Edelgaskomponente festgestellt, die sich durch eine starke Anreicherung von He^3 und Ne^{21} relativ zu atmosphärischem Helium und Neon auszeichnet: $\text{He}^3/\text{He}^4 = 5 \cdot 10^{-4}$, $\text{Ne}^{21}/\text{Ne}^{20} = 0.05$. Downing et al. [1] haben unabhängig einen Xe^{129} -Exzess in Josephinit festgestellt. Diese Anomalien deuten auf den Ablauf von nuklearen Prozessen im Erdinnern oder auf die Anwesenheit einer terrestrischen primordialen Edelgaskomponente mit einer neuartigen Isotopenzusammensetzung hin.

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Anwendungen einer digital gesteuerten Protonen-Mikrosonde

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[Communication retirée.]

Ein quasistatisches Modell für die homogene Keimbildung in einem geschlossenen, übersättigten System

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Ein Satz von gekoppelten Differential- und Integralgleichungen wird aufgestellt, der die Dynamik der homogenen Keimbildung in einem geschlossenen System beschreibt. Die Abnahme der Übersättigung der Atmosphäre durch die Kondensation ist berücksichtigt. Die Gleichungen wurden numerisch integriert und die Tröpfchenzahl und der totale Flüssigkeitsgehalt des Kondensates sowie die Übersättigung der Atmosphäre als Funktionen der Zeit berechnet.

Interprétation des résonances magnétiques nucléaires en présence d'un champ non-résonant

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Resumé. Pour interpréter les résonances à quanta multiples décrites dans une note précédente [1] nous avons utilisé la méthode de la résolvente. Elle nous a permis de prédire les fréquences propres du système (proton de H_2O) avec une précision satisfaisante; précision d'autant meilleure que l'on est plus près de la résonance. Outre le calcul des fréquences propres pour les transitions à 2 et 3 quanta, nous donnons également le calcul de la transition à 1 quanta. De plus nous avons fait le calcul de la largeur de raie en fonction de l'angle θ entre le champ directeur B_0 et l'aimantation M_0 .

Introduction

Les interactions entre un champ $B_2 \cos \Omega t$ non-résonant et un système de noyaux de spin $\frac{1}{2}$ (essentiellement les protons de l'eau) ont montré un grand nombre de phénomènes intéressants. L'ensemble de ceux-ci ayant déjà été décrits [1] nous nous bornerons ici à en donner l'interprétation à l'aide du formalisme dit de la résolvente. P. R. Fontana et P. Thomann [2] ont développé une méthode de calcul originale dont les fondements se trouvent dans les travaux de W. Heitler [3]. Appliquée à notre cas (système à 2 niveaux), cette méthode conduit à un accord excellent entre courbe théorique et points expérimentaux dans le cas des transitions à 1 quantum. Son extension aux transitions à quanta multiples – transitions que l'on a pu observer expérimentalement – nécessite l'utilisation d'un bon ordinateur et conduit rapidement à des expressions peu maniables. Cela se complique encore un peu plus si l'on introduit le paramètre θ (l'angle entre le champ non-résonant B_2 (Ω) et le champ statique B_0).

La méthode de la résolvente, plus simple, fournit une bonne description des expériences et cela même dans le cas où l'on choisit pour énergie des deux niveaux l'énergie commune E_0 au point de croisement. En fait cette approximation convient bien puisque du point de vue expérimental, c'est dans le voisinage immédiat des

points de croisement que l'on observe aisément les transitions à quanta multiples. En s'éloignant de ce point les déplacements de fréquence deviennent importants et l'on est confronté à une délicate opération pour accorder le système de réception.

Resumé de la méthode de la résolvante⁴⁾

Le problème est donc de décrire l'évolution des transitions entre deux niveaux discrets représentés par les vecteurs d'état $|a\rangle$ et $|b\rangle$ d'un hamiltonien H_0 correspondant au champ statique $\mathbf{B}_0$, sous l'effet de la perturbation $\mathbf{B}_2 \cos \Omega t$ décrite par l'hamiltonien $H_2 = \lambda \mathbf{J}(a + a^+)$.

L'évolution au cours du temps est régie par l'opérateur d'évolution U . Le calcul de U nous conduit donc aux amplitudes de probabilités recherchées. Nous admettons sans la justifier, en priant le lecteur de se reporter à la référence (4), l'expression :

$$U(t) = \frac{1}{2\pi i} \int_c e^{-i(E/\hbar)t} G(E) dE \quad (1)$$

où

$$G(E) = \frac{1}{E - H} \quad (2)$$

est par définition la résolvante de l'hamiltonien H , de même

$$G_0 = \frac{1}{E - H_0} \quad (2')$$

est la résolvante de H_0 , C étant un contour fermé dans le plan complexe.

La pratique montre qu'il est souvent plus facile de calculer les éléments de matrice de $G(E)$ que ceux de $U(t)$. De plus, seule la connaissance d'un petit nombre d'entre eux suffit dans de nombreux cas à résoudre le problème.

On est donc conduit à calculer la projection $\bar{G}(E)$ de $G(E)$ sur le sous-espace $\mathcal{P}_{H_0}$ d'états propres de H_0 , ce qui donne :

$$\bar{G}(E) = \frac{1}{E - H_0 - \bar{R}(E)} \quad (3)$$

$\bar{R}(E)$ étant l'opérateur 'shift'. Il peut se développer :

$$\bar{R}(E) = PH_2P + PH_2 \frac{Q}{E - H_0} H_2P + \dots \quad (4)$$

P étant le projecteur sur $\mathcal{P}_{H_0}$ et Q le projecteur sur le sous-espace complémentaire à $\mathcal{P}_{H_0}$. On peut alors donner une expression de $\bar{G}(E)$ en fonction de $\bar{R}(E)$ sous la forme :

$$\bar{G}_{ba}(E) = \frac{\bar{R}_{ba}(E)}{[E - E_a - \bar{R}_{aa}][E - E_b - \bar{R}_{bb}] - |\bar{R}_{ba}(E)|^2} \quad (5)$$

Expression permettant de calculer directement les transitions entre $|a\rangle$ et $|b\rangle$. Dans la mesure où l'on ne veut calculer que les énergies perturbées (ou la réponse en fréquence du système) ce qui est notre cas ici, ces énergies (ou fréquences) seront

fournies par les pôles de $\bar{G}(E)$. Nous avons donc ramené le calcul des éléments de matrice de $\bar{G}(E)$ (et donc de $U(t)$), au calcul des différents $\bar{R}$, en prenant, comme indiqué plus haut la valeur commune E_0 pour E .

Applications

1^o) Transitions à un quantum

Avec $\mathbf{B}_0$ selon l'axe Oz et $\mathbf{B}_2 \cos \Omega t$ selon l'axe Ox , nous trouvons – si l'on exprime par $\delta\omega_0^\pm$ l'écart de fréquence par rapport à la fréquence de Larmor $\omega_0 - 2$ composantes :

$$\delta\omega_0^\pm = \pm \Delta\omega \pm \sqrt{\Delta\omega^2 + \frac{q\omega_2^2}{(\omega_0 + 3\Omega)} \Delta\omega + \left[\frac{\omega_2^2}{(\omega_0 + 3\Omega)} \right]^2 + \omega_2^2} \quad (6)$$

avec $\Delta\omega = (\omega_0 - \Omega)$ et $\omega_2 = \lambda\sqrt{N}$, où λ est la constante de couplage et $\bar{N}$ le nombre moyen de modes du champ. Sur la Figure 1 nous avons reporté la courbe issue de l'expression (6) et les points expérimentaux. Comme prévu, l'accord est bon sauf lorsqu'on est loin du centre de la résonance. Ici nous avons $\omega_2 = 100$ Hz. La divergence s'accroît si l'on choisit des valeurs supérieures à 100 Hz, néanmoins l'allure générale donne une description tout à fait satisfaisante.

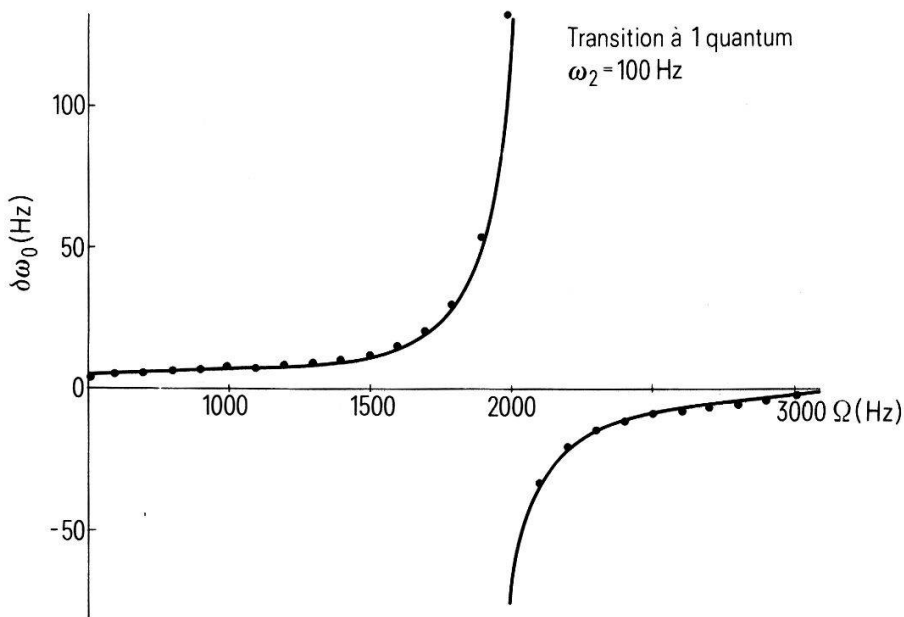


Figure 1

2^o) Transition à 3 quanta

Nous calculerons tout d'abord le cas particulier où comme ci-dessus $\mathbf{B}_0$ et $\mathbf{B}_2 \cos \omega t$ sont perpendiculaires.

Puis nous calculerons la largeur de la raie (distance entre les 2 transitions) et son évolution en fonction de l'angle θ que font les deux champs entre eux. Aux effets de relaxation près (négligeables ici) la largeur de la raie est essentiellement fournie par $\bar{R}_{ba}$.

Dans le cas où B_0 et $B_2(\Omega)$ sont perpendiculaires nous obtenons les 'fréquences de Larmor déplacées':

$$\delta_{\omega_0}^{\pm} = \Delta\omega \pm \sqrt{\Delta\omega^2 + \omega_2^2 \left[\frac{3\Omega + \omega_0}{(\Omega + \omega_0)(5\Omega + \omega_0)} \right] \Delta\omega + \omega_2^4 \left[* \right]^2 + \frac{\omega_2^6}{(\Omega + \omega_0)^4}} \quad (7)$$

où ici $\Delta\omega = (\omega_0 - 3\Omega)$.

Sur la figure 2 nous comparons la courbe donnée par l'expression (7) et les points expérimentaux pour $\omega_2 = 400$ Hz au voisinage de $\Omega = 3\omega_0$.

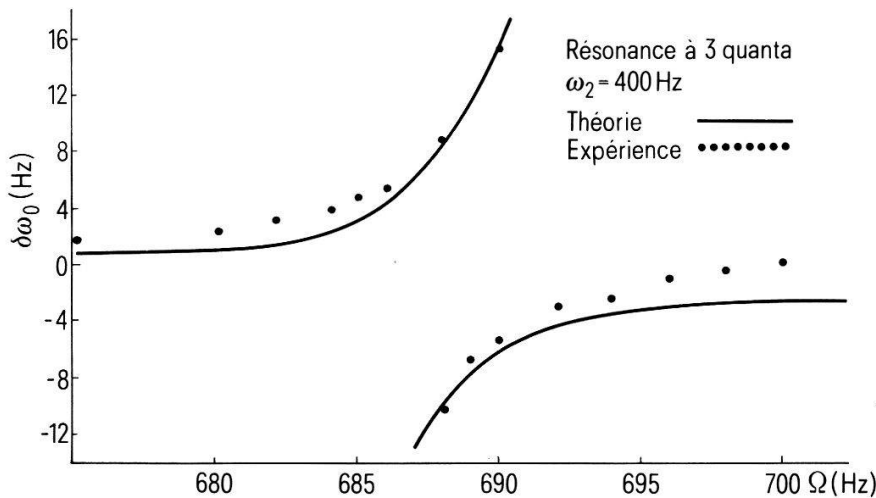


Figure 2

3⁰) Transitions à 2 quanta

Les 'fréquences de Larmor déplacées' dans le cas des transitions à 2 quanta sont données par:

$$\delta_{\omega_0}^{\pm} = \Delta\omega \pm \sqrt{\Delta\omega^2 + \omega_2^2 \left[\dagger \right] \Delta\omega + \frac{\omega_2^4}{4} \left[\dagger \right]^2 + \frac{\omega_2^4}{\omega_0^2} \sin^2 2\theta} \quad (8)$$

avec ici $\Delta\omega = (\omega_0 - 2\Omega)$.

Dans (8) apparaît au dernier terme une fonction simple de θ . Le calcul de (8) a donc été fait pour le cas général où B_0 et $B_2(\Omega)$ font un angle θ quelconque entre eux. La figure 3 permet de comparer la courbe fournie par (8) et les points expérimentaux. L'accord est excellent au voisinage immédiat du croisement de niveaux (centre de la résonance à 2 quanta). La divergence qui apparaît lorsqu'on s'éloigne de ce point tient pour une part à la restriction signalée plus haut et à la forte dépendance angulaire alors qu'il est expérimentalement difficile d'évaluer θ à mieux que 5°.

$$\left[* \right] = \left[\frac{3\Omega + \omega_0}{(\Omega + \omega_0)(5\Omega + \omega_0)} \right].$$

$$\left[\dagger \right] = \left[\frac{\omega_0 - 2\Omega}{\omega_0(\omega_0/4 - \Omega)} \cos^2 \theta - \frac{\omega_0 + 2\Omega}{\omega_0(\omega_0/4 + \Omega)} \sin^2 \theta \right].$$

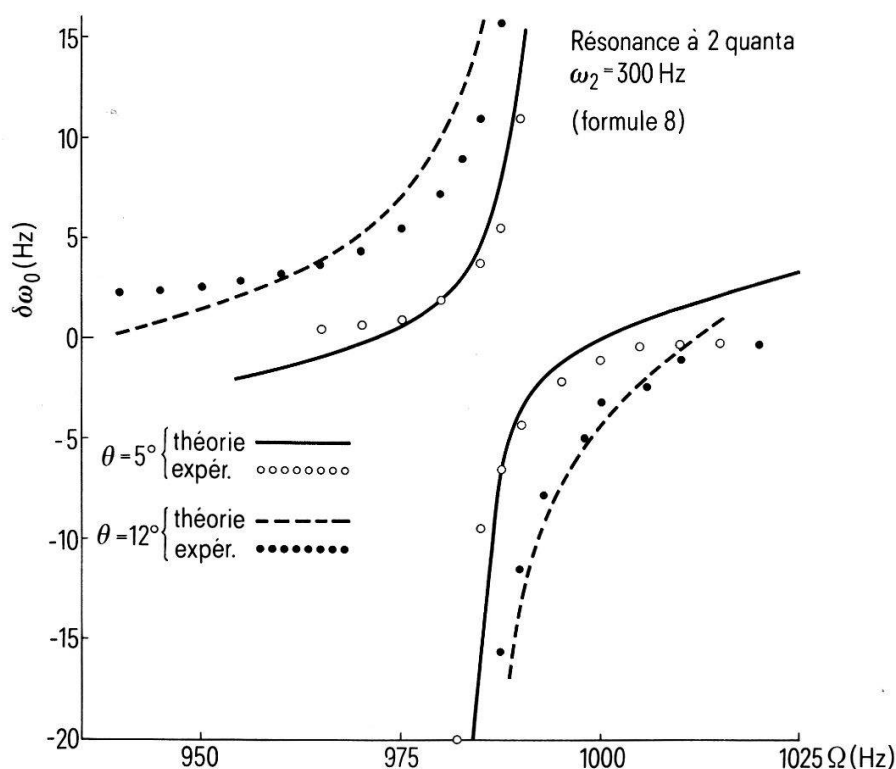


Figure 3

4⁰) Largeur de la raie pour les transitions à 2 et 3 quanta

Le calcul montre que la largeur de la raie évolue en fonction de θ selon les fonctions:

$\sin 3\theta$ pour la transition à 3 quanta et
 $\sin 2\theta$ pour la transition à 2 quanta.

Le calcul détaillé en a été donné ailleurs [1] nous nous contenterons ici de remarquer que ce calcul est en accord avec les points expérimentaux et confirme l'hypothèse faite dans une publication précédente [5].

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Relaxation magnétique nucléaire protonique de liquides pleuraux en champ faible. Application au diagnostic médical

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Abstract. The measurements of relaxation times in the weak field of the protons in the water of pleural liquids, obtained by punctions on patients under treatment, distinguishes clearly between two kinds of liquids. In the first category 'the transudates' the relaxation rate increases linearly with the concentration of proteins. Thus, knowing the proportionality factor, we can evaluate the proportion and the correlation time of the water molecules fixed to proteins.

In the second category 'the exudates', produced in particular by cancers, this linear law is not obeyed. In this case, the pleural liquid contains relatively high concentrations of leucocytes and lipoproteins.

Because of the weak amplitude of the applied fields, this technique could be applied in situ without danger for the patients.

I. Introduction

Il est bien connu que l'amplitude du signal, la grandeur des temps de relaxation qui caractérisent l'évolution de l'aimantation de noyaux atomiques (ici, des protons) dépendent étroitement de la concentration de ces nuclides dans l'échantillon et de leur environnement statique et dynamique.

La plupart des tissus biologiques des êtres vivants contiennent de l'eau en proportion plus ou moins grande. Particulièrement, les liquides du corps humain sont des solutions aqueuses d'ions minéraux et de protéines pouvant contenir en suspension plus ou moins de cellules.

On comprend donc que les grandeurs que permettent de déterminer les diverses techniques du magnétisme nucléaire (intensité du signal, temps de relaxation, constantes de diffusion . . .) soient très sensibles à la composition de tels liquides et à son évolution dans les cas pathologiques.

Le magnétisme nucléaire – surtout lorsqu'il est observé en champ faible – apparaît donc comme une approche simple, sans danger, permettant l'identification in situ d'un tissu vivant donné: elle est toute indiquée pour le diagnostic médical [1].

L'objet de cette communication est de présenter, à titre d'illustration, la mesure de temps de relaxation de liquides humains – pour la plupart, extraits par ponction de la plèvre de malades – leur analyse, en vue de l'identification possible d'états pathologiques.

II. Technique expérimentale

La mesure est faite par la technique bien connue de précession libre des protons dans le champ magnétique terrestre (0.05 m Tesla) après prépolarisation [2] dans un champ perpendiculaire de plus haute intensité (5 m Tesla). Le signal de précession libre, dans nos conditions expérimentales, est une exponentielle de laquelle on peut directement déduire le temps de relaxation spin-spin T_2 dans le champ terrestre.

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L'amplitude maximum M de ce signal dépend de la durée t de polarisation, passant évidemment d'une valeur voisine de 0 pour $t = 0$ à M_p (aimantation d'équilibre dans le champ polarisant) pour $t \gg T_1$ temps de relaxation spin-milieu. La variation $M(t)$ est encore une exponentielle dont la constante de temps est précisément le temps de relaxation spin-milieu dans le champ polarisant. On sait que la grandeur de T_1 , mesuré pour une valeur H_0 du champ magnétique, dépend étroitement de la grandeur relative du (ou des) temps de corrélation τ_i , caractérisant le milieu et de la période de Larmor ν_0^{-1} des noyaux dans le champ H_0 [3]. Or les solutions aqueuses de protéines ont des molécules d'eau dont le temps de corrélation se trouve précisément dans le domaine ν_p^{-1} des périodes de Larmor correspondant aux valeurs H_p du champ de polarisation. Nous disposons donc de deux paramètres (T_1 et T_2) très sensibles à la nature et à la concentration des protéines en solution.

III. Résultats expérimentaux

La figure 1 résume les résultats les plus significatifs portés sur un graphique où on a représenté $(T_1)^{-1}$ et $(T_2)^{-1}$ en fonction de la concentration en protéines des liquides étudiés.

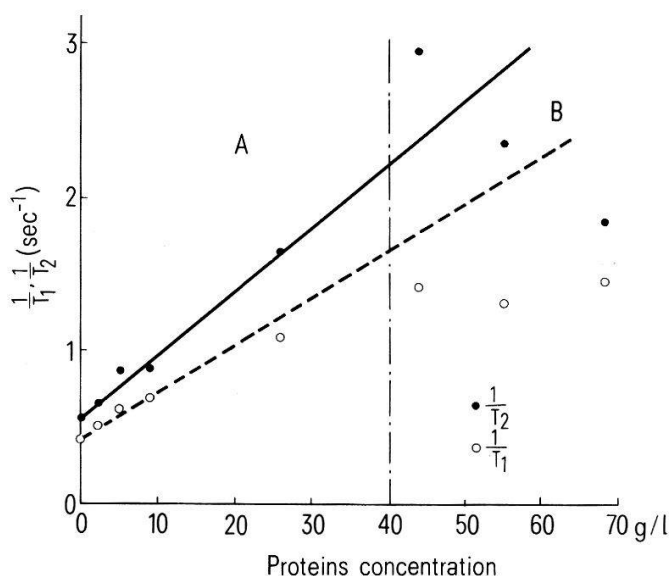


Figure 1

Le tableau I précise la nature des échantillons étudiés et leur concentration en protéines. Pour la clarté de l'analyse, on a porté sur le diagramme :

- les résultats obtenus avec les liquides pleuraux,
- les temps de relaxation de la solution ionique isotonique de base qui est le 'solvant' de tous les liquides examinés,
- et, à titre de comparaison, les résultats obtenus avec deux autres liquides du corps humain (liquide péritonéal et liquide amniotique).

Tableau I

Nature du liquide	Concentration en protéines (grammes/litre)
Solution ionique isotonique	0
Liquide amniotique sain [4]	2
Liquide pleural (insuffisance cardiaque)	4.5
Liquide péritonéal (cirrhose)	8.75
Liquide pleural (décompensation cardiaque)	26
Liquide pleural (cancer du sein)	44
Liquide pleural (tumeur de la plèvre)	55
Liquide pleural (lymphome malin avec chylothorax)	68

IV. Interprétation

On voit clairement sur la Figure 1 deux zones:

I. Une zone A dans laquelle on note une proportionnalité entre les taux de relaxation et la concentration en protéines. Cette zone correspond, en ce qui concerne les liquides pleuraux, à des *transudats*, et pour l'ensemble des liquides à de simples solutions de protéines dans la solution ionique de base.

Cette proportionnalité a été mise en évidence par O. K. Daszkiewicz et al. [5] pour des solutions aqueuses d'ovalbumine. Ces auteurs interprètent leurs résultats par l'existence de deux types de molécules d'eau:

- (a) celles du solvant,
- (b) celles qui sont liées aux protéines et qui ont le même temps de corrélation que ces dernières.

Comme il y a un échange très rapide entre les molécules d'eau des catégories (a) et (b), on n'observe qu'un seul temps de relaxation. De la pente des droites obtenues, on peut déduire la grandeur du temps de corrélation τ_c ainsi que la proportion W des molécules d'eau fixées aux protéines.

Avec des hypothèses de travail analogues à (5) on trouve

- $T_c = 1.6 \cdot 10^{-7}$ sec, valeur en bon accord avec les résultats obtenus pour des protéines de poids moléculaire 70 000 environ
- $W = 4\%$.

II. La zone B correspond à des *exsudats* de liquide pleural:

- plus concentré en protéines
- riche en leucocytes
- pouvant contenir des lipo-protéines. Dans ce cas, les taux de relaxation s'écartent des droites de la zone A.

Des travaux sont en cours pour une analyse détaillée des temps de relaxation de cette zone.

Un dispositif, en cours de construction, permettra la mesure 'in situ' des temps de relaxation des liquides du corps humain avec une précision suffisante [6].

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Physique Nucléaire et Corpusculaire

Comparison of π^+ and π^- elastic scattering off ^{40}Ca and ^{48}Ca showing the possibility to extract a neutron radius for ^{48}Ca

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Elastic and inelastic angular distributions at 130 MeV for π^+ and π^- incident on ^{48}Ca as well as ^{40}Ca were obtained with the SIN pion spectrometer. Significant differences in the positions of the minima and the values of the cross sections in the vicinity of these minima were observed by comparison of π^+ and π^- elastic scattering off ^{48}Ca . The ^{48}Ca elastic data is shown in Figure 1.

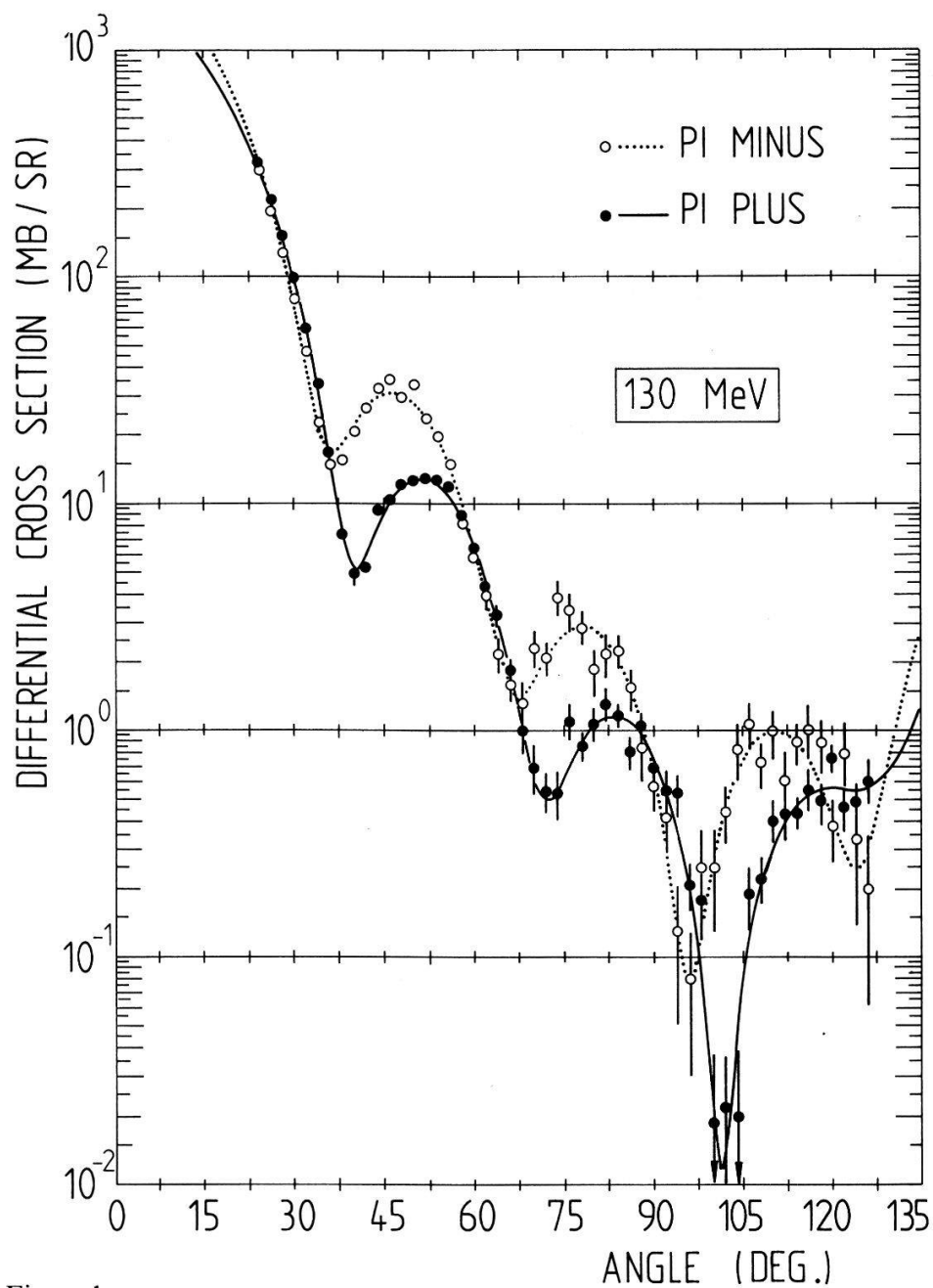


Figure 1

Comparison of π^+ and π^- — ^{48}Ca elastic scattering differential cross sections at 130 MeV versus the pion scattering angle in the lab system. The curves result from a fit of the scattering amplitude depending on the complex zeros, the forward value and a slope parameter.

Search for the metastable $2s$ state in muonic hydrogen

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Results of an experiment to search for the metastable $2s$ state in muonic hydrogen at 150 and 600 Torr are presented. About 6% of all μp are initially formed in the $2s$ state. Most of these $(\mu p)_{2s}$ decay in less than 100 nsec; assuming a calculated lifetime of 2 μ sec at 150 Torr (0.5 μ sec at 600 Torr) for a long-living fraction f of all $2s$ states, f is less than 4% (16%).

Charged particle emission following negative pion absorption in nuclei

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Stopped negative pions are absorbed by an ensemble of few correlated nucleons of the target nucleus. Therefore, the study of stopped pion reactions may provide information about nucleon clustering in a nucleus as well as pair correlations. Two experimental approaches are employed, firstly, the identification of the residual nucleus by its de-excitation γ -rays, and secondly, measurements of single and coincidence spectra of neutral and charged fragments emitted after the absorption.

Using the $\pi E3$ beam at SIN we have measured the single spectra of p , d , t , $He3$ and $He4$ emitted from $C12$, $CO59$ and $Au 197$ targets, as well as the spectrum of Li in the case of $C12$. The spectra have been corrected for self absorption in the target. Angular correlations of charged particles measured in coincidence will also be presented.

Pion-nucleus scattering in the alpha-particle model

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Pion elastic scattering by 4N-nuclei in the region of the first pion-nucleon resonance is calculated within the optical limit of Glauber theory on the basis of the naive alpha-particle model [1]. The pion-alpha scattering amplitudes needed as input are obtained from data by an amplitude analysis depending primarily on the nearby complex zeros [2]. One-body nuclear form factors and short range correlations between pairs of alpha particles are parametrized to reproduce electron scattering data. When account is taken of the Coulomb distortion and focussing effects [3] the

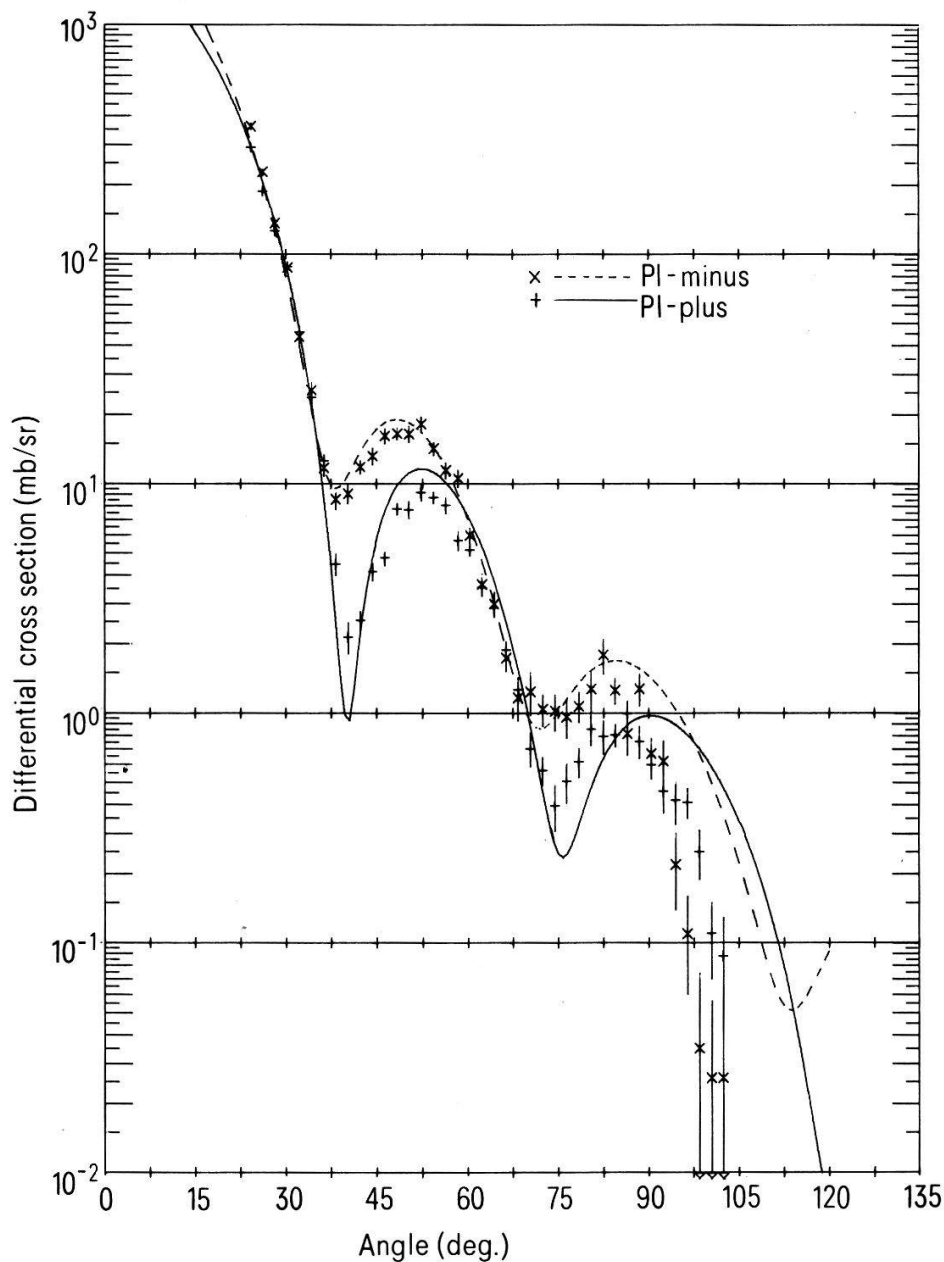


Figure 1
 $\pi - {}^{40}\text{Ca}$ elastic scattering at 130 MeV, the experimental points from the Neuchâtel group [4] being compared with predictions of the α -particle model within Glauber theory.

predicted cross sections agree quite well with the new accurate π^+ and π^- SIN data on ^{40}Ca [4, 5]. For ^{16}O the agreement of experiment [5] with theory cannot be considered as satisfactory, the discrepancy being as large as 50% in the diffraction peak. Except for a possible uncertainty in the absolute normalisation of the data [5], this suggests that the Glauber theory used is not valid for such a light nucleus. Therefore further data on 4N-nuclei between ^{16}O and ^{40}Ca are still needed to confirm that π -nucleus scattering can be consistently described by an alpha-cluster model.

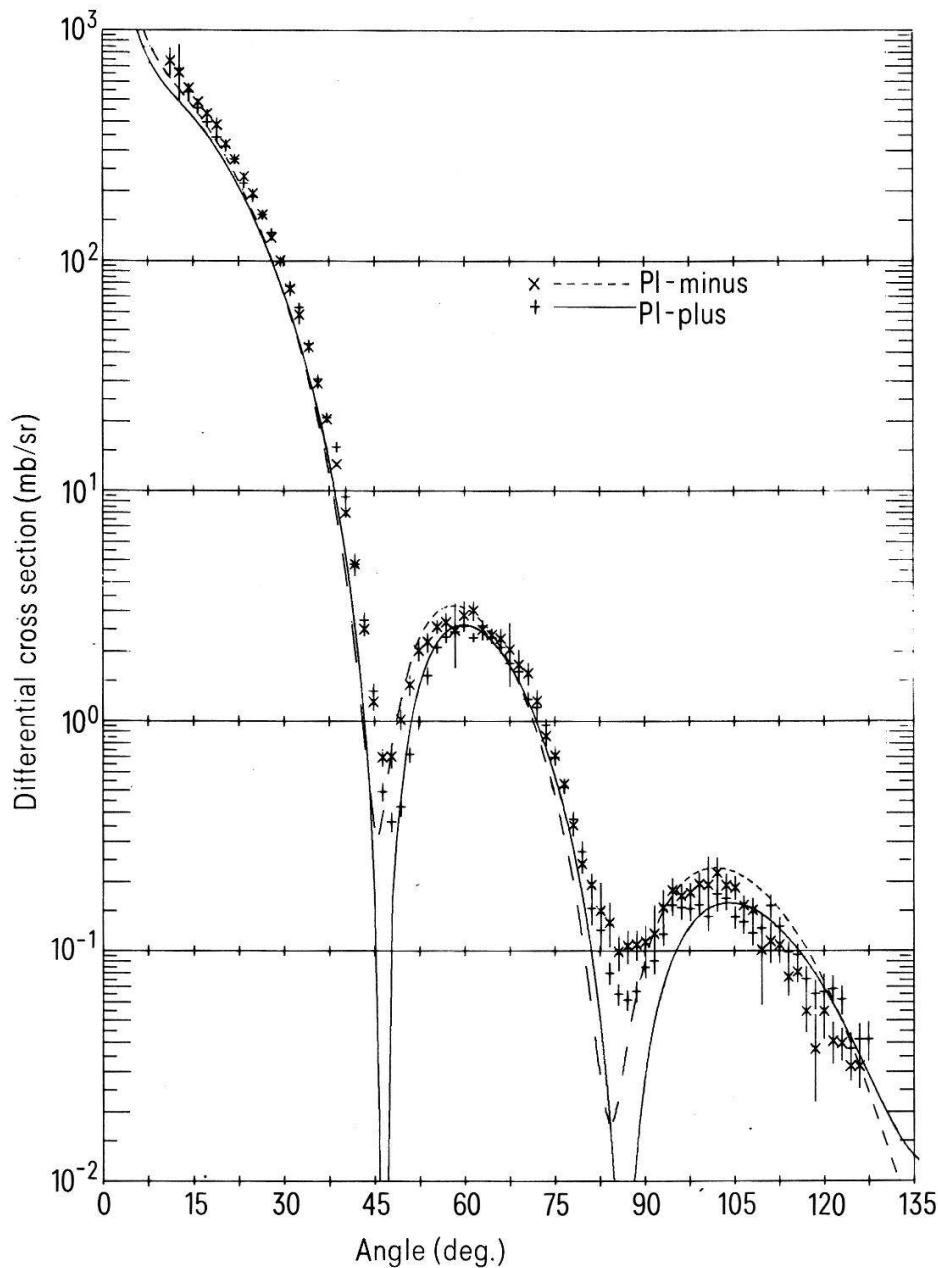


Figure 2

$\pi - ^{16}\text{O}$ elastic scattering at 163 MeV, the preliminary experimental points from the Grenoble – SIN – Karlsruhe collaboration [5] being compared with predictions of the α -particle model within Glauber theory.

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Observation of the ^{18}Ne ground state in the pion double charge exchange reaction $^{18}\text{O}(\pi^+, \pi^-)^{18}\text{Ne}$

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Les premiers résultats ont été présentés dans *Phys. Letters* 69B, 301 (1977).

Détermination des paramètres de l'interaction neutron-neutron a_{nn} et r_{nn} , à partir du spectre de photons de la réaction $\pi^- + {}^2\text{H} \rightarrow \gamma + 2n$

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Le spectre des photons issus des 2 réactions: $\pi^- + {}^1\text{H} \rightarrow \gamma + n$ et $\pi^- + {}^2\text{H} \rightarrow \gamma + 2n$ ont été mesurés à l'aide du spectromètre à paires du SIN et d'une cible remplie, alternativement, d'hydrogène et de deutérium liquide. La réponse du spectromètre est déterminée à partir de la raie de photons de capture par l'hydrogène ($E_\gamma = 129.4$ MeV, résolution : 0.9 MeV fwhm). La comparaison, près de l'énergie maximale ($E = 131.4$ MeV), du spectre expérimental de capture par le deutérium avec les spectres théoriques convolués par la réponse du spectromètre permet de déterminer les valeurs des paramètres a_{nn} et r_{nn} . L'expérience a été réalisée dans des conditions telles que l'incertitude expérimentale, σ , sur l'extraction de a_{nn} soit de l'ordre de 0.3 fm, améliorant sensiblement les résultats antérieurs ($\sigma > 1$ fm).

En l'état actuel de l'analyse, la valeur obtenue de a_{nn} est comprise entre 17.8 et 18.1 fm suivant le domaine d'énergie des γ considérés pour la comparaison. L'évaluation de l'erreur théorique correspondante étant en cours, il n'est pas possible de donner une valeur définitive. Les auteurs de la référence [1] indiquent une erreur théorique inférieure à 0.3 fm. Cette erreur est établie pour une expérience complète

dans laquelle seuls sont considérés les événements dont l'impulsion relative des 2 neutrons est inférieure à 35 MeV/c. Dans notre cas la résolution du spectromètre nous impose d'échantillonner des impulsions relatives plus grandes (45 MeV/c au minimum).

De même, l'erreur expérimentale sur la valeur de r_{nn} est de l'ordre de 0.1 fm mais celle-ci varie entre 2.8 et 3.1 fm suivant le domaine d'énergie considéré.

Le remarquable accord (Figure 1) entre le spectre théorique et expérimental, jamais obtenu avec les modèles antérieurs, permet d'espérer que l'erreur théorique restera proche de celle indiquée.

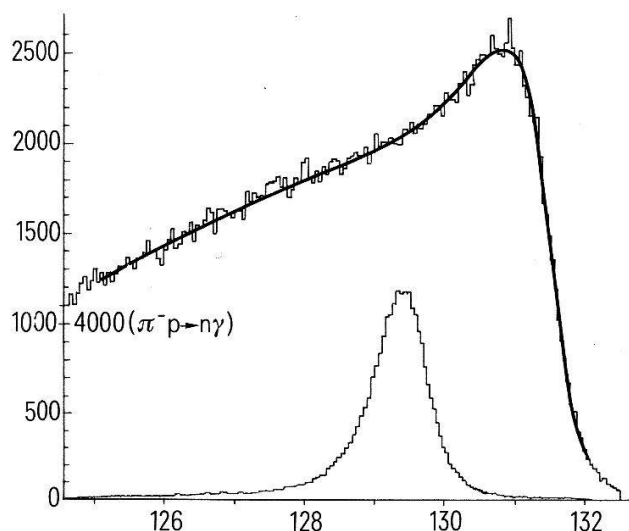


Figure 1

Spectre des photons de la réaction $\pi^- d \rightarrow nn\gamma$ avec ajustement réalisé entre 129 et 132 MeV du spectre théorique (référence 1) Spectre des photons de la réaction $\pi^- p \rightarrow n\gamma$.

Ces résultats toujours préliminaires remplacent ceux ($16.6 < a_{nn} < 17.2$ fm) publiés dans les Proceedings de la 2e Conférence Internationale de Vancouver (27-30 juin 1977) sur l'interaction nucléon-nucléon. La différence provient d'une erreur dans la valeur du champ magnétique et d'une approximation de l'ordre de E_γ/m_π dans les spectres théoriques.

Ces résultats sont en accord avec la valeur de a_{nn} que l'on peut dériver de la valeur de a_{pp} , après correction des effets électromagnétiques et après correction de la brisure de la symétrie de charge par le mélange des mésons $\rho - \omega$ et $\pi - \eta$. Ils diffèrent par contre de la valeur expérimentale actuellement admise pour a_{nn} (-16.4 ± 0.9 fm).

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K-Röntgenwirkungsquerschnitte bei Schwerionenkollisionen

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Die Wirkungsquerschnitte der schwerioneninduzierten K-Röntgenstrahlung wurden als Funktion der Projektilenergie für verschiedene symmetrische ($Z_{\text{Proj}} =$

Z_{Target}) und asymmetrische ($Z_{\text{Proj}} \leq Z_{\text{Target}}$) bestimmt. Ein Vergleich mit den verschiedenen Theorien zeigt, dass der Lochproduktionsmechanismus immer noch nicht vollständig verstanden wird.

Experimentelle Untersuchungen an Spiegelreaktionen zur Bestimmung einer möglichen Ladungssymmetrieverletzung

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Für die Spiegelreaktionen $D(d, p)T$ und $D(d, n)^3\text{He}$ wurden bei sieben Energien zwischen 2,5 und 11,5 MeV die Analysationsvermögen iT_{11} , T_{20} , T_{21} und T_{22} mit einem Fehler von weniger als 0,005 bestimmt. Die Messungen an der Reaktion $D(d, n)^3\text{He}$ wurden durch Detektion der Rückstoss- ^3He -Teilchen ausgeführt. Zwischen beiden Reaktionen treten kleine, aber signifikante Unterschiede auf, deren Energieabhängigkeit für die einzelnen Komponenten verschieden ist. Eine genaue Analyse dieser beiden Reaktionen, unter Berücksichtigung des langreichweitigen Teils der Coulomb-Wechselwirkung, sollte zeigen, ob die gemessenen Unterschiede allein durch Coulomb-Effekte zu erklären sind, oder ob sie durch eine mögliche Ladungssymmetrieverletzung entstehen.

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Neutronenemission bei Spaltung hochangeregter Kerne

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Es werden Messungen von Pre- und Post-Spaltneutronen in Abhängigkeit der Fragmentmasse und deren kinetischer Energie beschrieben. Man erhält Kenntnisse über die Energiebilanz am Abtrennpunkt und über $\Gamma n/\Gamma f$.

Preequilibrium-alpha-emission in the $^{27}\text{Al}(p, \alpha x)$ -reaction at $E_p = 50$ MeV

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Preequilibrium processes in nuclear reactions have first been explained by Griffin [1] with the so-called Exciton Model and this idea has been further developed and extended by other authors [2]. The energy distribution of the emitted nucleons and recently even their angular distribution [3] have been predicted satisfactorily. The problem of the emission of complex particles is in the focus of recent investigations, experimentally as well as theoretically. Such studies are expected to deliver valuable information on the cluster structure of nuclei.

We report on preliminary results of our program of investigations on α -particle emission induced by proton bombardment. We have measured the α -particle spectra induced by 49.36 MeV protons on an ^{27}Al -target using the proton beam of the SIN injector cyclotron. The experimental arrangement is shown schematically in Figure 1.

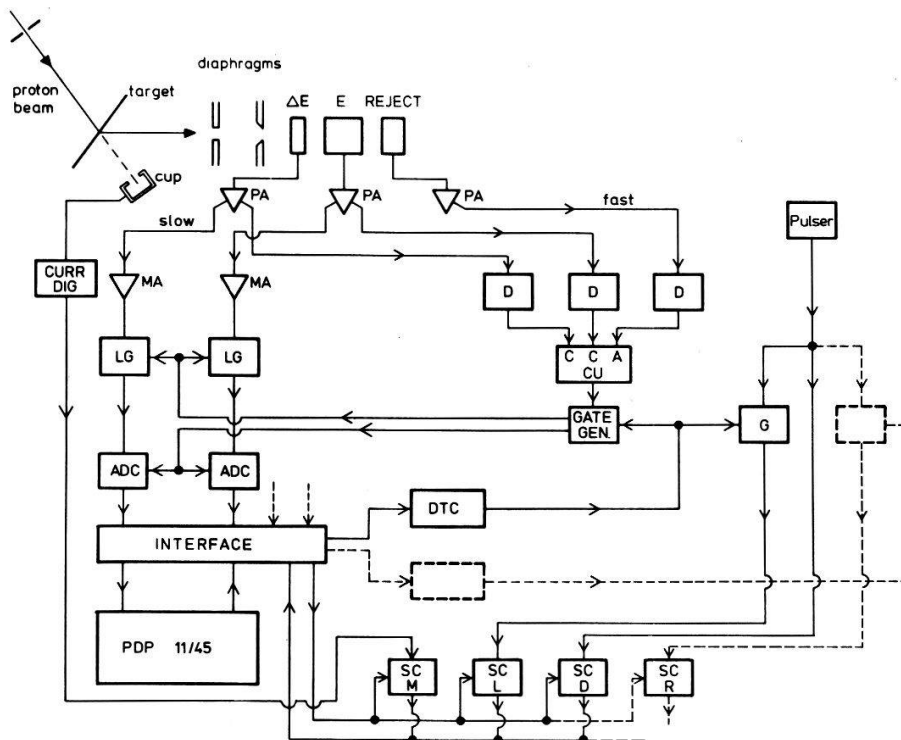


Figure 1

Experimental arrangement. Only one Si-telescope is shown. The distance between target and solid-angle defining diaphragm is 176 mm.

The proton beam was focussed onto the $250 \mu\text{g}/\text{cm}^2$ thick aluminium target with a spot diameter of approx. 3 mm. Two Si-detector telescopes were mounted on arms which could be moved independently in a scattering chamber. The particles are identified by their signals produced in the ΔE - and E -detectors with a thickness of $50 \mu\text{m}$ (or $100 \mu\text{m}$) and $2000 \mu\text{m}$, respectively. Each of the Si-detectors was equipped with two α -sources for permanent check and energy calibration. These α -sources together with the discrete transitions to known (p, α) final states allowed the calibration of the whole energy range of interest.

The electronic system was designed for high count rates and was corrected for dead-time losses. The detector signals were routed into a fast and a slow branch, the fast channels controlling the slow ones. After AD-conversion the signals of both telescopes were analysed on-line by a PDP-11 computer. A simple and fast procedure was used for particle identification. The α - and τ -spectra were stored in a two-dimensional array, recorded on magnetic tape and analysed off-line. The measure-

ments were performed at 9 angles from 20° to 145° . The stability and the reproducibility in this experiment were very good. Typical results are shown in Figure 2, where the spectra of the τ -particles at 20° and of the α -particles at 20° and 145° are shown. The spectra have a pronounced structure in the high energy region.

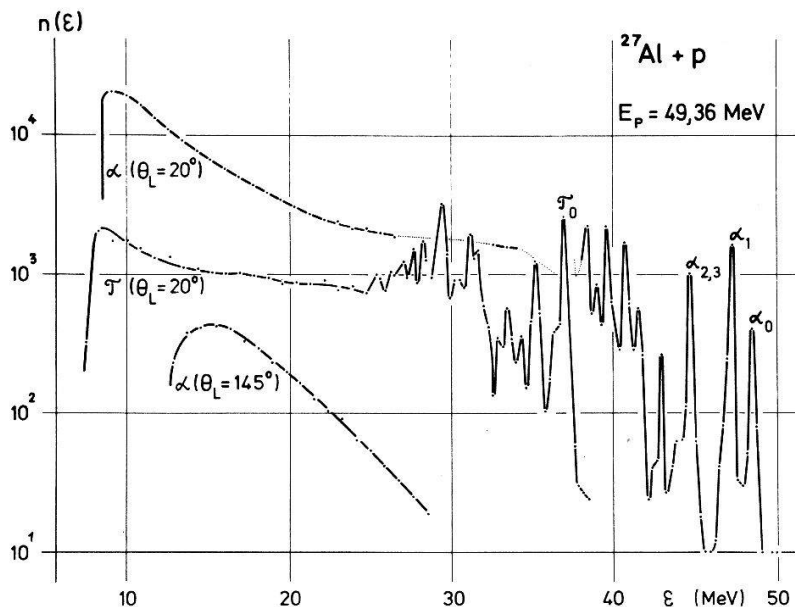


Figure 2
Typical α - and τ -spectra. The ordinate values are given in relative units.

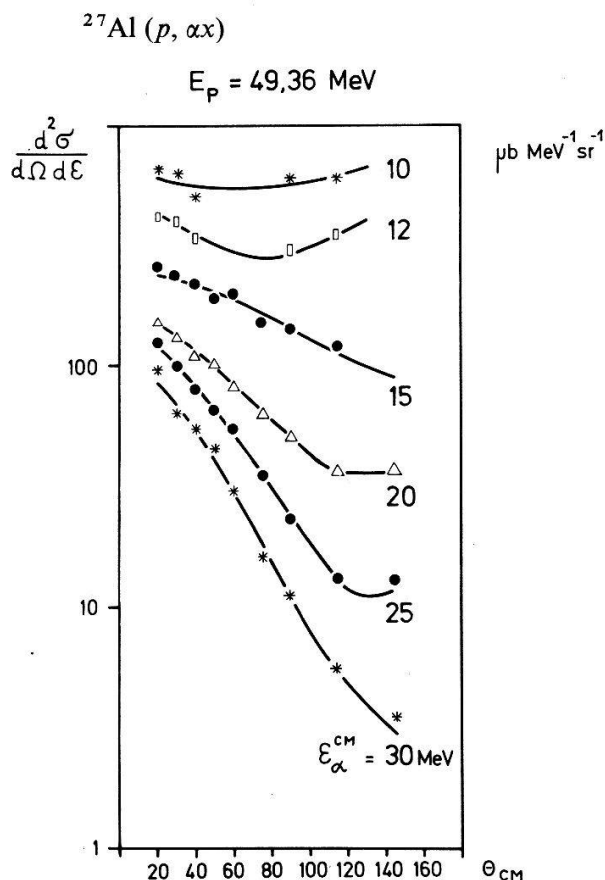


Figure 3
Typical example of angular distributions at six α -energy sections. The numbers give the energy of the 1 MeV section considered. The curves are Legendre-polynomial fits.

The spectra were averaged over a 1 MeV range, transformed to the CM system and the corresponding 1 MeV sections from each angle were fitted with Legendre-polynomials. Some examples of angular distributions of these energy sections are shown in Figure 3. Differential cross-sections at 26 α -energies between 10 and 35 MeV have been used in the final analysis to obtain the angle-integrated cross section for α -emission.

This data is compared with the prediction of the Quasi-Free Scattering Model (QFS) [4] and with the results of a Monte Carlo calculation for an Exciton Model [5]. For the QFS calculation an α -particle break-up probability of 50% and an α -Fermi-energy of 4 MeV were assumed. In the Monte Carlo approach the preequilibrium as well as the evaporation component is considered. Successive emission of up to 8 (in the preequilibrium stage up to 4) particles, n , p and α , is taken into account. For both models best agreement for a cluster factor of $\varphi = 0.10$ is obtained. Figure 4 shows the comparison of our results with the theoretical predictions.

The agreement is good except at the high energy end of the spectrum where direct reactions predominate and the models loose their validity. The cluster factor from our analysis coincides with the one reported by L. Milazzo-Colli et al. [6] ($\varphi = 0.11$) for the same nucleus at a proton energy of 36.5 MeV.

We would like to thank Dr. W. Scobel from the University of Hamburg for the QFS-calculation and Prof. E. Gadioli from the University of Milano for stimulating discussions and for providing us with his MC-computer code.

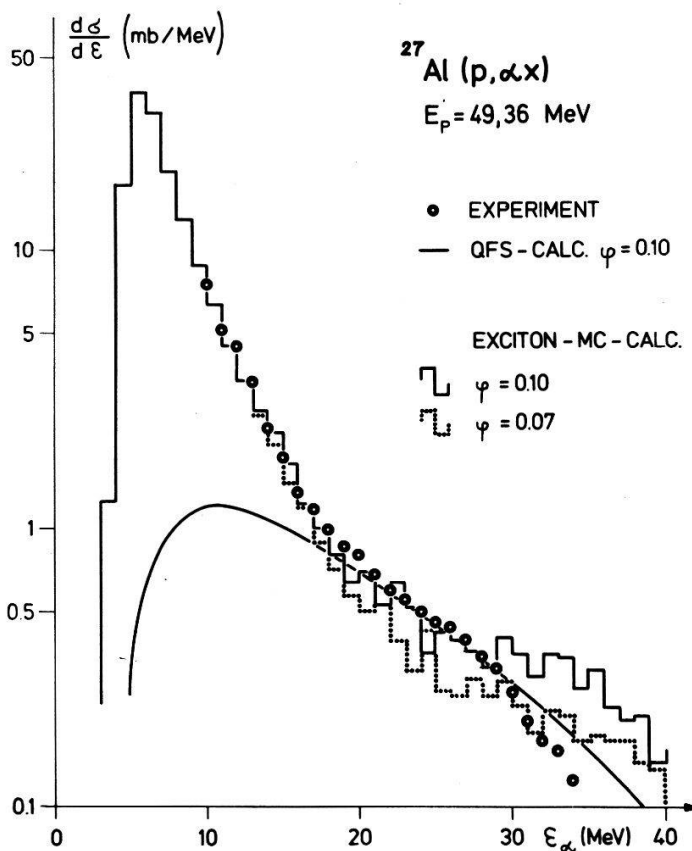


Figure 4
Angle-integrated $^{27}\text{Al}(p, \alpha x)$ cross section. The QFS-calculation gives the preequilibrium component only, no evaporation term has been added. The Monte-Carlo Exciton calculation is shown for two values of the α -cluster factor.

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Dispersionsrelationen und Kernreaktionen

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Es hat sich als äusserst fruchtbar erwiesen, Streuamplituden von Kernreaktionen als analytische Funktionen der (komplexen) kinematischen Variablen aufzufassen. Aus der dispersionstheoretischen Analyse empirischer Reaktionsamplituden erhält man unter recht schwachen Annahmen (Kausalität) praktisch modellunabhängig spektroskopische Information (Kopplungskonstanten) über leichte Kerne. Wir diskutieren den dabei auftretenden Problemkreis anhand der $p + d$ und $p + {}^3\text{He}$ Streuamplituden [1].

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Bedeutung von Hadron-Kern Wirkungsquerschnitten in der Bestimmung von Kern-Neutronenverteilungen

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Kürzlich sind in vermehrtem Mass Hadron-Kern Streu- und Reaktionsdaten zur Bestimmung der radialen Neutronenverteilung $\rho_n(r)$ des Targetkerns beigezogen worden. Dabei wird vorausgesetzt: (i) bekannte Protonenverteilung $\rho_p(r)$, (ii) Reaktionsmodell, ausgehend von $\rho_n(r)$, $\rho_p(r)$ und bekannten Projektil-Nukleon-Amplituden (z.B. Glauber-Modell). Wir untersuchen im Rahmen des Glauber-Modells, welche Eigenschaften von $\rho_n(r)$ in die Observablen (insbesondere totale Wirkungsquerschnitte σ_{tot}) eingehen. Es wird gezeigt, dass der 'r.m.s.-Radius' von $\rho_n(r)$ aus σ_{tot} nur dann bestimmt werden kann, wenn Annahmen über die Form von $\rho_n(r)$ gemacht werden. Diese Einschränkung muss physikalischer Natur sein. Falls die Form von $\rho_n(r)$, wie üblich, durch die Wahl einer Fermi-Verteilung eingeschränkt wird, ist der bestimmte r.m.s.-Radius abhängig von der gewählten Parametrisierung.

Radiale Verteilung von Valenz-Neutronen in Kernen

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Wir zeigen, dass elastische, magnetische Elektronenstreuung hoher Multipolarität (M7, M9) zur Bestimmung der radialen Wellenfunktionen von Neutronen (und Protonen) benutzt werden kann. Im Gegensatz zu den bisher mit stark-wechselwirkenden Sonden durchgeführten Untersuchungen von Neutronendichten $\rho_n(r)$ kann damit zum ersten Male eine saubere und präzise Information betreffend $\rho_n(r)$ erhalten werden. Wir diskutieren die in Zusammenarbeit mit der Saclay-Gruppe durchgeführten Messungen am ^{87}Sr , ^{49}Ti , die daraus bestimmten radialen Wellenfunktionen $R(r)$ und den Vergleich mit DDHF-Rechnungen. Die Implikationen der zwischen $1f_{7/2}$, $1g_{9/2}$ Protonen und Neutronen gefundenen Unterschiede in $R(r)$ bezüglich der Coulombenergie-Anomalie werden ebenfalls untersucht.

Nuclear charge radii from muonic X-ray transitions in F, Al, Si, P and K

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Energies of muonic X-rays of the K -series of five light natural elements (F, Al, Si, P and K) have been determined with an accuracy of 15 to 30 eV. Root mean square radii $\langle r^2 \rangle^{1/2}$ and equivalent charge radii R_k have been deduced, employing refined calculations of the Lamb shift and the nuclear polarization. As an example, we find for Si $\langle r^2 \rangle^{1/2} = 3.129(3)$ fm and $R_k = 4.017(4)$ fm. The results are in good agreement and of higher accuracy than comparable electronic scattering data. The experiment has been carried out at the SIN muon channel.

A microscopic analysis of the low lying levels in ^{86}Sr with the reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$

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¹⁾ Work supported by the Swiss National Science Foundation.

²⁾ Work supported by Fonds zur Förderung der wissenschaftlichen Forschung.

Abstract. The reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ has been measured at $E_p = 34$ MeV. DWBA-calculations using microscopic form factors have been performed for the transitions to the ground and first excited state in ^{86}Sr . Wave functions with configuration mixing were investigated. It is shown that not only the amplitudes but also the relative signs of the components in these wave functions are important to obtain good fits to the measured angular distributions.

Introduction

As was shown in References [1, 2] the microscopic analysis of (p, α) reactions is very sensitive to the wave functions used. Not only the amplitudes but also the relative signs of the components of different configurations in wave functions can be tested by the coherence properties of the (p, α) reaction [3]. This feature is utilized in our study of the $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ reaction to obtain information about the nuclear structure of the residual nucleus ^{86}Sr .

Little is known about the ground and first excited state of ^{86}Sr . Shell model calculations have been performed in the Sr-region but in a restricted $2p_{1/2} - 1g_{9/2}$ subspace only [4, 5]. However, some studies of reactions, leading to the residual nucleus ^{86}Sr , i.e. the $(^3\text{He}, d)$ reaction by Schneider et al. [6], the (p, t) reaction by Ball et al. [7] and the (p, α) reaction by Fulmer and Ball [8], indicate that the wave function of the ^{86}Sr ground state is more complicated than these shell model calculations predict. The most detailed wave function is given by Morten et al. [9] who deduced the amplitudes for $2p_{3/2}$, $2p_{1/2}$ and $1g_{9/2}$ configuration from the observed excitation strengths of pure neutron hole states by the reaction $^{86}\text{Sr}(d, p)^{87}\text{Sr}$. But this one-particle-transfer reaction does not test the relative signs of the corresponding components in the wave function.

Our experiment was performed with the SIN injector cyclotron using an energy-analysed proton beam of 12 keV resolution at an energy of 34 MeV. This energy was chosen to keep compound nucleus contributions as low as possible. Self-supporting targets of ^{89}Y with a thickness of $300 \mu\text{g}/\text{cm}^2$ were used.

DWBA analysis

We relate the experimental to the theoretical cross section as follows

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{exp}} = \varepsilon \left(\frac{d\sigma}{d\Omega}\right)_{\text{DWBA}}.$$

The DWBA-analysis has been performed with the code DWUCK [10]. In the zero range approximation the DWBA transition matrix element for the reaction $p + A = \alpha + B$ can be written as [11]

$$T_{p\alpha}(M_B, M_\alpha = 0, M_A, M_p) = \sum_{\substack{LJ\gamma \\ MM_J}} (J_B M_B J M_J | J_A M_A) (L M S_p - M_p | J M_J) S_{AB}^{1/2}(\gamma, L, S_p, J)$$

$$\int d\mathbf{R} \chi_{k'}^{(-)*} \left(\frac{M_B}{M_A} \mathbf{R} \right) F_M^{\gamma, L}(R) \gamma_L^M(\theta, \phi) \chi_k^{(+)}(\mathbf{R}).$$

$F_M^{\gamma L}$ is the three-nucleon form factor which we have calculated with the code written by Falk [12]. The nuclear structure information is contained in the three-nucleon spectroscopic amplitude $S_{AB}^{1/2}$. Using shell model wave functions – as will be discussed below for the target and residual nuclei – we have calculated these spectroscopic amplitudes $S_{AB}^{1/2}$ following the methods of Glendenning [13] and Towner and Hardy [14] formulated for the two-nucleon transfer processes. Since absolute cross sections are not correctly predicted by the zero-range DWBA calculation an overall absolute normalization factor was deduced from the requirement that the average value of the enhancement factor ε becomes unity.

For the proton and alpha optical potential we used the same parameter sets as in our study of the reaction $^{92}\text{Zr}(p, \alpha)^{89}\text{Y}$ [1] reproducing the elastic scattering cross section data of $^{90}\text{Zr}(p, p)$ at $E_p = 40$ MeV and $^{92}\text{Zr}(\alpha, \alpha)$ at $E_\alpha = 34.4$ MeV respectively. The bound nucleon wave functions $\phi_m^{nlj}(\mathbf{r}) = (u^{nlj}(r)/r)y_l^m(\theta, \varphi)$ were calculated for Saxon-Woods potentials with parameters reproducing the neutron and proton separation energy of ^{89}Y and the level splitting of the $p_{1/2}$ and $p_{3/2}$ single particle states in the Zr-region [15]. In our calculations we used the convention that the single particle wave functions have a positive slope at the origin.

Results and discussion

A typical alpha-particle spectrum of the reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ at $E_p = 34$ MeV and $\theta_{\text{Lab}} = 30^\circ$ is shown in Figure 1.

The wave function of the ^{89}Y ground state is fairly well known from the calculations of Vergados and Kuo [16]. The proton configuration is a nearly pure $2p_{1/2}$ one particle state, whereas the neutron configuration is taken as a closed shell with $N = 50$.

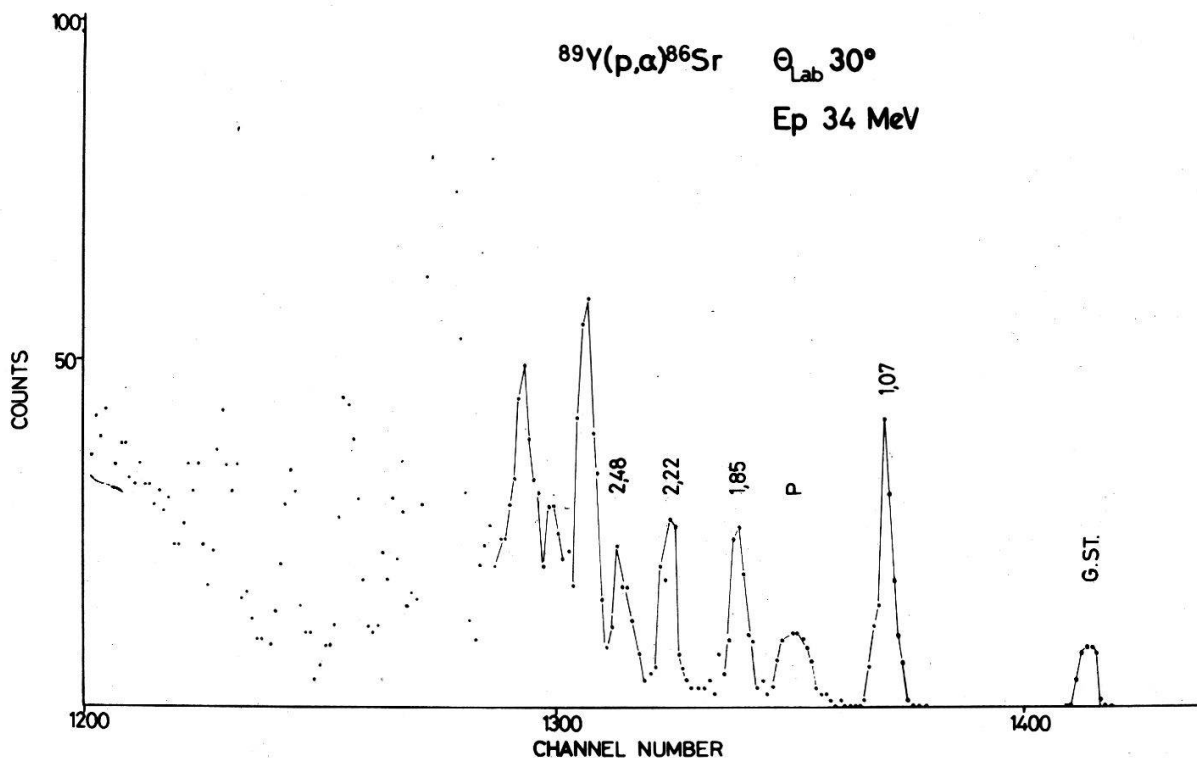


Figure 1
Alpha-particle spectrum of the reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ at $E_p = 34$ MeV and $\theta_{\text{Lab}} = 30^\circ$.

For the ^{86}Sr ground state Morton et al. [9] give the following term for the neutron part of the wave function:

$$|^{86}\text{Sr}\rangle_{0^+ \text{g.s.}} = a|1g_{9/2}^{-2}\rangle + b|2p_{3/2}^{-2}\rangle + c|2p_{1/2}^{-2}\rangle.$$

The protons are assumed to fill the $2p_{3/2}$ subshell. With the values of Morton et al. [9] (Set I) $a = +0,93$, $b = 0,25$ and $c = +0,27$ we have calculated the cross sections and the results are shown in Figure 2 (curve -·-·-). Set II for the amplitudes of Morton et al. [9] does not alter the results significantly. We tried to improve the fits by varying the signs of the amplitudes in the wave function. As demonstrated in Figure 2 (curve ---) the choice of $a = 0,93$, $b = -0,27$ and $c = -0,25$ gives a very good fit to the experimental data.

For the first excited state of ^{86}Sr (2^+ , $E_x = 1.07$ MeV) Ball et al. [7] and Schneider et al. [6] suggest that the main component in the neutron wave function is $\nu(g_{9/2}^{-2})_{2^+}$ configuration. However, it follows from our analysis that by including a $(2p_{3/2}^{-2})_{2^+}$

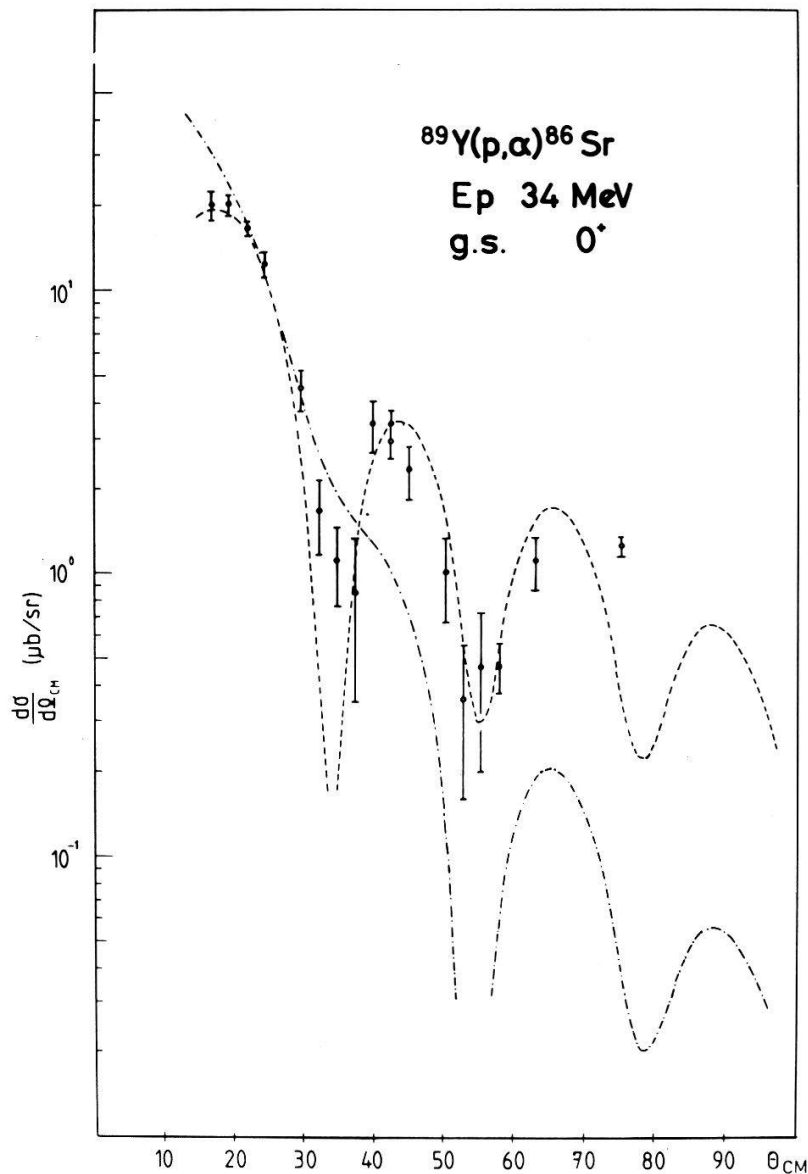


Figure 2

Angular distribution of the reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ leading to the ground state of ^{86}Sr with the wave function $a|1g_{9/2}^{-2}\rangle + b|2p_{3/2}^{-2}\rangle + c|2p_{1/2}^{-2}\rangle$ for the neutron configuration. $a = +0.93$, $b = +0.25$, $c = +0.27$: curve (-·-·-); $a = +0.93$, $b = -0.25$, $c = -0.27$: curve (---).

configuration of the same strength as in the ground state wave function for the $(2p_{3/2}^{-2})_{0+}$ – part a better agreement between theory and experiment can be obtained. The cross section calculated with the wave function

$$|^{86}\text{Sr}\rangle_{2+} = a'|1g_{9/2}^{-2}\rangle_{2+} + b'|2p_{3/2}^{-2}\rangle_{2+}$$

where $a' = 0.97$, $b' = -0.25$ and $a'^2 + b'^2 = 1$ is shown in Figure 3 (curve —). Again, a positive sign for the $(2p_{3/2}^{-2})$ – amplitude gives a bad fit to the data (Figure 3, curve - - - -). The enhancement factors ε extracted for the transitions to the ground and first excited states in ^{86}Sr are 0.7 and 1.3 respectively. We conclude from these values that we have taken the main components of the wave function into account. For further progress it would be important to have wave functions available which were calculated in an enlarged subshell-space.

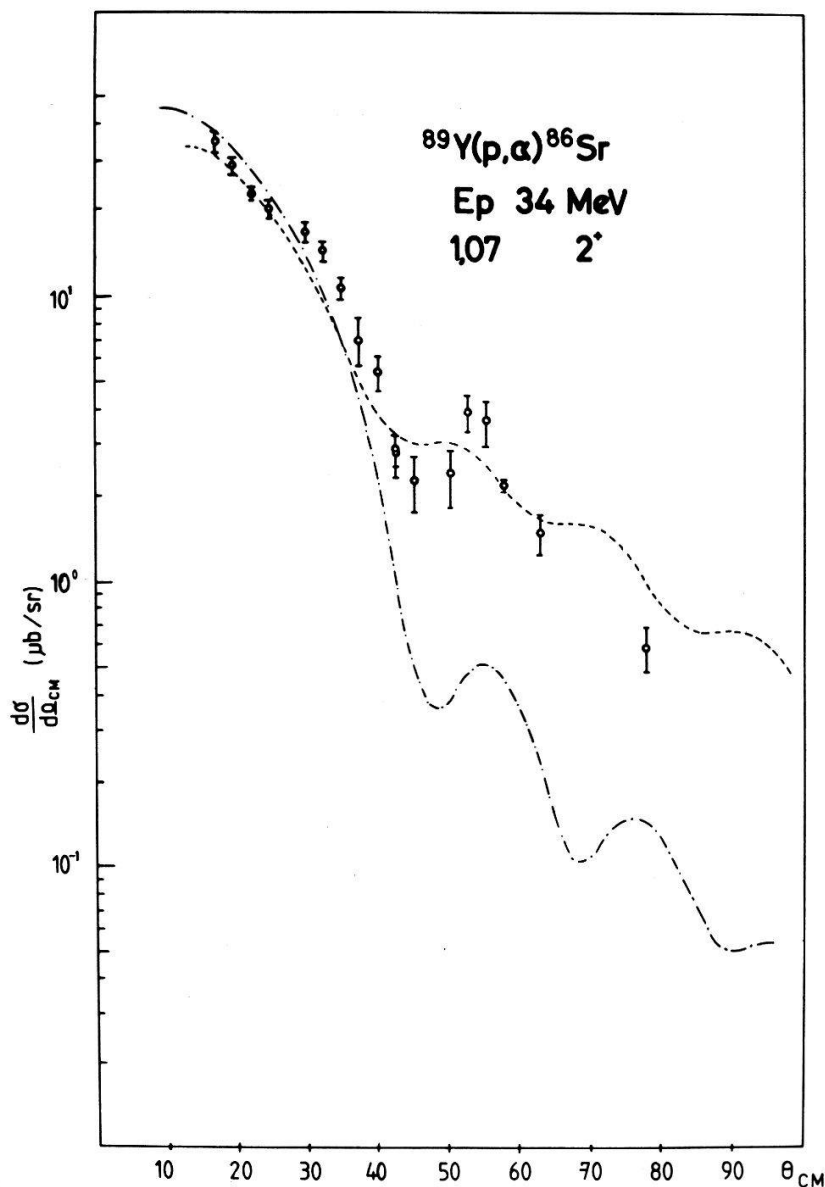


Figure 3

Angular distribution of the reaction $^{89}\text{Y}(p, \alpha)^{86}\text{Sr}$ leading to the first excited 2^+ state at $E_x = 1.07$ MeV with the wave function $|^{86}\text{Sr}\rangle_{2+} = a'|1g_{9/2}^{-2}\rangle + b'|2p_{3/2}^{-2}\rangle$ for the neutron configuration. $a' = +0.97$; $b' = +0.25$: curve (- - - -); $a' = +0.97$; $b' = -0.25$: curve (—).

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Anomale Rückwärtsstreuung von α -Teilchen

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Gegenstand unserer Untersuchung ist das vor allem bei ^{40}Ca beobachtete starke Ansteigen des Wirkungsquerschnitts für elastische Streuung von α -Teilchen bei Rückwärtswinkeln. Am SIN-Injektor haben wir Winkelverteilungen der elastischen α -Streuung an ^{40}Ca und ^{58}Ni im Bereich 27 bis 49 MeV in 0.5 bzw. 1 MeV-Schritten gemessen. Die Streuwinkel liegen zwischen 120° und 172° für $^{58}\text{Ni} + \alpha$ und zwischen 30° und 172° für $^{40}\text{Ca} + \alpha$ und drei ausgewählte Energien von $^{58}\text{Ni} + \alpha$. Die gemessenen Wirkungsquerschnitte zeigen bei grossen Streuwinkeln eine starke Energieabhängigkeit. Gegenwärtig versuchen wir diese Daten mit Hilfe eines schwach von der Energie abhängigen optischen Potentials und eines zusätzlichen Regge-Pols zu beschreiben.

Suche nach dem seltenen Myonen-Zerfall $\mu^+ \rightarrow e^+ \gamma$

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C. PETITJEAN, H. PFEIFFER, H. P. POVEL, M. SALZMANN, A. VAN DER SCHAAF, F. SCHLEPÜTZ,
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Der Zerfall $\mu^+ \rightarrow e^+ \gamma$ ist wegen der Erhaltung der Myonzahl verboten. Die Messmethode basiert auf der folgenden kinematischen Bedingung: e^+ und γ müssen kollinear sein und gleiche Energien $E_e = E_\gamma = m_\mu/2 = 52.8$ MeV haben. Das Experiment wurde am Myonenkanal des SIN durchgeführt. Der Aufbau des Experiments und die Analyse der Daten wird beschrieben. Eine neue obere Grenze für

das Verzweigungsverhältnis $\Gamma(\mu - e^+ \gamma)/\Gamma(\mu - e^+ \nu e \bar{\nu}_\mu)$ von 1.1×10^{-9} (90% confidence) wird angegeben.

Théorie

Sur l'énergie de cohésion du Jellium

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Pour le modèle du Jellium tridimensionnel avec conditions aux bords périodiques, on recherche pour quelles valeurs du paramètre r_s l'état cristallin est énergétiquement plus favorable que l'état de densité électronique uniforme. Pour cela, on évalue la valeur moyenne de l'hamiltonien du Jellium dans différents déterminants de Slater formés soit à partir d'ondes planes, soit à partir d'ondes de Bloch construites par combinaison linéaire d'orbitales électroniques.

Soliton-like properties of heat pulses

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Y. HIWATARI

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Using the molecular-dynamics technique, we establish soliton-like properties of heat pulses in a lattice-dynamic model when the ambient temperature is in the second-sound regime.

On the dielectric response of the one dimensional OCP

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(Laboratoire de Physique Théorique EPF-L – Department de Physique EPF-L – C.R.P.P.)

(Communication retirée).

Threshold-phase transition analogy

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Abstract. We demonstrate close analogy between the threshold properties of an overdamped Brownian particle in a periodic potential and applied field, being far from thermal equilibrium and the critical properties close to continuous phase transitions.

In recent years it has been corroborated that phase transition-like phenomena are not the privilege of systems in thermal equilibrium, but may also occur in open

systems. Examples are the laser [1, 2], instabilities in fluid dynamics [3] (the Bénard and Taylor problems), the Gunn instability near threshold [4], the tunnel diode [5], and chemical reactions [6]. For a review of the analogies between phase transitions occurring in thermal equilibrium and transitions in non-equilibrium systems, we refer to Reference [3].

In this work, we study the threshold phase-transition analogy on the basis of a Langevin equation, describing the Brownian motion of a particle in a periodic potential, driven by a static external field. The corresponding equation is

$$\dot{p} = -B \sin ax + E + \Gamma p - \eta(t), \quad (1)$$

where p is the momentum, x the position, Γ the damping, and E the external field. B and a are model parameters. We assume that the random force $\eta(t)$ has a white spectrum, i.e.,

$$\langle \eta(t) \rangle = 0 \quad (2)$$

$$\langle \eta(t)\eta(t') \rangle = 2\Gamma M k_B T \delta(t - t'),$$

where M denotes the mass of the particle. Equation (1) also describes a Josephson junction in series with a large external resistance and battery [7, 8]. Moreover, it represents a simple model for superionic conductance [9].

Our aim will be to derive the mean velocity as a function of temperature and field E . It will enable us to calculate the threshold phase-transition analogy. To derive exact results, we consider the overdamped situation only. In this case, the Langevin equation (1) reduces to

$$\Gamma p = -\sin x + E + \eta(t) = \Gamma \dot{x}, \quad (3)$$

where we have chosen $B = a = M = 1$. The associated Fokker-Planck equation for the distribution function $w(x, t)$ is the Smoluchowski equation

$$\frac{\partial w}{\partial t} = \frac{1}{\Gamma} \frac{\partial}{\partial x} \left(\frac{\partial u}{\partial x} w \right) + \frac{k_B T}{\Gamma} \frac{\partial^2 w}{\partial x^2} = -\frac{\partial A}{\partial x}, \quad (4)$$

where

$$u = -\cos x - Ex. \quad (5)$$

We only need to consider the value of x modulo 2π , so we may restrict x to $0 \leq x \leq 2\pi$, and adopt periodic boundary conditions to equation (4).

The steady-state solution of equation (5) must have A constant. Furthermore, if

$$\int_0^{2\pi} w_0(x) dx = 1, \quad (6)$$

then A^{-1} is the average time for the particle to diffuse one periodicity length, i.e., 2π . The mean velocity is then given by

$$\langle \dot{x} \rangle = 2\pi A. \quad (7)$$

The required solution of equation (4) is then [7]

$$w_0(x) = \frac{A\Gamma}{k_B T} \frac{f(x)}{f(2\pi) - f(0)} \left[f(0) \int_0^x \frac{dx'}{f(x')} + f(2\pi) \int_x^{2\pi} \frac{dx'}{f(x')} \right], \quad (8)$$

where

$$f(x) = \exp \left[\frac{1}{k_B T} (\cos x + Ex) \right]. \quad (9)$$

The normalization of $w_0(x)$ [equation (6)] then requires

$$1 = \frac{A\Gamma}{k_B T} \left[\left(\exp \left(\frac{2\pi E}{k_B T} \right) - 1 \right)^{-1} \left(\int_0^{2\pi} f(x) dx \int_0^{2\pi} \frac{1}{f(x)} dx \right) - \int_0^{2\pi} f(x) dx \int_x^{2\pi} \frac{dx'}{f(x')} \right]. \quad (10)$$

The curves plotted in Figure 1 have been obtained from a numerical integration of this formula. Analytic forms revealing the threshold phase-transition analogy can be obtained in several limiting cases:

$$\Gamma \langle \dot{x} \rangle = (E + 1)^{1/2} (E - 1)^{1/2} : T = 0, \quad (11)$$

$$\Gamma \langle \dot{x} \rangle = 3^{5/6} \frac{\Pi^{1/2}}{\Gamma(1/6)} T^{1/3} : E - E_c = 1, \quad (12)$$

$$\Gamma \frac{d\langle \dot{x} \rangle}{dT} = 0 : T = 0, E \neq E_c \quad (13)$$

$$\Gamma \frac{d^2 \langle \dot{x} \rangle}{dT^2} = (E^2 + 1/4)(E^2 - 1)^{-5/2} : T = 0, E \gtrsim 1 \quad (14)$$

$$\Gamma \langle \dot{x} \rangle = (E^2 - 1)^{1/2} \left[1 - \frac{1}{8} \frac{T^2(4E^2 + 1)}{(E^2 - 1)^3} \right]^{-1} : \begin{cases} T \approx 0 \\ E \gtrsim 1 \end{cases}, \quad (15)$$

$$\Gamma \langle \dot{x} \rangle = \sqrt{2} (1 - E)^{1/2} \exp \left[-\frac{4\sqrt{2}}{3} (1 - E)^{3/2} \frac{1}{T} \right] : \begin{cases} T \approx 0 \\ E \lesssim 1 \end{cases}, \quad (16)$$

where $k_B = 1$. From the power laws [equations (11)–(14)], it is obvious that close to the threshold $E = E_c = 1$, there is a close analogy to a continuous phase transition. Confining this analogy to a magnetic system, identifications can be made as shown in Table 1.

Table 1

Threshold model	Magnet	
$\langle \dot{x} \rangle / T = 0 \sim (E - E_c)^{1/2}$	$M H = 0 \sim (T - T_c)^\beta$	Order parameter
$E - E_c$	$T - T_c$	
E : field	T : temperature	
E_c : threshold	T_c : critical temperature	
T	H	Field conjugate to order parameter
$\langle \dot{x} \rangle E = E_c \sim T^{1/3}$	$M/M T = T_c \sim H^{1/\delta}$	Equation of state
$\frac{d\langle \dot{x} \rangle}{dT} \Big _{E = E_c} = 0$	$\frac{dM}{dH} \Big _{H = 0} \sim T - T_c ^{-\gamma}$	Susceptibility
$\frac{d^2 \langle \dot{x} \rangle}{dT^2} \Big _{E = E_c} \sim (E - E_c)^{-5/2}$	$\frac{d^2 M}{dH^2} \Big _{H = 0} \sim (T - T_c)^{\beta - 2\beta\delta}$	

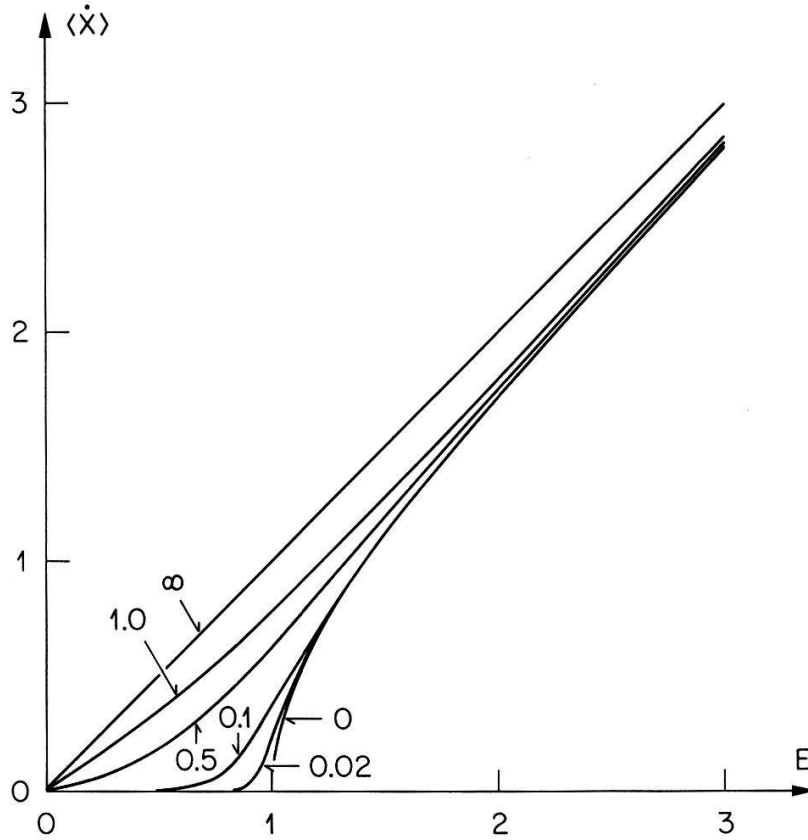


Figure 1
Velocity-field characteristic for various temperatures. $E = E_c = 1$ is the threshold field.

β , γ and δ are critical exponents. The above comparison reveals that the analogy between the behavior close to threshold and the critical phenomena close to a continuous phase transition is very close. Note, however, that the threshold model is far from thermal equilibrium for finite temperatures. Moreover, the present model exhibits classical critical exponents, i.e., $\beta = 1/2$, $\delta = 3$. To complete the analogy, we also note that according to equations (15) and (16), the prediction of the scaling hypothesis, [10]

$$\langle \dot{x} \rangle = |E - E_c|^\beta F_\pm(T|E - E_c|^{-\beta\delta}), \quad (17)$$

is also satisfied ($\beta = 1/2$, $\beta\delta = 3/2$). The subscript $\pm$ denotes $E - E_c > 0$ and $E - E_c < 0$, respectively. Equation (17) predicts that if we scale $\langle \dot{x} \rangle$ by $|E - E_c|^{1/2}$, and if we scale T by $|E - E_c|^{3/2}$, then plots $\langle \dot{x} \rangle |E - E_c|^{1/2}$ versus $T|E - E_c|^{-3/2}$ should be the same for all values of T . The analytic [equations (15) and (16)] and numerical [equations (7) and (10)] results are shown in Figure 2, for $E > E_c$, $E = E_c = 1$ and $E < E_c$. It is seen that by approaching E_c from $E = E_c + 0$, or $E = E_c - 0$, equation (17) is satisfied. We also note that $F_+(x) \sim x^2$ and $F_-(x) \sim \exp(-\alpha/x)$ for small x [see equations (15) and (16)], so that the susceptibility vanishes. This differs from $F_\pm(x) = ax + bx^2 + \dots$, expected in equilibrium phase transitions [10].

This analysis can be extended to more general periodic potentials, where the $\sin x$ term in equation (3) is replaced by $g(x)$, satisfying the conditions $g(x) = -g(-x)$ and $g(z/2 - x) = g(x)$, where z is the period. The analytical behavior near the absolute maximum of $g(x)$ determines the critical exponents. In fact, if $g(u) = g_{\max}$ has a zero of p th order, we find $\beta = (p - 1)/p$ and $\delta = (p + 1)/p - 1$.

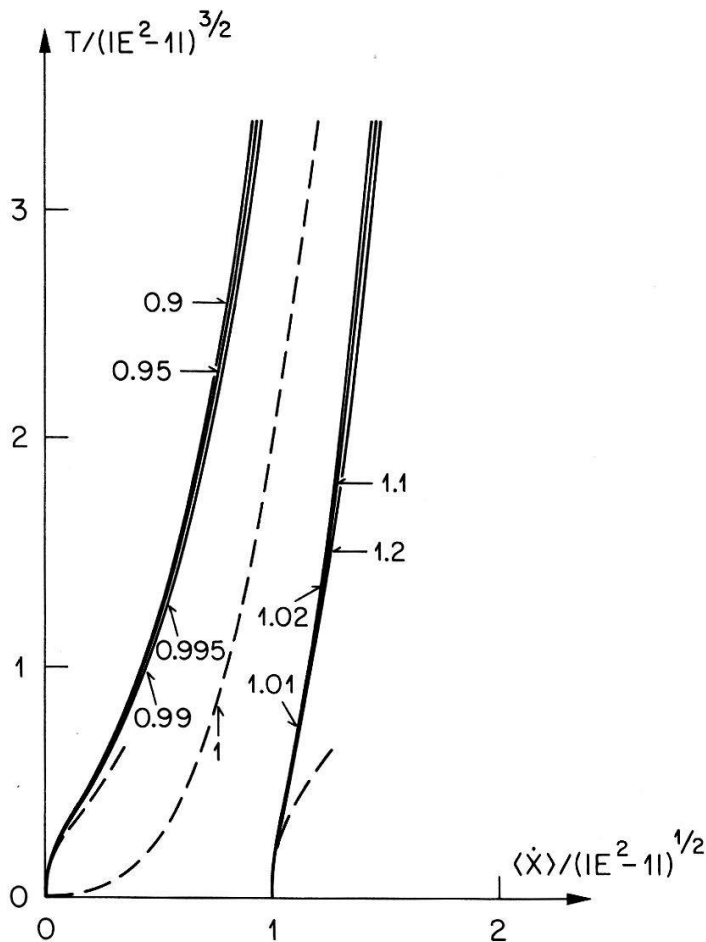


Figure 2

Scaled mean velocity is plotted against scaled temperature to demonstrate the scaling properties of equation (18). The dashed curves represent the asymptotic behavior given by equations (13), (16) and (17).

In conclusion, we have shown that the overdamped Brownian motion of a particle in a periodic potential, driven by a static external field, exhibits threshold phenomena in close analogy to critical behavior close to continuous phase transitions. It should be emphasized however that this system is far from equilibrium. It also represents a simple model for superionic conduction and a Josephson junction. We hope to deal with the underdamped and interacting version of the model in a future paper.

This work was initiated by discussions with A. R. Bishop. We also acknowledge stimulating discussions with C. P. Enz, W. Dietrich, H. Haken, R. Morf, and R. Risken.

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Aspect dynamique de la transition de phase supraconductrice

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On étudie la dynamique du système de BCS en interaction faible avec un bain thermique par la théorie des équations maîtresses. L'analyse des équations différentielles (non linéaires) qui régissent l'évolution des observables macroscopiques dans la limite thermodynamique met en évidence une bifurcation qui se produit à la température critique. La construction explicite d'une fonction de Liapounov permet une description détaillée du comportement des solutions.

End effects in long current-carrying superconducting channels

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Our continuing study of one-dimensional solutions of the time-dependent Ginzburg–Landau equations revealed new features beyond those reported earlier [1].

- (1) If the relaxation time ratio $u = \tau_\psi/\tau_j < 1$, no sharp transition between boundary superconductivity and localized phase slip occurs at the current J_c corresponding to a stationary SN boundary; the dc I–V characteristic is reversible and smooth above the maximum supercurrent $J_m \simeq J_c$.
- (2) If the channel connects two bulk superconductors with sufficiently lower T_c , J_m may be depressed below J_c even if $u > 1$; phase slips localized near the ends occur if between J_m and J_c .

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Real space renormalization group for the 1 and 2 dimensional XY model

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The critical behaviour of the 1 and 2 dimensional classical XY model is investigated using a variational renormalization group approach. Albeit easily solvable, the 1 dimensional case is interesting because it exhibits some of the particularities present in 2 dimensions. For the 2 dimensional case, we found a critical fixed point describing the onset of long-short-range order in the system. The results for the critical temperature and the critical indices are compared with those from series expansion and other renormalization group approaches. Finally, the possibility to produce in our framework a line of fixed points describing the low temperature phase is discussed.

Upper bounds to the free energy of the one-component plasma

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H. de Witt has recently proposed a simple interpolation formula for the free energy of the OCP on the basis of J. P. Hansen's Monte-Carlo data. We apply here a newly developed method of optimal cell decomposition to calculate upper bounds to the free energy of this system. In the strong coupling limit, the best bound turns out to lie a few 0/00 above De Witt's values [1].

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Evaluation of path integrals for the semiclassical propagator

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The Green's function (or propagator) for a non relativistic quantum mechanical system is written as a path integral. The semiclassical approximation is evaluated by expansion around the classical paths. Algorithms are presented to compute the absolute value of the reduced propagator associated with each classical path [1], as well as their relative phases [2]. As a tool, general product formulas for eigenvalues of differential operators are derived [3].

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Theoretical investigation of the local and the global components of the neutron noise field in a boiling water reactor

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In view of recent experimental work the neutron noise in a BWR is believed to be separable into a local and a global component. It is the existence of the local com-

ponent which makes possible the measurement of steam velocity by correlating the signals of axially placed incore neutron detectors. In the present work (Nucl. Sci. Eng. 63, 306 (1977)) we use a one dimensional (axial) core model and solve the two group diffusion equations satisfied by the neutron noise. The solution is shown to be composed of two terms which can be identified as the theoretical counterparts of the components found in experiments. The properties of the two terms are discussed in the special case of an axially propagating disturbance of the moderator density (steam content).

Percolation : Scaling et exposants critiques

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En utilisant une image de blocs de clusters inspirée des blocs de spin de Kadanoff, nous dérivons diverses relations de scaling pour la percolation, en particulier pour la distribution de taille et de surface des clusters. Un groupe de renormalisation inspiré de cette image nous permet de calculer approximativement les exposants critiques à deux et à trois dimensions.

Winkelverteilungen der Nukleonen nach Pion-Absorption an ^3He

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Mit dem Modell von Phillips und Roig [1] haben wir die differentielle Uebergangsrate des Prozesses $\pi^- + ^3\text{He} \rightarrow p + n + n$ berechnet. Die Ergebnisse sind in qualitativer Uebereinstimmung mit der Theorie von Helwig [2], weichen jedoch dramatisch von einer Phasenraumverteilung ab. Wir zeigen, dass diese Abweichung klassisch erklärt werden kann. Dabei ist die einzige Annahme die, dass das Pion an einem Nukleonenpaar absorbiert wird. Das dritte (Spectator-) Nukleon hat dann vor und nach dem Ereignis eine Impulsverteilung, die durch die ^3He -Wellenfunktion bestimmt ist. Durch Aufzwingen dieser Nebenbedingung erhält man eine modifizierte Phasenraumverteilung, welche qualitativ mit dem Ergebnis der vollen Rechnung des Prozesses übereinstimmt. Wir schliessen daraus, dass die differentielle Uebergangsrate vor allem auf den Absorptionsmechanismus empfindlich ist.

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Proof of spontaneous magnetization in the d -dimensional Ising, XY- and Heisenberg- models, $d > 2$

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The existence of spontaneous magnetization has been proven for Ising- XY- and Heisenberg models in $d \geq 2$ dimensions. The results are achieved by the fact that the partition functions are monoton increasing in the coupling constants.

The Josephson motor

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Abstract. The Josephson equation for a local junction including the interference term is

$$\beta\ddot{\varphi} + \dot{\varphi}(1 + \gamma \cos \varphi) + \sin \varphi = \alpha,$$

which, in a mechanical analogue, can be interpreted as a torqued (α) pendulum (mass β) with anisotropic (γ) Stokes damping or driving. For the junction model, BCS theory gives $|\gamma| \leq 1$, i.e., a nowhere negative phase-dependent conductivity. Here the rotational solutions are discussed for $|\gamma| > 1$. It is shown that the pendulum behaves like a motor if $\gamma < -1$, i.e., α becomes negative (torque output) in a steady state.

The Josephson equation assuming a linear quasiparticle characteristic (Stoke's friction in the pendulum analogue) can be written as

$$\beta\ddot{\varphi} + \dot{\varphi}(1 + \gamma \cos \varphi) + \sin \varphi = \alpha, \quad (1)$$

where β is the capacitance (reciprocal friction), γ the ratio interference to quasiparticle current (anisotropic Stoke's friction parameter) and α the input current (torque) of a local junction (pendulum).

Its solution has been discussed in many papers (case $\gamma = 0$) [1–5], (case $|\gamma| \leq 1$ mostly) [6–9]. Here we should like to extend the solutions to $|\gamma| > 1$. In this case, the dissipative friction term becomes a driving term for certain angles. This condition has to be excluded in the Josephson-junction model since the BCS theory gives $|\gamma| \leq 1$, such that the phase-dependent conductivity is nowhere negative.

The mechanical analogue based on the Josephson equation still has a meaning and will shortly be called the Josephson motor (JM) because of a possible torque output. Falco [6] found an oscillatory solution for the JM if $\gamma < -1$. Under this condition, the stationary solution $\varphi_\alpha = \arcsin \alpha$ becomes instable for $|\alpha| < \alpha_\gamma = \sqrt{1 - 1/\gamma^2}$ and a limit cycle exists in the phase plane around the repulsor (point) $\dot{\varphi} = 0$, $\varphi = \varphi_\alpha$. Then the JM only vibrates within $-\varphi_\alpha - \pi < \varphi_{\min} < \varphi < \varphi_{\max} < \pi - \varphi_\alpha$, and the time average $\langle \dot{\varphi} \rangle$ becomes 0.

Here we are interested in rotational solutions of (1) with nontrivial characteristic $\omega = \langle \dot{\varphi} \rangle = \omega(\alpha, \beta, \gamma)$ or by inversion in $\alpha = \alpha(\omega, \beta, \gamma)$.

I. The general symmetry properties are $\alpha(-\omega, \beta, \gamma) = -\alpha(\omega, \beta, \gamma)$ (proved from (1) by $\varphi \rightarrow -\varphi$, $t \rightarrow t$) and $\alpha(-\omega, -\beta, -\gamma) = -\alpha(\omega, \beta, \gamma)$ (proved from (1) by $\varphi \rightarrow \varphi + \pi$, $t \rightarrow -t$).

II. The asymptotic expansions for $\beta \gg 1$ are

$$\alpha(\omega, \beta, \gamma) = \omega + \frac{\gamma}{2\beta\omega} + \frac{1}{2\beta^2\omega^3} - \frac{1}{\beta^3} \left[\frac{\gamma^3 + 4\gamma}{8\omega^3} + \frac{3\gamma}{16\omega^5} \right] - \frac{1}{\beta^4} \left[\frac{1}{2\omega^5} + \frac{9}{32\omega^7} \right] \dots \quad (2)$$

and the critical current $\alpha_c(\beta, \gamma) = \alpha(0, \beta, \gamma)$,

$$\alpha_c(\beta, \gamma)$$

$$= \frac{4}{\pi\sqrt{\beta}} \left[1 + \frac{\gamma}{3} - \frac{1}{\beta} (0.24751 + 0.07370\gamma + 0.04846\gamma^2 + 0.03688\gamma^3) \right] \dots, \quad (3)$$

where (3) is derived from Zilberman [10]. For $\gamma = -3$ especially, $\alpha_c(\beta, -3) = 32/(15\pi\beta^{3/2}) = 0.67906/\beta^{3/2}$.

III. The energy balance per cycle follows by integrating (1) multiplied by $\dot{\varphi}/T$ over one period giving $\langle \dot{\varphi}^2 \rangle + \gamma \langle \dot{\varphi}^2 \cos \varphi \rangle = \omega\alpha$, since in a steady state, kinetic energy and potential are unchanged during one cycle. If $|\gamma| \leq 1$, then $\langle \dot{\varphi}^2 \rangle + \gamma \langle \dot{\varphi}^2 \cos \varphi \rangle \leq (1 \pm |\gamma|) \langle \dot{\varphi}^2 \rangle$, since $|\cos \varphi| \leq 1$. Therefore, α becomes positive for all $|\gamma| \leq 1$.

IV. The characteristic for $\beta = 0$ and $|\gamma| \leq 1$ is $\omega = \sqrt{\alpha^2 - 1}$ independent of γ , but for $|\gamma| > 1$ it is (see Appendix)

$$\frac{1}{\omega} = \frac{1}{\sqrt{\alpha^2 - 1}} \left(1 - \frac{1}{\pi} \arctg \frac{\sqrt{\alpha^2 - 1}}{\alpha} \sqrt{\gamma^2 - 1} \right) + \frac{\gamma}{\pi} \text{ArTh} \frac{\sqrt{\gamma^2 - 1}}{\alpha\gamma} \quad (4)$$

depending on $|\gamma|$ but not on its sign (see reference [9]). The steady-state motion corresponds to a relaxation oscillation [1] (mass = 0) with discontinuity in the velocity (discontinuous oscillations [2]). The limit slope is $(\lim_{\alpha \rightarrow \infty} \omega/\alpha) = 1/(1 + (1/\pi)(\sqrt{\gamma^2 - 1} - \arctg \sqrt{\gamma^2 - 1}))$, tending from 1 to 0 for $|\gamma|$ from 1 to ∞ .

V. There exist steady-state rotational solutions with $\alpha_c(\beta, \gamma) < 0$, as a consequence of the asymptotic expansions; for example, if β is sufficiently large and $\gamma < -3$, α_c and therefore also a range of α values become negative. Since for $\beta \gg 1$ according to (2) all characteristics tend to the line $\omega = \alpha$ and because of continuity there exists an intersection $\omega_0(\beta, \gamma)$, where $\alpha = 0$.

VI. The characteristic has been computed numerically for $\beta = 10$ and γ 's ranging from -20 to 20 and the results are compiled in Figure 1. For $|\alpha| > 1$ a rotational solution always exists since $\alpha = \pm 1$ is the torque to overcome the restoring force maximum at $\varphi = \pm \pi/2$. For $\alpha \in (\alpha_\gamma, 1)$ and $(-1, -\alpha_\gamma)$ there are two solutions: the trivial stationary one with $\varphi = \varphi_\alpha$ (stable) and the rotational one; for $\alpha \in (-\alpha_\gamma, \alpha_\gamma)$ there are also two solutions: the oscillatory one with $\omega = 0$ (see reference 6) and the rotational solution.

The different behavior for $\gamma > 1$, where always $\alpha > 0$, and for $\gamma < -1$, where $\alpha < 0$ (JM) is possible can be understood qualitatively. If $\gamma > 1$, a Stoke's friction takes place for $\varphi \in (-\varphi_\gamma, \varphi_\gamma)$ ($\varphi_\gamma > \pi/2$), i.e., in the whole lower semicircle (SC), whereas driving acts only in the part $\varphi \in (\varphi_\gamma, 2\pi - \varphi_\gamma)$ of the upper SC and vice-versa for $\gamma < -1$. Therefore, the motion which by gravitation tends to be fast in the lower SC will there be drastically braked for $\gamma > 1$ and be accelerated by a driving force

which is proportional to the velocity for $\gamma < -1$. Since the acting φ range for the driving force increases from 0 (for $\gamma = -1$) to $(-\pi/2, \pi/2)$ for $\gamma = -\infty$, the JM will start moving (i.e., $\alpha < 0$), if γ is less than a critical value $\gamma_0(\beta)$ (for $\beta = 10$, $\gamma_0 \simeq -3.1$; from (3) follows, quite general, $\gamma_0(\beta) \simeq -3 - 1.6/\beta$, if $\beta \gtrsim 100$).

For $\gamma = \beta$ the characteristic coincides with the solution of $\beta = 0$, $\gamma = 0$ since (1) can then be written as $\beta(\varphi + \sin \varphi)^* + \varphi + \sin \varphi = \alpha$, which has the solution $\dot{\varphi} + \frac{\sin \varphi}{\sqrt{\alpha^2 - 1}} = \alpha$, i.e., (1) for $\beta = 0$, $\gamma = 0$ with the hyperbolic characteristic ($\omega = \sqrt{\alpha^2 - 1}$ dotted curve). For $\gamma > \beta$ the characteristics are beyond this hyperbola.

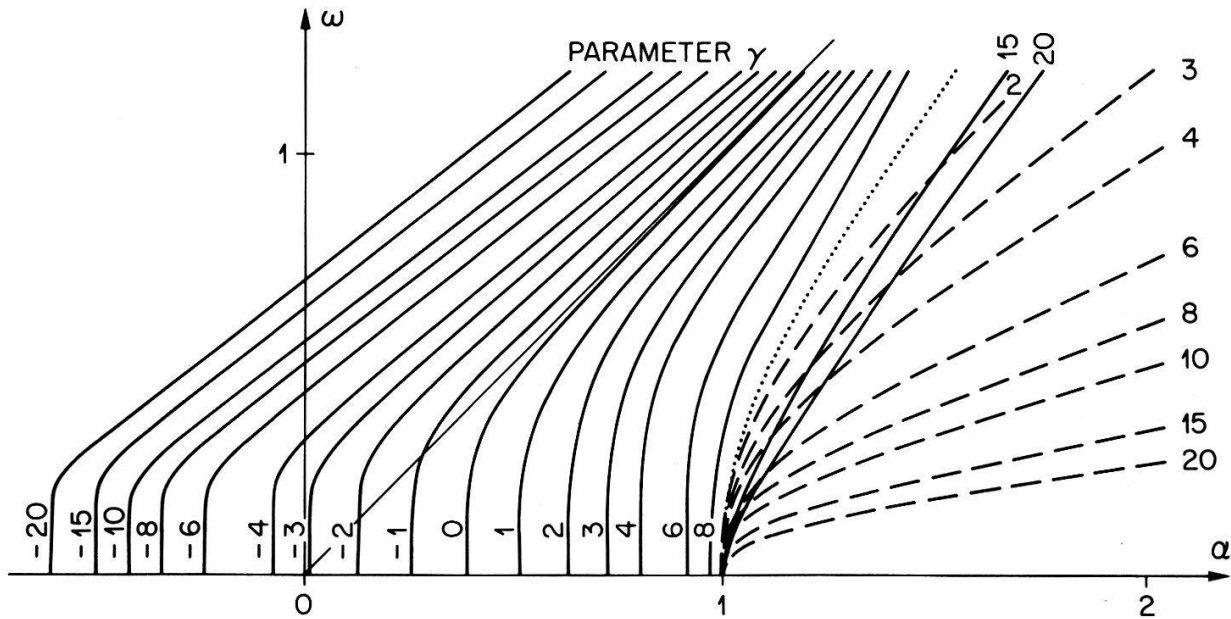


Figure 1

Characteristics of the Josephson equation for $\beta = 10$ and $\gamma = 0, \pm 1, \pm 2$, etc. For $\gamma < -3.1$, rotational solutions with $\alpha < 0$ corresponding to an output torque (Josephson motor) exist. The characteristic for $\beta = 0$ for $|\gamma| = 1, 2$, etc. is drawn by broken curves. In the range $|\alpha| < 1$ trivial solutions exist; $\omega = 0$ being either stationary (for $\gamma > 1$ and all $|\alpha| < 1$; for $\gamma < -1$ only for $\sqrt{1 - 1/\gamma^2} < |\alpha| < 1$) or oscillatory (only for $\gamma < -1$ in $|\alpha| < \sqrt{1 - 1/\gamma^2}$). The dotted hyperbola $\omega = \sqrt{\alpha^2 - 1}$ is the characteristic for $\beta = 0$, $|\gamma| \leq 1$ and $\beta = \gamma \geq 0$.

Appendix

The characteristic for $\beta = 0$ and $|\gamma| > 1$

The Josephson equation becomes

$$\dot{\varphi}(1 + \gamma \cos \varphi) + \sin \varphi = \alpha \quad (\text{A.1})$$

$$\Rightarrow \dot{\varphi} = \infty \quad \text{for } \varphi_\gamma = \arccos(-1/\gamma) \in (0, \pi).$$

If

$$\begin{aligned} \gamma > 1: \quad \dot{\varphi} > 0 & \quad \text{for } \varphi \in (-\varphi_\gamma, \varphi_\gamma), \\ \dot{\varphi} < 0 & \quad \text{for } \varphi \in (\varphi_\gamma, 2\pi - \varphi_\gamma). \end{aligned}$$

If

$$\begin{aligned} \gamma < -1: \quad \dot{\varphi} > 0 & \quad \text{for } \varphi \in (\varphi_\gamma, 2\pi - \varphi_\gamma), \\ \dot{\varphi} < 0 & \quad \text{for } \varphi \in (-\varphi_\gamma, \varphi_\gamma). \end{aligned}$$

Since $\beta = 0$ (mass = 0), the velocity can jump instantaneously (no inertia). Assuming $\beta = 0 + 0$ (small positive) the $\dot{\phi} < 0$ ranges have to be replaced by $\dot{\phi} = \infty$. The time during one period in ϕ is therefore

$$T = \frac{2\pi}{\omega} = \int_{-\phi_\gamma}^{2\pi - \phi_\gamma} \frac{d\phi}{\dot{\phi}} = \int_{-\phi_\gamma}^{\phi_\gamma} d\phi \frac{1 + \gamma \cos \phi}{\alpha - \sin \phi} \quad \text{for } \gamma > 1 \quad (\text{A.2})$$

or

$$T = \int_{\phi_\gamma}^{2\pi - \phi_\gamma} d\phi \frac{1 + \gamma \cos \phi}{\alpha - \sin \phi} \quad \text{for } \gamma < -1. \quad (\text{A.3})$$

The transformation $\phi \rightarrow \pi - \phi$ in (A.3) brings the integrand and boundaries into the form (A.2) with $-\gamma \rightarrow \gamma$, showing that $\omega(\alpha, 0, \gamma)$ depends on $|\gamma|$ only.

Indefinite integration of

$$\int d\phi \frac{1 + \gamma \cos \phi}{\alpha - \sin \phi} = \frac{2}{\sqrt{\alpha^2 - 1}} \operatorname{arctg} \frac{\alpha \operatorname{tg}(\phi/2) - 1}{\sqrt{\alpha^2 - 1}} - \gamma \ln(\alpha - \sin \phi) \quad (\text{A.4})$$

yields

$$\begin{aligned} \frac{2\pi}{\omega} = \frac{2}{\sqrt{\alpha^2 - 1}} & \left[\operatorname{arctg} \frac{\alpha \sqrt{\frac{\gamma + 1}{\gamma - 1}} - 1}{\sqrt{\alpha^2 - 1}} \right. \\ & \left. + \operatorname{arctg} \frac{\alpha \sqrt{\frac{\gamma + 1}{\gamma - 1}} + 1}{\sqrt{\gamma^2 - 1}} \right] + \gamma \ln \frac{\alpha\gamma + \sqrt{\gamma^2 - 1}}{\alpha\gamma - \sqrt{\alpha^2 - 1}} \quad (\text{A.5}) \end{aligned}$$

and (4) if the addition theorem is applied.

The last term in (A.5) (and (4)) is missing in reference 9. The authors [9] also claim the lowest order of the asymptotical formula (3) for $\alpha_c(\beta, \gamma)$ to be valid only for $\gamma > -3$ which according to its derivation (see reference 8) is not true. It obtains for large β but, in principle, for all γ of the order 1, which generally excludes extreme values (too large or too near to zero). For α_c this only means $|\gamma|$ should not be too large (comparable to $\sqrt{\beta}$). This fact becomes more obvious when many higher terms in $1/\beta$ are available as, for example, in expansion (2).

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les 7 et 8 octobre 1977

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