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Investigations on the odd-odd isotopes Ga^{64}, Ga^{66}, Ga^{68} and the odd-even isotope Ga^{67}

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(4. II. 1952.)

Summary. Disintegrations of the odd-odd isotopes Ga^{64}, Ga^{66} and Ga^{68} have been investigated by spectrometer and coincidence measurements. The positron activity assigned in literature to Ga^{64} has to be attributed to some impurities. Disintegration schemes for Ga^{66} and Ga^{68} have been established. Spin assignments to ground states of Ga^{66} and Ga^{68} have been compared to those predicted by Nordheim's rules regarding odd-odd nuclei.

Some new γ-radiations observed during the course of disintegration of Ga^{67} are reported in the supplement and a disintegration scheme proposed. The findings are difficult to reconcile with the predictions from shell model in its simplest form.

1. Introduction.

It is well known that all the odd-odd nuclei with the exception of ${}_1H^2$, ${}_3Li^6$, ${}_5B^{10}$ and ${}_7N^{14}$ are unstable. They, therefore, disintegrate into the corresponding stable even-even isobars. The nuclear spins of the nuclei with odd mass numbers can be predicted with the help of the nuclear shell model[1–4]) in its simplest form of single particle picture. For the odd-odd nuclei Nordheim[5]) has formulated an empirical rule about the manner in which the odd proton and the odd neutron combine together to give the total spin of a nucleus of this type in its ground state. But till now only relatively few instances are known to verify the validity of this rule and it seems to be too premature to draw conclusions from it. Otherwise also, it seems important to know more about the combinations of spins of the odd-odd nuclei, as it is hoped that valuable informations about the properties of bound states of nuclei may be obtained from such studies. The precise investigation of the disintegration schemes of radioactive isotopes is a good method for assigning spins of the nuclear energy levels.

The work reported here is a part of a bigger programme of investigations that is being carried out in this laboratory to study the modes of decay of odd-odd nuclei and consequently, the excited states of the even-even nuclei.

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The element gallium has quite a number of unstable isotopes which have been produced by various means in different laboratories since the earliest days of artificial radioactivity. Their half-lives have been well determined and well assigned. On the other hand, prior to the beginning of the investigations reported here, for most of the isotopes the radiations emitted had been rather poorly studied and practically no proper attempts had been made to arrive at their disintegration schemes. It is the special purpose of this paper to work out the disintegration schemes of some of the odd-odd isotopes of gallium which can be produced by proton bombardment of zinc. As the effects due to the isotope Ga^{67} entered into all our measurements, a general investigation of it was done, although not in direct connection to the problem proposed.

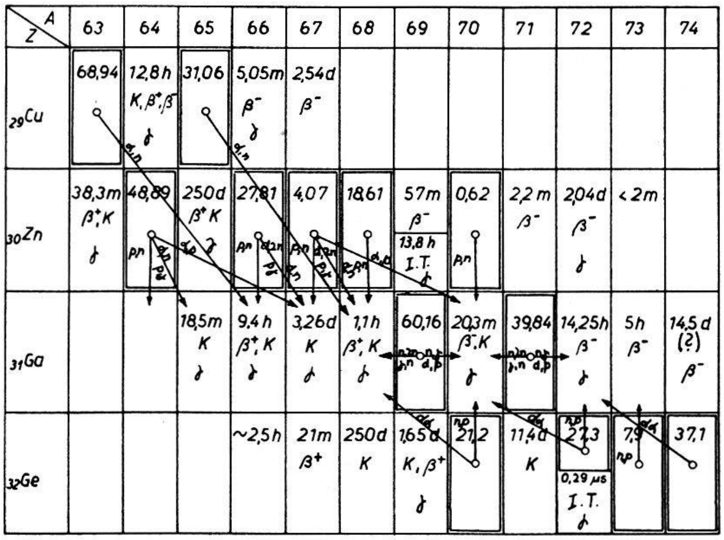


Fig. 1.
Relevant part of nuclear chart.

The relevant part of the nuclear chart is reproduced in Fig. 1 which provides a rapid orientation about the nuclear properties of the isotopes with which we are concerned here.

2. Method of Irradiation and Preparation of the Sample.

Irradiations necessary for all the experiments described in this paper were carried out in the following way. A thin layer of zinc was melted on a directly cooled copper probe target which was then bombarded by the 6.8 MeV internal proton beam of the E.T.H. cyclotron. The layer of activated zinc was then dissolved off the target by immersion in hot 6 N HCl. Gallium was chemically separated from zinc by extracting this solution with ether. On evaporating ether, carrier-free gallium remained behind and could be brought on a formvar or mica foil with the use of a few drops of water or dilute acid as the case may be. Our sincerest thanks are due to Dr. E. JACOBI who carried out the chemical separations for all the samples prepared during the course of these investigations.

3. The Isotope Ga^{64}.

By bombarding zinc with 6.3 MeV protons BUCK[6]) observed a positron activity of 48 m half-life in the gallium fraction. The threshold of production was determined by him to be 4.1 MeV and from it the maximum energy of the positrons was deduced to be 2.3 MeV. He assigned the activity to Ga^{64}.

Since the energy of the proton-beam used by us exceeded 6.3 MeV, it should have been sufficient to produce this activity. However, we were unable to detect any trace of the same, both by measurements with *G-M* counters and beta-ray spectrometer. Hence, we are led to conclude that Buck's findings of an activity of 48 m half-life were due to some impurities present in the gallium sample used by him, probably Zn^{63}. The activity due to Ga^{64} must have either a very much shorter half-life or the threshold of production exceeds our proton energy.

4. The Isotope Ga^{66}.

a) *Summary of Previous Investigations.*

When these investigations were undertaken, of this isotope only the half-life of 9.4 h and the maximum energy of the positrons were known. A number of workers[6–14]) have observed this activity from different reactions and hence the assignment of the same to the gallium isotope of mass number 66 may be considered as unique.

MANN[8]) studied the positron spectrum in the cloud-chamber and also determined the maximum energy of the positrons by absorption in aluminium. This value is given in the literature[15]) as 3.9 MeV. Simultaneously with the present authors[16]) reporting their findings, MOFFAT and LANGER[17]) announced the results of their investigations on this isotope and later they[18]) published a detailed description. The results of our experiments lead us to different conclusions so that a report giving full particulars of the experiments was considered to be of some interest.

The main components of the disintegration are the high energy positron spectrum with an upper limit at 4.15 MeV and two intense

γ-lines of 2.75 and 1.05 MeV. By coincidence measurements we have established that these two γ-radiations are emitted in cascade and that the main part of the positrons are emitted without being in coincidence with a nuclear γ-line. We are thus led to conclude that the main positron spectrum goes over directly to the ground-level. The disintegration scheme of MOFFAT and LANGER[18]) is in contradiction to the fact that the threshold for production by (p, n) reaction is less than 6.8 MeV. The recent determination of the threshold for (p, n) reaction in this laboratory[19]) at 6.3 MeV confirmed our conclusion. The proposed disintegration scheme will be discussed in detail after having described the experimental investigations.

b) *Positron-Spectrum and the Fermi-Kurie Plot.*

The positron spectrum was measured with the lens spectrometer of the laboratory using a resolving power of 2.5%. The upper limit of the spectrum was found to be (4.15 ± 0.05) MeV. At the lower energy end the internal conversion lines due to Ga^{67} of 78 h half-life were superimposed on the spectrum and their effects had to be corrected for. The Fermi-Kurie plot indicates a complex spectrum which can be decomposed into four partial spectra with the end-points at 4.15, 1.38, 0.90 and 0.40 MeV. Their relative intensities are given in Table I.

Table I.

	End Energies of the Partial Positron Spectra in MeV	Relative Intensities
1	4.15 ± 0.05	87.0
2	1.38 ± 0.05	4.2
3	0.90 ± 0.05	6.9
4	0.40 ± 0.05	1.8

The form of the main spectrum is that of an allowed transition. The results of the positron spectrum measurement are in excellent agreement with those of MOFFAT and LANGER[18]). From the end points of the partial spectra we are able to conclude on the excited levels of Zn^{66} at 2.75, 3.3 and 3.8 MeV.

c) *γ-Radiations.*

To look for the presence of nuclear γ-radiations besides the strong annihilation, measurements of Compton and photo-electrons in the lens spectrometer were undertaken. The high energy positrons were

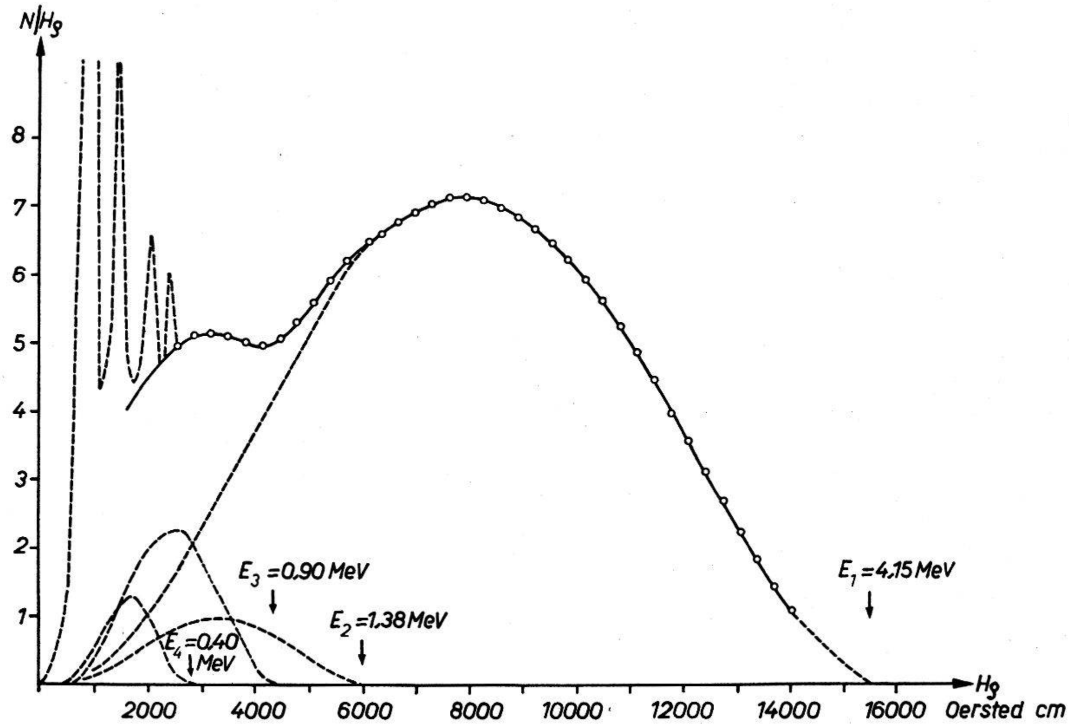


Fig. 2.
Positron spectrum of Ga^{66}.

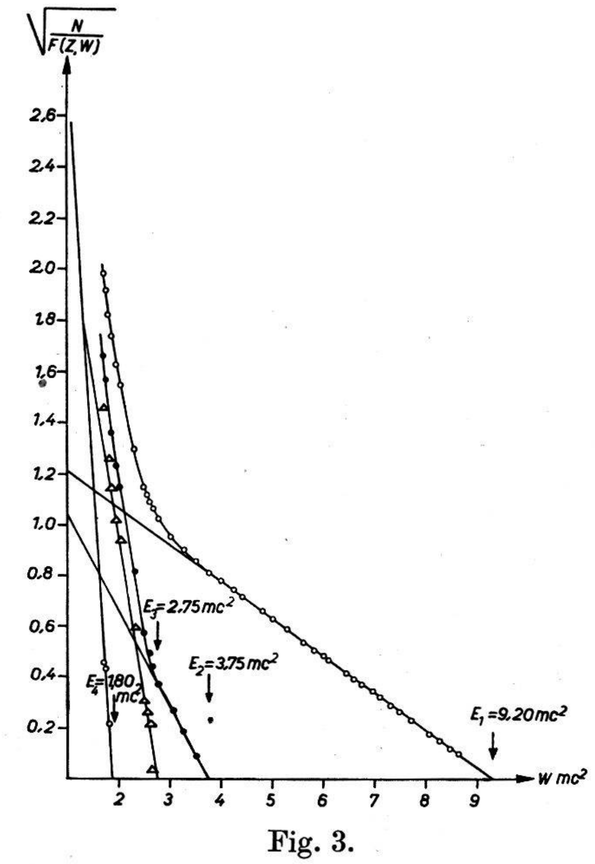


Fig. 3.
Fermi-Kurie plot of the positron spectrum of Ga^{66}.

absorbed in capsules of gold and brass and the Compton electrons were ejected from that of aluminium.

The Compton electron distribution curve indicates several components. Besides the annihilation, there are two very intense γ-rays of energies 1.05 and 2.75 MeV present. Calibration measurements undertaken for a check with Na^{24} using identical geometry of the arrangement showed clearly that there are weaker components present with energies between these two. These could be resolved into two components of 1.7 and 2.2 MeV. Further, at the higher energy end the presence of a 3.3 MeV γ-component is apparent. The tail of the curve shows the presence of even higher energy γ-radiations which could not however be measured with our spectrometer. Moffat and Langer[18]) have observed a γ-line of 4.8 MeV. Hofstadter and McIntyre[20]) were able by the NaI scintillation counter method to resolve a γ-line at 4.27 MeV. The energies of the two principal γ-lines were determined more accurately by measuring the photo-electrons ejected from a lead radiator and was found to be

$$h\nu_1 = (1.05 \pm 0.02) \text{ MeV}$$
$$h\nu_2 = (2.75 \pm 0.02) \text{ MeV}$$

To determine the shapes of the Compton electron distribution curves as well as the efficiencies of the radiator used for the γ-ray energies with which we were concerned, calibration measurements were done with the 0.511 MeV γ-ray of C^{11}, the 1.17 and 1.33 MeV γ-rays of Co^{60} and the 1.38 and 2.758 MeV γ-rays of Na^{24}. For these calibration measurements the geometry of the arrangements employed was identical as in the case of Ga^{66}. In the light of these measurements it was possible to resolve the Compton electron distribution curve into its various components and then to estimate their relative intensities. The values so obtained are given in Table II.

Table II.

	Energies of the γ-Radiations in MeV	Relative Intensities
1	0.511	12.0
2	1.05	3.7
3	1.7	0.3
4	2.2	0.5
5	2.75	2.9
6	3.3	0.5
7	4.25	0.2
8	4.8	0.2

From Hofstadter and McIntyre's[20]) measurements we estimate the relative intensities of the 4.25 and 4.8 MeV components to be approximately equal. The relative intensities of the low energy components might have been a little over-estimated on account of the

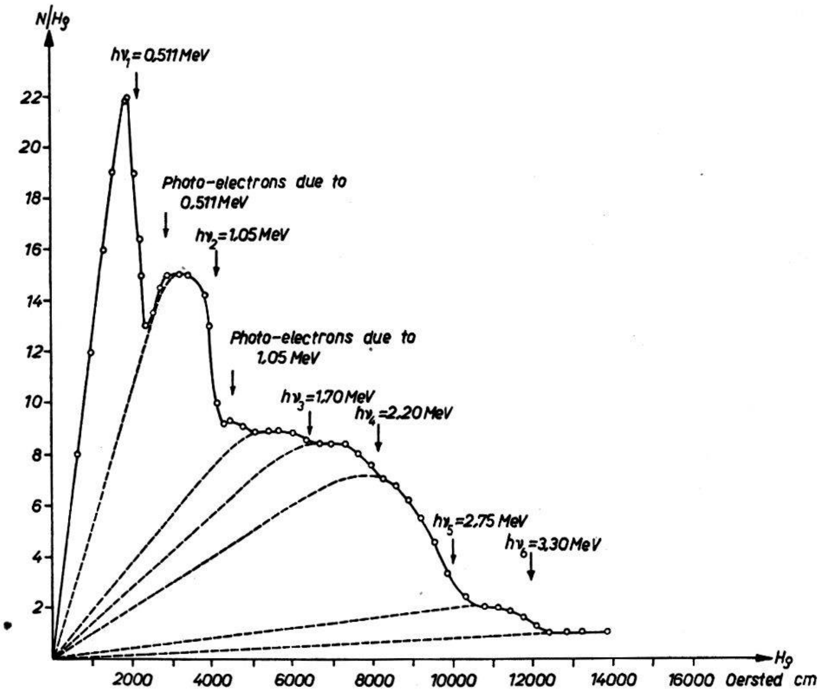


Fig. 4.
Compton electron distribution curve of Ga^{66}. The sample taken inside capsules of gold and brass and the Compton electrons ejected from that of aluminium.

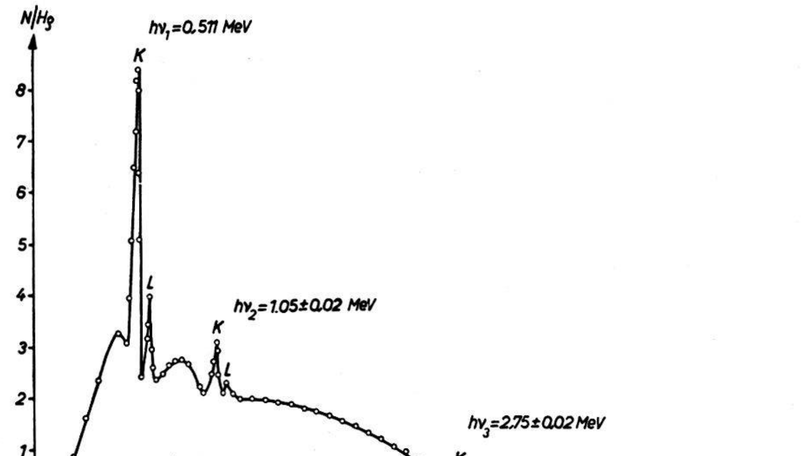


Fig. 5.
Photo- and Compton electron distribution curve of Ga^{66}.
17 μ-thick Pb foil used as radiator.

continuous spectrum of Bremsstrahlung and the hard annihilation radiation due to the stopping of the high energy positrons in the capsule (*vide* Investigations on Zn^{63})[21]).

d) γ-γ-*Coincidences.*

γ-γ-coincidences may take place in the case under discussion besides those between annihilation radiations, between the annihilation radiation and the nuclear γ-rays or between any two nuclear γ-rays. To clear up the mode of disintegration process it was important to determine the energies of the γ-radiations which were in coincidence. For this purpose measurement of coincidence absorption curve was considered very helpful.

Two anthracene crystals together with photo-multipliers were used for registering the counts. They were so arranged that the two crystals subtended a right angle at the source. The source was taken inside a brass capsule round which sufficient lead foils were wrapped

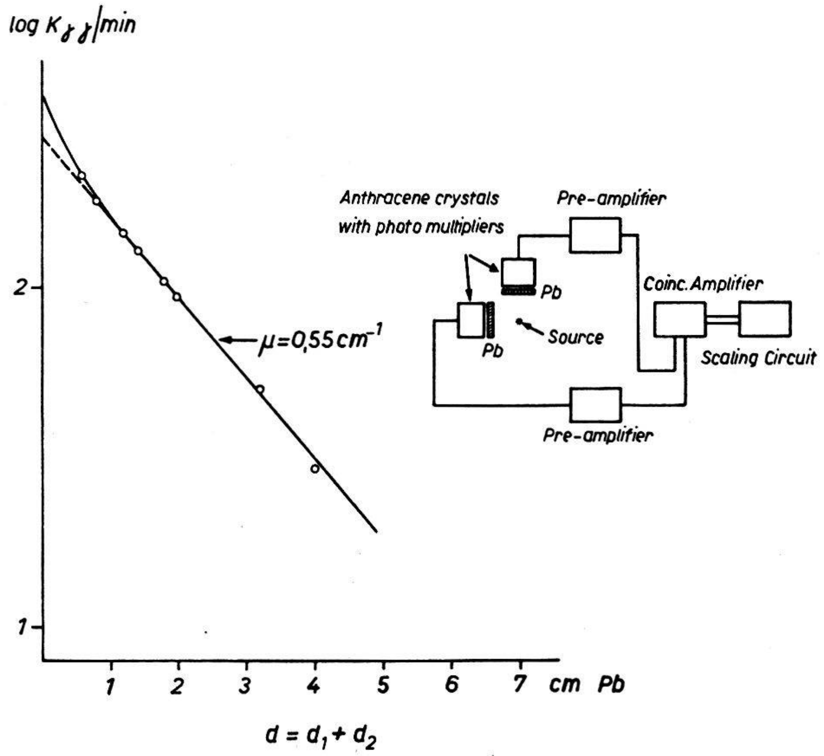


Fig. 6.
$\gamma-\gamma$ coincidence absorption curve of Ga^{66} obtained using anthracene crystals together with photo-multipliers.

to completely absorb the extremely hard positrons due to this isotope. Fig. 6 shows the coincidence absorption curve in lead obtained by every time placing equal absorbers in front of each counter. When two γ-radiations are emitted in cascade, the number of recorded coincidences is given by

$$K = 2\ N\ \varepsilon_1\ \varepsilon_2\ e^{-(\mu_1+\mu_2)\,d}\ \omega^2$$

where N is the number of nuclei disintegrating per minute, ε_1, ε_2

the efficiencies of the anthracene crystals for the energies of the γ-radiations in coincidence, μ_1 and μ_2 the absorption coefficients in lead for these γ-rays, ω the solid angle subtended and d the thickness of the absorber. The absorption curve plotted in logarithmic scale is mainly a straight line with an absorption coefficient $\mu = 0.55\ cm^{-1}$.

If we admit that the two most intense γ-lines 1.05 and 2.75 MeV are emitted in cascade, then the expected $\mu = 0.6\ cm^{-1}$. The exprimentally determined value is in excellent agreement with this assumption taking into account the correction which has to be applied for the geometry of the arrangement.

It is apparent from the measured curve that some coincidences are also due to a softer component presumably the annihilation radiation being emitted in cascade with one of the principal nuclear γ-rays. But the intensity is much less than it would have been if the main positron spectrum were in coincidence with a γ-radiation as concluded by MOFFAT and LANGER[18]).

e) *Transition Probabilities.*

The ratio of disintegrations by K-captures to those by positron emissions has been experimentally determined by MOFFAT and LANGER[18]) by comparing the area of the measured curve of the Auger electrons to that of the positron spectrum. They found that 34% of all the nuclear disintegrations are due to K-captures.

The theoretical ratios for the probabilities of K-captures to positron emissions for all the partial transitions taking them to be allowed, have been calculated by us and are given in Table III.

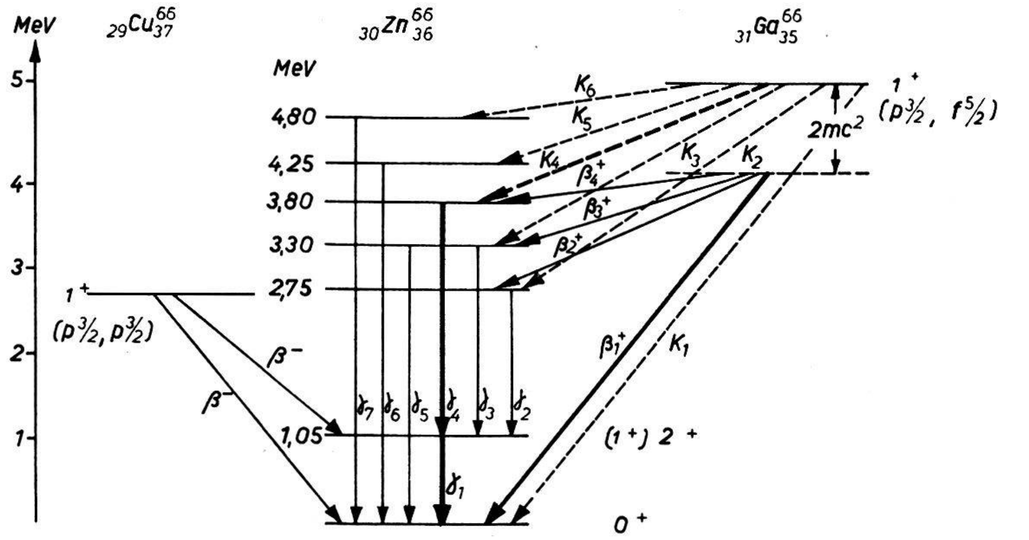
Table III.

	Excited Energy Levels of Zn^{66} in MeV	End Energies of the β^+-Spectra in MeV	Rel. Probabilities of K-Captures and β^+-Emissions
1	0	4.15	8.7×10^{-3}
2	2.75	1.38	1.8×10^{-1}
3	3.30	0.90	1.1
4	3.80	0.40	24.0

The transitions to the levels indicated by 4.25 and 4.8 MeV γ-lines can only be possible by orbital electron-captures. These transitions may be taken as approximately 4% of the total disintegrations from the relative intensities of the following γ-lines to the positron spectrum.

f) *Discussion of the Disintegration Scheme.*

As has been mentioned before we came to the conclusion that the main part of the positron emissions leads directly to the ground level of Zn^{66}. Decomposition of the positron spectrum revealed excited levels at 2.75, 3.3 and 3.8 MeV. The conclusion that the de-excitation of the 2.75 MeV level takes place by the emission of the strong γ-radiation of the corresponding energy would be most natural. However, then the difficulty would be in fitting the 1.05 MeV γ-radiation to the scheme as it is the strongest component



$\beta_1 = 4.15$ MeV	56.0%	K_1	0.5%	$\gamma_1 = 1.05$ MeV	35.0%
$\beta_2 = 1.38$ MeV	2.7%	K_2	0.5%	$\gamma_2 = 1.7$ MeV	3.2%
$\beta_3 = 0.90$ MeV	4.5%	K_3	4.5%	$\gamma_3 = 2.2$ MeV	4.5%
$\beta_4 = 0.40$ MeV	1.1%	K_4	26.2%	$\gamma_4 = 2.75$ MeV	27.3%
		K_5	2.0%	$\gamma_5 = 3.3$ MeV	4.5%
		K_6	2.0%	$\gamma_6 = 4.25$ MeV	2.0%
				$\gamma_7 = 4.8$ MeV	2.0%

Fig. 7.

Proposed disintegration scheme of Ga^{66}.

and is emitted in coincidence with the 2.75 MeV one. This difficulty might be solved by assuming that the 1.05 MeV component consists really of two γ-radiations of nearly the same energy and only one of them precedes the 2.75 MeV. This interpretation would be rather artificial as no splitting of the 1.05 MeV line has been observed.

This problem can be cleared up when we take into account the other weaker γ-components and the excited energy level of Zn^{66} at 1.05 MeV revealed by the beta-decay of Cu^{66} which has recently been found out by MEYERHOF et al.[22])* and MAEDER and PREISWERK[23]). The energy of the γ-radiation in the case of Ga^{66} is the same and therefore most probably emanates from this lowest excited level. The 2.75 MeV radiation which is in cascade with this radiation then originates from the 3.8 MeV excited level revealed by the Fermi-Kurie plot of the positron spectrum. The de-excitation of the 2.75 MeV level takes place by the emission of the weak 1.7 MeV γ-ray also in cascade with the 1.05 MeV line. The 2.2 MeV component too can be easily fitted into this scheme. The higher energy components are to be taken as leading directly to the ground level. The total disintegration energy would not allow of any other conclusions. The disintegration scheme so arrived at is reproduced in Fig. 7. The assumed scheme explains fully the relative intensities of the γ-lines (cf. Tables I, II and III).

We are now in a position to calculate the ft values for the different transitions which are given in Table IV.

Table IV.

	Excited Energy Levels of Zn^{66} in MeV	Transition Energies in MeV	log ft
1	0	$4.15+2\ mc^2$	7.8
2	2.75	$1.37+2\ mc^2$	7.1
3	3.30	$0.90+2\ mc^2$	5.9
4	3.80	$0.40+2\ mc^2$	4.9
5	4.25	0.92	5.7
6	4.80	0.37	4.9

In attempting to assign spins to the excited levels we take into consideration the disintegration of ${}_{29}Cu^{66}_{37}$. According to the shell model in the region of 29 to 37 nucleons, the orbits $f_{5/2}$ and $p_{3/2}$ are filled up. From the data on odd-even nuclei, we know that the orbits $f_{5/2}$ and $p_{3/2}$ have approximately the same energy but the orbit $f_{5/2}$ is preferentially filled up in pairs. Between N or $Z = 29$ to 37 inclusive, spins of 3/2 have been measured for every odd number, with the exception of ${}_{37}Rb^{85}_{48}$ and ${}_{30}Zn^{67}_{37}$ which show $f_{5/2}$ orbits. The transitions from Cu^{66} to Zn^{66} are of zero order according to the ft values evaluated. Hence, for ${}_{29}Cu^{66}_{37}$ a configuration $(p_{3/2}, p_{3/2})$ or $(f_{5/2}, f_{5/2})$ may be assumed. We admit the first configuration as

*) The authors wish to express their thanks to Prof. W. E. MEYERHOF for private communications on the subject of Cu^{66}.

the spins of odd-even copper nuclei are 3/2. This gives for the filling up of the outer proton and neutron orbits in ${}_{30}Zn^{66}_{36}$

$$P \quad (f_{7/2})^8 \; (f_{5/2})^0 \; (p_{3/2})^2$$

$$N \quad (f_{7/2})^8 \; (f_{5/2})^6 \; (p_{3/2})^2$$

For ${}_{29}Cu^{66}_{37}$ NORDHEIM[24]) has assigned a configuration $(p_{3/2}, f_{5/2})$ which was based on the results of old measurements. The beta transitions were supposed to lead to an excited state of ${}_{30}Zn^{66}_{36}$. In the configuration assumed by us a resultant spin greater than 1 of the nucleus would be expected following NORDHEIM's[5]) rule. But it seems unreasonable to assume a configuration $(p_{3/2}, p_{1/2})$ just to fit this rule.

A spin of 1^+ or 2^+ is to be assigned to the excited level at 1.05 MeV taking into consideration the ft value of the transition according to G—T selection rules. A spin assignment of 2^+ is however more probable according to GOLDHABER and SUNYAR's[25]) classification that the first excited states of the majority of till now investigated even-even nuclei have spins 2^+.

The transition from ${}_{31}Ga^{66}_{35}$ to the ground level of ${}_{30}Zn^{66}_{36}$ is characterised by an allowed form of the positron spectrum coupled with exceptionally high ft value, $\log ft = 7.8$. Such transitions following NORDHEIM[5]) are of the type $\Delta L = 2, \Delta I = 1$. We conclude, therefore, for ${}_{31}Ga^{66}_{35}$ a configuration $(p_{3/2}, f_{5/2})$ with a total spin of 1 and even parity. For the other excited levels no definite spin assignments can be made. Spins of 1^+ or 2^+ only, are possible for the excited levels at 4.8, 4.25 and 3.3 MeV which de-excite directly to the ground from a consideration of the ft values.

Measurements of the angular correlation of the γ-rays made in this laboratory (M. RÜETSCHI, unpublished) show that it depends on a small negative $\cos^2\Theta$ term $[W(\Theta) = 1 + A \cos^2\Theta, A = -0.06 \pm 0.03]$. A conclusive spin assignment for the levels involved in the principal cascade transition cannot be made till the contributions to the coincidences due to the weaker components can be eliminated.

The disintegration scheme shows an interesting point, the missing transition from Ga^{66} to the 1.05 MeV level. In the case of Cu^{66} there are transitions of the same order to both this level and the ground by β^--decay. It seems that to explain this fact, a further assumption has got to be introduced. The following is an attempt to explain it.

The resultant state is assumed as a mixture of several configurations, one out of which is given below as an example:

	${}_{29}Cu^{66}_{37}$	${}_{30}Zn^{66}_{36}$	${}_{31}Ga^{66}_{35}$
P	$(f_{7/2})^8\ (f_{5/2})^0\ (p_{3/2})^1$	$(f_{7/2})^8\ (f_{5/2})^0\ (p_{3/2})^2$	$(f_{7/2})^8\ (f_{5/2})^0\ (p_{3/2})^3$
N	$(f_{7/2})^8\ (f_{5/2})^6\ (p_{3/2})^3$	$(f_{7/2})^8\ (f_{5/2})^6\ (p_{3/2})^2$	$(f_{7/2})^8\ (f_{5/2})^5\ (p_{3/2})^2$

It can now be presumed that the excited level at 1.05 MeV is due to the excitations of the two protons or two neutrons in the unfilled $p_{3/2}$ sub-shells or to a mixture of both. During the disintegration of Cu^{66}, the transition takes place between two nucleons of the unfilled sub-shells and may be therefore excited. In the case of Ga^{66} on the other hand, the transition is to a sub-shell which is filled up by the process and therefore, only the living proton shell can be excited. If the 1.05 MeV level is owing to the mixture excitations of both the unfilled proton and neutron sub-shells, the probability of transitions to this level is reduced. This idea is put forward with reservations and only as an attempt to explain the missing transition. It has to be looked for if other similar cases can be found.

5. The Isotope Ga^{68}.

a) *Summary of Previous Investigations.*

A positron activity with 68 m half-life assigned uniquely to Ga^{68} is known since a very long time and a large number of investigators [6–8][10][13][27–38] have produced it by various means.

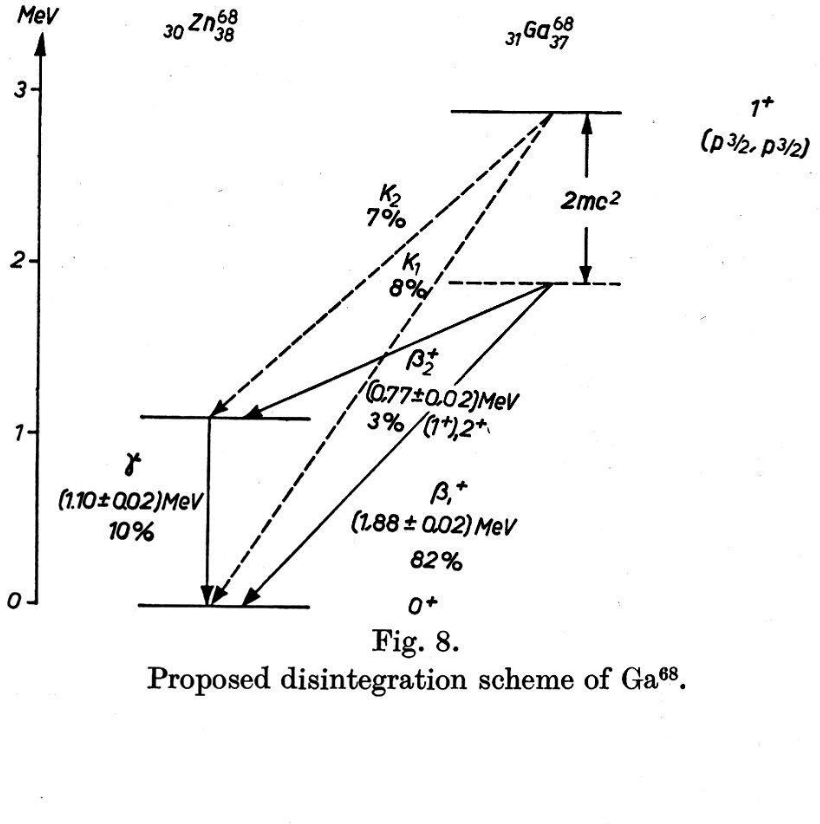


Fig. 8.
Proposed disintegration scheme of Ga^{68}.

Mann[8])[15]) and Ridenour and Henderson[10]) determined the maximum energy of the positrons by absorption in aluminium as 1.9 MeV. Further, Mann[8]) and Sagane and Miyamoto[33]) investigated the positron spectrum with the help of cloud chambers.

We studied the mode of decay of Ga^{68} more thoroughly and arrived at the disintegration scheme given in Fig. 8. The results of measurements performed, details of which follow, support the scheme.

b) *Positron Spectrum and Fermi-Kurie Plot.*

Investigations on the positron spectrum were made with the lens spectrometer employing the same procedure as described in the case of Ga^{66}. To avoid any disturbances due to the shorter period isotopes Ga^{65} and Ga^{70}, the samples were allowed to age for sufficient long time before the measurements were started. Effects due to Ga^{66} had to be subtracted. The spectrum obtained in this way and the corresponding Fermi-Kurie plot are reproduced in Figs. 9 and 10. The straight line form of the Fermi-Kurie plot in the higher energy region indicates a transition of zero order. The extrapolation of this straight line gives the upper limit of the positron spectrum as

$$E_1 = (1.88 \pm 0.02) \text{ MeV}.$$

The deviation from the straight line form of the Fermi-Kurie plot starting at 2.5 mc^2 indicates a second partial spectrum with an upper limit of

$$E_2 = (0.77 \pm 0.02) \text{ MeV}.$$

The second deviation must be taken as due to back scattering from the source and its mounting. The relative intensities of the two spectra were obtained by comparing their areas after decomposing the Fermi-Kurie plot following the theoretical forms and are given in Table V.

Table V.

	End Energies of the Partial Positron Spectra in MeV	Relative Intensities
1	1.88 ± 0.02	26
2	0.77 ± 0.02	1

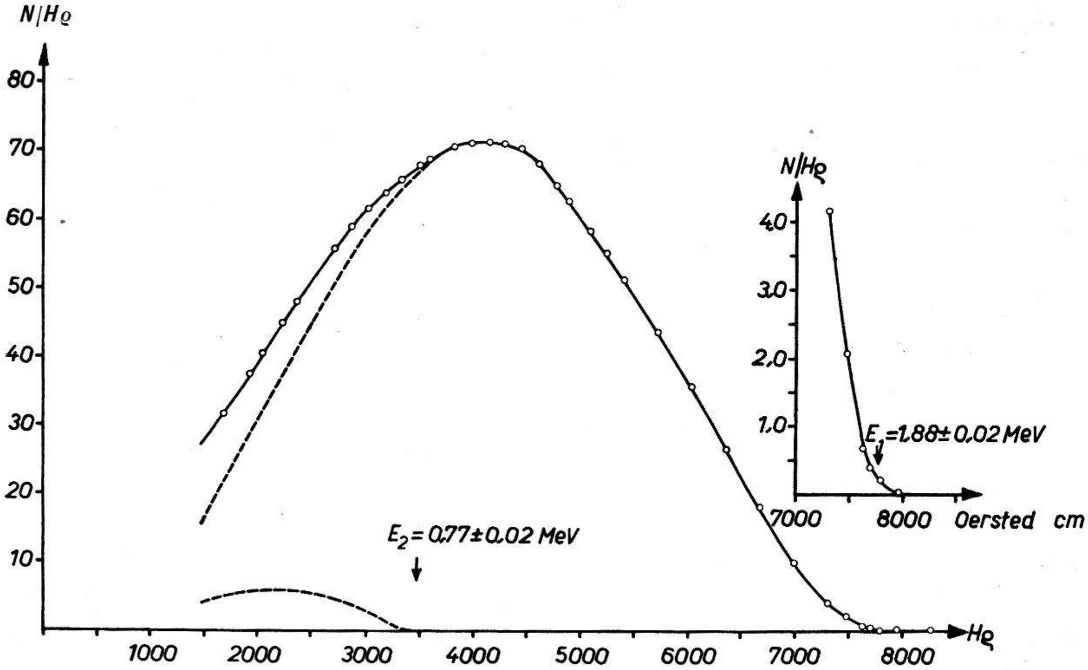


Fig. 9.
Positron spectrum of Ga^{68}.

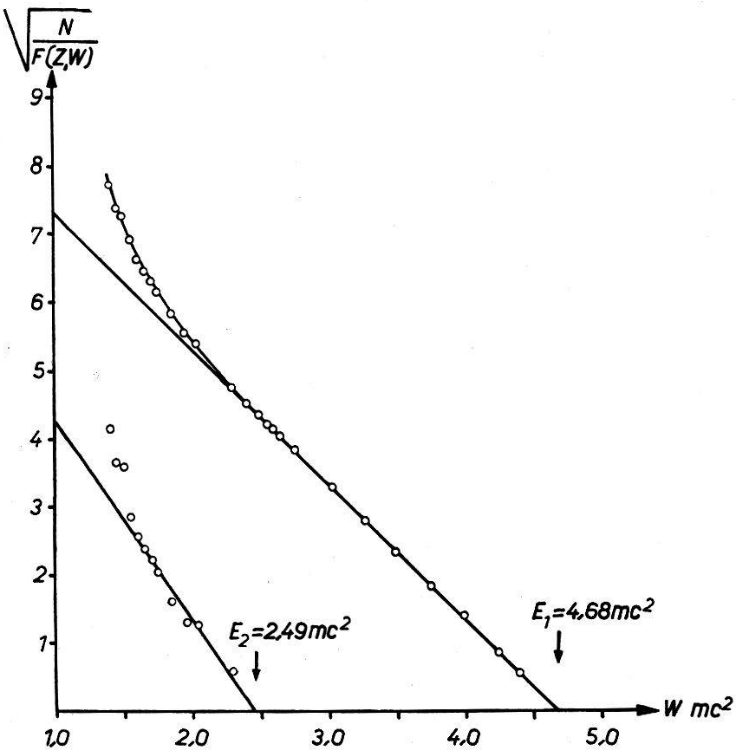


Fig. 10.
Fermi-Kurie plot of the positron spectrum of Ga^{68}.

c) γ-*Radiations.*

Search was made for the presence of internal conversion lines. Groups of electrons at $H\varrho = 295$, 312 and 321 Oersted cm were observed (Fig. 11) which correspond to Auger electrons emitted following K orbital electron captures.

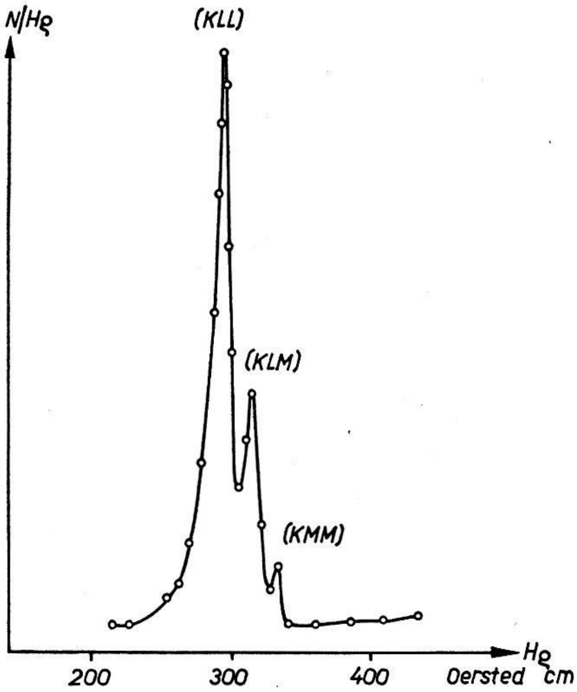


Fig. 11.
Auger electrons emitted following K-orbital electron captures during the process of disintegration of Ga^{68}.

To look for higher energy γ-radiations measurements of Compton and photo-electrons were undertaken with the same arrangements as described in the case of Ga^{66}. Besides the intense annihilation radiation, a higher energy component of 1.10 MeV is revealed in the Compton electron distribution curve (Fig. 12). The more accurate value of

$$h\nu = (1.10 \pm 0.02)\ \text{MeV}$$

was obtained from photo-electron measurement (Fig. 13). The photo-electron peak of this line was rather broad when measured after an interval of about an hour after bombardment. The time decay of the peak was followed. Gradually instead of one, two peaks were clearly discernible. The second peak decayed with a half-life much longer than 68 m and as has been shown earlier belongs to Ga^{66}.

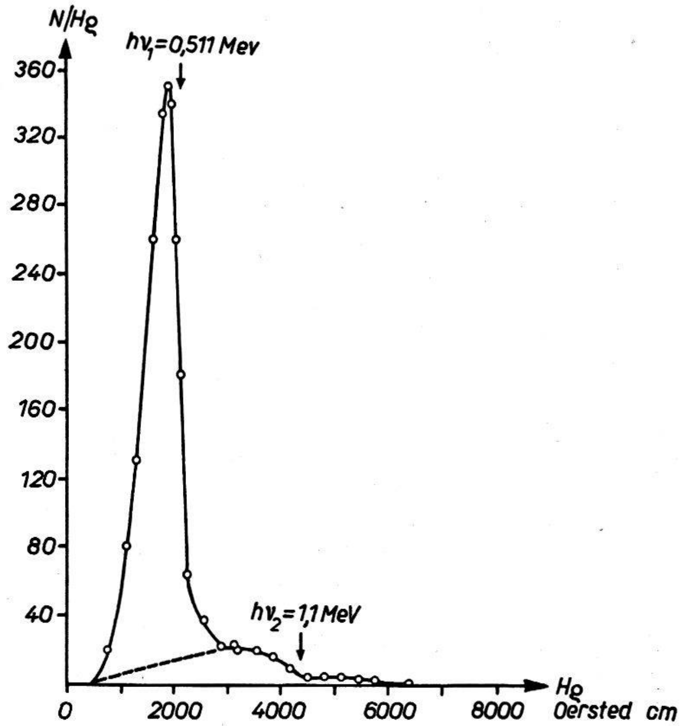


Fig. 12.
Compton electron distribution curve of Ga^{68}. The sample taken inside capsules of gold and brass and the secondary electrons ejected from that of aluminium.

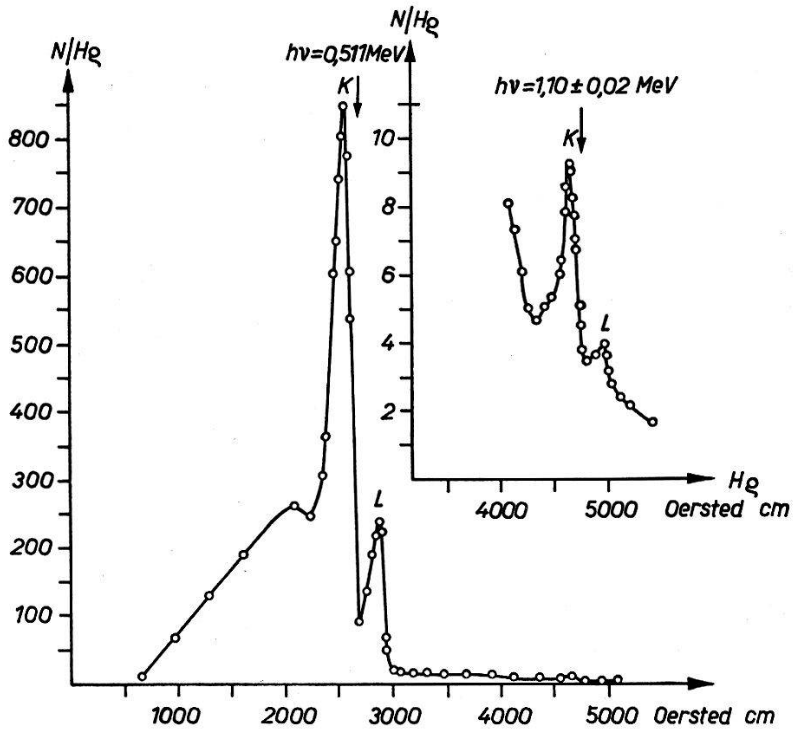


Fig. 13.
Photo- and Compton electron distribution curve of Ga^{68}.
17 μ-thick Pb foil used as radiator.

*

The intensity of the 1.10 MeV γ-line relative to the annihilation radiation was obtained by comparing the heights of the photoelectron peaks in the light of previous calibration measurements performed with 0.511 MeV annihilation radiation of C^{11} and 1.17 and 1.33 MeV γ-radiations of Co^{60}. The results are given in Table VI.

Table VI.

	Energies of the γ-Radiations in MeV	Relative Intensities
1	0.511	17.6
2	1.10	1.0

d) *Transition Probabilities.*

It has been shown above that the positron decay is complex consisting of two partial spectra with $E_1 = 1.88$ MeV and $E_2 = 0.77$ MeV. The second spectrum leads to an excited level at 1.10 MeV which is revealed also by the γ-line of the corresponding energy. The theoretical ratios for the probabilities of transitions by orbital electron captures and positron emissions have been calculated and are given in Table VII.

Table VII.

	Excited Energy Levels of Zn^{68} in MeV	End Energies of the Partial Positron Spectra in MeV	Rel. Probabilities of K-Captures to β^+-Emissions	log ft
1	0	1.88	0.1	5.1
2	1.1	0.77	2.2	4.8

The ft values for both the transitions are of the same order and correspond to zero order transitions as have been revealed by the forms of the spectra.

e) *Discussion of the Disintegration Scheme.*

From the fact that the spin of the even-even nucleus ${}_{30}Zn^{68}_{38}$ is obviously 0^+ and that the form of the main positron spectrum is that of an allowed transition we are to assign a spin of 1^+ to the disintegrating nucleus ${}_{31}Ga^{68}_{37}$ in its ground state according to G—T selection rules.

The following is the one chosen out of the possible configurations which may be assigned to the nuclei ${}_{31}Ga^{68}_{37}$ and ${}_{30}Zn^{68}_{38}$ on the shell model:

	${}_{31}Ga^{68}_{37}$	${}_{30}Zn^{68}_{38}$
P	$(f_{7/2})^8\ (f_{5/2})^0\ (p_{3/2})^3$	$(f_{7/2})^8\ (f_{5/2})^0\ (p_{3/2})^2$
N	$(f_{7/2})^8\ (f_{5/2})^6\ (p_{3/2})^3$	$(f_{7/2})^8\ (f_{5/2})^6\ (p_{3/2})^4$

The transition to the ground state of ${}_{30}Zn^{68}_{38}$ would be of the type $\Delta L = 0$ and $\Delta I = 1$ which is normal allowed according to NORDHEIM[5]) and the experimental value of log ft is compatible to it. It must however be pointed out that for such a configuration for ${}_{31}Ga^{68}_{37}$, the total spin according to NORDHEIM's[5]) rule should be high which as has been mentioned above does not agree with the measured form of the positron spectrum. If we assume a configuration $(p_{3/2}, f_{5/2})$ for ${}_{31}Ga^{68}_{37}$, as has been done by NORDHEIM[22]), the transition to the ground level of ${}_{30}Zn^{68}_{38}$ would be of the type $\Delta L = 2, \Delta I = 1$. Such a transition would have an exceptionally high ft value which again is not compatible to our experimental results. The parallel case is that of ${}_{29}Cu^{66}_{37}$ which has been discussed along with Ga^{66}.

Following G—T selection rules a spin assignment of 1^+ or 2^+ to the excited level of ${}_{30}Zn^{68}_{38}$ is possible. However, a spin 2^+ would agree with the general characteristics of even-even nuclei[25]). The configuration of this level may be taken as that of the ground level, the spins of the last two $(p_{3/2})$ protons have combined to a spin 2^+.

6. Supplement: The Isotope Ga^{67}.

a) *Summary of Previous Investigations.*

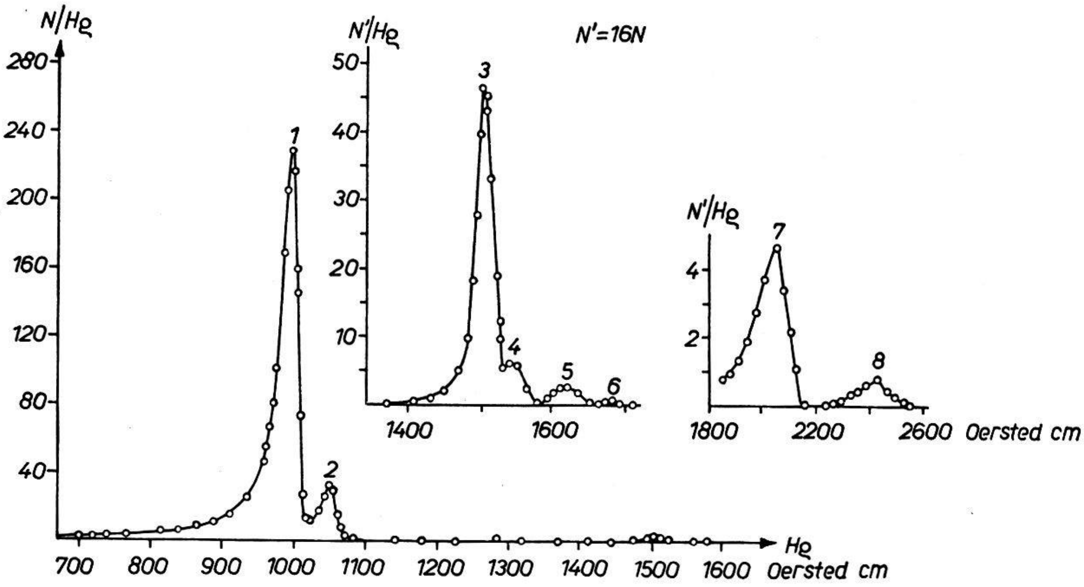
Effects due to the long period isotope of gallium, ${}_{31}Ga^{67}_{36}$ had to be taken into account for all the measurements during the course of our investigations on the odd-odd isotopes of gallium. As we obtained by this occasion some new results, we are presenting them here as a supplement.

The long life activity uniquely assigned to Ga^{67} has been the subject of study of a large number of workers[13])[37])[39–42]). According to the latest determinations by McCOWN, WOODWARD and POOL[43]) the half-life is given as 3.26 ± 0.02 d. The radiations emitted have been studied with great care by ALVAREZ[44]) whose work on this isotope established for the first time the occurance of orbital electron captures and the consequent emission of corresponding x-rays in course of nuclear disintegrations. The γ-transitions have been investigated further by VALLEY and McCREARY[36]), HELMHOLZ[45]), GUTHRIE[46]) and CORK et al[9]).

The mode of decay seems to be much more complicated than was thought till these investigations were undertaken. A number of new γ-components have been found out by us which lead to the conclusion of new energy levels.

b) *Measurement of Internal Conversion Lines.*

For measuring the internal conversion lines samples were prepared by evaporating a drop of the active solution on a 0.4 mgr/cm² thick and 3 mm dia. gold-foil mounted on a thin formvar foil. The window of the *G—M* counter used in the lens spectrometer



1	*K* line due to 92 KeV transition.	5	*K* line due to 205 KeV transition.
2	*L* line due to 92 KeV transition.	6	*L* line due to 205 KeV transition.
3	*K* line due to 182 KeV transition.	7	*K* line due to 298 KeV transition.
4	*L* line due to 182 KeV transition.	8	*K* line due to 388 KeV transition.

Fig. 14.
Internal conversion lines of Ga^{67}.
Extended source and slit system used for the measurement of the weaker lines.

was a formvar-zaponlack foil and was 0.188 mgr/cm² thick. The resolving power of the spectrometer was set at 1.4% for the measurement of the stronger lines and for the weaker components an extended source was employed which increased the luminosity at the cost of resolving power. These measurements were started after the effects due to shorter period isotopes had died down to negligible intensities. By applying a steady potential of 6 KV to the source, search was made for the presence of low energy electron lines till zero KeV. In this region, only the groups of Auger electrons were observed. Thanks are due to Dr. H. SCHNEIDER for help with this measurement.

The energies of the internal conversion lines corresponding to five transitions which have been observed are given in Table VIII.

Table VIII.

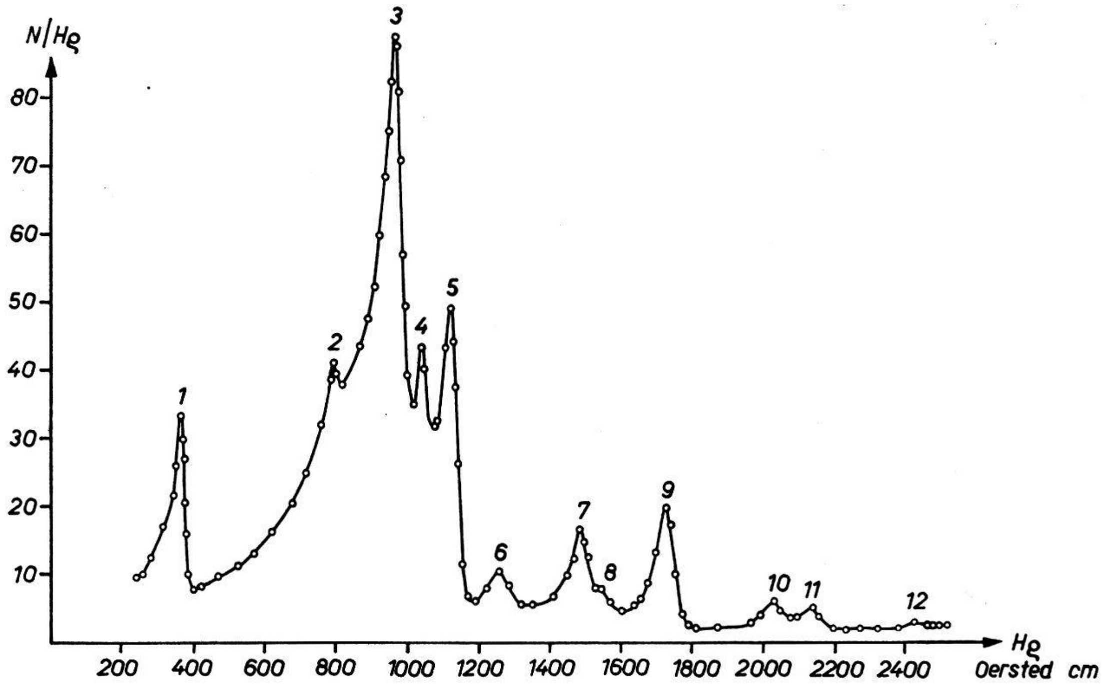
	Energies of the Electron Lines observed in KeV	Interpretations KeV
1	82 ± 1	K int. conv. line of $h\nu = 92$
2	91 ± 1	L int. conv. line of $h\nu = 92$
3	172 ± 2	K int. conv. line of $h\nu = 182$
4	181 ± 2	L int. conv. line of $h\nu = 182$
5	195 ± 2	K int. conv. line of $h\nu = 205$
6	205 ± 2	L int. conv. line of $h\nu = 205$
7	288 ± 3	K int. conv. line of $h\nu = 298$
8	378 ± 5	K int. conv. line of $h\nu = 388$

93, 180 and 297 KeV lines have been previously reported by HELMHOLZ[45]), CORK et al.[9]) reported later a further γ-transition of 174 KeV which was not observed by us.

c) *Measurement of the Unconverted Parts of the γ-Transitions.*

To investigate the unconverted parts of the γ-transitions measurement of the photo-electrons were undertaken with the lens spectrometer. The radiator consisted of a thin gold foil of 5 mgr/cm^2 thickness. The hardest of the converted electrons were completely absorbed by taking the sample inside an aluminium capsule of 3 mm wall thickness. The measured electron distribution curve is reproduced in Fig. 15. The energies of the lines observed with the corresponding interpretations are tabulated below.

To determine the relative intensities of the lines the heights of the photo-electron peaks were compared employing the coefficients given by GRAY[47]). Corrections for the effects of the absorptions of the soft electrons involved, in the radiator itself were made by taking help of some previous calibration measurements performed under identical geometrical arrangements (*vide* PREISWERK and STÄHELIN[48])).



1 K due to 92 KeV line.
2 KLL Auger electrons from Au.
3 L due to 92 KeV line.
4 M due to 92 KeV line.
5 K due to 182 KeV line.
6 K due to 205 KeV line.
7 L due to 182 KeV line.
8 L due to 205 KeV line.
9 K due to 298 KeV line.
10 L due to 298 KeV line.
11 K due to 388 KeV line.
12 L due to 388 KeV line.

Fig. 15.

Photo-electron lines of Ga^{67}. 5 mgr./cm^2 gold foil used as radiator.

Table IX.

Energies of the Photo-Electron	Interpretations	KeV	Relative Intensities of the *Total*
11.3	K photo-electrons from Au $h\nu =$	92	62
51.9	KLL Auger electrons from Au		
77.6	L photo-electrons from Au $h\nu =$	92	
88.6	M photo-electrons from Au $h\nu =$	92	
101.3	K photo-electrons from Au $h\nu =$	182	21
124.3	K photo-electrons from Au $h\nu =$	205	2
168	L photo-electrons from Au $h\nu =$	182	
191	L photo-electrons from Au $h\nu =$	205	
218	K photo-electrons from Au $h\nu =$	298	12
284	L photo-electrons from Au $h\nu =$	298	
307	K photo-electrons from Au $h\nu =$	388	3
374	L photo-electrons from Au $h\nu =$	388	

d) *The Internal Conversion Coefficients and the Multipole Orders of the γ-Transitions.*

The internal conversion coefficients for the different γ-components can be determined from the relative intensities of the converted and the unconverted parts when the absolute number of quantas emitted from both the samples used for these measurements are known. These values were obtained in our case by measuring the number of counts from the samples with a special gold cathode counter placed at a fixed distance, the efficiencies of the counter for the different γ-components being already known[49]. Corrections were made for the absorptions of the various components in the walls of the counter.

The values of the internal conversion coefficients deduced in this manner are compared with the theoretical values for different multipole orders in Table X. The theoretical values were calculated from the tables recently published by Rose et al.[50]. For γ-energies less than 150 KeV, the values computed using the approximate formulas of Hebb and Uhlenbeck[51] and Dancoff and Morrison[52] were corrected by factors obtained by comparing the values from these formulas with those from Rose et al's[50] tables for higher energy γ-rays.

Table X.

E in KeV	K/L exp.	Electrical 2^l-pole Radiation			Magnetic 2^l-pole Radiation			Exptl.
		$l = 1$	$l = 2$	$l = 3$	$l = 1$	$l = 2$	$l = 3$	
92	6.5	6.8×10^{-2}	7.5×10^{-1}	7.0	5.5×10^{-2}	6.6×10^{-1}	6.7	2.8×10^{-1}
182	9	8.8×10^{-3}	6.0×10^{-2}	3.9×10^{-1}	1.1×10^{-2}	7.3×10^{-2}	4.1×10^{-1}	9.1×10^{-3}
205		6.5×10^{-3}	3.6×10^{-2}	2.3×10^{-1}	8.1×10^{-3}	4.9×10^{-2}	2.6×10^{-1}	5.5×10^{-3}
298		2.1×10^{-3}	9.5×10^{-3}	3.8×10^{-2}	3.3×10^{-3}	1.5×10^{-2}	6.0×10^{-2}	2.5×10^{-3}
388		1.0×10^{-3}	3.9×10^{-3}	1.4×10^{-2}	1.8×10^{-3}	6.7×10^{-3}	2.4×10^{-2}	1.7×10^{-3}

The 182, 205, 298 and 388 KeV transitions can either be electrical or magnetic dipole radiations as the differences between the conversion coefficients for these two types of radiations in this region are not big enough to decide conclusively. Determination of K/L conversion ratio was only possible for the 92 and the 182 KeV radiations. For the former a ratio of 6.5 ± 0.5 and for the latter 9 ± 1 were found. From the empirical values given by Goldhaber and Sunyar[25], in the case of 92 KeV radiation this ratio would be 5 for an electrical quadrupole and 8 for a magnetic dipole. Considering this radiation to be a mixture of magnetic dipole and electrical quadrupole would fit with our experimental results. In the case of 182 KeV radiation theoretical non-relativistic values of K/L conversion ratio according to Hebb and Nelson or Tralli and Lowen[51]) differ widely from the experimental, both for electrical and magnetic dipoles. But these values are known to differ in other cases also. Extrapolation of the empirical curves of Goldhaber and Sunyar shows that the experimental value is compatible to a magnetic dipole radiation.

e) β—γ *Coincidence Measurement.*

In order to arrive at a disintegration scheme supplementary indications had to be obtained from coincidence measurement. β—γ coincidence absorption curve was obtained by registering the electrons with a window type (mica foil of 2.5 mgr/cm²) counter and the gammas with a 40 μ-thick gold cathode G-M counter. A 0.22 mm

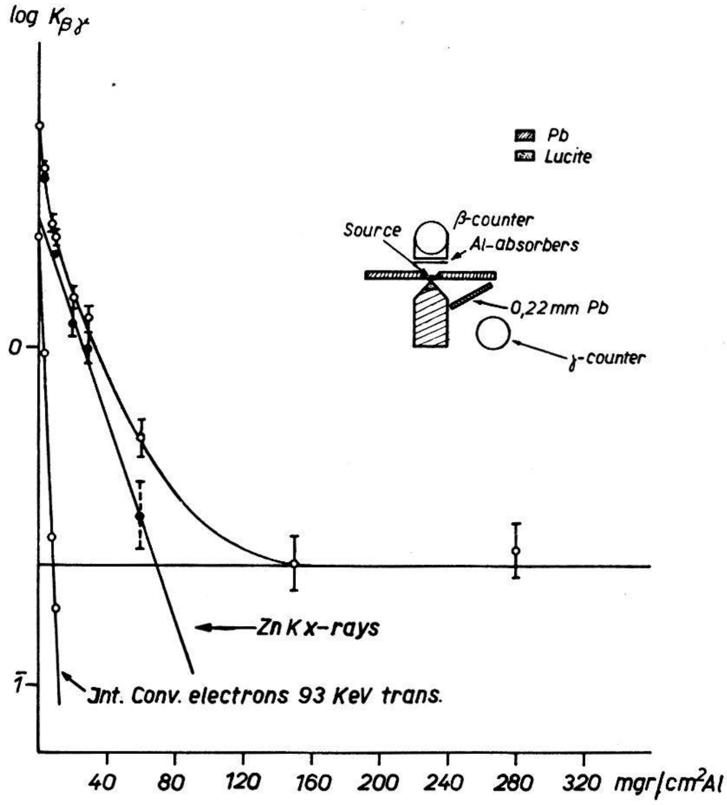


Fig. 16.
$\beta-\gamma$-coincidence absorption curve of Ga^{67}.

lead foil was placed in front of the γ-counter to absorb the x-rays and the hardest electrons. Schematic sketches of the geometrical arrangement used for this purpose is given in Fig. 16 along with the measured curve. To get the curve variable absorbers of aluminium were placed in front of the window type counter. The decomposition of the absorption curve shows that the K x-rays of zinc are emitted in coincidence with some γ-rays and further, that the 92 KeV transition is also emitted in cascade with a harder component or components.

f) *Ratio of X-Rays to γ-Radiations.*

Since Ga^{67} is known to have no continous beta spectrum and decays only by orbital electron captures and γ-transitions, it was considered important to know the ratio of x-rays to γ-radiations emitted, for deciding whether there were any direct transitions from the ground state of Ga^{67} to that of Zn^{67}.

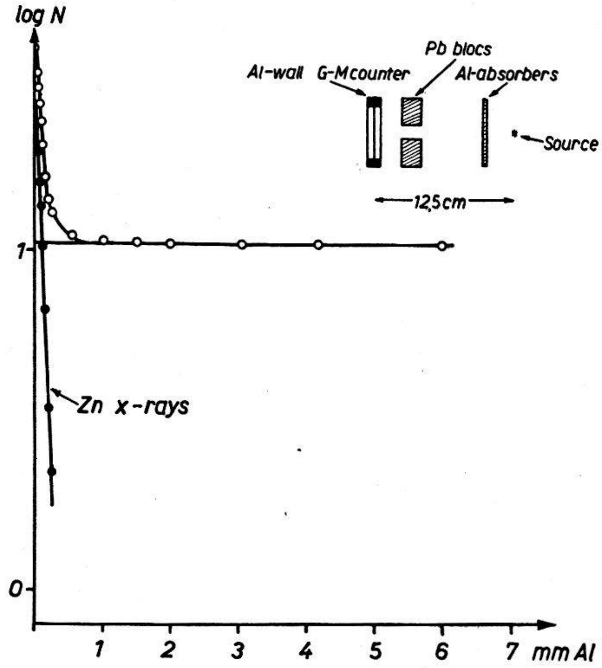


Fig. 17.

Absorption of the x-rays of Ga^{67} measured in a magnetic field.

A schematic sketch of the arrangement used is given in Fig. 17 along with the measured curve. This arrangement was placed in a magnetic field of sufficient strength to deviate all the electrons from reaching the counters. For recording the x-rays a calibrated aluminium G—M counter (*vide* SAURER[53]) and for the gammas a gold cathode one were used.

Admitting the disintegration scheme given in Fig. 18 the total number of K x-quantas emitted may be evaluated taking into account those due to K-captures as well as internal conversions in the K-shells. This value has got to be corrected for the fluorescence yield. The total γ-transitions are also known from the disintegration scheme and the internal conversion coefficients of the different components have been determined. Hence, the theoretical ratio between γ-quantas and x-quantas emitted can be calculated and has been found to be

$$N_\gamma / N_x = 2.9.$$

Experimentally this ratio has been determined and is given below

$$N_\gamma / N_x = 2.8 \pm 0.2.$$

The accuracy of the measurement does not permit to conclude that no direct transitions to the ground level occur. But these results show that if there are any such transitions, they cannot be in an appreciable amount and cannot certainly be of zero order in which case a ratio

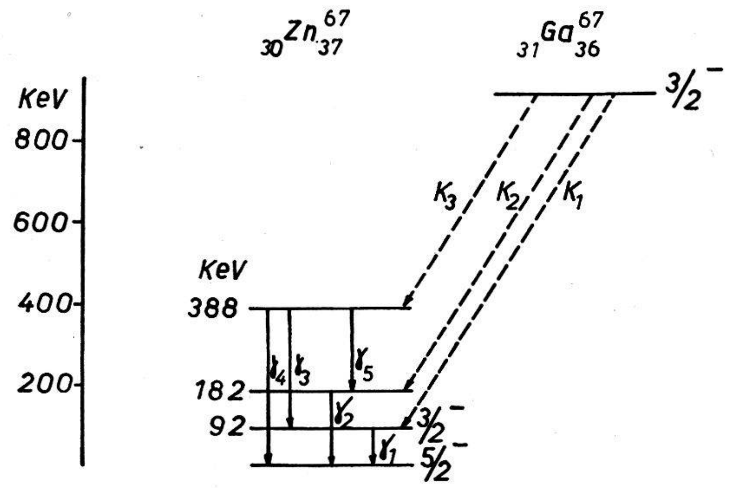
$$N_\gamma / N_x = 1.1$$

should have been expected.

g) *Discussion of the Disintegration Scheme.*

From the relative intensities of the different transitions as given in Table IX, it is obvious that the conclusion reached by HELMHOLZ[45]) viz. the three principal γ-transitions were in threefold cascade and the 298 KeV transition followed the 182 KeV, cannot be correct. As the energy of the 388 KeV γ-transition fits well with the energy of a cross-over transition when the 298 KeV and 92 KeV transitions and the 205 KeV and 182 KeV transitions are in cascade, we propose the scheme in Fig. 18. The relative intensities of the γ-transitions indicate that there are direct transitions by orbital electron captures to all the three excited levels of Zn^{67}. It is evident that most of the transitions by this process are to the 92 KeV level. Therefore, assuming these to be normal allowed (log $ft = 5$), the total disintegration energy can be estimated thereby fixing the position of the energy level of Ga^{67} in its ground state. The assumption of forbidden transition is ruled out as in that case the energy difference would allow of positron emission. The ft values for the transitions to the two other energy levels come out to be of the same order.

Possible spin assignments to the energy levels have been made taking into account the internal conversion coefficients of the γ-transitions and the K-capture probabilities. The fact that the intensity of the 298 KeV transition is much greater than that of the 388 KeV, although they seem to be of the same multipole order, is not considered as a big obstacle.



K_1		48%	γ_2	(182 KeV)	30%
K_2		28%	γ_3	(298 KeV)	18%
K_3		24%	γ_4	(388 KeV)	4%
γ_1	(92 KeV)	70%	γ_5	(205 KeV)	2%

Fig. 18.
Proposed disintegration scheme of Ga^{67}.

We now discuss the disintegration scheme following the shell model. The spin of $_{30}Zn^{67}_{37}$ in its ground state is known to be 5/2. We have pointed out before that this nucleus is one of the two exceptions in the region of N or $Z = 29$ to 37 inclusive, which have spins 5/2 and all the other nuclei have spins 3/2. This indicates that the pairing energy is not enough to make the 3/2 level lower. Therefore, we expect the following configuration

$$_{30}Zn^{67}_{37}$$
$$P \quad (f_{7/2})^8 \ (f_{5/2})^2 \ (p_{3/2})^0$$
$$N \quad (f_{7/2})^8 \ (f_{5/2})^5 \ (p_{3/2})^4.$$

For $_{31}Ga^{67}_{36}$ in its ground state we assume a spin of 3/2 as the stable gallium nuclei are known to have this spin. Therefore, we can ascribe a configuration

$$_{31}Ga^{67}_{36}$$
$$P \quad (f_{7/2})^8 \ (f_{5/2})^2 \ (p_{3/2})^1$$
$$N \quad (f_{7/2})^8 \ (f_{5/2})^6 \ (p_{3/2})^2$$

From these configurations it is obvious that no allowed transition can take place to the ground level of Zn^{67} from Ga^{67}.

A spin of 3/2 may be assigned to the lowest excited level at 92 KeV as the transitions by K-captures to this level are of zero order and the corresponding γ-transition from this level is a mixture of magnetic dipole and electrical quadrupole radiations. A level of this type following single particle model is expected to have a configuration

$$\begin{array}{ll} P & (f_{7/2})^8 \; (f_{5/2})^2 \; (p_{3/2})^0 \\ N & (f_{7/2})^8 \; (f_{5/2})^6 \; (p_{3/2})^3 \end{array}$$

An allowed transition from the ground level of Ga^{67} can take place to a level having the configuration

$$\begin{array}{ll} P & (f_{7/2})^8 \; (f_{5/2})^2 \; (p_{3/2})^0 \\ N & (f_{7/2})^8 \; (f_{5/2})^6 \; (p_{3/2})^2 \; (p_{1/2})^1 \end{array}$$

But then no direct transition from such a level with spin 1/2 to the level with spin 5/2 should occur as such a transition involves interchanges of orbits of two particles.

Our experimental findings show that three transitions of zero order occur to the low lying excited states of Zn^{67}. It follows that two of these states at least have same values of spins. This is a very interesting case as this fact is not compatible to the single particle picture of shell model. One cannot avoid taking into consideration other excitations than that of a single particle. A three-particle state of neutrons is however ruled out in the case of Zn^{67}. In extending the single particle model a plausible assumption seems to be that in the other excited states the two protons in the outermost unfilled orbit $(f_{5/2})$ take part.

Our sincerest thanks are due to Prof. Dr. P. Scherrer for the continued interest that he has taken in this work. One of us (A. M.) is also deeply indebted to him for being given the opportunity of working in this laboratory.

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