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Autor: Singh, A.K. / Manuel, A.A. / Jarlborg, T.

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Study of the many-body correlation effects in nickel by positron annihilation

By A. K. Singh, A. A. Manuel, T. Jarlborg, Y. Mathys, E. Walker and M. Peter

Département de Physique de la Matière Condensée, Université de Genève, CH-1211 Genève 4, Switzerland.

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Abstract. We characterize for the first time the nature of the many-body correlations in nickel by analyzing measurements of 2D- angular correlation of the positron annihilation radiation. We find that the correlations are very different for *d*-bands than the usual enhancement found for *s*-bands: when the *d*-states are near E_F the correlations give a decrease of the positron–electron wavefunction overlap calculated within the independent particle approximation.

Positron annihilation measurements are usually interpreted in terms of the independent particle model (IPM) and it is generally thought, at least in angular correlation experiments, that the many-body correlation effects have a minor influence on the results. In this paper we present preliminary results concerning the many-body effects in 2D- angular correlation of the positron annihilation radiation (2D-ACAR) distributions measured in nickel. We show that these effects are large enough to significantly alter the interpretation of the data in terms of the IPM. An attempt is made to characterize the deviations from the IPM by use of a modified version of the description proposed by Mijnarends and Singru [1]. The results show first that the two-photon momentum density greatly depends on the nature of the correlation effects and second that the nature of the correlations is sensitive to the character of the electronic states. While a positive enhancement factor is found for *s*-bands, in agreement with the results obtained in alkali metals [2], a negative factor is required for the *d*-bands.

It is well known that the 2D-ACAR distributions have to be calculated taking into account the wavefunctions of both electrons and positrons. Nevertheless, it has been shown during the last few years that 2D-ACAR can well be interpreted in terms of the Fermi surface topology in a wide range of transition metals, rare earths, disordered alloys and intermetallic compounds [3]. In many cases the analysis was based on the Lock–Crisp–West (LCW) theorem [4] which, in spite of its well known approximations appears to give satisfactory results.

Rabou and Mijnarends [5] have recently pointed out the limitations of the LCW theorem by calculating 2D-ACAR for paramagnetic Ni: substantial

deviations from the occupation number are found when the positron wavefunction is taken into account while calculating the momentum distribution of the two-photon annihilation pairs (TPMD). The same conclusion was drawn from our previous work [6] where we reported the first 2D-ACAR measurement in Nickel and compared it with TPMD calculations. In the present work we show, for data obtained in two other crystallographic planes ([100] and [111]), that TPMD calculations are necessary but not sufficient to interpret the experimental results. Moreover, we show that the agreement can be significantly improved by introducing a rather simple description of the many-body correlations.

The 2D-ACAR distributions have been measured at 4.2K in the ferromagnetic state with our 2D-apparatus based on high density proportional chambers [7]. Further experimental details are given in [6]. Although the partial polarisation of the positron permits a study of the spin density distribution by taking the differences between the 2D-ACAR measured with opposite magnetic field directions, we will restrict ourself in this paper to the analysis of the sum of these two 2D-ACAR.

We analyze our data in terms of the relative momentum density [8]. In general this method has the advantage to be independent of the 3γ correction term which is difficult to evaluate precisely due to the large uncertainty of the experimental coefficient given by Berko and Mills [9]. We found that, for nickel, the relative momentum density approach does not influence significantly the predicted 2D-ACAR distributions.

We show in Fig. 1 for the [100] plane and in Fig. 2 for the [111] plane some lines extracted from the distributions remapped according to the LCW procedure. The error bars of the experimental points are of the order of the size of the triangles. In the two figures, the lines are labelled in terms of the points in the Brillouin zone (BZ), projected in the appropriate plane (either [100] or [111]).

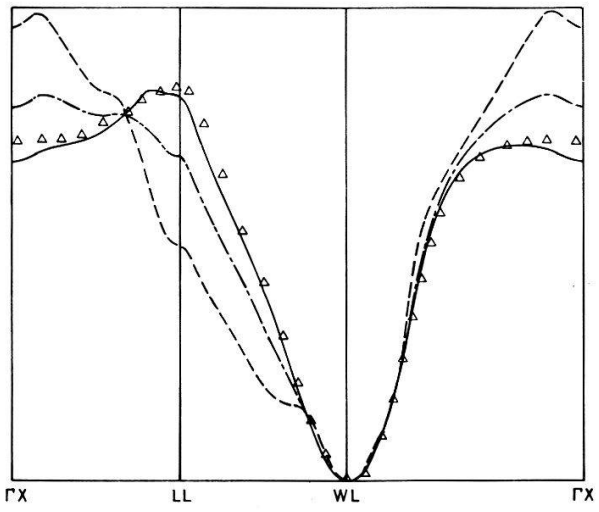

Figure 1
2D-ACAR from ferromagnetic nickel along some lines in the [100] plane. The triangles are the experimental data. The dashed line is the calculated Fermi surface cross-section. The chain line is the electron–positron momentum distribution calculated with the IPM. The solid line is the result including the many-body correlations.

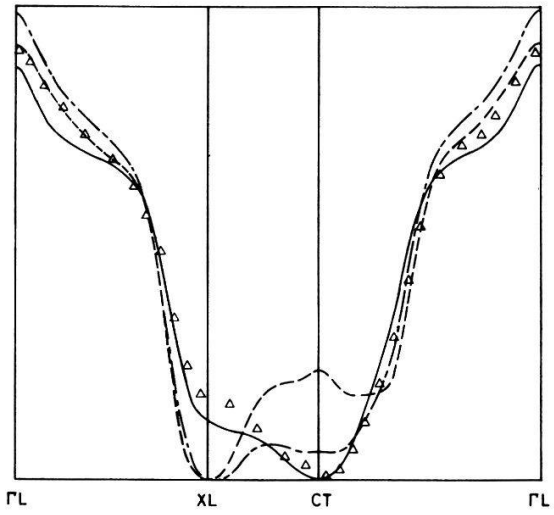

Figure 2
2D-ACAR from ferromagnetic nickel along some lines in the [111] plane. The conventions used in Fig. 1 hold here also.

Each point has two labels because, for these high symmetry planes, two different points of the BZ project at the same place in the plane. A special mention has to be given for the point CT in the [111] plane which is not part of the conventional set of points for the fcc BZ. This point lies at the center of an equilateral triangle defined by the projections of W points. Consequently, the path extracted from the [111] distribution and shown in Fig. 2 follows the limits of the projection of the irreducible BZ in the [111] plane. Our data are the first 2D-ACAR measurements in nickel, therefore a comparison with results from other groups is not possible.

There are three calculated curves on Figs. 1 and 2. They were obtained from a self-consistent Linear Muffin Tin Orbital (LMTO) band structure calculation for ferromagnetic nickel [10]. All these three curves are extracted from complete 2D distributions convoluted with the experimental resolution. The normalisation to the experiment has been performed for the distributions on the irreducible part of the projected BZ, after subtraction of a constant background. The dashed lines in Figs. 1 and 2 show the contribution of the Fermi surface (FS) only. The chain line is the TPMD calculated using the formalism developed by Singh and Jarlborg [11] in the IPM approximation. The positron wavefunction has been computed by changing the sign of the potential and removing the exchange-correlations term. The solid line is the result obtained from the TPMD calculation including many-body correlations. This is done by replacing equation (11) of Singh and Jarlborg [11] for $X_{\vec{G}}^{\vec{k},j}$, defined in terms of spherical harmonic wavefunctions, by

$$X_{\vec{G}}^{\vec{k},j} = 4\pi \sum_{l,m} a_{lm}^{\vec{k},j} Y_{lm}(\vec{k}+\vec{G}) \sqrt{\varepsilon_l(\gamma)} \int^{S} R_l(E_j^{\vec{k}}, r) R^{+}(r) j_l(|\vec{k}+\vec{G}|\, r) r^2\, dr \qquad (1)$$

In this formula $a_{lm}^{\vec{k},j}$ are the tail-decomposed eigenvectors and R_l (R^+) the electron (positron) l-state radial wavefunctions. The $\varepsilon_l(\gamma)$ coefficient is introduced to account for the many-body correlations. The square root has been introduced at

this level because the TPMD is given by the square of some linear combination of $X_{\vec{G}}^{\vec{k},j}$. We have used the following form for $\varepsilon_l(\gamma)$:

$$\varepsilon_l(\gamma) = a_{l0} + a_{l1}\gamma + a_{l2}\gamma^2 + a_{l3}\gamma^3 \tag{2}$$

where

$$\gamma = (E - E_{\text{bottom}})/(E_F - E_{\text{bottom}}); \quad \gamma \leqslant 1 \tag{3}$$

where E_{bottom} is the energy of the lowest band at $\vec{k} = 0$. It is worthwhile to point out that this description of the many-body correlations is similar to the one introduced by Mijnarends and Singru [1]. We have introduced two modifications of this model. First, we have included the γ^3 term because the formula of Kahana [12], which is the root of the Mijnarends and Singru's formula, is known to give a poor description of the enhancement near p_F [13]. Second, the range of our correlation formula is not restricted to the first BZ only. This is an important fact because, in contrast to the simple metals, the high momentum components play a major role in transition metals. Another very useful characteristic of the present formalism is that it is possible to have a partial enhancement coefficient $\varepsilon_l(\gamma)$ for each l-state. This has proved to be a very valuable property for the present work because it permits to state that these many-body correlations may either increase or decrease the positron-electron overlap estimated within the IPM, on the basis of the l-character of the electron states.

We have studied in detail the effect of the $\varepsilon_l(\gamma)$ for various sets of parameters a_{li}. The parameters selected for the result shown by the solid lines in Figs. 1 and 2 are shown in Table 1. This set is the best of more than 20 different sets of a_{li}. They represent a first effort to reveal the many-body effects and the most important finding is the parameter $a_{23} = -1$, showing that the d-states near E_F are not enhanced but effectively depressed. We have not tried a least square fit determination of the 16 a_{li}-parameters for several reasons: First, the p and f content of the wavefunction is small so the a_{1i} and a_{3i} parameters are not easily determined. Second, a least square fit would suffer from the polynomial assumption in equation (2). Other de-enhancement shapes such as exponentials or inclusion of tailing states above E_F would not be represented. Third, as will be shown later, changes in the exchange splitting are advantageous for χ^2, and such a parameter should be included in the fit too. Finally, considering the errors in the experimental and theoretical procedures, it is doubtful whether a more

Table 1
The a_i coefficients (see equation (2)) used for the enhanced TPMD calculation shown in Figs. 1 and 2.

l \ i	0	1	2	3
0 s-	1.0	0.4	0.0	0.0
1 p-	1.0	0.0	0.2	0.0
2 d-	1.0	0.0	0.0	−1.0
3 f-	1.0	0.0	0.0	0.0

optimized set of a_{li}-parameters would contain more useful information than what has been obtained here.

One sees from Figs. 1 and 2 that, beyond the improvement of the fit obtained by the TPMD calculation (chain line) over the *FS* (dashed line), some features are only reproduced when $\varepsilon_l(\gamma)$ is introduced in the calculation. In Fig. 1 the positive slope of the experimental data between ΓX and LL is only reproduced by the model including correlations. This means that the trend along this line has been reversed with the introduction of the $\varepsilon_l(\gamma)$ coefficient. A similar effect can be observed for the XL–CT line in Fig. 2. The FS and TPMD curves are minimum at XL, in disagreement with the experiment which is minimum at the point CT. By including the many-body correlation coefficient $\varepsilon_l(\gamma)$, the theoretical calculation reproduces well the experimental trends. It is also seen in Fig. 2 that the distributions in the vicinity of the ΓL point are not largely affected by the use of the $\varepsilon_l(\gamma)$ factor. The situation is different for the [100] plane (Fig. 1) where the major changes introduced by $\varepsilon_l(\gamma)$ occur around the ΓX point. This is because de-enhancements effects are more pronounced where *d*-electrons are present near E_F.

In order to characterize more precisely the improvement obtained by our empirical description of the many-body correlations, we report in Table 2 some values of χ^2 between experiment and the theoretical models. These values are not restricted to the lines shown in Fig. 1 and 2 but have been calculated from the total 2D distributions. One sees that χ^2 decreases by a factor of 1.8 to 5.8 from the FS cross-section to the TPMD calculation. This significant reduction reflects the limited validity of the LCW theorem in Nickel as stated above. But beyond this first decrease of χ^2, we gain a factor of 5.3 to 25 when we use the TPMD modified for the correlations (equation (1)). This shows very clearly that the model we have introduced to describe the many-body correlations plays an important role in the precise interpretation of the 2D-ACAR in Nickel. It is also important to note that the negative value of a_{23} quoted in Table 1 play a major role in this reduction of χ^2: One can see from Table 2 that a positive or zero value of a_{23} leads to a significantly larger value of χ^2. One sees also that the enhancement of

Table 2
The values of $\chi^2 \times 10^{10}$ calculated for different models of the 2D-ACAR remapped in $\vec{k}$-space.

plane:	[100]	[110]	[111]
Fermi surface model	662	1423	439
TPMD without the many body term	113	369	249
TPMD with many-body term:			
– $a_{\neq i}$ given in Table 1	4.5	55	47
– same except $a_{23} = 0.0$	58	210	261
– same except $a_{23} = 1.0$	167	510	236
– $a_{01} = a_{12} = 0.0$ $a_{23} = -1.0$	11	68	47
TPMD (without many-body) exchange splitting = 167 mRy (magnetic mom. = 1.1_B)	34	96	127

the s- and p-bands plays only a minor role compared to the large effect of the de-enhancement of the d-bands. At the present stage it is difficult to say if this effect is due to positron-electron correlations. This problem has only been theoretically investigated for the simple metals (for a recent review see reference [14]). It is possible that the important cubic term for the d-bands is due to the electron–electron correlations. It is known (see reference [15]) that these interactions tend to decrease break of the occupation number at the Fermi energy.

We have investigated the possibilities that the observed features we attribute to the many-body correlations have an other origin. The first possibility is that our calculation scheme for the TPMD from LMTO band structure is not sufficiently precise. But it has already been successfully tested for many different systems. The results obtained for alkali metals are in good agreement with experiment and with KKR calculations [2]. In vanadium, theoretical results have been compared with experimental 2D-ACPAR and the agreement is better than with other calculations [16]. The method of calculation has also been successfully tested for A15 compounds [17]. Another possibility is the exchange splitting. It is obvious that the effect of strongly reduced electron–positron overlap for d-states near the (minority) Fermi energy, can be modeled by a lowering of the minority band E_F. In Ni this can be achieved by an increase of the exchange splitting ξ. However, only a very large increase of ξ will lead to χ^2 values which are not as reduced as in our de-enhancement calculations. A rigid band increase of the exchange splitting of 120 mRy (lowering the minority E_F with 20 mRy and increasing the majority E_F with 100 mRy to keep the number of electrons constant) diminishes χ^2 values by a factor of 3, compared with a mean factor of 10 for the de-enhancement calculation. It is likely that a decrease of the minority E_F of 30 mRy would lead to a good χ^2 value, but then the majority E_F has to be increased accordingly in order to fulfill electron conservation, and both ξ and the magnetic moments are far too large. From this we exclude the possibility that an incorrectly calculated ξ is the cause of the discrepancy between experiment and IPM calculations. Moreover, we have to conclude that it is not possible to extract a precise value for ξ from the sum of the experimental spin up and spin down spectra. The calculated value is about 47 mRy for ξ, in agreement with other calculations. Smaller ξ has been obtained from (spin-polarized) photoemission [18, 19] for temperatures near and above room temperature.

In conclusion, we think that the many-body correlations may play a major role in the interpretation of positron annihilation data. The situation we have encountered in nickel could probably also be found in nickel compounds. Some very recent results we have obtained in the hexagonal compound $CeNi_5$ seems to show similar effects and one can think that they will also be present in other systems like Ni_3Ga. This mechanism should also work in other transition metals where E_F is near the top of the d-band. (Maybe in Pd, with the advantage of a zero ξ over Ni?) Moreover, there are recent 2D-ACAR measurements on Cr [20] which so far could not be interpreted in term of the FS topology, we suggest that de-enhancements may be of importance here too. The theory of Fujiwara *et al.*

[21] predicts a possible de-enchancement of the partial positron annihilation rate. Our results give a strong credit to their work. Despite the simplicity of their model, these authors predicted both an effect depending on the type of electron and a decrease or the wavefunctions overlap in filled or nearly-filled band structures like Ge, Sn, Bi, Be and so on. Very recently, Sormann and Puff [22] have also investigated the possibility of de-enhancement of high momentum components in alkali metals. We hope that the present analysis will inspire further theoretical developments to determine if the de-enhancement effects we observe originate from electron–positron or electron–electron correlations.

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