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Positron angular correlation in copper

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Abstract. The two photon pair momentum density and the angular correlation curves for positron annihilation radiation from copper have been obtained by a band structure calculation, employing Hubbard's approximation scheme. A comparison of the calculated curve for the long slit geometry with the experimental results shows that the theory rather overestimates the contributions in the high momentum region. An angular correlation curve for a point slit geometry is also calculated.

Keywords. Positron annihilation; band structure calculations; copper.

1. Introduction

In recent years positron annihilation studies in metals have provided us with interesting information about the electronic momentum densities in solids. In particular, the angular correlation of the positron annihilation radiation from copper single crystals has been measured employing long slit (Berko and Plaskett 1958, Berko *et al* 1968, Mijnarends 1969, Lock *et al* 1973, Singru 1974), point slit (Williams *et al* 1968, Senicki *et al* 1973, Singru 1973), spark chamber (Howells and Osmon 1972) and multicounter (Mader *et al* 1973) geometries. Copper provides an ideal test case for positron annihilation studies because its Fermi surface is well studied by other conventional methods. In this paper we present positron angular correlation curves computed from band structure calculations and compare them with the experimental results.

Berko and Plaskett (1958) employed the Wigner-Seitz method for obtaining the positron and electron wavefunctions in their calculations of the two-photon pair momentum density $\rho(\boldsymbol{p})$ in copper. However their results did not agree with the experimental curves in the high momentum region because the '3d' "core" electrons were described in a crude approximation. Gould *et al* (1972) have used symmetrised plane wave expansion for the positron wavefunction to calculate the core contribution to the momentum density $\rho(\boldsymbol{p})$. The results by their method when compared with the point slit measurements (Senicki *et al* 1973) do not give a satisfactory agreement thus emphasizing the need for a more sophisticated description of electron states.

A better approach to this problem is therefore to perform a band structure calculation (Loucks 1966, Mijnarends 1973 *a*, *b*) of the momentum density $\rho(\boldsymbol{p})$. Besides taking the $\boldsymbol{k}$-dependence of the overlap matrix element into account, such calculations can also give a more realistic description of the Fermi surface and the (outer d + conduction) electron states in the transition metals. Such calculations employing Hubbard's approximation scheme have already been

reported by Mijnarends (1973 *a*) for copper. We have extended these calculations by incorporating additional features which are outlined in the next section.

2. Calculation

The procedure of present calculations closely follows the theory discussed by Mijnarends (1973 *a*). The extra features of the present work are as follows: (*a*) The positron wavefunction ϕ_+ was calculated by employing a symmetrised plane wave expansion (Gould *et al* 1972) in which

$$\phi_+(r) = (1/\tau)^{1/2} \sum_{K} A_K \exp(i\, \boldsymbol{K}.\boldsymbol{r}) \tag{1}$$

The coefficients A_K's were obtained by solving the secular equation

$$\sum_{K} [(K^2 - E)\,\delta_{KK'} + V_{K-K'}]\, A_K = 0 \tag{2}$$

where the Fourier coefficients $V_{K-K'}$ of the crystal potential were obtained from the muffin-tin potential of Burdick (1963) using partial waves up to $l = 12$ in the following formula

$$V_{K-K'} = (1/\tau) \int_{\tau} V(r) \exp[i\,(\boldsymbol{K} - \boldsymbol{K'}) \cdot \boldsymbol{r}]\, d\boldsymbol{r} \tag{3}$$

In eq. (1) the sum was carried out for the first 89 reciprocal lattice vectors and the eigenvalue (for the $\boldsymbol{k} = 0$ state) of the positron was found to be 0·559 Ry with respect to the constant potential in the interstitial region. The first Fourier coefficient A_K was adjusted to make $\phi_+(\boldsymbol{r} = 0) = \tau^{-1/2} \sum_{K} A_K = 0$ subject to $\sum_{K} |A_K|^2 = 1$. The resulting $\phi_+(\boldsymbol{r})$ along three (h, k, l) directions is shown in

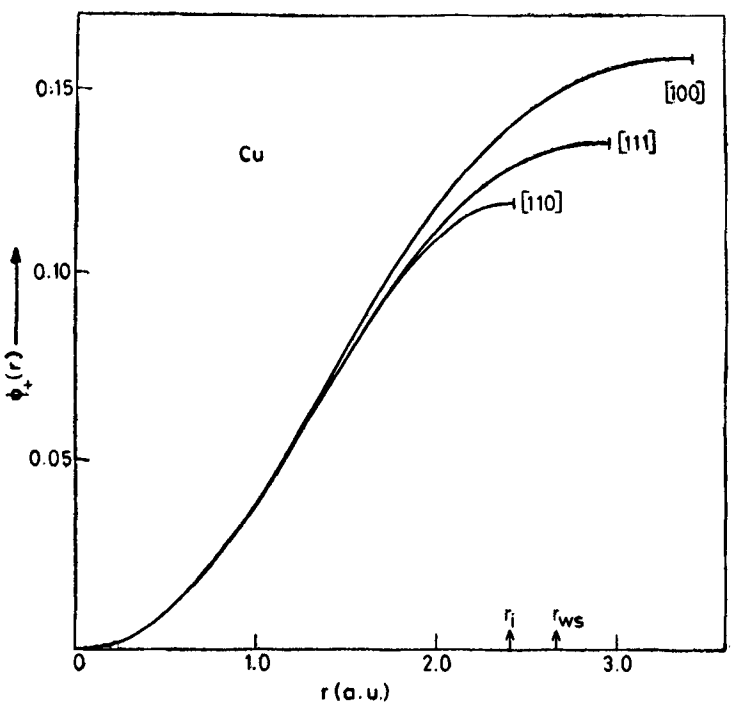

Figure 1. Positron wavefunction along three directions in copper.

figure 1. In subsequent computations of $\rho(p)$ a spherically averaged positron wavefunction calculated from the formula (Betts *et al* 1956)

$$\phi_+^{sph}(r) = \frac{1}{35}[10\,\phi_+^{100}(r) + 16\phi_+^{110}(r) + 9\phi_+^{111}(r)] \tag{4}$$

was used throughout.

(*b*) In the calculation of the overlap matrix element between the positron and the (3d + conduction) electrons [eq. 7 of Mijnarends (1973 *a*)] by the Hubbard's approximation scheme, terms up to $l = 3$ were considered for the electronic wavefunction inside the muffin-tin sphere.

(*c*) The contribution to $\rho(p)$ by the core (in our case the " core " consisted of the 1s 2s 2p 3s 3p orbitals) electrons was included by calculating it in the spherical approximation following Gould *et al* (1972) and using Herman-Skillman (1963) free atom wavefunctions.

3. Results and discussion

The band structure of copper calculated by us is in good agreement with the results of Burdick (1963). The results for $\rho(p)$ along the ⟨100⟩, ⟨110⟩ and ⟨111⟩ directions in copper looked very similar to those of Mijnarends (1973 *a*) with some minor adjustments brought in because of the additional features in the present calculations.

To compare our results with experimental data we calculated the theoretical angular correlation curves for a long slit and point slit geometry with a polycrystal-

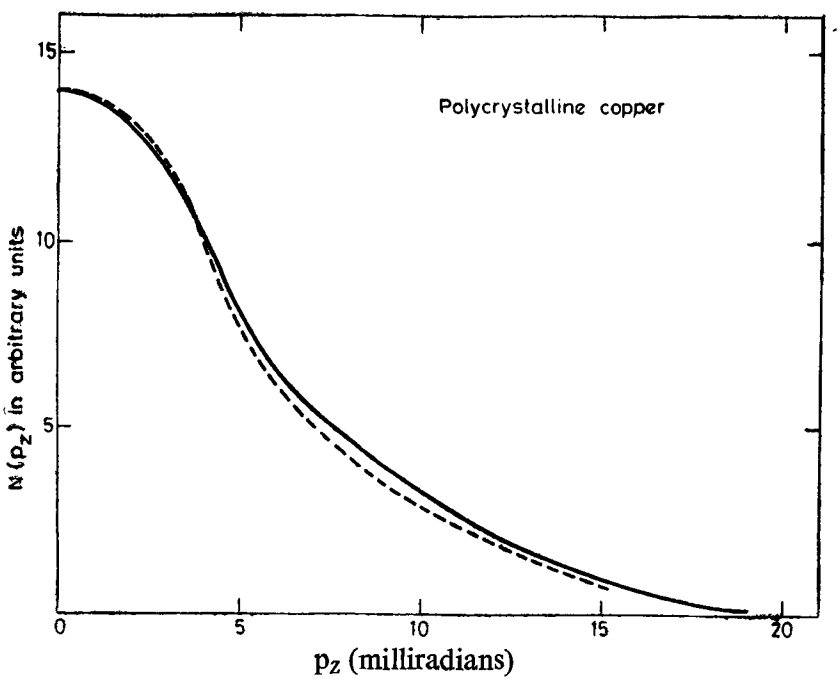

Figure 2. Comparison between the calculated and experimental long slit angular correlation curves for polycrystalline copper. — — — Berko *et al* (1968) and ————Theory.

line copper sample. The long slit curve was obtained by using the relation

$$N(p_z) = \int_{|p_z|}^{\infty} \rho_{\text{sph}}(p)\, p\, dp \tag{5}$$

where the spherically averaged $\rho_{\text{sph}}(p)$ was obtained by averaging the ρ_{100}, ρ_{110} and ρ_{111} with a formula similar to eq. (4). The experimental curves for $N_{100}(p_z)$, $N_{110}(p_z)$ and $N_{111}(p_z)$ by Berko *et al* (1968) were similarly spherically averaged and the resulting curve is shown in figure 2 along with the theoretical $N(p_z)$ curve. The comparison in figure 2 indicates that the theory slightly underestimates $N(p_z)$ at low momenta while it overestimates $N(p_z)$ at high momenta. These results are similar to those obtained for nickel (Singru and Mijnarends 1974). After completing the present work we received the report describing the modified augmented plane wave (MAPW) calculations of $\rho(p)$ in copper by Bross and Stöhr (1974). Although these authors find a good agreement with the measured (Berko *et al* 1968, Cushner *et al* 1970) anisotropy $[N_{111}(p_z) - N_{110}(p_z)]$ the agreement with $N(p_z)$ itself is not equally satisfactory. They suggest that a momentum-dependent enhancement factor arising out of many-body correlations might be necessary to explain this disagreement. Our results in figure 2 seem to support this suggestion.

The point slit angular correlation curve was calculated using the formula

$$N(p_y, p_z) = 2 \int_{p_0}^{\infty} \rho_{\text{sph}}(p)\,(p^2 - p_0^2)^{-1/2}\, p\, dp \tag{6}$$

where $p_0 = (p_y^2 + p_z^2)^{1/2}$. For computational convenience (*i.e.*, to avoid the singularity in the integrand at $p = p_0$) eq. (6) was rewritten, following Gould (1973), as

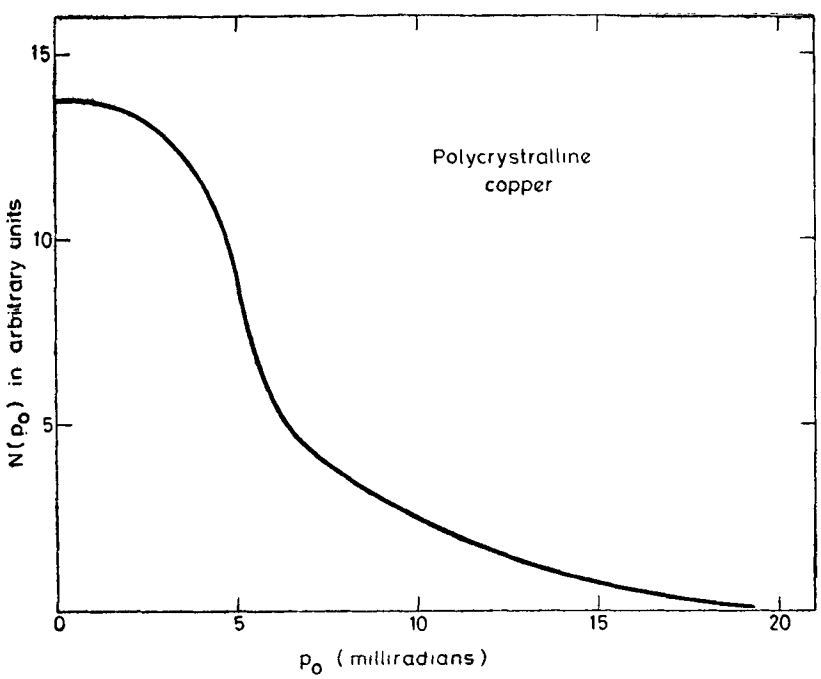

Figure 3. Calculated point slit angular correlation curves for polycrystalline copper.

$$N(p_y, p_z) = 2\rho_{\text{sph}}(p_0) \int_{p_0}^{p_0+\Delta} (p^2 - p_0^2)^{-1/2}\, p\, dp + 2 \int_{p_0+\Delta}^{\infty} \rho_{\text{sph}}(p)\,(p^2 - p_0^2)^{-1/2}\, p\, dp \quad (7)$$

where the interval $\triangle$ was chosen as 0·25 mrad, a value such that $\rho_{\text{sph}}(p)$ changes very little from p_0 to $p_0 + \triangle$. The first integral was evaluated analytically, its value being $2\rho_{\text{sph}}(p)\,(2p_0\triangle + \triangle^2)^{1/2}$. The resulting curve, shown in figure 3, cannot be *directly* compared with the results of Senicki *et al* (1973) because they measure the variation of $N(p_y, p_z)$ with p_z with four sets of coincidence detectors while we have obtained the plot of $N(p_0)$ against p_0 (where $p_0 = (p_y^2 + p_z^2)^{1/2}$). Nevertheless, we feel that the theoretical curve in figure 3 can be used, with the relation $p_0 = (p_y^2 + p_z^2)^{1/2}$, as a *basis* to compare the experimental results for polycrystalline copper obtained by point slit, spark chamber or multi-counter geometry. For a more complete description of single crystal results obtained by any of the above geometries, a calculation of $\rho(\boldsymbol{p})$ as well as subsequent integration over one or two components of $\boldsymbol{p}$ throughout the $\boldsymbol{k}$-space will have to be carried out.

In conclusion, the results of the present work as well as that of the MAPW calculations (Bross and Stöhr 1974) show that the band structure calculations give a good description of the two photon pair momentum densities in copper and can provide a basis to understand the effects of enhancement.

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