Positron Annihilation in Copper – Comparisons of Different Results*

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Different positron annihilation measurements of the momentum distributions in copper are compared. A disagreement and some indications about the nature of the electron wave functions are found.

Positron annihilation studies of the momentum distribution of electrons in single crystals of copper using point detectors,[1] rectangular slits,[2,3] and wide slits[4–6] have been reported. Annihilations of positrons with conduction electrons whose momenta are in the necks of the copper Fermi surface are detected. A comparison of these results reveals a disagreement and points, with some simple considerations, to the possible nature of the 4*s* and 3*d* wave function of metallic copper.

In the point detector[1] and rectangular slit measurements,[2] the single-crystal sample is turned so that various directions, including the [111], are in turn aligned with the detectors. An increase in γ-coincidence counting rate at [111] due to the Fermi-surface necks is observed.

Taking into account resolution, the magnitude of this increase measured by these two techniques differs by over a factor of 2. The angular resolution of 1 mrad of the point detectors[1] corresponds to 13° width at the Fermi surface. The neck width is about 21°. In the rectangular slit case in one direction the resolution is 0.80 mrad, but in the other it is 5 mrad. This measurement samples a thin slice of momentum space about 2.5 times wider than the necks of the Fermi surface and about half as wide as its belly. In effect, the counting rate due to the background is doubled, and the relative increase in counting rate due to the necks would be about half of that observed with point detectors.

With point detectors, the increase in counting rate due to the necks is about 4%.[1] So with the rectangular slits one would expect about 2%. The observed increase,[2] however, is 6%.

A detailed interpretation of positron annihilation results, using the independent particle approach,[4,7–9] centers around the calculation of $\rho(\vec{p})$, that is, the probability that the center-of-mass momentum of the annihilating pair is $\vec{p}$. For conduction electrons in a metal in the nearly free-electron approximation[6] or in the orthogonalized-plane-wave (OPW) approximation[8] (with small orthogonalization terms), $\rho(\vec{p})$ is nearly constant over the inner part of the Brillouin zone but falls to half its zone-center value at a zone boundary (Fig. 1); $\rho(\vec{p})$ is zero in unoccupied parts of momentum space and small in neighboring Brillouin zones (in the periodic or repeated zone scheme).

The collinear point detector apparatus[1] measures a quantity (ignoring finite resolution)

$$\nu = \int_{-\infty}^{\infty} \rho(p_x, p_y, p_z)\big|_{p_z = p_y = 0}\, dp_x\,, \tag{1}$$

where the crystal is rotated so that various directions, including the [111], are in turn aligned with the p_x axis.

Now, for the conduction 4s electrons in Cu, the occupied part of momentum space is well known[10,11] and, for the present purposes, can be approximated by a sphere with necks (Fig. 1). If the equatorial cross section of the sphere is taken to be the average of the measured belly areas[10,11] (which do not differ by more than 4%), then taking $\rho(\vec{p}) = \rho(0)$ over the sphere and using the symmetry of $\rho(\vec{p})$ in the necks, the ratio of ν_{111} (for p_x in the [111] direction) to the isotropic conduction electron background ν_B can be estimated. Thus, $\nu_{111}/\nu_B = 1.13$ or $(\nu_{111} - \nu_B)/\nu_B = 0.13$.

The experimental counting rate includes a contribution from core (mainly 3d) electrons. This has been estimated to be about 0.75 of the 4*s*

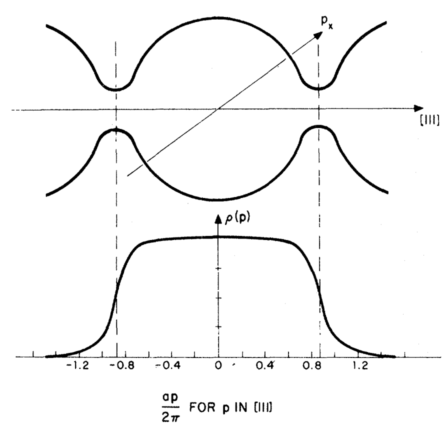

FIG. 1. Schematic of the copper Fermi surface and of the function $\rho(\vec{p})$.

counting rate.[2] With the core contribution subtracted from the rectangular slit results[2] the increase in counting rate due to the necks above the *conduction* electron background would be about 11%. Inclusion of the factor of 2 arising from the resolution in the estimate $(\nu_{111} - \nu_B)/\nu_B = 0.13$ reduces it to 0.065 which is considerably smaller than the measured 0.11.

Two considerations might account for the gross failure of the simple estimate:

(a) The function $\rho(\vec{p})$ might be larger in the necks than expected from the nearly free-electron model (Fig. 1). The small 1*s* orthogonalization part of the conduction electron wave function in Li causes a small increase of $\rho(\vec{p})$ above the value of $\frac{1}{2}$ at the zone boundary.[8] In Cu such orthogonalization terms to the conduction electron wave function would be much larger and might make $\rho(\vec{p})$ larger in the necks.

(b) The many-body electron-positron correlation[12] produces a large enhancement of the annihilation probability with conduction electrons in a metal. This enhancement is larger near the Fermi momentum than in the interior of the Fermi surface. What the enhancement might be for the necks is open to question. The calculations were done for a free-electron gas[12] and do not seem applicable to the neck electrons which have one effective-mass component negative.

Berko *et al.*[5] also suggest the non-plane-wave character of the wave function and enhancement as possible explanations for the difference between the anisotropy measured with wide slits and that expected from the Fermi-surface geometry alone. By fitting a Kubic harmonic expansion of $\rho(\vec{p})$ to experimental wide slit measurements of $N(p_z)$, Mijnarends[6] obtains an experimental function $\rho(\vec{p})$. Although due probably to enhancement there is some peaking of the function near the Fermi radius, in the necks $\rho(\vec{p})$ seems to follow the nearly free-electron model[4] and falls to $\frac{1}{2}$ as shown in Fig. 1. Nevertheless, because of the finite number of terms in the expansion and because of the angular resolution, $\rho(\vec{p})$ does not show the expected discontinuity as a function of angle that results in going from occupied states in the necks to unoccupied states outside. Therefore, the determination of $\rho(\vec{p})$ in the necks by this expansion technique[6] is probably not sensitive enough to rule out the two possible causes of the high annihilation rate in the necks proposed above.

We return briefly to the ratio of core electron to conduction electron annihilation in ν. This can also be estimated from the wide slit results[4,5] which measure

$$N(p_z) = \int\int_{-\infty}^{\infty} dp_x\, dp_y\, \rho(\vec{p}) \quad . \tag{2}$$

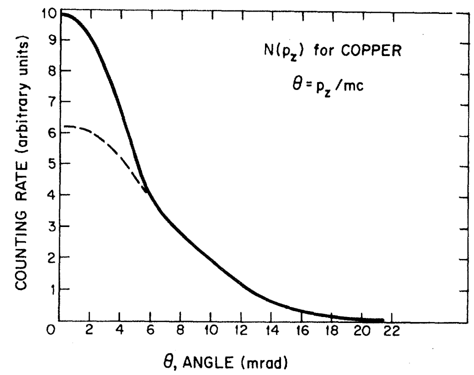

FIG. 2. Wide slit angular correlation results for copper. Solid line is the average of the measured results for the three symmetry directions (Ref. 4, Fig. 11 or Ref. 5). Dashed line is the extrapolation of the core contribution deduced by a comparison with Ref. 2.

$\rho(\vec{p})$ can here be assumed isotropic, i.e., a function of only $q = (p_x^2 + p_y^2 + p_z^2)^{1/2}$. In this case,

$$N(P_z) = 2\pi \int_{P_z}^{\infty} \rho(q)\, q\, dq \quad . \tag{3}$$

From this follows the relation

$$\nu = -\frac{1}{\pi}\int_0^{\infty} \frac{1}{p_z}\,\frac{dN}{dp_z}(p_z)\, dp_z\,. \tag{4}$$

If we separate the contributions of conduction band and core electrons to $N(p_z)$ for Cu (Fig. 11, Ref. 4) and use Eq. (4) to calculate the separate contributions to ν, we find that to get agreement with the estimated[2] ratio $\nu_{\mathrm{core}}/\nu_{\mathrm{cond}} = 0.75$ we must extrapolate the core distribution below 6 mrad as shown in Fig. 2. Thus the core contribution is much more peaked than that predicted from the atomic *M* shell[4] (mostly 3*d*) functions,[13] indicating the 3*d* electrons to be more spread out in real space.

The three main observations in this note are (a) the relative increase in counting rate due to the necks of the Cu Fermi surface measured by rectangular slits[2,3] is more than twice as large as that measured by the point detectors[1]; (b) this increase measured by the rectangular slits[2,3] is almost twice as large as a simple theoretical estimate possibly because of large departures of the conduction electron wave function from a plane wave and/or to enhancement[12]; and (c) the core electron contribution to the wide slit results at zero angle is larger than expected from atomic *M* shell wave functions.[4]

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