Anisotropic Positron Annihilation with the High-Momentum Components of the Conduction Band in Copper*

S. CUSHNER, J. C. ERSKINE, AND S. BERKO
Brandeis University, Waltham, Massachusetts 02154
(Received 18 July 1969)

High-precision measurements of the angular correlation of annihilation radiation from oriented single crystals of copper are reported, showing marked anisotropies even at large angles. A comparison with the predictions of a simple theoretical model indicates that most of the large-angle anisotropy can be attributed to positron annihilation with the high-momentum components of the conduction band in copper.

POSITRON annihilation experiments can yield, in principle, valuable information about electron momentum distribution in metals and alloys.[1] In recent years several investigators have used copper as a test case to study the extent to which such experiments are relevant to the shape of the Fermi surface.[2–6] In this paper we report measurements representing the first direct observation of the existence of high-momentum component annihilation with the conduction band in copper.

The two-quantum angular distribution, as measured by the standard long-slit geometry apparatus, is given in the independent particle model, as a function of the angle $\theta=p_z/mc$, by

$$N_{\hat{n}}(p_z)=\int_{-\infty}^{+\infty}\int \rho(\mathbf{p})dp_x dp_y=\int_{-\infty}^{+\infty}\int dp_x dp_y \times \sum_{\substack{k\\ \text{occupied states}}} \left|\int_{\text{xtal}} d^3\mathbf{r}e^{-i\mathbf{p}\cdot\mathbf{r}}\psi_+(\mathbf{r})\psi_k(\mathbf{r})\right|^2, \quad (1)$$

where $p_z=\theta mc=\mathbf{p}\cdot\hat{n}$, $\hat{n}$ is a unit vector describing the crystal orientation, $\psi_+(\mathbf{r})$ is the positron ground-state wave function, and $\psi_\mathbf{k}(\mathbf{r})$ is the wave function of a Bloch electron in state $\mathbf{k}$; $\hbar=1$. Expanding the wave function product in plane waves, one obtains

$$\rho(\mathbf{p})\propto \sum_{\substack{k\\ \text{occupied states}}} \sum_{\mathbf{G}} \delta(\mathbf{p}-\mathbf{k}-\mathbf{G})|A_\mathbf{G}(\mathbf{k})|^2, \quad (2)$$

where $\mathbf{G}$ is a reciprocal lattice vector, and the $A_\mathbf{G}(\mathbf{k})$ are the Fourier components

$$A_\mathbf{G}(\mathbf{k})=\int_{\text{cell}} d^3\mathbf{r}\psi_+(\mathbf{r})\psi_k(\mathbf{r})e^{-i(\mathbf{G}+\mathbf{k})\cdot\mathbf{r}}. \quad (3)$$

Thus, each electron with wave number $\mathbf{k}$ contributes to the annihilation not only at $\mathbf{p}=\mathbf{k}$, but at every $\mathbf{p}=\mathbf{k}+\mathbf{G}$ with a relative weight $|A_\mathbf{G}(\mathbf{k})|^2$.

In the first paper[2] (BP), on annihilation from single crystals of copper, the importance of these high-momentum component (HMC) annihilations was studied in various independent particle approximations. Although since the BP paper several complete band computations[7–9] have been performed, which include the effect of the HMC's, the statistical accuracy of the existing annihilation data for large angles (high momenta) have so far been insufficient to indicate clearly the existence of the HMC ("umklapp") annihilations.[10]

Our two-photon angular distributions were obtained using a standard apparatus[1] with detector slits subtending 1 by 150 mrad at the sample. A source of approximately 150 mCi of Co^{58} was used with a superimposed 15-kG focusing field. The samples were oriented single-crystal cylinders of copper ($\hat{n}$ along the axis of the cylinder), 0.3 in. in diameter and 0.25 in. long, slowly annealed prior to the experiment. Data were taken with the samples at room temperature, in vacuum. To test for possible instrumental errors, the experiments were also performed on a second correlation apparatus with 1- by 90-mrad slits, and at 77°K. No significant differences were observed between the two independent sets of data.

As discussed in BP, the angular correlation curves in copper include, besides the annihilation with the conduction band, a broad distribution resulting from the annihilation with core electrons, particularly with the $3d$ band. In order to exhibit the anisotropies of the correlation curves most sensitively, we have plotted in Fig. 1 the differences between the experimental points taken with $\hat{n}$ along the [111], [110], and [100] directions [see Eq. (1)]. Thus all isotropic contributions cancel out. We have marked on the $\theta=p_z/mc$ axes the intersections with the relevant first zone faces as $Z_{[ijk]}(Z_{[ijk]}=\frac{1}{2}(\hat{n}\cdot\mathbf{G}_{[ijk]}))$.

Well-defined oscillations are evident for angles corresponding to p_z outside the first zone, which we attribute to HMC annihilations. The amplitude of the oscil-

* Work supported by the National Science Foundation and the U. S. Army Research Office (Durham).

[1] *Positron Annihilation*, edited by A. T. Stewart and L. O. Roellig (Academic Press Inc., New York, 1967).
[2] S. Berko and J. S. Plaskett, Phys. Rev. **112**, 1877 (1958).
[3] K. Fujiwara and O. Sueoka, J. Phys. Soc. Japan **21**, 1947 (1966).
[4] D. L. Williams *et al.*, Phys. Rev. Letters **20**, 448 (1968).
[5] S. Berko, S. Cushner, and J. C. Erskine, Phys. Letters **27A**, 668 (1968).
[6] P. E. Mijnarends, Phys. Rev. **178**, 622 (1969).
[7] J. Melngailis and S. DeBenedetti, Phys. Rev. **145**, 400 (1966).
[8] R. W. Williams and A. R. Mackintosh, Phys. Rev. **168**, 679 (1968).
[9] D. Stroud and H. Ehrenreich, Phys. Rev. **171**, 399 (1968).
[10] See, for example, J. J. Donaghy and A. T. Stewart, Phys. Rev. **164**, 391 (1967).

lations corresponds to approximately 2% of the total counting rate at high momenta. We expect that the sharp oscillations within the first zone reflect mainly the asphericity of the Fermi surface of copper, as discussed by us in a previous paper.[5] However, even at p_z close to 0, HMC's can and do contribute.[11] In order to establish whether or not the observed anisotropies come from the conduction band of copper, we have calculated theoretical difference curves using the following extremely simple approximation: (a) We assumed that the $A_G(\mathbf{k})$ in Eq. (3) are $\mathbf{k}$-independent. This assumption (Wigner-Seitz) is expected to be, of course, unrealistic for copper in regions of $\mathbf{k}$ close to the (111) face, where the large energy gap will cause an appreciable $\mathbf{k}$ dependence of the $A_G(\mathbf{k})-s$, even in the nearly free electron approximation, as discussed in Ref. 2. (b) We centered about each $\mathbf{G}$, with the proper orientation, the known copper Fermi surface,[12] and obtained the theoretical $N_{\hat{n}}(p_z)$ by weighting each of these surfaces by the proper $|A_G|^2$ and computing the cross-sectional areas of this system of weighted Fermi surfaces for the three $\hat{n}$ directions. Only the one $\mathbf{G}_{[000]}$, the eight $\mathbf{G}_{[111]}$, and the six $\mathbf{G}_{[200]}$ vectors were used. (c) These theoretical cross-sectional areas were then mathematically folded into the resolution of the apparatus. (d) The theoretical difference curves were finally obtained and were normalized for best fit by a single normalization factor to the data (plotted in arbitrary count-rate units). Two such sets of theoretical difference curves are plotted on Figs. 1(a), 1(b), and 1(c), together with the experimental points. The curves marked A correspond to $|A_{[000]}|^2=0.9082$, $|A_{[111]}|^2=0.0161$, and $|A_{[200]}|^2=0.0076$. These Fourier components are identical to those computed for copper in the Wigner-Seitz approximation in BP. For the curves marked B we used $|A_{[000]}|^2=0.890$, $|A_{[111]}|^2=0.010$, $|A_{[200]}|^2=0.005$. We notice that the difference curves do not change substantially between these two sets of Fourier components; on the other hand, the values of set B were chosen because the $N_{\hat{n}}(\theta)$ obtained with them fit better the individual shapes of the observed angular distribution data, after a Gaussian had been subtracted for the core contribution.[13] (The difference curves do not, of course, depend on such a subtraction.) The over-all fit in shape and even in magnitude to the experimental points is surprisingly good in view of the obvious deficiencies of our theoretical model. The largest discrepancy is observed in the region between 4–8 mrad, particularly for the $N_{[110]}(\theta)-N_{[100]}(\theta)$ curve, and can probably

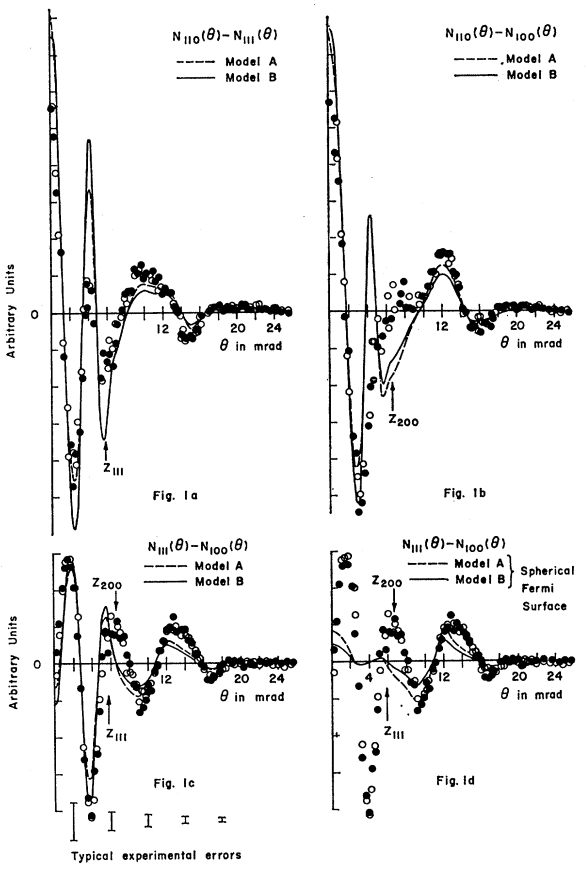

FIG. 1. Experimental and theoretical difference curves of two-photon angular correlations from oriented copper single crystals. The theoretical models used are described in the text. The experimental points are folded about $p_z=0$, open circles representing data for $\theta<0$. Typical errors are indicated at the bottom of Fig. 1(c).

be attributed to anisotropies[14,15] in the $3d$ band due to $(s-d)$ mixing. In order to demonstrate that within the first zone in the difference curves reflect mainly the topology of the Fermi surface, we have plotted in Fig. 1(d) the theoretical difference curves, using the same Fourier components as for Figs. 1(a), 1(b), and 1(c), but with a spherical Fermi surface. The other difference curves exhibit the same large discrepancies for p_z inside the first zone.

In order to check that the observed high-momentum anisotropies do not stem from Laue diffraction of the annihilation photons traversing the crystalline samples, we have performed an independent check: We measured the angular distribution exhibiting a narrow positronium peak[16] from annihilation in an Al_2O_3 sample,

[11] We do not agree with the assertion of Sueoka [O. Sueoka, J. Phys. Soc. Japan **23**, 1246 (1967)] and of Donaghy and Stewart (Ref. 10) that HMC's cannot introduce anisotropies at $p_z=0$. This holds strictly only for the nearly free electron approximation, if we take only one zone face into account.

[12] D. J. Roaf, Phil. Trans. Roy. Soc. London **A255**, 135 (1962).

[13] The direct comparison between the experimental and theoretical $N_{\hat{n}}(\theta)$ indicates that a k-dependent "enhancement factor" would not necessarily improve the fit.

[14] Such an anisotropy has been noted by P. E. Mijnarends (Ref. 6).

[15] The sharp peak at around 16 mrad observed recently in a "rotation" measurement by Sueoka [O. Sueoka, J. Phys. Soc. Japan **26**, 864 (1969)], and attributed by him to core anisotropy, cannot be easily compared with our present results, because of the specialized geometry of his measurement.

[16] J. Løvseth, Phys. Norveg. 1, 145 (1963).

with a copper crystal placed between the Al_2O_3 sample and our movable counter, oriented as in the copper annihilation experiments. No Laue peaks were observed in this "correlation-absorption" curve, taken with the same statistical accuracy as the original copper $N_{\hat{n}}(\theta)$.

We conclude therefore that the observed high-momentum anisotropies in copper can be attributed to a large extent to HMC annihilations with the conduction band and that the observed order of magnitude of these HMC is in agreement with simple independent particle computations. It is clear that a far better theoretical calculation is needed to include the following effects neglected in our model: (a) k-dependent Fourier components obtained from a "first-principle" band computation, which includes s-d hybridization both for the "s" and the "$3d$" bands and the effect of the large energy gaps on the (111) faces, combined with anisotropic positron wave functions[17]; (b) k-dependent enhancement factors due to positron-electron correlation.[18]

The authors thank A. Thompson and J. M. Weingart for technical assistance during the experiments.

[17] Similarly to the computation of Ref. 9 for Si.
[18] J. P. Carbotte and S. Kahana, Phys. Rev. **139**, A213 (1965).

PHYSICAL REVIEW B VOLUME 1, NUMBER 7 1 APRIL 1970

Straggling of Heavy Charged Particles: Comparison of Born Hydrogenic-Wave-Function Approximation with Free-Electron Approximation*

HANS BICHSEL†
Lawrence Radiation Laboratory, University of California, Berkeley, California 94720
(Received 26 September 1969)

Statistical fluctuations in the energy loss of heavy charged particles in thin absorbers resulting from collisions with atomic electrons are determined for collision cross sections obtained from the first Born approximation calculated with hydrogenic wave functions. A comparison is given with the results calculated with a $1/\epsilon^2$ collision spectrum.

1. INTRODUCTION

STRAGGLING functions describe the statistical fluctuations of the energy losses of fast charged particles. Landau[1] has introduced a transport equation describing the behavior of the straggling function $f(x,\Delta)$ for energy losses Δ small compared to the initial energy T of the incident particle:

$$\frac{\partial f(x,\Delta)}{\partial x} = \int_0^\infty w(\epsilon) f(x, \Delta-\epsilon) d\epsilon - f(x,\Delta)\sigma_t, \quad (1)$$

where $f(x,\Delta)$ is the probability density function of particles that have penetrated a thickness x of the absorber and have experienced an energy loss Δ; $w(\epsilon)d\epsilon$ is the differential collision cross section for single collisions, with an energy loss ϵ; and $\sigma_t = \int_0^\infty w(\epsilon)d\epsilon$ is the total collision cross section. This equation has recently been discussed by Tschalär,[2] and Kellerer.[3]

In experiments, for example, N_0 particles penetrate an absorber of a given thickness x. The number dN of particles emerging with energy losses between Δ and $\Delta+d\Delta$ is given by $dN = N_0 f(\Delta) d\Delta$. Often, only the mean energy loss $\langle\Delta\rangle = \int f(\Delta)\Delta d\Delta$ is of interest. It is closely related to the stopping power S: $\langle\Delta\rangle \simeq xS$.

The collision cross section $w(\epsilon)$ is of great importance in the solution of Eq. (1). The simple approximation $w(\epsilon) = k/\epsilon^2$ used so far[1,2,4] and a more realistic function obtained from calculation in first Born approximation using hydrogenic wave functions are therefore discussed in Sec. 2. It may be noted, though, that the true collision cross section $w(\epsilon)d\epsilon$ for single atoms is zero below an energy ϵ_I equal to the difference in energy between the lowest possible excited state and the ground state of the atoms, and also vanishes rapidly for $\epsilon > \epsilon_m \simeq 2mv^2$. Similarly, $f(x, \Delta-\epsilon)$ must be equal to zero for $\epsilon > \Delta$. The limits of integration introduced by Vavilov have to be understood from these conditions.

The solution of the transport equation using the Laplace transform[1,4] is

$$f(x,\Delta) = \frac{1}{2\pi i}\int_{c-i\infty}^{c+i\infty} \exp\left[p\Delta - x\int_0^\infty w(\epsilon)(1-e^{-p\epsilon})d\epsilon\right] dp. \quad (2)$$

* Work supported in part by the U. S. Atomic Energy Commission and Public Health Service Research Grant No. CA-08150 from the National Cancer Institute.
† Now at Department of Radiology, University of Washington, Seattle, Wash. 98105.
[1] L. Landau, USSR J. Phys. **8**, 201 (1944).
[2] C. Tschalär, Nucl. Instr. Methods **61**, 141 (1968).
[3] A. Kellerer, G. S. F. Bericht *B*-1 Strahlenbiologisches Institut der Universität München.
[4] P. V. Vavilov, Zh. Eksperim. i Teor. Fiz. **32**, 920 (1957) [English transl.: Soviet Phys.—JETP **5**, 749 (1957)]. Extensive tabulations of the Vavilov functions are given by Seltzer and Berger, Natl. Acad. Sci.—Natl. Res. Council, Publ. **1133**, 187 (1967), 2nd printing.