Positron Annihilation and Orthogonalized Plane Waves in Lithium*

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Anisotropies in the momentum distribution of positron annihilation radiation have in the past been observed in several single-crystal metals, most recently in Li. Interpretation of the results in terms of Fermi-surface structure is complicated. The angular correlation pattern is affected not only by the Fermi-surface shape but also by the necessary departure of the conduction-electron wave function from a plane wave. The magnitude of these two effects is evaluated in Li. The orthogonalized-plane-wave method with 19 waves is used to obtain the lowest band, $E(\mathbf{k})$, and the corresponding conduction-electron wave functions. The numerically calculated angular correlation curves show anisotropies in agreement with the measured results if both the effect of the Fermi-surface shape and the effect of the wave function are included. If the Fermi-surface shape alone is considered, the observed anisotropies cannot be explained. Quantitative agreement with experiment can be obtained if, in addition to all other effects, the enhancement factor due to electron-positron attraction calculated by Kahana is included.

INTRODUCTION

ONE of the aims of positron-annihilation, two-photon, angular correlation measurements has been to study the momentum distribution of electrons in solids.[1] This aim has perhaps to some extent been fulfilled in light metals. When a positron annihilates with an electron in a solid, the center-of-mass momentum of the two particles appears as a small deviation from collinearity of the two gamma rays emitted. This momentum is due mostly to the electron since the positron can be assumed thermalized.[2] The conduction-electron momenta (on a free-electron model) are less than or equal to the Fermi momentum, k_F. Thus one would expect a sharp drop in the number of γ pairs emitted with a deviation from collinearity corresponding to a momentum larger than k_F. This, in fact, has been observed for many metals.[3]

One might, therefore, hope that angular-correlation studies on oriented metal single crystals would yield information on Fermi surface structure. Such studies have been performed on several materials, for example, graphite,[4] Cu,[5] Al,[5,6] Ge,[6] Si,[6,7] Be,[8] Mg,[8] and, most recently, Na and Li.[9] Although in most of these cases the angular-correlation curves show clear anisotropies, interpretation in terms of Fermi-surface structure is difficult. As Berko pointed out,[8] the angular-correlation curves in a real metal are affected not only by the fact that the maximum electron momentum k_F depends on direction in the crystal but also by the departure of the electron wave function from a plane wave which must accompany the nonspherical Fermi surface.

In the present paper the orthogonalized plane-wave method is applied to single-crystal Li to obtain conduction-electron wave functions which correspond to a nonspherical Fermi surface. These wave functions are then used to calculate the long slit angular correlation curve $N(p_z)$ for [100], [110], and [111] orientations. The extent to which anisotropies in the curves and departures from parabolas can be due to the details of the wave functions and the extent to which they can be due to the Fermi surface shape are determined. Many-body effects are neglected except as discussed at the end.

1. CONDUCTION-ELECTRON WAVE FUNCTION

The orthogonalized plane wave method[10] (OPW) has been used to obtain the conduction-electron energy bands in several metals.[11–13] Callaway most recently applied the method to Li.[14] We follow his calculation

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† Supported by Westinghouse Electric Corporation.

[1] The most recent review is P. R. Wallace, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Vol. 10.

[2] G. E. Lee-Whiting, Phys. Rev. **97**, 1557 (1955).

[3] G. Lang, S. DeBenedetti, and R. Smoluchowski, Phys. Rev. **99**, 596 (1955).

[4] S. Berko, R. E. Kelley, and J. S. Plaskett, Phys. Rev. **106**, 824 (1957).

[5] S. Berko and J. S. Plaskett, Phys. Rev. **112**, 1877 (1958).

[6] P. Colombino, B. Fiscella, and L. Trossi, Nuovo Cimento **31**, 950 (1964).

[7] J. C. Erskine and J. McGervey, Bull. Am. Phys. Soc. **9**, 478 (1964).

[8] S. Berko, Phys. Rev. **128**, 2166 (1962); A. T. Stewart, J. B. Shand, J. J. Donaghy, and J. H. Kusmiss, *ibid.* **128**, 118 (1962).

[9] (a) J. J. Donaghy and A. T. Stewart, Bull. Am. Phys. Soc. **9**, 238 (1964); (b) A. T. Stewart, J. J. Donaghy, J. H. Kusmiss, and D. M. Rockmore, *ibid.* **9**, 238 (1964); (c) J. J. Donaghy and A. T. Stewart (private communication); (d) J. J. Donaghy, A. T. Stewart, D. M. Rockmore, and J. H. Kusmiss, in *Proceedings of the Ninth International Conference on Low Temperature Physics, Columbus, Ohio, 1964*, edited by J. G. Daunt, D. V. Edwards, F. J. Milford, and M. Yagub (Plenum Press, Inc., New York, 1965), p. 835.

[10] C. Herring, Phys. Rev. **57**, 1169 (1940).

[11] J. Callaway, Phys. Rev. **97**, 933 (1955).

[12] T. O. Woodruff, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1957), Vol. 4, p. 367.

[13] J. Callaway, *Energy Band Theory* (Academic Press Inc., New York, 1964), p. 68.

[14] J. Callaway, Phys. Rev. **124**, 1824 (1961); **131**, 2839(E) (1963). The periodic potential used by Callaway is the semi-empirical Seitz potential as quoted by W. Kohn and N. Rostoker, Phys. Rev. **94**, 1111 (1954). The wave function u_{1s} was calculated (Ref. 16) to be the ground state in this potential which is seen by the *conduction* electrons. It is therefore different from the *atomic* u_{1s}.

in an abbreviated form to obtain the filled part of the $2s$ band in Li in 5 directions of $\mathbf{k}$, and then, in addition we determine the wave function at the same points. (Atomic units with $e=m=\hbar=1$ are used.)

The conduction-electron wave function is expanded as follows:

$$\psi_{\mathbf{k}}=\sum_{\mathbf{K}} a_{\mathbf{K}}(\mathbf{k})\chi_{\mathbf{k}+\mathbf{K}}. \tag{1.1}$$

The sum extends over reciprocal lattice vectors $\mathbf{K}$ which are inside a sphere about $\mathbf{K}=0$, and the χ's are the orthogonalized plane waves given by

$$\chi_{\mathbf{k}}=V^{-1/2}e^{i\mathbf{k}\cdot\mathbf{r}}-\mu_{\mathbf{k}}\varphi_{\mathbf{k}}, \tag{1.2}$$

where V is the volume of the crystal and $\varphi_{\mathbf{k}}$ is the tight-binding approximation to the core state in Li,

$$\varphi_{\mathbf{k}}=N^{-1/2}\sum_{\mathbf{R}} e^{i\mathbf{k}\cdot\mathbf{R}}u_{1s}(\mathbf{r}-\mathbf{R}). \tag{1.3}$$

u_{1s} is the core $1s$-type wave function, and the sum is over the N lattice sites in the crystal located at points $\mathbf{R}$. The coefficient $\mu_{\mathbf{k}}$ is determined so that $\chi_{\mathbf{k}}$ is orthogonal to $\varphi_{\mathbf{k}}$.

The expansion coefficients $a_{\mathbf{K}}(\mathbf{k})$ are determined variationally so that $\psi_{\mathbf{k}}$ in Eq. (1.1) is the best approximation to a solution of the one-electron Schrödinger equation with an eigenvalue $E(\mathbf{k})$. If $\varphi_{\mathbf{k}}$ is assumed to be an eigenstate with energy E_{1s} of the same Hamiltonian as $\psi_{\mathbf{k}}$,[11,15] then the $(\chi^*,H\chi)_{\mathbf{K}\mathbf{K}'}$ and $(\chi^*,\chi)_{\mathbf{K}\mathbf{K}'}$ matrix elements are given by simple expressions [in our notation, Ref. 13, Eqs. (2.52) and (2.53)]. Only the Fourier coefficients of the potential enter in these expressions. We have used the values tabulated by Callaway.[14] If for the core state we take $u_{1s}=\lambda^{3/2}\pi^{-1/2}e^{-\lambda r}$, then the orthogonality coefficient $\mu_{\mathbf{k}}$ is given explicitly by

$$\mu_{\mathbf{k}}=8\pi^{1/2}\Omega^{-1/2}\lambda^{-3/2}(1+\mathbf{k}^2/\lambda^2)^{-2}, \tag{1.4}$$

where Ω is the volume of a unit cell, $N\Omega=V$. Glasser and Callaway[16] computed the ground state in the conduction-electron Hamiltonian, $\varphi_{\mathbf{k}}$, by numerical integration. With the u_{1s} found in this way they computed a table of orthogonality coefficients $\mu_{\mathbf{k}}$. We can reproduce this table to within 3% by choosing $\lambda=2.37$ in the above expression for $\mu_{\mathbf{k}}$. The corresponding energy E_{1s} is[16] -3.765 Ry.

According to a variational calculation[17] of the *atomic* $1s$ state, λ should be 2.69. Thus the u_{1s} wave function in the crystal potential is somewhat broader as seen in Fig. 1. (See Ref. 14.)

With these considerations the $E(\mathbf{k})$ and $a_{\mathbf{K}}(\mathbf{k})$ were determined numerically using 19 waves. The results obtained for $E(\mathbf{k})$ shown in Fig. 2 are in good agreement (within 3%) with Callaway's more accurate 43 wave OPW band calculation for [100], [110], and [111] directions. The 19 expansion coefficients $a_{\mathbf{K}}(\mathbf{k})$ were then determined[18] at each $\mathbf{k}$ by the homogeneous linear equations [the vanishing of whose determinant of coefficients determined $E(\mathbf{k})$],

$$\sum_{\mathbf{K}'}[(\chi^*,H\chi)_{\mathbf{K}\mathbf{K}'}-E(\mathbf{k})(\chi^*,\chi)_{\mathbf{K}\mathbf{K}'}]a_{\mathbf{K}'}(\mathbf{k})=0,$$

and the normalization condition on $\psi_{\mathbf{k}}$: $(\psi_{\mathbf{k}}^*,\psi_{\mathbf{k}})=1$.

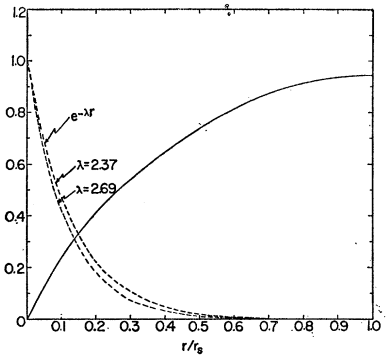

FIG. 1. The positron wave function ψ_+ in the Wigner-Seitz spherical cell approximation (unnormalized). $r_s=3.19$ (atomic units) is the radius of the cell. The core u_{1s} wave function used in the OPW calculation (the ground state in the potential seen by the conduction electrons) is plotted—upper dashed curve, $\lambda=2.37$. The atomic u_{1s} wave function with $\lambda=2.69$ is shown by the lower dashed curve.

2. APPLICATION TO POSITRON ANNIHILATION

The momentum distribution of the two photons produced when a positron annihilates with electrons

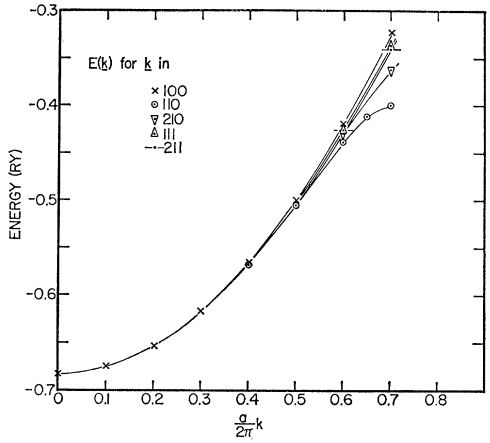

FIG. 2. The filled part of the conduction-electron energy band in Li for 5 different directions of $\mathbf{k}$, obtained with 19 orthogonalized plane waves.

[15] V. Heine, Proc. Roy. Soc. (London) **A240**, 354 (1957).
[16] M. L. Glasser and J. Callaway, Phys. Rev. **109**, 1541 (1958).
[17] P. M. Morse, L. A. Young, and E. S. Haurwitz, Phys. Rev. **48**, 948 (1935).
[18] Complete tables are published in J. Melngailis, Ph.D. thesis, Carnegie Institute of Technology, 1965 (unpublished). Because of the use of an incorrect formula for the potential seen by a positron, the positron wave function calculated in the thesis is wrong. However, it is qualitatively similar to the correct one, Fig. 2, and the subsequent results, which have here been recalculated, are almost unchanged.

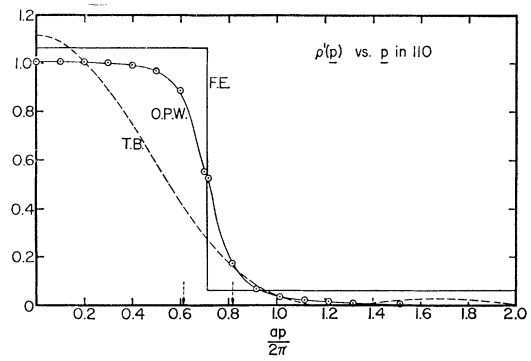

FIG. 3. The quantity $\rho'(\mathbf{p})$ versus $\mathbf{p}$ in the [110] direction as computed by three different methods: OPW, free-electron (FE) and tight-binding (TB) (using atomic $2s$ wave function, Ref. 16). The behavior of $\rho'(\mathbf{p})$ in other directions is similar. The vertical scale is arbitrary.

in a given band in a solid is[1,5,19]

$$\rho(\mathbf{p})=C\sum_{\mathbf{k}\leq\mathbf{k}_F}\left|\int_V\psi_{\mathbf{k}}(\mathbf{r})\psi_+(\mathbf{r})e^{-i\mathbf{p}\cdot\mathbf{r}}d^3r\right|^2, \quad (2.1)$$

where $C=\alpha^3/4\pi^2$, $\alpha=1/137$, ψ_+ is the positron wave function, and the sum over $\mathbf{k}\leq\mathbf{k}_F$ extends over all $\mathbf{k}$ within the Fermi surface. The positron is taken as thermalized[2] with wave number zero. Hence $\psi_+(\mathbf{r})$ by Bloch's theorem has the periodicity of the lattice. Substituting the expressions Eqs. (1.1) and (1.2) for $\psi_{\mathbf{k}}$ in Eq. (2.1), using the periodicity of $\psi_+(\mathbf{r})$, and neglecting $u_{1s}(\mathbf{r})$ outside the central cell, we have

$$\rho(\mathbf{p})=C\sum_{\mathbf{k}\leq\mathbf{k}_F}\left|\sum_{\mathbf{K}}a_{\mathbf{K}}(\mathbf{k})\sum_{\mathbf{R}}\exp(i(\mathbf{k}-\mathbf{p})\cdot\mathbf{R})\right.$$
$$\left.\times[V^{-1/2}G(\mathbf{k}-\mathbf{p}+\mathbf{K})-N^{-1/2}\mu_{\mathbf{k}+\mathbf{K}}F(\mathbf{p})]\right|^2, \quad (2.2)$$

where G and F are given by integrals over the unit cell Ω;

$$G(\mathbf{K})=\int_\Omega e^{i\mathbf{K}\cdot\mathbf{r}}\psi_+(\mathbf{r})d^3r, \quad (2.3)$$

and

$$F(\mathbf{p})=\int_\Omega e^{-i\mathbf{p}\cdot\mathbf{r}}u_{1s}(\mathbf{r})\psi_+(\mathbf{r})d^3r. \quad (2.4)$$

For the sum over $\mathbf{R}$ in Eq. (2.2) we use

$$\sum_{\mathbf{R}}e^{i\mathbf{k}\cdot\mathbf{R}}=N\sum_{\mathbf{L}}\delta(\mathbf{k}-\mathbf{L}), \quad (2.5)$$

where the sum over $\mathbf{L}$ extends over all reciprocal lattice sites, to obtain

$$\rho(\mathbf{p})=C\sum_{\mathbf{k}\leq\mathbf{k}_F}\left|\sum_{\mathbf{KL}}a_{\mathbf{K}}(\mathbf{k})N\delta(\mathbf{k}-\mathbf{p}-\mathbf{L})\right.$$
$$\left.\times[V^{-1/2}G(\mathbf{K}+\mathbf{L})-N^{-1/2}\mu_{\mathbf{p}+\mathbf{L}+\mathbf{K}}F(\mathbf{p})]\right|^2. \quad (2.6)$$

[19] R. A. Ferrel, Rev. Mod. Phys. **28**, 308 (1956). Each $\mathbf{k}$ value is occupied by 2 electrons.

Performing the sum over all $\mathbf{k}$ inside the Fermi surface we have

$$\rho(\mathbf{p})=NC\sum_{\mathbf{L}}\left|\sum_{\mathbf{K}}a_{\mathbf{K}}(\mathbf{p}+\mathbf{L})[\Omega^{-1/2}G(\mathbf{K}+\mathbf{L})\right.$$
$$\left.-\mu_{\mathbf{p}+\mathbf{L}+\mathbf{K}}F(\mathbf{p})]\right|^2 \text{ for } \mathbf{p}+\mathbf{L}\leq\mathbf{k}_F \quad (2.7)$$

$\rho(\mathbf{p})=0$ otherwise.

The sum over $\mathbf{K}$ extends over the 19 sites corresponding to the 19 OPW's chosen at the beginning, but the sum over $\mathbf{L}$ is over all reciprocal lattice sites. The above inequality means that $\mathbf{p}+\mathbf{L}$ must lie inside the Fermi surface. In Eq. (2.7) the terms in the sum over $\mathbf{L}$ are spatially separated, namely, at a given $\mathbf{p}$ only one (if any) term contributes to $\rho(\mathbf{p})$.

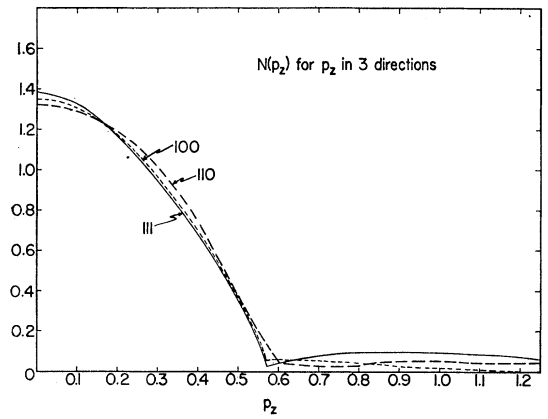

FIG. 4. Calculated angular distribution $N(p_z)$ for annihilation with conduction electrons only for [100], [110], and [111] orientations of p_z.

For later convenience define $\rho'(\mathbf{p})$ by

$$\rho'(\mathbf{p})=\rho(\mathbf{p})\Omega/CN[G(0)]^2, \quad (2.8)$$

so that $\rho'(0)$ is of order unity.

The momentum distribution of the annihilation photons as given by Eq. (2.7) can be determined at the points $\mathbf{p}$ (and $\mathbf{p}+\mathbf{L}$) at which the $a_{\mathbf{K}}(\mathbf{p})$ have been calculated. The quantity $\mu_{\mathbf{p}}$ is given by Eq. (1.4), and $G(\mathbf{K})$ and $F(\mathbf{p})$ by Eqs. (2.3) and (2.4). The latter depend on the positron wave function.

The positron wave function was calculated in the Wigner-Seitz cell in the usual manner.[5,20,21] Instead of using numerical integration of the Schrödinger equation, the positron wave function was expanded in a Fourier series which automatically satisfies the boundary condition that $d\psi_+/dr=0$ at $r=r_s$, the cell radius:

$$\psi_+(r)=\sum_{n=1,3,\cdots}a_n\sin\frac{n\pi r}{2r_s}. \quad (2.9)$$

The coefficients a_n were determined by the variational

[20] B. Donovan and N. H. March, Phys. Rev. **110**, 582 (1958).
[21] K. L. Rose, Ph.D. dissertation, Carnegie Institute of Technology, 1964 (unpublished).

method. The potential was calculated using $u_{1s}(r)$ with $\lambda=2.69$ (atomic $1s$ state) for the wave function of the core electrons and assuming the conduction-electron charge density constant over the cell. If terms up to $n=11$ are included, then $a_1=1$, $a_3=0.0743$, $a_5=0.0307$, $a_7=0.0177$, $a_9=0.0086$, and $a_{11}=0.0034$. The positron wave function ψ_+ is not normalized and is plotted in Fig. 2.

With this positron wave function the $G(\mathbf{K})$ were determined. The results are given in Table I. $F(\mathbf{p})$ was likewise determined using the OPW core-wave function, $u_{1s}(r)$, this time with $\lambda=2.37$.

We now have all the quantities needed to evaluate $\rho(\mathbf{p})$, Eq. (2.7). The sum over the reciprocal lattice sites Eq. (2.7) was performed on the computer. The resulting $\rho'(\mathbf{p})$, Eq. (2.8), has the rotational symmetry of the lattice. If the restriction that $\mathbf{p}+\mathbf{L}\leq\mathbf{k}_F$ is set aside for the moment, then $\rho'(\mathbf{p})$ is a continuous function of $\mathbf{p}$ with a maximum at $\mathbf{p}=0$ and a sharp drop at the zone boundary. This is illustrated for $\mathbf{p}$ in the [110] direction in Fig. 3. The $\rho'(\mathbf{p})$ obtained by the OPW method is intermediate between the tight-binding and the free-electron results.

3. ANGULAR-CORRELATION RESULTS

The standard thin slit apparatus measures only the distribution of the z component of momentum of the

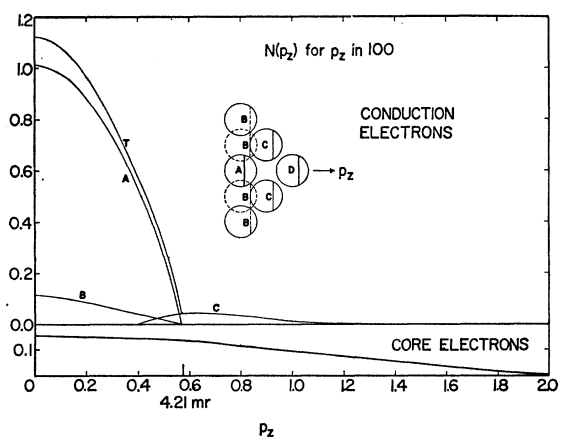

FIG. 5. Calculated angular correlation for p_z in [100] as made up of contributions from $\rho'(\mathbf{p})$ near $\mathbf{K}=0$, A all cells with $K_z=0$, B, and other cells C. Sum of A+B+C is T. The arrangement of the Fermi surfaces drawn about each $\mathbf{K}$ on a (100) plane is shown schematically in the center; the dashed surfaces are on a different plane. The contribution from core annihilation is shown at the bottom.

annihilating pair. The distribution function of this component is given by

$$N(p_z)=\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}dp_x dp_y \rho'(p_x,p_y,p_z), \qquad (3.1)$$

where $\rho'(\mathbf{p})$ is given by Eq. (2.8) evaluated as described above. The restriction on Eq. (2.7) that $\mathbf{p}$ or $\mathbf{p}+\mathbf{L}$ must be inside the Fermi surface can be replaced by a restriction on the integration limits on Eq. (3.1). Consider the Fermi surface (for Li almost spherical and not touching the zone boundary) drawn around each reciprocal lattice site, $\mathbf{L}$. The integration in Eq. (3.1) is then taken over the inside of the almost circular intersections of these surfaces with the plane perpendicular to the z direction at p_z. The function $\rho'(\mathbf{p})$ is of order unity in the central surface surrounding $\mathbf{L}=0$ and falls very rapidly in the neighboring zones.

TABLE I. Values of the Fourier coefficient $G(\mathbf{K})$ of the positron wave function. $\mathbf{K}=(2\pi/a)\mathbf{n}$.

$\lvert\mathbf{n}\rvert^2$	$G(\mathbf{K})/\Omega$	$\lvert\mathbf{n}\rvert^2$	$G(\mathbf{K})/\Omega$
0	0.8609	10	0.0295
2	−0.0203	12	0.0035
4	−0.0983	14	−0.0167
6	−0.0103	16	−0.0222
8	0.0360		

$N(p_z)$ was determined by numerical integration on the computer. The equation of the Fermi surface is $E(\mathbf{k})=E_F$. Callaway[14] has expanded $E(\mathbf{k})$ in a series with Kubic harmonics and determined the Fermi energy E_F to be −0.427 Ry. We have modified[22] his expansion to improve the fit to the bands, Fig. 1, near $|\mathbf{k}|=k_F$ at the expense of other regions. The surface defined by $E(\mathbf{k})=E_F$ is then almost spherical with 5% bulges in [110] directions. Analytic functions were fitted to $\rho'(\mathbf{p})$ (by trial and error) in the central, nearest, and next nearest zones. The integral over p_x and p_y was performed for the three types of sites with limits of integration given by $E(p_x,p_y,p_z)=E_F$.

The resulting $N(p_z)$ are shown in Fig. 4 for p_z in 100, 110, and 111 directions. These curves are made up of contributions from various sites as illustrated for the [100] direction in Fig. 5. Figure 6 shows the angular-correlation curves obtained including only the Fermi surface shape but not the effect of the wave functions, i.e., integrating Eq. (3.1) over the surface $E(\mathbf{p})=E_F$ with $\rho'(\mathbf{p})=$constant. The experimental results ob-

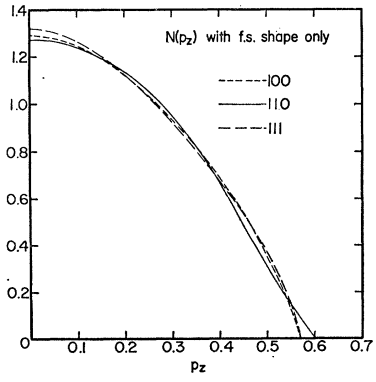

FIG. 6. The angular correlation calculated using the Fermi-surface shape as the only source of distortion, i.e., assuming $\rho(\mathbf{p})$ =constant.

[22] The coefficients given in Table IV, Ref. 14 were changed as follows: $E_2=0.781$; $E_4^{(2)}=-0.0148$; $E_6^{(2)}=0.095$; $E_6^{(3)}=0.13$; the others remained the same.

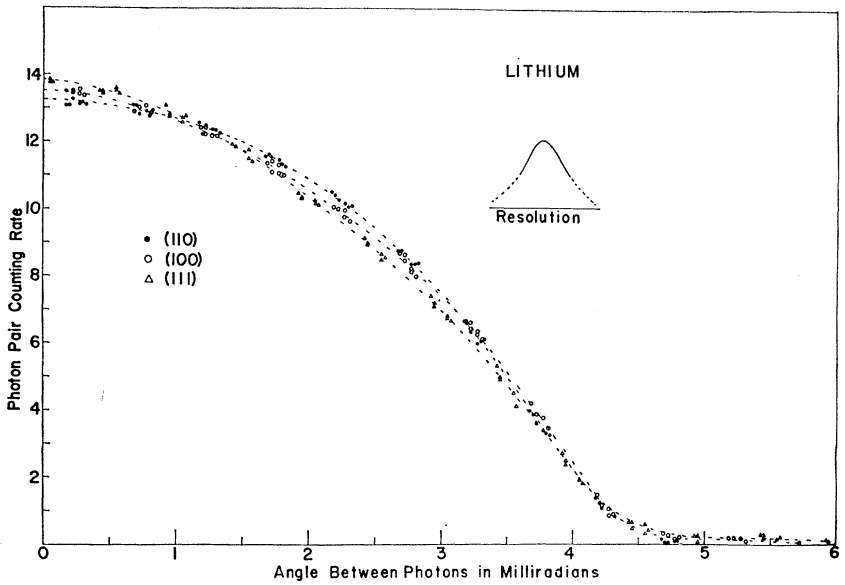

FIG. 7. [Taken from Ref. 9(d).] "The angular-correlation data from lithium after folding about the center and subtracting a smoothed approximation to the background and core effects. The dashed lines are visual fits to the data for the three directions."

tained by Donaghy *et al.*[23] for Li in the same orientations are shown in Fig. 7. A comparison of Figs. 4 and 6 to Fig. 7 indicates that the Fermi-surface shape as computed here alone can only account for part of the observed anisotropy (except perhaps at small angles) whereas when the effect of nonplane-wave conduction-electron wave functions is included, the calculated anisotropies in the curves closely resemble the experimental ones. However, the calculated curves are all (together) shifted downward from the experimental ones for momenta $p_z > 0.2$. A possible explanation will be discussed in the next section.

The momentum distribution of annihilations with the two core $1s$ electrons in Li can be easily computed assuming the tight-binding functions for the core state.[5] The result is, in agreement with Berko and Plaskett,[5]

$$\rho(\mathbf{p}) = 2CN|F(\mathbf{p})|^2. \tag{3.2}$$

In this case $F(\mathbf{p})$ is computed using the atomic $u_{1s} = \pi^{-1/2}\lambda^{3/2}e^{-\lambda r}$ with $\lambda = 2.69$. Integration of Eq. (3.2) to get $N(p_z)$ leads to a flat broad curve shown in Fig. 5, which is about an order of magnitude smaller than the conduction electron contribution.

4. LIFETIME AND ENHANCEMENT

The annihilation rate of positrons with conduction electrons is given by the integral of $\rho(\mathbf{p})$, Eq. (2.1), over all momenta. Since the above calculation is a small correction to the free-electron model, the annihilation rate would not differ significantly from the rate obtained from a free-electron calculation. This rate is[1,5] $\pi\alpha^3 n/\Omega$, where n is the number of free electrons per atom. The mean life is the reciprocal of this rate, and for Li is 2.7×10^{-9} sec. The experimental value obtained by Bell and Jorgensen[24] is 0.3×10^{-9} sec. Even if the calculated rate were decreased 10–20% by including core annihilations, the value of the lifetime corresponding to the above angular-correlation results would still be larger than that measured by a factor of 7 to 8. This discrepancy is probably for the most part due to the fact that electron-positron correlation was neglected.

Kahana[25] has calculated positron annihilation rates including a screened Coulomb attraction between metallic conduction electrons and the positron. The results are in the form of a momentum-dependent enhancement factor $\epsilon(\gamma)$, $(\gamma = p/k_F)$, which is a measure of the increased density of conduction electrons of momentum p at the positron. As shown by Kahana, integration of $\epsilon(\gamma)$ over all conduction-electron momenta gives annihilation rates in agreement with experiment[24] for electron densities corresponding to Na, Li, and Al.

The enhancement factor for Li is[25]

$$\epsilon(\gamma) = 7.28 + 1.65\gamma^2 + 1.33\gamma^4, \tag{4.1}$$

where the coefficients were interpolated from Table IV of Kahana's paper. Assuming that the enhancement somehow does not affect the center-of-mass momentum distribution of the annihilating pair, we can include the momentum-dependent enhancement in our calculation by multiplying the integrand for the central, $\mathbf{L}=0$, integration in Eq. (3.1) by $\epsilon(\gamma)$ (using $k_F = 0.6$). This does not affect the anisotropy but shifts the curves slightly outward (normalized to the same height as before) as shown in Fig. 8 for p_z in [100], and brings them into better quantitative agreement with experiment.

[23] Reference 9(d). We thank Dr. A. T. Stewart and Dr. J. J. Donaghy and the Plenum Press for permission to use these results prior to publication.

[24] R. E. Bell and M. H. Jørgensen, Can. J. Phys. **38**, 652 (1960).

[25] S. Kahana, Phys. Rev. **129**, 1622 (1963).

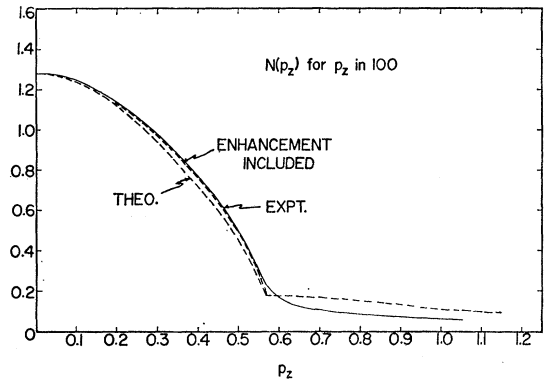

FIG. 8. A comparison of the measured (solid line) and calculated (dashed) angular correlation curves for p_z in the [100] direction. The lower dashed curve is the same as in Fig. 4 (for [100]) except that the core contribution, Fig. 5, has been added to the conduction electron contribution. The upper dashed curve results from including Kahana's enhancement factor (Ref. 25) and normalizing to the same height. The measured curve is from Ref. 9(d), with no core or background corrections subtracted. The situation in the [110] and [111] directions is similar: the agreement is improved by including the enhancement, and the calculated tails are too big.

5. DISCUSSION AND CONCLUSIONS

The model of the conduction electrons in Li chosen here leads to a Fermi-surface shape which agrees with other, more careful, calculations.[14,26,27] The wave function associated with this Fermi surface was obtained and was incorporated into a positron-annihilation angular-correlation, calculation. Before Kahana's enhancement factor[25] is introduced, this calculation predicts anisotropies in the angular distribution curves, $N(p_z)$, which for the three directions are qualitively in good agreement with experiment. Furthermore, annihilation with conduction electrons of momentum $\mathbf{p}+\mathbf{L}$ is shown to affect the anisotropies in the central "parabolic" peak and to contribute almost as much to the tails ($p_z > k_F$) as expected from core annihilations. This conduction-electron contribution to the tails also shows a dependence on the direction of p_z. In particular it is largest for p_z in the [111] direction. This seems to be due to the fact that we consider only 19 sites in the reciprocal lattice, and they all lie on only three (111) planes. Thus the contributions from six sites add to give a large tail in $N(p_z)$. So far there seems to be no experimental evidence that the tails in the angular-correlation curves in metals might show structure or anisotropy, such as seen in Ge and Si.[6]

[26] F. S. Ham, Phys. Rev. **128**, 2524 (1962).
[27] H. Schlosser, Ph.D. thesis, Carnegie Institute of Technology, 1960 (unpublished).

The angular distribution due to annihilations with core electrons was calculated assuming $u_{1s}=\pi^{-1/2}\lambda^{3/2}e^{-\lambda r}$, with $\lambda=2.69$.[16] (See Fig. 5.) This result is quite sensitive to the choice of u_{1s}. A 10% decrease in λ, for example, doubles the core contribution. The simple form[16] for the core $1s$ state used agrees to better than 5% with the self-consistent calculation[28] over the important part of the range. Nevertheless, the difference between the tails of the calculated and measured curves (Fig. 8) could easily be attributed to our choice of core state. Another reason for the disagreement in the tails might be provided by the enhancement.

Although the introduction of Kahana's enhancement factor improves the agreement between the calculation and experiment, it raises new problems and questions. We have no rigorous justification for simply multiplying the single particle $\rho(\mathbf{p})$ by the many-body enhancement factor $\epsilon(p/k_F)$. Doing so also destroys any relation between the parabolic peak and the tails. For example, if the main peak is raised by a factor of about 7, by how much should the core contribution be enhanced? On the other hand, one might speculate as follows: The momentum distribution of the annihilating pair is determined to a fair approximation by the one-particle calculation, while the correlation affects only the probability of annihilation with an electron of a certain momentum but does not change the center-of-mass momentum of the two annihilating particles. If the two particles could be considered isolated, for example, then their center-of-mass momentum, even when they are strongly attracted, would be the same as when they were too far apart to interact.

In any event, the effect of positron-electron correlation as treated by Kahana[25] is isotropic. The one-electron calculation seems to account for the experimentally observed angular-correlation anisotropy if both the effect of Fermi-surface departure from a sphere and the associated departure of the conduction-electron wave function from a plane wave are included.

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[28] V. Fock and M. J. Petrashan, Physik. Z. Sowjetunion **8**, 547 (1935).