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Comments on the Lock-Crisp-West Theorem with Experimental Examples* **

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Abstract. The application of the Lock-Crisp-West (LCW) theorem to positron-annihilation angular-correlation data for copper and germanium as well as to Compton profile data for germanium is presented and discussed. Deviations from the LCW prediction are observed for copper and for germanium, attributable to the positron's presence. In germanium the Compton results indicate near agreement with the LCW expectation; the sensitivity of the theorem is analyzed.

It has been known for a long time [1] that the angular correlation of annihilation radiation measures the real momentum distribution $\varrho(\boldsymbol{p})$ of electrons in a crystal (as modulated by the positron momentum distribution), and not the electronic distribution in $\boldsymbol{k}$ space. Thus one can interpret long-slit angular distributions in a metal as cross sectional cuts of the Fermi surface only in an approximation that neglects the positron contribution as well as the higher momentum components (HMC) of the electron wavefunctions [2]. Assumptions must also be made about the shape of the contribution due to electrons in filled shells. The same problems of HMC and of core subtraction arise in the interpretation of Compton profile measurements [3], although positron effects are of course absent.

In a recent paper [4] Lock *et al.* have derived an important mathematical connection, the LCW theorem, that allows a simple transformation of the momentum distribution obtained either by Compton profile or positron angular correlation measurements into the electronic distribution in $\boldsymbol{k}$ space. More specifically, if $N_{ijk}(p_z)$ represents the momentum distribution along the direction normal to the (ijk) planes of the reciprocal lattice ($p_z = mc\theta$, where θ is the angle between the long-slit gamma counters in the positron case) the LCW theorem states that, in a certain approximation, the function

$$F_{ijk}(p_z) = L_{ijk} \sum_{n=-\infty}^{\infty} N_{ijk}(p_z + nL_{ijk}) \quad (n \text{ integer}) \qquad (1)$$

should reproduce the functional variation of the cross sectional areas of the actual Fermi surface in a repeated zone scheme. The contribution to $F_{ijk}(p_z)$ from full zones is a constant independent of p_z. In (1) L_{ijk} is the spacing of the (ijk) planes; for convenience we have modified the LCW definition by multiplication with L_{ijk} – in our expression the average,

$$\langle F_{ijk} \rangle = 1/L_{ijk} \int_{-L/2}^{L/2} F_{ijk}(p_z) dp_z ,$$

is equal to the area of $N_{ijk}(p_z)$, independent of the orientation $[ijk]$. The LCW theorem holds in the following approximation: a) The electron wavefunctions and the annihilation probability must be computed in the independent particle approximation (IPM); b) the sum $\Sigma_{\boldsymbol{G}} |C_{\boldsymbol{G}}(\boldsymbol{k})|^2$, where the $C_{\boldsymbol{G}}(\boldsymbol{k})$ are the Fourier coefficients of the electron-positron wavefunction product, must be $\boldsymbol{k}$-independent, as discussed in [4] (a single plane-wave positron wave-

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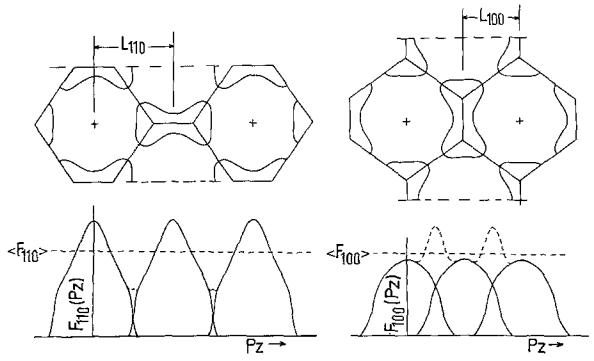

Fig. 1. The projection of the Fermi surface of copper onto $p_x p_z$ plane for p_z along the [100] and [110] direction, in the repeated zone scheme, and the sketch of the respective $F_{ijk}(p_z)$. The region of the reduced zone scheme contributing to the LCW construction is indicated. Note that only where the surfaces centered in different zones do not overlap is the $F_{ijk}(p_z)$ proportional to a single cross section of the reduced-zone Fermi surface. (The dashed lines in F_{ijk} show the region of overlap)

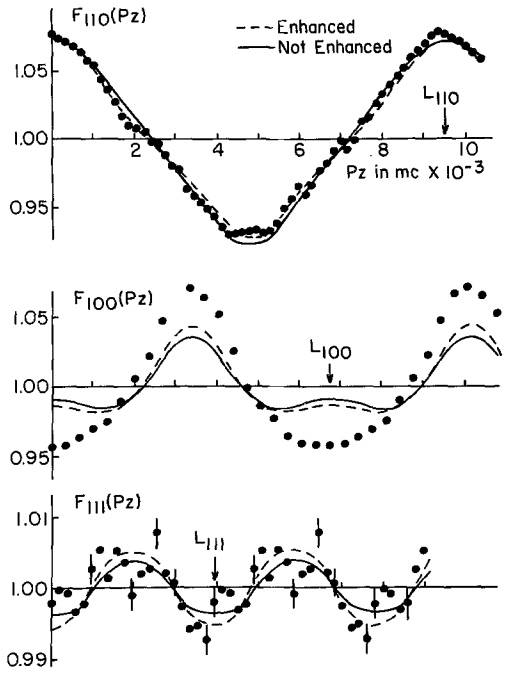

Fig. 2. The function $F_{ijk}(p_z)$ for the [110], [100], and [111] directions constructed by the LCW prescription from positron angular correlation curves (data points), and from the cross sections of the copper Fermi surface with (dashed line) and without (solid lines) many-body correction. Note the expanded scale of the [111] plot compared to the other two curves. Typical statistical uncertainties are smaller that the size of the data points except in the [111] plot where they are indicated by error bars

function is a sufficient condition). In this paper we study the applicability of the theorem to positron data for various orientations in copper and to Compton and positron measurements in germanium.

In Fig. 1 we have indicated qualitatively the parts of the Fermi surface that can be obtained from the F_{ijk} function, if the LCW theorem holds true in copper. It is to be noted that even for the best orientation (i.e. [110]), when the stacking along p_z is least dense, only part of the actual Fermi surface can be obtained unambiguously. In other directions the oscillations of F_{ijk} decrease and smaller parts of the Fermi surface are left unsuperimposed. For example, F_{111} for copper exhibits less than 1% oscillations and is nowhere directly related to a single Fermi surface cut.

In Fig. 2 we have compared the F_{ijk} curves for the three principal directions in copper obtained by the LCW prescription from the experimental $N_{ijk}(p_z)$ with those obtained using the cross sectional area variation of the parametrized copper Fermi surface of Halse [5], superimposed on a constant corresponding to the closed shells. In order to compare F_{ijk} amplitudes we have assumed the same 16% conduction electron contribution used by LCW. The experimental points for N_{ijk} were taken in our laboratory by Thompson [6] (the curves along [110] and [100], with 0.5 mrad resolution) and Cushner [7] ([111] direction, with 0.3 mrad resolution). We have not folded the experimental curves about $p_z = 0$, in order to exhibit the symmetry of the data. We note that although good agreement is obtained between experiment and theory along the [110] direction, in agreement with the results of LCW, the experimental F_{100} exhibits a much larger oscillation than predicted by theory. We can obtain better agreement in the [100] direction if we use 25% for the conduction electron contribution, a percentage more in accord with previous estimates [7, 8], but only at the expense of worsening the [110] fit. This discrepancy between experiment and theory can be due either to a breakdown of the LCW theorem because of a non-plane wave positron wavefunction or due to positron-electron correlation effects (or both). (It is to be noted that the LCW prediction of a constant, orientation independent $F(p_z)$ for the filled bands does *not* require isotropic momentum distributions.) In order to check the importance of the latter effect, we have also plotted in Fig. 2 the F_{ijk} curves based on a theoretical $N_{ijk}(p_z)$, worked out by Cushner [7],

that includes the anisotropy of the Fermi surface [parameterization by Roaf [9] in this case, but the Roaf and the Halse curves yield indistinguishable $F_{ijk}(p_z) - s$], multiplied by the momentum-dependent enhancement factor due to positron-electron correlation predicted by Kahana [10], using the appropriate r_s for copper. Again a 16% conduction band contribution has been assumed. It is noted that these curves improve somewhat the fit between the [110] and [100] curves, but not enough to resolve the major discrepancy noted above. Correlation effects based on a more realistic two-band model, rather than on the empty lattice as in Kahana's case, have been studied by Fujiwara *et al.* [11], but the enhancement effect at the Fermi surface, relative to that at the center of the zone, is found to be of the same magnitude.

The fit of the copper F_{ijk} curves was, of course, based on the use of a constant for the filled bands, as predicted by the LCW theorem. It is therefore important to study experimentally the extent to which an insulator does yield a constant for F_{ijk}. Most insulators, however, give rise to a Gaussian-like, bell-shaped N_{ijk} curve with very small anisotropies. We show in an appendix that the F_{ijk} curves formed from Gaussian distributions – using arbitrary but physically reasonable values of the shift distance L – display very small oscillations, independently of the LCW theorem. This implies that data of extremely high precision will be required to demonstrate the vanishing of oscillations at a particular value of L (L_{ijk}) that the LCW theorem predicts.

On the other hand the LCW theorem leads also to a constant $F(p_z)$ in the case of semiconductors, where $N(p_z)$ is non-Gaussian and highly anisotropic. We have thus decided to test the applicability of the theorem to germanium, where both Compton [12] and positron data [13, 14] are available.

In Fig. 3 we have plotted $F_{110}(p_z)$ generated using the Compton data of Reed and Eisenberger (from which a calculated isotropic core momentum distribution has been subtracted), as well as that obtained from positron data taken recently in our laboratory [14]. As in the case of copper, the [110] direction has the largest shift distance and thus provides the most sensitive test. We notice that the positron curve (which includes a small core electron contribution) exhibits a nonconstant $F(p_z)$ with a $\approx 2\%$ oscillation amplitude, as compared to the Compton curve which has a maximum $\approx 0.8\%$ amplitude. The Compton curve was obtained from a recent re-

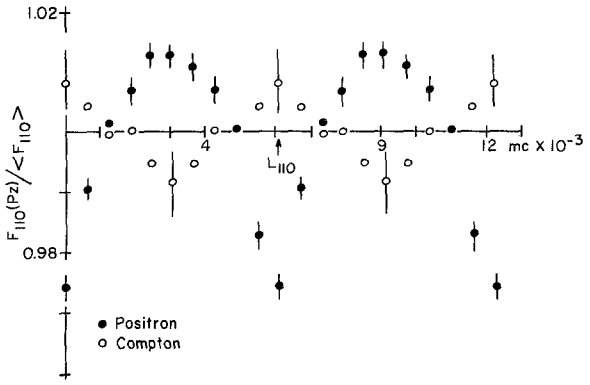

Fig. 3. $F_{110}(p_z)$ for germanium, constructed from positron (solid points) and Compton (open circles) momentum distributions

evaluation by Reed and Eisenberger of their data published in [12]. The original data contains some unphysical oscillations in $N(p_z)$, due to an unfolding technique, that can substantially alter the shape of $F(p_z)$. We find that the positron result does not follow the LCW theorem, which would require a constant $F(p_z)$ for germanium. The nonconstancy of $F(p_z)$ obtained from the Compton experiment is smaller; it either reflects the effect of departures from the IPM due to electron-electron correlations, or it could be due to a small remaining uncertainty in the deconvolution problem. [Unlike the positron angular correlation, where the effective angular resolution is small (0.5 mrad), the Compton data, because of a much wider inherent resolution, requires deconvolution.] In order to test the sensitivity of the LCW theorem for the nearly constant $F(p_z)$ Compton data, we have also computed $F(p_z)$ while varying L about its actual value as given by the lattice parameter of germanium. If $F(p_z)$ is LCW theorem governed, the oscillation amplitude should have a minimum at the physical L. A purely Gaussian-like distribution should have of course a continuously decreasing amplitude as $L \to 0$. In Fig. 4 we plot

$$[F_{\max}(p_z) - F_{\min}(p_z)]/[F_{\max}(p_z) + F_{\min}(p_z)]$$

for the Compton data as a function of L and find indeed a clear minimum close to the physical value given by the lattice constant. We conclude that in spite of the observed small deviation from a constant value, the $F(p_z)$ for the Compton data indicates the near applicability of the LCW theorem to Compton momentum distributions. It would be important to obtain high precision Compton data for a metal

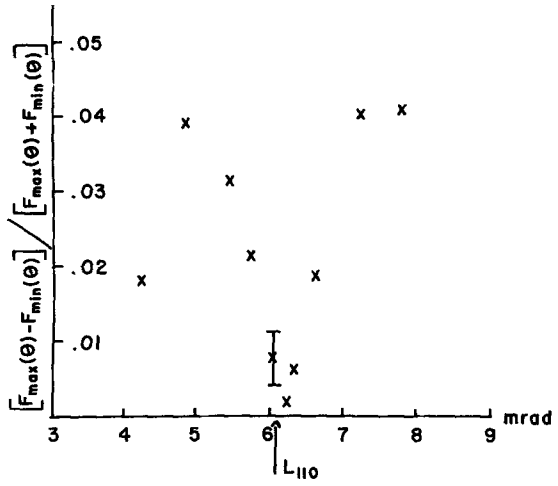

Fig. 4. The amplitude of oscillations in $F_{110}(p_z)$ from the Compton data as a function of the shift distance L_{110}, see (1). The arrow shows the physical L_{110} obtained from the lattice parameter of germanium

like copper in order to test the applicability of the LCW theorem to metal systems, as was done for positrons.

In conclusion, the LCW theorem provides one, in the case of the Compton profile, with a powerful tool to test the validity of the IPM, to obtain the shape of at least parts of the Fermi surface from momentum distributions and, in the case of the angular correlation of annihilation radiation, to check the importance and influence of the positron wavefunction.

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Appendix

$F(p_z)$ is an even periodic function and may be expanded in the Fourier cosine series:

$$F(p_z) = a_0/2 + \sum_{n=1}^{\infty} a_n \cos(2n\pi p_z/L) .$$

It is easily shown from the definition (1) of $F(p_z)$, that

$$a_n = 2R(2n\pi/L) ,$$

where $R(x)$ is the Fourier integral transform of $N(p_z)$. If we assume the normalization

$$\int_{-\infty}^{\infty} N(p_z) dp_z = 1 ,$$

then $\langle F \rangle = a_0/2 = 1$. If N is the Gaussian function

$$N(p_z) = (p_0\sqrt{\pi})^{-1} \exp[-(p_z/p_0)^2] ,$$

we obtain $a_n = 2\exp[-(n\pi p_0/L)^2]$. Suppose we now form $F(p_z)$ using the shift distance $L = p_0$ (i.e. $\approx 1.2 \times$ HWHM); this leads to $a_1 = 2\exp(-\pi^2) \simeq 10^{-4}$, and to much smaller values for the higher Fourier coefficients. Any function having a nearly Gaussian shape will have similarly small coefficients.

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