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PHYSICAL REVIEW B

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## Determination of the Total Momentum Distribution by Positron Annihilation

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Current experimental techniques of two-photon angular correlation in positron-annihilation studies measure the probability distribution of one or two components of the total momentum of the electron-positron pair. We discuss the problem of deducing the probability distribution of the total momentum itself from the experimental data. It is shown that information about two components of the total momentum can in principle determine the probability distribution of the total momentum.

## INTRODUCTION

Several different geometries are used for two-photon angular-correlation experiments in studying positron annihilation in various substances. Let  $\rho(\vec{p})$  denote the probability that the total momentum of the two  $\gamma$  rays (and therefore of the annihilating electron-positron pair) is  $\vec{p}$ . Now the experiments do not determine  $\rho(\vec{p})$  directly, but only the probability distribution of some component of  $\vec{p}$ , obtained by suitably integrating over  $\rho(\vec{p})$ .

The most common setup uses two long slits.<sup>1</sup> This geometry measures the probability that the photon pair has the  $z$  component of momentum equal to  $p_z$ :

$$N(p_z) = \int_{-\infty}^{\infty} \int_{-\infty}^{\infty} dp_x dp_y \rho(\vec{p}). \quad (1)$$

A second type of setup uses two point counters moving in one plane<sup>2</sup>; this gives

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

A variation of this geometry, proposed by Fujiwara,<sup>3</sup> uses two pairs of crossed slits, and can give useful information about the Fermi-surface anisotropies.

A new technique recently introduced<sup>4</sup> uses a point counter together with a spark chamber. The line joining the sample to the point counter defines the  $z$  axis; the plates of the spark chamber are parallel to the  $xy$  plane. This geometry yields

$$N(p_x p_y) = \int_{-\infty}^{\infty} \rho(\vec{p}) dp_z. \quad (3)$$

In each case, one can try to determine  $\rho(\vec{p})$  itself from the experimental data. In this paper we shall discuss the mathematical solution of this problem. In particular, we describe how the data supplied by Eq. (3) can be analyzed to yield  $\rho(\vec{p})$ .

This problem for the two long slits was discussed

by Mijnaerends.<sup>5</sup> He deduced an integral equation and found its solution on the assumption that  $N(p_z)$  as given by (1) is differentiable. This assumption does not hold for the Fermi distribution at 0°K. It is possible, however, to find a solution to Mijnaerends's integral equation without making this differentiability assumption. We shall show that there is an inherent drawback of the long-slit geometry, in that the solution for  $\rho(\vec{p})$  cannot, in general, be unique. The determination of  $\rho(\vec{p})$  from Eq. (2) with two point counters leads again to an integral equation, which can be solved. However, this method also suffers from lack of uniqueness.

The two-dimensional information available from Eq. (3) can be inverted through an integral equation to get  $\rho(\vec{p})$ . We shall argue that the solution can be made unique from the point of view of physics. The reason for the advantage is roughly this: The knowledge of the distribution of two components of momentum provides information about the azimuthal quantum number ( $m$  values), so that one can put a limit on the number of partial waves ( $l$  values) present and thus reduce the problem to a determination of a finite number of quantities. Nothing can be said about the azimuthal quantum number from the experimental results on the distribution of one component of momentum. Finally, we make some remarks on the practical aspects of the various methods.

### I. INFORMATION OF ONE MOMENTUM COMPONENT

We shall confine our discussion to cubic crystals throughout this paper. Two coordinate systems are defined as follows: (i)  $R_c$ , fixed to the crystal with coordinates  $\xi\eta\zeta$  along the crystal axes, and (ii)  $R$ , fixed suitably by the experimenter to the measuring apparatus, with coordinates denoted by  $xyz$ . The subscript  $c$  denotes the system  $R_c$ ; thus  $\Omega_c$  will denote the orientation of  $\vec{p}$  in the coordinate system  $R_c$ . All experimental numbers, of course, refer to the system  $R$ .

In  $R_c$ , the distribution function  $\rho(\vec{p})$  can be expanded in cubic harmonics  $K_{l\nu}(\Omega_c)$  of angular momentum  $l$  and type  $\nu$ ,

$$\rho(\vec{p}) = \sum_{l\nu} \rho_{l\nu}(p) K_{l\nu}(\Omega_c) \quad (4)$$

$$= \sum_{l\nu m} a_{l\nu m}^{\nu} \rho_{l\nu}(p) C_{l\nu m}(\Omega_c), \quad (5)$$

where

$$C_{l0}(\Omega_c) = Y_{l0}(\Omega_c), \quad (6)$$

$$C_{lm}(\Omega_c) = (1/\sqrt{2})[Y_{lm}(\Omega_c) + Y_{l-m}(\Omega_c)] \quad (m \neq 0).$$

$a_{l\nu m}^{\nu}$  can be obtained from the work of Mueller and Priestley.<sup>6</sup> Note that  $l$  is even ( $l=0, 4, 6, 8, 10, \dots$ ) and  $m=0 \pmod{4}$  and positive. Let  $\omega = (\alpha\beta\gamma)$  be the rotation that brings  $R_c$  into coincidence with  $R$ , so that  $(\beta\alpha)$  are the polar and azimuthal angle of the  $z$  axis with respect to  $R_c$ . The spherical har-

monics transform according to the well-known relation<sup>7</sup>

$$Y_{lm}(\theta_c\phi_c) = \sum_{m'} \mathcal{D}_{m'm}^l(\omega) Y_{lm'}(\theta\phi). \quad (7)$$

Utilizing (4), (5), and (7), Mijnaerends shows that Eq. (1) for the orientation  $(\beta\alpha)$  of the  $z$  axis with respect to the crystalline axes  $R_c$  becomes

$$N_{\beta\alpha}(p_z) = 2\pi \sum_{l\nu} K_{l\nu}(\beta\alpha) g_{l\nu}(p_z), \quad (8)$$

with

$$g_{l\nu}(p_z) = \int_{p_z}^{\infty} \rho_{l\nu}(p) P_l(p_z/p) p dp \quad (p_z > 0). \quad (9)$$

To determine  $\rho_{l\nu}(p)$ , one first obtains the functions  $g_{l\nu}(p_z)$  from (8), and then solves (9). The solution to (9) given by Mijnaerends is

$$\rho_{l\nu}(p) = -\frac{1}{p} \left[ \frac{dg_{l\nu}(p)}{dp} - \frac{l(l+1)}{2p} g_{l\nu}(p) + \frac{1}{p^2} \int_0^p g_{l\nu}(z) P_l''(z/p) dz \right]. \quad (10)$$

$P_l''(x)$  is the second derivative of the Legendre polynomial  $P_l(x)$ . In particular,

$$\rho_{01}(p) = -\frac{1}{p} \frac{dg_{01}(p)}{dp}. \quad (11)$$

It is, however, easy to see that for the simple Fermi distribution at 0°K,  $g_{01}(p)$  is not differentiable at the Fermi momentum.  $N(p_z)$  is the well-known inverted parabola in this case,

$$N(p_z) = 2\pi(k_F^2 - p_z^2), \quad p_z \leq k_F \\ = 0, \quad p_z \geq k_F. \quad (12)$$

As the experiments are done at finite temperature, this usually does not matter, since the Fermi distribution at finite temperature is differentiable. It is possible, though, to solve (9) without this restrictive assumption.

Consider the Mellin transform<sup>8</sup> of  $g_{l\nu}(p_z)/p_z^2$  with  $\text{Res} > 2$  (the transform is indicated by a caret on top):

$$\begin{aligned} \hat{g}_{l\nu}(s) &= \int_0^{\infty} p_z^{s-3} g_{l\nu}(p_z) dp_z \\ &= \int_0^{\infty} p_z^{s-3} dp_z \int_{p_z}^{\infty} \rho_{l\nu}(p) P_l(p_z/p) p dp \\ &= \int_0^{\infty} dp \rho_{l\nu}(p) p^{s-1} \int_0^1 x^{s-3} P_l(x) dx \\ &= \int_0^{\infty} dp \rho_{l\nu}(p) p^{s-1} \frac{\pi^{1/2} \Gamma(s-2) 2^{2-s}}{\Gamma(\frac{1}{2}(s-l-1)) \Gamma(\frac{1}{2}(s+l))}. \end{aligned} \quad (13)$$

By Mellin inversion formula we get ( $\sigma > 2$ )

$$\rho_{l\nu}(p) = \frac{1}{2\pi i} \int_{\sigma-i\infty}^{\sigma+i\infty} \frac{ds}{p^s} \hat{g}_{l\nu}(s) \times \frac{\Gamma(\frac{1}{2}(s-l-1))\Gamma(\frac{1}{2}(s+l))}{\pi^{1/2} 2^{2-s} \Gamma(s-2)} \quad (14)$$

It is easy to check that the inverted parabola presents no difficulty here.

Let us now examine the solution of (8) more carefully. Notice that the set of quantities  $g_{l\nu}(p_z)$  is to be determined from a highly underdetermined system. Mijnaerends proceeds as follows: He collects the data for several directions ( $\beta\alpha$ ). Then he *arbitrarily* truncates the summation at convenient  $l$  values such that he has just as many  $g_{l\nu}$ 's as  $N_{\beta\alpha}(p_z)$ 's, or even fewer  $g_{l\nu}$ 's. In the latter contingency, he uses the method of least squares to fit the data. The procedure is arbitrary and cannot lead to unique solutions for  $\rho(\vec{p})$ , unless, of course, one guesses luckily the correct number of  $l$ 's present.

We shall briefly consider the inversion problem for Eq. (2). Substituting (4), (5), and (6) into (2), and using (7), we get

$$N(p_z) = \sum_{l\nu m} \sum_{m'=0}^l b_{l\nu m m'} g_{l\nu}^{m'}(p_z), \quad (15)$$

with

$$g_{l\nu}^{m'}(p_z) = \int_{p_z}^{\infty} \frac{dp}{(1-p_z^2 p^{-2})^{1/2}} \rho_{l\nu}(p) P_l^{-m'}(p_z/p). \quad (16)$$

The solution of (16) can be obtained by Mellin transform. Note that the solution of (15) suffers from the same nonuniqueness difficulty as described above. We shall later return to the practical aspects of the problem.

## II. INFORMATION OF TWO MOMENTUM COMPONENTS

We shall now turn to the problem of determining  $\rho(\vec{p})$  from Eq. (3). For simplicity, let us take first

the "fixed configuration" measurement, in which the axes of the cubic crystal are coincident with the  $xyz$  axes of the measuring apparatus. Then Eqs. (4) and (5) hold in the  $R$  system as well; using the explicit form for spherical harmonics,<sup>7</sup> we get

$$\rho(\vec{p}) = \sum_{l\nu m} b_{l\nu}^m P_l^{-m}(\cos\theta) \cos m\phi \rho_{l\nu}(p) \quad (m > 0). \quad (17)$$

We express  $N(p_x p_y)$  in polar coordinates ( $R\phi$ ) and write the Fourier series

$$N(p_x p_y) \equiv g(R\phi) = \sum_m g_m(R) \cos m\phi. \quad (18)$$

$g_m(R)$  can be obtained from a Fourier cosine transform of the experimental data. On the other hand, by substituting (18) and (17) into the left- and right-hand sides of (3), respectively, we get

$$g_m(R) = 2 \sum_{l\nu} b_{l\nu}^m f_{l\nu}^m(R), \quad (19)$$

where

$$f_{l\nu}^m(R) = \int_R^{\infty} P_l^{-m} [(1-R^2 p^{-2})^{1/2}] \times \rho_{l\nu}(p) (1-R^2 p^{-2})^{-1/2} dp. \quad (20)$$

The solution of the integral equation can be easily obtained. Consider the integral

$$\hat{f}_{l\nu}^m(s) = \int_0^{\infty} R^{s-2} f_{l\nu}^m(R) dR, \quad (21)$$

with  $\text{Res} > 1$ , so that the integral converges at  $R=0$ , when  $f_{l\nu}^m(R)$  is finite at the origin. In practice  $f_{l\nu}^m(R)$  will fall off very fast as  $R \rightarrow \infty$ ; even if  $f_{l\nu}^m(R)$  decays as slowly as  $R^{-1}$ , we can restrict  $\text{Res} < 2$ .

Now,

$$\begin{aligned} \hat{f}_{l\nu}^m(s) &= \int_0^{\infty} dR R^{s-2} \int_R^{\infty} dp P_l^{-m} [(1-R^2 p^{-2})^{1/2}] \\ &\quad \times \rho_{l\nu}(p) (1-R^2 p^{-2})^{-1/2} \\ &= \int_0^{\infty} dp p^{s-1} \rho_{l\nu}(p) \int_0^1 (1-x^2)^{(s-3)/2} P_l^{-m}(x) dx. \end{aligned} \quad (22)$$

But as  $\text{Res} > 1$ ,<sup>9</sup>

$$\int_0^1 (1-x^2)^{(s-3)/2} P_l^{-m}(x) dx = \frac{\pi^{1/2} \Gamma(\frac{1}{2}(s+m+1))}{2^{1+m} \Gamma(1+m) \Gamma(\frac{1}{2}(s+m))} {}_3F_2(\frac{1}{2}(l+m+1), -\frac{1}{2}(l-m), \frac{1}{2}(s+m+1); 1+m, \frac{1}{2}(s+m); 1). \quad (23)$$

${}_3F_2(abc; de; z)$  is a standard higher hypergeometric function. As  $\frac{1}{2}(l-m)$  is an integer, the function is always cut off into a rational function in  $s$ , which in fact has real zeros and poles. Equation (23) is then solved by the Mellin inversion formula:

$$\rho_{l\nu}(p) = \frac{1}{2\pi i} \int_{\sigma-i\infty}^{\sigma+i\infty} \frac{ds}{p^s} \frac{2^{1+m} \Gamma(1+m) \Gamma(\frac{1}{2}(s+m)) \hat{f}_{l\nu}^m(s)}{\pi^{1/2} \Gamma(\frac{1}{2}(s+m+1)) {}_3F_2(\frac{1}{2}(l+m+1), -\frac{1}{2}(l-m), \frac{1}{2}(s+m+1); 1+m, \frac{1}{2}(s+m); 1)}, \quad (24)$$

where  $\sigma > 1$ .

Notice, however, that  $f_{l\nu}^m(R)$  are, as before, underdetermined in (19). But we now have some addi-

tional information about the  $m$  values in using (18) on the experimental data. We shall examine the situation in detail. Take first the extreme case,

when

$$g_0(R) = 2(1 - R^2)^{1/2}, \quad R \leq 1 \\ = 0, \quad R \geq 1. \quad (25)$$

All higher  $g_m(R)$ 's are zero, as obtained experimentally. In (19) for any  $m$  on the left side,  $l \geq m$  on the right. Hence one can argue that all  $\rho_{l\nu} = 0$  except  $\rho_{01}$ , and we have a single equation

$$g_0(R) = 2b_{00}^1 f_{01}^0(R). \quad (26)$$

So we have

$$f_{01}^0(R) = [4\pi(1 - R^2)]^{1/2}$$

and

$$\hat{f}_{01}^0(s) = \frac{1}{2} \pi \Gamma(\frac{1}{2}(s - 1)) / \Gamma(\frac{1}{2}s + 1). \quad (27)$$

Then

$$\rho_{01}(p) = \frac{(4\pi)^{1/2}}{2\pi i} \int_{\sigma-i\infty}^{\sigma+i\infty} \frac{ds}{s} \frac{1}{p^s}, \quad (28)$$

which, by elementary contour integration, gives rise to the standard Fermi distribution.<sup>10</sup> Except in this trivial spherically symmetric case, the situation is not so simple. Consider, for instance, the case when only  $g_0(R)$  and  $g_4(R)$  are finite and the rest are zero. We may argue that all  $l$ 's higher than 6 are absent. We have to determine  $f_{01}^0$ ,  $f_{41}^0$ ,  $f_{61}^0$ ,  $f_{41}^4$ , and  $f_{61}^4$ , but  $f_{41}^4$  and  $f_{61}^4$  can be calculated through (22) if  $f_{41}^0$  and  $f_{61}^0$  are known. Even then three quantities,  $\rho_{01}$ ,  $\rho_{41}$ , and  $\rho_{61}$ , are to be determined from two equations for  $g_0(R)$  and  $g_4(R)$ . The solution cannot be unique.

This ambiguity can be resolved by making measurements with the crystal rotated. We now use Eqs. (4)–(7) and (3), and get the equations ( $n$  even)

$$g_0^\omega(R) = 2 \sum_{l\nu} K_{l\nu}(\beta\gamma) f_{l\nu}^0(R), \quad (29)$$

$$g_m^\omega(R) = 2 \sum_{l\nu m} d_{l\nu m} f_{l\nu}^m(R) \quad (m \neq 0), \quad (30)$$

where

$$d_{l\nu 0n} = a_{l0}^\nu \left[ \frac{(l+n)!}{(l-n)!} \right]^{1/2} [Y_{l-n}(\beta\alpha) + Y_{ln}(\beta\alpha)], \\ d_{l\nu mn} = \frac{1}{\sqrt{2}} a_{lm}^\nu \left[ \frac{(2l+1)(l+n)!}{4\pi(l-n)!} \right]^{1/2} \\ \times [\mathcal{D}_{nm}^l(\omega) + \mathcal{D}_{n-m}^l(\omega) + \mathcal{D}_{nm}^l(\omega) + \mathcal{D}_{n-m}^l(\omega)]. \quad (31)$$

The  $f_{l\nu}^m(R)$ 's are the same as before. By making enough measurements for various rotations one can always get unique solutions, once the  $l$  values have

been delimited. For example, in the case discussed above, using (29) for three orientations, one can find  $\rho_{01}$ ,  $\rho_{41}$ , and  $\rho_{61}$  uniquely. The consistency of  $l$  values appearing in the sum can also be checked.

The use of Mellin transform raises a practical problem, since  $\hat{f}_{l\nu}^m(s)$  has to be determined analytically. One solution is to fit the experimental data in terms of linear combinations of simple functions like  $(1 - R^2)^a$ ,  $e^{-aR}$ ,  $e^{-aR^2}$ , etc., which are suggested by the physics of the problem [compare (25) above], and have known Mellin transforms. Then  $\hat{f}_{l\nu}^m(s)$  can be found in terms of known functions and the analysis can be completed. Notice that this feature is also present in our solution (14) for the one-momentum-component problem and thus makes it somewhat less practical than the solution (10) given by Mijnaerends. For the spark-chamber analysis, we have not been able to find a solution in terms of real variables only.

### III. DISCUSSION

We have shown how the data on two components of the total momentum can be analyzed to yield  $\rho(\vec{p})$  and that this method, in principle, has some advantages. In practice, the gains may be less gratifying. Consider Eq. (18);  $g_m(R)$ 's are obtained by Fourier cosine transform of experimental data. The state of affairs will often be less clear-cut than what is assumed in the two examples above. In many cases—particularly if the momentum distribution has strong anisotropies—besides the large Fourier components, we shall have a number of small Fourier components, and these will be very sensitive to experimental errors. Obviously, even here one will have to use one's judgment on experimental errors to truncate the  $m$  series so that the number of  $l$  values becomes delimited. The only advantage is this: While in the one-component case the truncation is arbitrary or dependent on clever guess and can be improved by trial and error, in the two-component problem, experimental information is used as guide for the truncation. In a systematic study of  $\rho(\vec{p})$  for many cases, this may prove to be helpful. Hopefully this new spark-chamber technique will provide interesting information about anisotropies in the momentum distribution of metals and other substances.

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## Nuclear-Magnetic-Resonance Studies of the Semiconductor-to-Metal Transition in Chlorine-Doped Cadmium Sulfide\*†

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Spin-lattice relaxation times ( $T_1$ ) and Knight shifts were measured for  $\text{Cd}^{113}$  nuclei in 12 CdS crystals doped with various amounts of chlorine. Hall coefficients were measured in order to estimate conduction-electron concentrations. Data were obtained for all samples at 300 °K and for some highly doped samples at 77, 4.2, and 2.13 °K. Metallic properties were observed in all samples having electron concentrations  $n > 2 \times 10^{18} \text{ cm}^{-3}$ . At 300 °K, we find  $1/T_1 \propto n$  for nonmetallic samples and  $1/T_1 \propto n^{2/3}$  when samples are metallic. The latter proportionality continues to hold at lower temperatures. The dependence of  $T_1$  on  $n$  becomes increasingly less pronounced at lower temperatures in the nonmetallic samples indicating that the nuclear relaxation becomes at least partially dependent on mechanisms other than conduction electrons, such as spin-diffusion coupling to paramagnetic impurity sites. In the metallic samples, the Knight shift  $K \propto n^{1/3}$  and the Korringa product is a constant:  $T_1 K^2 = 3.3 \times 10^{-6} \text{ sec } ^\circ\text{K}$ . Both the Knight shift and Korringa product decrease sharply for  $n < 2 \times 10^{18} \text{ cm}^{-3}$ . Our analysis shows that the Mott transition (formation of an impurity conduction band or transition to "free" conduction) occurs in a region  $5 \times 10^{17} < n < 1.6 \times 10^{18} \text{ cm}^{-3}$  and that the impurity conduction band and the CdS conduction band become merged (i. e., the Fermi level crosses into the CdS conduction band) in a region  $1.6 \times 10^{18} < n < 2.4 \times 10^{18} \text{ cm}^{-3}$ .

### I. INTRODUCTION

This paper reports on the experimental nuclear-magnetic-resonance (NMR) behavior of  $\text{Cd}^{113}$  nuclei in chlorine-doped CdS. We have measured the spin-lattice relaxation times ( $T_1$ ) and Knight shifts ( $K$ ) in CdS:Cl having a wide range in the doping concentration. These data were complemented by measurements on the electrical properties and used to investigate the semiconductor-to-metal transition.

Pure CdS is a 2.5-eV band-gap photoconductor which becomes an  $n$ -type semiconductor when

donor impurities are present. Chlorine is a donor impurity for CdS and thus evidently goes into the crystalline lattice substitutionally for sulfur.<sup>1</sup>

The electrical conductivity increases with impurity concentration and at a rather high level the doping will effect a semiconductor-to-metal transition. This phenomenon may be studied by NMR via the hyperfine interaction with conduction electrons which affects both  $T_1$  and  $K$ .

Impurity conduction phenomena in the group IV semiconductors and the III-V compounds have been studied by several investigators using NMR.<sup>2-5</sup> Cadmium oxide is the only II-VI compound reported