

case of very thin crystals, where both should give identical results. Our examples have shown that such is indeed the case for reflections of the first order and of low indices, but that in general there is no agreement for other types of reflections. The discrepancy in the second case can be explained as follows: The diffracted beam under consideration is more strongly coupled with one or more other diffracted beams than it is with the incident beam; in other words, the contribution of electron paths with successive collisions on different atoms is predominant and is the cause of the discrepancy between both theories, since these paths are neglected in the PKT.

In the study of a given diffracted beam the number of other beams of equal or higher intensity is roughly the same as the order of the system of Eqs. (8); therefore DT calculations for weak beams of high indices would be very lengthy and, at the same time, strongly dependent on azimuthal orientation. No agreement with the PKT should then be expected in the limit of thin crystals. Furthermore, it will be clear from the previous discussion that the commonly accepted notion that integrated intensities are proportional to $|F_h^B|$ in the DT, in contrast to the values $|F_h^B|^2$ in the KT, is only correct insofar as the two-waves formula (11) is valid.

In weighting the relative merits of the PKT and the DT, it should be remembered that our comparison

has been based on the scattering by a given potential field, and has therefore overlooked a main feature of the PKT, namely that, unlike the DT, it can take up in a straightforward way *all* single-atom scattering effects, including even those which cannot be strictly described by a potential (polarization, electron exchange for instance). There is yet no experimental evidence, however, that these effects are actually important.

Finally, it should be emphasized that the possibility of crystal structure determinations from Fourier inversion of diffraction data (a procedure which is based upon the validity of the KT, but can still be carried out if the PKT is valid¹¹), is in fact brighter than it would appear from our discussion. Our calculations apply to perfect crystals of simple shape for which dynamical effects are most conspicuous. Since mosaic structure destroys the coherence between the waves scattered by the various mosaic blocks, the effective scatterer size may well correspond to the range of validity of the KT. This effect, combined with the small weight of the atoms present, is the probable reason of recent successful crystal structure determinations by electron diffraction.

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Spin and Polarization Effects in the Annihilation of Triplet Positronium*

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The annihilation rate of triplet positronium has been calculated without summing at once over spin and polarization. The results, quoted without proof, apply to some recent measurements of spin and polarization effects.

INTRODUCTION

THE calculation by Ore and Powell¹ and others^{2,3} of the angular correlations and spectrum of the annihilation quanta from triplet positronium has been repeated without averaging over spin or summing over polarization. The results then apply to recent measurements of the relative polarization of a single annihila-

tion quantum⁴ and the effect of the $m=0$ spin state on the angular distribution of the annihilation quanta.^{5,6}

The calculation follows readily from expressions given by Ore and Powell; the results will be quoted without proof.

MATRIX ELEMENT

The transition probability for a positronium atom in the lowest triplet S -state to annihilate with the production of three quanta of energies k_1 , k_2 , k_3 and

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¹ A. Ore and J. Powell, Phys. Rev. **75**, 1696, 1963 (1948).

² J. Radcliffe, Phil. Mag. **42**, 1334 (1951).

³ R. Ferrell, doctoral thesis, Princeton University, 1951 (unpublished).

⁴ Leipuner, Siegel, and DeBenedetti, Phys. Rev. **91**, 198 (1953).

⁵ J. Wheatley and D. Halliday, Phys. Rev. **88**, 424 (1952).

⁶ Marder, Hughes, and Wu, Phys. Rev. **95**, 611 (1954), also **98**, 1840 (1955).

momenta $k_1\mathbf{n}_1$, $k_2\mathbf{n}_2$, $k_3\mathbf{n}_3$ with k_1 , k_2 in the ranges k_1 , k_1+dk_1 , and k_2 , k_2+dk_2 , respectively, and with \mathbf{n}_1 lying in the solid angle $d\Omega_1$ and with the plane of the three quanta making an angle ϕ_2 with the plane determined by \mathbf{n}_1 and a unit vector \mathbf{z} specifying the axis of quantization, with ϕ_2 lying in the range ϕ_2 , $\phi_2+d\phi_2$ is given by⁷

$$\omega = \frac{\kappa^3}{\pi} \frac{e^6}{m^2} \frac{dk_1}{m} \frac{dk_2}{m} \frac{d\Omega_1}{4\pi} \frac{d\phi_2}{2\pi} k_1 k_2 k_3 |H_{FA}|^2, \quad (1)$$

where $1/\kappa$ is twice the Bohr radius and H_{FA} is the matrix element.

The matrix element H_{FA} is easily evaluated from the expression following Eq. (3) in reference 1 for a given initial spin state and given polarizations of the quanta. The result is

$$k_1 k_2 k_3 |H_{FA}|^2 = t_z^2 \quad \text{triplet } m=0 \quad (2a)$$

$$= \frac{1}{2} (t_x^2 + t_y^2)$$

$$\text{triplet } m=+1 \text{ or } m=-1, \quad (2b)$$

where

$$\mathbf{t} = \sum_{\text{cyc}} [(\mathbf{e}_2 \cdot \mathbf{e}_3) - (\mathbf{e}_2' \cdot \mathbf{e}_3')] \mathbf{e}_1 + \{(\mathbf{e}_2' \cdot \mathbf{e}_3) + (\mathbf{e}_2 \cdot \mathbf{e}_3')\} \mathbf{e}_1' \quad (3)$$

is the vector defined in reference 1. Here \mathbf{e}_1 is a unit vector describing the linear polarization of quantum 1 and $\mathbf{e}_1' = \mathbf{e}_1 \times \mathbf{n}_1$, etc. The summation on the right hand side of Eq. (3) is over the three cyclic permutations of the indices 1, 2, 3.

On setting

$$\mathbf{e}_1^+ = (\mathbf{e}_1 + i\mathbf{e}_1')/\sqrt{2}, \quad \mathbf{e}_1^- = (\mathbf{e}_1 - i\mathbf{e}_1')/\sqrt{2} = (\mathbf{e}_1^+)^*,$$

Eq. (3) becomes

$$\mathbf{t} = \sqrt{2} \sum_{\text{cyc}} [(\mathbf{e}_2^+ \cdot \mathbf{e}_3^+) \mathbf{e}_1^- + (\mathbf{e}_2^- \cdot \mathbf{e}_3^-) \mathbf{e}_1^+]. \quad (4)$$

POLARIZATION OF THE ANNIHILATION QUANTA

The form of \mathbf{t} in Eq. (4) implies that if two quanta are right circularly polarized, the third must be left circularly polarized. This selection rule, a consequence here of first-order perturbation theory, was found by Fumi and Wolfenstein⁸ using general group theoretic arguments. Their result is restricted to the symmetric event in which $k_1 = k_2 = k_3 = \frac{2}{3}m$ and therefore the quanta emerge at angles of 120° with one another. Their result is further restricted to the $m = \pm 1$ states; they could not rule out terms of the form $RRR + LLL$ for the $m=0$ state.

The simultaneous measurement of the circular polarizations of all three annihilation quanta is not experimentally feasible, but the polarization of a single quantum with respect to the plane of the quanta has

been measured.⁴ For such an experiment, in which the polarizations, say, of quanta 2 and 3 are not measured, the relative probability that quantum 1 is linearly polarized at an angle α with respect to the normal to the plane of the quanta is then

$$\begin{aligned} k_1 k_2 k_3 \sum_{\text{spin } e_2, e_3} |H_{FA}|^2 \\ = 4(1 - \mathbf{n}_1 \cdot \mathbf{n}_2)(1 - \mathbf{n}_1 \cdot \mathbf{n}_3)(1 - \mathbf{n}_2 \cdot \mathbf{n}_3) \cos 2\alpha \\ + 4 \sum_{\text{cyc}} (1 - \mathbf{n}_1 \cdot \mathbf{n}_2)^2. \end{aligned} \quad (5)$$

The polarizations of quanta 2 and 3 and the initial spin states of the atom have been summed over in this expression.

If the plane of the quanta is perpendicular to the direction of quantization, the contribution of the $m=0$ state is:

$$\begin{aligned} k_1 k_2 k_3 \sum_{e_2 e_3} |H_{FA}|^2_{m=0} = 4(1 - \mathbf{n}_1 \cdot \mathbf{n}_3)(1 - \mathbf{n}_1 \cdot \mathbf{n}_2) \cos 2\alpha \\ + 2 \sum_{\text{cyc}} (1 - \mathbf{n}_2 \cdot \mathbf{n}_3)^2. \end{aligned} \quad (6)$$

The $m=+1$ and -1 states contribute equally. Their total contribution may be obtained by subtracting Eq. (6) from Eq. (5). The $m=\pm 1$ states favor polarization of quantum 1 perpendicular to or in the plane of the quanta according as $\mathbf{n}_2 \cdot \mathbf{n}_3$ is negative or positive. The $m=0$ state favors polarization perpendicular to the plane of the quanta for all angles, so that a magnetic field perpendicular to the plane of the quanta will always lower the polarization ratio, by quenching the $m=0$ state.

For the symmetric case ($k_1 = k_2 = k_3 = \frac{2}{3}m$) mentioned above, Eq. (5,6) imply that the ratio of the number of quanta polarized perpendicular to the plane of the quanta to the number polarized in the plane of detection of the quanta is 5:1 when the positronium atom annihilates from the triplet $m=0$ state, 2:1 for the $m=\pm 1$ states, and 3:1 for unpolarized positronium (corresponding to the experiment of reference 4, in which the spin state of the atom was not measured).

For completeness, the contribution of the $m=0$ state for an arbitrary orientation of the plane of the quanta is

$$\begin{aligned} k_1 k_2 k_3 \sum_{e_2 e_3} |H_{FA}|^2_{m=0} \\ = 4(1 - \mathbf{n}_1 \cdot \mathbf{n}_2)(1 - \mathbf{n}_1 \cdot \mathbf{n}_3) [(\mathbf{n} \cdot \mathbf{z})^2 \\ - (\mathbf{n} \times \mathbf{n}_2 \cdot \mathbf{z})(\mathbf{n} \times \mathbf{n}_3 \cdot \mathbf{z})] \cos 2\alpha \\ + 4(1 - \mathbf{n}_1 \cdot \mathbf{n}_2)(1 - \mathbf{n}_1 \cdot \mathbf{n}_3)(\mathbf{n} \cdot \mathbf{z}) \\ \times \{(\mathbf{n} \times \mathbf{n}_3 \cdot \mathbf{z}) + (\mathbf{n} \times \mathbf{n}_2 \cdot \mathbf{z})\} \sin 2\alpha \\ + 2 \sum_{\text{cyc}} (1 - \mathbf{n}_2 \cdot \mathbf{n}_3)^2 \{1 - (\mathbf{n}_1 \cdot \mathbf{z})^2\}, \end{aligned}$$

where \mathbf{n} is the unit normal vector to the plane of the quanta.

⁷ Reference 1, Eqs. (2) and (5).

⁸ F. G. Fumi and L. Wolfenstein, Phys. Rev. **90**, 498 (1953).

ANGULAR CORRELATIONS, ANGULAR DISTRIBUTION, AND ENERGY SPECTRUM

If no polarizations are measured, the relative probability of the quanta emerging with momenta $k_1\mathbf{n}_1$, $k_2\mathbf{n}_2$, $k_3\mathbf{n}_3$ is obtained by summing Eq. (2) over the polarizations of all three quanta:

$$4 \sum_{\text{cyc}} (1 - \mathbf{n}_1 \cdot \mathbf{n}_2)^2 [1 - (\mathbf{n}_3 \cdot \mathbf{z})^2], \quad m=0 \quad (8a)$$

$$k_1 k_2 k_3 \sum_{e_1 e_2 e_3} |H_{FA}|^2 = 4 \sum_{\text{cyc}} (1 - \mathbf{n}_1 \cdot \mathbf{n}_2)^2 [1 + (\mathbf{n}_3 \cdot \mathbf{z})^2], \quad m=+1 \text{ and } m=-1 \quad (8b)$$

$$8 \sum (1 - \mathbf{n}_1 \cdot \mathbf{n}_2)^2, \quad \text{all three spin states.} \quad (8c)$$

Equations (8a) and (8b) show that the angular correlations between the quanta depend on the initial spin state. Thus, in general, quenching the $m=0$ state alters the correlations. The angular correlations of the quanta emerging in the plane perpendicular to the direction of quantization ($\mathbf{n}_1 \cdot \mathbf{z} = \mathbf{n}_2 \cdot \mathbf{z} = \mathbf{n}_3 \cdot \mathbf{z} = 0$) are independent of initial spin state. Of these quanta, half (rather than $\frac{1}{3}$ as might be expected from statistical weights) are contributed by the $m=0$ state. This phenomenon has been detected.⁵

Equation (8c) gives, in agreement with Eq. (4) of reference 1, the angular correlations of the quanta from unpolarized positronium.

Integration of Eq. (8) over dk_2 and $d\phi_2$ gives the relative probability per unit solid angle and per unit energy interval for detecting quantum 1 with energy k_1 at an angle θ with the direction of quantization:

$$16[F(k_1/m) \sin^2\theta + f(k_1/m)(3 \cos^2\theta - 1)], \quad \text{triplet } m=0 \quad (9a)$$

$$8[F(k_1/m)(1 + \cos^2\theta) - f(k_1/m)(3 \cos^2\theta - 1)], \quad m=+1 \text{ or } m=-1 \quad (9b)$$

$$32F(k_1/m), \quad \text{all three spin states} \quad (9c)$$

where

$$F(k) = \frac{2(2-k)}{k} + \frac{4(1-k)}{k^2} \ln(1-k) + \frac{2k(1-k)}{(2-k)^2} - \frac{4(1-k)^2 \ln(1-k)}{(2-k)^3}, \quad (10)$$

$$f(k) = \frac{k}{2(2-k)^2} - \frac{(1-k)}{(2-k)^3} \ln(1-k) - \frac{4(1-k)}{k^3} - \frac{(2-k)^2}{2k^3} - \frac{3(2-k)(1-k) \ln(1-k)}{k^4}, \quad (11)$$

$$\int_0^1 F(k) dk = \pi^2 - 9, \quad (12)$$

$$\int_0^1 f(k) dk = \frac{1}{6}. \quad (13)$$

Equations (9a) and (9b) indicate that the angular distribution of the quanta depends on the initial spin state of the atom. Equation (9c), representing the angular distribution of quanta from unpolarized positronium, is independent of the direction of quantization, as it must be, and represents the relative probability per unit energy interval (dk_1) of finding a quantum with energy k_1 . It is the energy spectrum given by Ore and Powell.¹

From Eqs. (12), (13) and Eq. (9) with an integration over dk_1 , the total number of quanta making an angle θ with the direction of quantization is, apart from a constant,

$$(\pi^2 - 9) \sin^2\theta + \frac{1}{6}(3 \cos^2\theta - 1), \quad (14a)$$

$$(\pi^2 - 9)(1 + \cos^2\theta) - \frac{1}{6}(3 \cos^2\theta - 1), \quad (14b)$$

$$2(\pi^2 - 9). \quad (14c)$$

For $\theta=90^\circ$, the $m=+1$ and -1 states contribute $(\pi^2 - 9 + \frac{1}{6})/[2(\pi^2 - 9)] = 59.6\%$ of the quanta. Thus, turning on a very strong magnetic field perpendicular to the direction of detection of the quanta will reduce the counting rate by about 40% (and not by the statistical share, 33%, of the $m=0$ state). Account of this has been taken in interpreting the results of magnetic field quenching measurements on triplet positronium.^{6,9}

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⁹ S. DeBenedetti and H. C. Corben, Ann. Revs. Nuclear Sci. 4, 191 (1954).