A thin sample of Na^{22} (1.4×10^6 dis/sec) was located in the center of a bell-shaped Al container (2.5 cm in diameter, 0.44 g/cm² thick) filled with a dense atmosphere of SF_6. Positrons from this source formed positronium[2] and provided a sufficiently intense source of three-quantum annihilation events. Three NaI(Tl) scintillation counters were used to detect the gamma-rays, and were placed as indicated in Fig. 1. Counters A and B detected

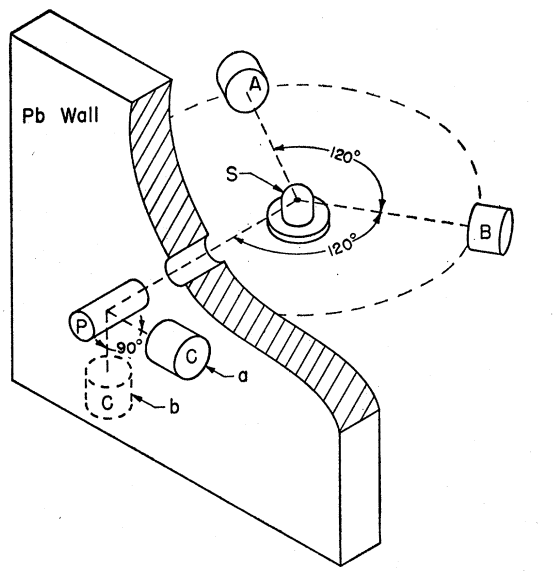

FIG. 1. Apparatus.

two of the three annihilation rays directly, while counter C detected the third ray only after it had been scattered by the polystyrene cylinder P. Triple coincidences were counted with counter C alternatively in positions a and b (Fig. 1). In such positions counter C detected rays scattered respectively parallel or perpendicular to the plane ABP. The background of triple coincidences was measured by lifting counter A at 45° away from the plane BPS, as described in the account of our previous work.[1] The geometric equivalence of positions a and b was checked by counting single scattered rays; the counting rates never differed by more than 10 percent in the two positions.

The experimental results, after subtraction of background are shown in Table I.

TABLE I. Experimental results.

Series	Position a (parallel) counts/hr	Position b (perpendicular) counts/hr	Ratio par./perp.
I	7.0 ± 1.2	3.6 ± 1.0	1.92 ± 0.55
II	9.4 ± 1.3	5.4 ± 1.2	1.73 ± 0.37
III	10.3 ± 1.3	5.2 ± 1.0	2.00 ± 0.43
IV	8.3 ± 1.3	4.7 ± 1.1	1.78 ± 0.44
Average			1.87 ± 0.23

The theory[3] predicts a polarization ratio 3/1 in favor of rays plane-polarized with the electric vector perpendicular to the plane of the three photons. A rough Monte-Carlo computation,[3] taking into account the geometry of the experiment and the anisotropy of Compton scattering for polarized photons, leads to the prediction of a ratio (parallel/perpendicular) 1.80 ± 0.15 for the result of the experiment, performed as described above. This is in agreement with the experimental value.

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[2] M. Deutsch, Phys. Rev. **82**, 455 (1951).
[3] R. Drisko (private communication).

Effect of the Electric Quadrupole Interaction on the $\gamma-\gamma$ Directional Correlation in Cd^{111}. II

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In a previous letter[1] we described experiments with metallic indium single crystals containing In^{111}, which indicate that the interaction which influences the angular correlation in this type of source is electric in origin. Furthermore, the magnetic decoupling experiments showed that the magnetic interaction cannot be responsible for the observed attenuations in polycrystalline sources, which were previously measured.[2] It is therefore reasonable to assume that these attenuations are also due to the coupling between the electric quadrupole moment of the nucleus and the inhomogeneous electric crystalline field.

In this letter we describe some new measurements confirming this hypothesis and we discuss the hypothesis in connection with older experiments.

Angular correlation measurements in sources in which no inhomogeneous electric fields exist at the radioactive nucleus should show the undisturbed correlation according to our quadrupole hypothesis. Experiments on three types of such sources have been carried out:

Type a: Cubic crystals. (The active atoms must be in lattice positions and the crystals must be sufficiently large and perfect. Nevertheless, cubic crystals do not exclude interaction with higher electric moments and therefore offer the possibility to be measured.)

Type b: Solutions. (Care must be taken to avoid hydrolysis and/or precipitation.)

Type c: Melts.

Experiments with sources of the first type were performed in the earlier work of the Zürich group.[2] The active atoms were embedded in different metals by the double stream evaporation method. Cubic crystallizing metals (Ag, Cu) showed close to the maximum anisotropy, $A=[W(180°)-W(90°)]/W(90°)=-0.20$, under certain conditions (sufficiently large and pure crystallites and sufficient thickness of the source), whereas crystals with lower symmetry than cubic (In, Te, Cd, Se) always gave considerably less than the maximum value of the anisotropy.

Experiments with sources of the second type have been reported.[3,4] Aqueous solutions of $InCl_3$, InI_3, $In_2(SO_4)_3$, $In(NO_3)_3$, and others gave $A=-0.20$, whereas crystalline sources of the same compounds showed nearly isotropic correlations. The disappearance of the attenuation of the angular correlation when a crystalline source is dissolved in water gives the most striking evidence for the quadrupole hypothesis.

We have recently measured sources of the third type. Indium metal containing In^{111} was carefully reduced with hydrogen and sealed into an evacuated thin-walled Pyrex glass capsule. As no special crystallization methods were used, the sources were poly-

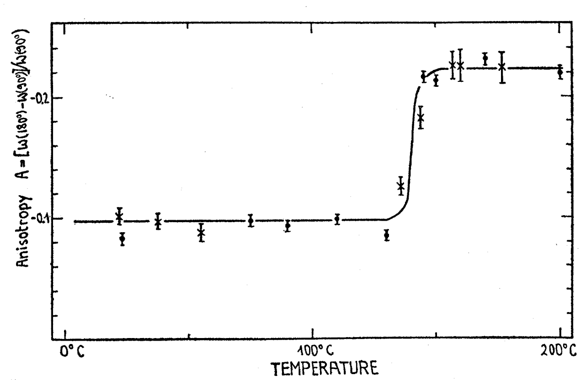

FIG. 1. Elimination of the quadrupole interaction by melting a polycrystalline metallic indium source. Anisotropy *vs* temperature. ● run 1, ✕ run 2-

crystalline. The anisotropy remained constant while the source was heated up to the melting point, where the anisotropy rose rapidly up to 22 percent and remained constant to higher temperatures. Figure 1 shows a typical run. The curve was measured twice with the same source.

These three types of experiments show that when no appreciable inhomogeneous electric fields are present, one obtains the undisturbed angular correlation. We therefore conclude, that in the case of Cd^{111} the experimental results for the various types of sources (excluding sources with neighboring paramagnetic ions) can be largely and perhaps entirely explained by the quadrupole interaction hypothesis.

The anisotropy measured with the "powder" sources of metallic indium, $A=-0.095\pm0.005$, will be used together with the results of the single crystal—and some other experiments (delayed coincidence, combined magnetic-electric field measurements) to determine the quadrupole coupling and the quadrupole moment by comparison with the theory developed by Alder.[5] This comparison will be treated in a following letter.

We thank Professor P. Scherrer for his continued interest in this work, K. Alder for the very helpful discussions of theory, and O. Braun for assistance during the experiments.

* National Science Foundation Postdoctoral Fellow, on leave from Argonne National Laboratory.

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$\gamma-\gamma$ Angular Correlation Measurements*

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In performing angular correlation measurements of nuclear radiations emitted in cascade, it is necessary to remember certain points which we shall discuss here mathematically.

(a) Time of measurements: Since the angular anisotropy expected is usually very small, it is necessary to keep small $\Delta W(\theta)$, the error in the measurement of $W(\theta)=G(\theta)/G(\pi/2)$; $G(\theta)$ and $G(\pi/2)$ being the genuine coincidence rates for angles θ and $\pi/2$ between the cascaded radiations. We have

$$\frac{\Delta W(\theta)}{W(\theta)}=\left[f^2(\theta)+f^2(\pi/2)+\frac{\bar{R}}{T^2(R)}\left(\frac{1}{G^2(\theta)}+\frac{1}{G^2(\pi/2)}\right)\right]^{\frac{1}{2}}$$

where $f(\theta)$ is defined by $f(\theta)^2=\bar{c}(\theta)/T^2(\theta)G^2(\theta)$, $\bar{c}(\theta)$ being the total number of coincidences recorded in time $T(\theta)$, and $\bar{R}$ is the total number of random coincidences recorded in time $T(R)$. With the error in counting random coincidences made very small and with $f(\theta)=f(\pi/2)$ we have $\Delta W(\theta)/W(\theta)\approx(2)^{\frac{1}{2}}f(\theta)$. We can show that the necessary time of observation $T(\theta)$ is connected to $f(\theta)$ by the relation

$$T(\theta)=\frac{1+\lambda}{f^2(\theta)}\cdot\frac{2\tau}{\epsilon_1\epsilon_2\omega_1\omega_2},$$

where τ is the resolving time of the coincidence circuit, ϵ_1 and ϵ_2 are the fractional efficiencies of the counters, ω_1 and ω_2 are the solid angles subtended by them (and expressed as fractions of 4π steradians), and λ is the ratio of the genuine to random coincidence rates ($=1/2N\tau$, N being the number of disintegrations per second). With this expression for $T(\theta)$ it is possible to plan the experiment for best results. For a typical set-up with scintillation counters where $\epsilon_1=\epsilon_2=0.10$ and with $\omega_1=\omega_2=2.5\times10^{-3}$ of 4π steradians, $\lambda=1.00$, $f=0.01$, the values of T for different values of τ are as follows:

τ(sec)	10^{-6}	10^{-7}	10^{-8}	10^{-9}	10^{-10}
T(hr)	178	17.8	1.78	0.18	0.02

(b) Angular resolution: In actual experiments, counters have definite angular spreads whose effects consist, we find, in modifying the coefficients occurring in the expansion of $G(\theta)$ in powers of $\cos^2\theta$. If the horizontal angular resolutions of the counters are $2\alpha_1$ and $2\alpha_2$ and the vertical angular resolutions are $2\beta_1$ and $2\beta_2$, and if the angles θ_0, ψ, ϕ, ξ, η are defined as in Fig. 1, then using $\cos\theta=\cos(\theta_0+\psi+\phi)+\sin\xi\sin\eta$ and the following expression for total number of genuine coincidence per sec for a separation of θ_0 between the centers of the counters:

$$G(\theta_0)=N\epsilon_1\epsilon_2\int_{-\alpha_1}^{\alpha_1}d\phi\int_{-\alpha_2}^{\alpha_2}d\psi\int_{-\beta_1}^{\beta_1}d\xi\int_{-\beta_2}^{\beta_2}d\eta\cdot g(\theta),$$

where $g(\theta)$ is the correlation function giving the probability of coincidence for an angle θ between the cascaded radiations $[g(\theta)=a_0+a_1\cos^2\theta+a_2\cos^4\theta]$, we have

$$G(\theta_0)=N\epsilon_1\epsilon_2\omega_1\omega_2[(a_0+a_0')+(a_1+a_1')\cos^2\theta_0+(a_2+a_2')\cos^4\theta_0].$$

Here a_0', a_1', and a_2' are the correction terms and with $\alpha_1=\alpha_2$, $\beta_1=\beta_2=\beta(<15°)$, and neglecting terms of order of magnitude $<10^{-4}$,

$$a_0'=\frac{a_1}{2}\left[1-\frac{\sin^2(2\alpha)}{4\alpha^2}\right]+a_2\left[-\frac{1}{2}\cdot\frac{\sin^2(2\alpha)}{4\alpha^2}(\sin^2\alpha-\sin^4\alpha)+\frac{3}{8}-\frac{3}{8}\cdot\frac{\sin^2(2\alpha)}{4\alpha^2}\right],$$

$$a_1'=a_1\left\{\frac{\sin^2(2\alpha)}{4\alpha^2}-1\right\}+a_2\left\{\frac{\sin^2(2\alpha)}{4\alpha^2}-\frac{\sin^2(4\alpha)}{16\alpha^2}\right\},$$

$$a_2'=a_2\left\{\frac{\sin^2(4\alpha)}{16\alpha^2}-1\right\}.$$

The corrections introduced by 2β, the vertical angular spread of the counters, are generally very small up to quite an appreciable value of 2β ($\sim15°$). The values in Table I, however, are calculated with the help of more complete expressions for a_0', a_1' and a_2'. We have assumed that $g(\theta)$ stops at $\cos^4\theta$, but there is no difficulty in extending the method to higher powers of $\cos\theta$. Finally, we have

$$W(\theta)=\frac{G(\theta_0)}{G(\pi/2)}=1+\frac{a_1+a_1'}{a_0+a_0'}\cos^2\theta_0+\frac{a_2+a_2'}{a_0+a_0'}\cos^4\theta_0.$$

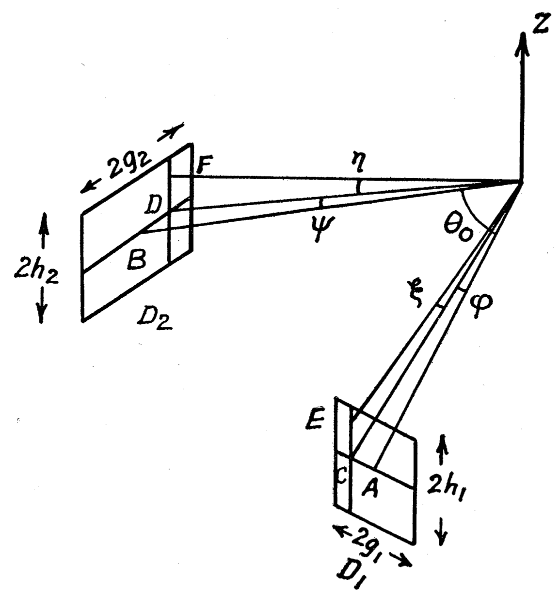

Fig. 1. Geometrical arrangement of the source and the crystal faces, showing the effect of finite angular resolution.