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Influence of the Time-Dependent Quadrupole Interaction on the Directional Correlation of the Cd^{111} Gamma Rays*†

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The influence of the time-dependent quadrupole interaction in various liquid sources of In^{111} on the directional correlation of the Cd^{111} gamma rays has been investigated quantitatively. By using the technique of delayed correlation measurements the characteristic interaction (relaxation) parameters λ_2 have been determined for aqueous $InCl_3$ solutions as a function of the macroscopic viscosity. These experiments revealed that λ_2 is not simply proportional to the macroscopic viscosity of the liquid as might be expected from simple considerations. The extranuclear interaction in indium metal sources was studied as a function of temperature. The relatively large attenuation of the Cd^{111} gamma-gamma correlation in polycrystalline indium metal was found to vanish completely upon reaching the liquid phase, a result which was confirmed by delayed correlation measurements. The "true," unperturbed, directional correlation of the Cd^{111} gamma rays was computed from these experimental data and is represented by $W(\Theta)=1-(0.180\pm0.002)P_2(\cos\Theta)+(0.002\pm0.003)P_4(\cos\Theta)$. This correlation is characteristic for a cascade of a mixed $M1+E2$ transition, with an $E2/M1$ intensity ratio of 0.021 ± 0.002 ($\delta=+0.146$) followed by a pure $E2$ transition, between states of angular momentum quantum number 7/2, 5/2, and 1/2.

1. INTRODUCTION

NUMEROUS theoretical as well as experimental investigations have been made of the influence of extranuclear fields on the directional correlation of nuclear radiation.[1] The interest in these extranuclear effects arises because of the following considerations:

(i) The interpretation of directional correlation measurements as to the angular momenta of the nuclear levels involved and as to the multipole character of the nuclear radiations[2–5] between these levels requires knowledge of the directional correlation which is unperturbed by any interaction of the nuclear moments with external fields. It is true, however, that in the majority of nuclear cascades the intermediate states involved have lifetimes τ_N of the order of 10^{-11} sec or less, which is too short a time for a correlation-affecting reorientation of the nucleus, even when the interaction of the nuclear moments with the almost always present atomic and crystalline fields is strong.[1] Nevertheless, the directional correlations of several nuclear cascades involving longer lived intermediate states are known only approximately or not at all due to the uncertainties concerning the influence of extranuclear effects.

(ii) On the other hand, quantitative information on parameters, which describe the interaction of the nuclear moments with extranuclear fields, makes it possible to compute the nuclear moments in the intermediate state, if the external fields at the sites of the nuclei are known.[1,6,7]

(iii) Once the nuclear moments are known, the dependence of directional correlations on the chemical and physical state of the source might provide information about the electric and magnetic fields prevailing at the decaying nucleus, and thus might give information on atomic and molecular structure and the structure of liquids and solids similar to the way that nuclear magnetic resonance absorption and nuclear induction do.

In this paper, which is the first one in a series of articles on the directional correlation of the Cd^{111} gamma-gamma cascade, we shall present investigations of the time-dependent interaction of the 247-kev Cd^{111} state in liquid sources of the parent nuclide In^{111}. From the results of delay experiments, the unperturbed directional correlation of the Cd^{111} gamma rays will then be extracted.

2. INFLUENCE OF EXTRANUCLEAR FIELDS ON DIRECTIONAL CORRELATIONS

2.1. General Considerations

An anisotropic intensity distribution of radiation emitted from a nuclear state of angular momentum $\mathbf{I}\hbar$ is a result of an unequal population of the $2I+1$ magnetic sublevels defined for some axis of quantization. In the absence of any extranuclear fields the state is $2I+1$ fold degenerate. The unequal population of the m-states can be introduced, for instance, by observation of the preceding radiation along a fixed direction z which leads to the formation of an ensemble of nuclear states with unequal population of the m-states defined with respect to z. The intensity distribution of the second radiation with respect to z is, in general, anisotropic and is termed the directional correlation

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* A brief account of this work was presented at the Washington Meeting of the American Physical Society, April 29–30, 1955 [Phys. Rev. **99**, 659 (1955)].

† Supported in part by the U. S. Atomic Energy Commission.

[1] For a review and references, see R. M. Steffen, Advances in Phys., Phil. Mag. Suppl. 4, 293 (1955).

[2] D. R. Hamilton, Phys. Rev. **58**, 122 (1940).

[3] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).

[4] H. Frauenfelder, article in K. Siegbahn's book on *Beta and Gamma Ray Spectroscopy* (North-Holland Publishing Company, Amsterdam, 1955), p. 531.

[5] M. Ferentz and N. Rosenzweig, Argonne National Laboratory Report ANL-5324, 1955 (unpublished).

[6] Alder, Albers-Schönberg, Heer, and Novey, Helv. Phys. Acta **26**, 761 (1953).

[7] A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

of the nuclear cascade. The relative probability that the two cascade quanta are emitted at an angle Θ is given by the correlation function $W^0(\Theta)$ which is most conveniently expressed as an expansion in even Legendre polynomials[8]:

$$W^0(\Theta)=1+\sum_{k=1}^{k_{\max}} A_{2k}{}^0 P_{2k}(\cos\Theta). \tag{1}$$

The coefficients $A_{2k}{}^0$ depend upon the angular momenta of the three nuclear states involved in the cascade and upon the multipole character of the radiation fields emitted.[2–4] Numerical values of these coefficients can be obtained from reference 3 or from the more extensive tables of Ferentz and Rosenzweig.[5]

2.2. Static Interactions in Solids

Interactions of the nuclear moments (magnetic dipole moment μ, electric quadrupole moment Q, and possibly moments of higher order) with external static fields cause a complete or partial lifting of the $2I+1$ fold degeneracy in the intermediate nuclear state and, depending upon the magnitude of the resulting energy splitting, the population of the m-sublevels as defined with respect to the axis of propagation of the first nuclear radiation, may change rapidly enough within the lifetime, $\tau_N=1/\lambda$, of the intermediate state to alter the directional correlation.[1,6]

Semiclassically, the situation in the case of the static interaction can be described adequately by the picture of a nucleus precessing with its axis (**I**) about the symmetry axis of the stationary field. The "precession frequencies" are the frequency equivalents of the energy splittings mentioned above. In this investigation we are concerned about the interactions of the electric quadrupole moment Q of the nucleus with the electric charge distribution of its environment. Magnetic interactions will be considered in the following paper.[9]

As far as the electric quadrupole interaction is concerned, the extranuclear fields are described by the electrostatic gradient, grad **E**. This tensor quantity reduces to a single essential component, $\partial E_z/\partial z$, $[\partial E_x/\partial x=\partial E_y/\partial y=-\frac{1}{2}(\partial E_z/\partial z)]$, in case of axial symmetry (e.g., about the z axis). The electric quadrupole interaction is then characterized by the fundamental precession frequency ω_e (corresponding to the smallest nonvanishing energy splitting):

$$\omega_e=\frac{3}{4I(2I-1)}\frac{eQ}{\hbar}\frac{\partial E_z}{\partial z}, \tag{2}$$

for integral nuclear spin I. It is twice as large for half-integral values of I.

In solids the extranuclear fields which give rise to an interaction with the nuclear moments are essentially static fields and depend upon the crystalline and atomic structure. In polycrystalline sources, without externally applied fields, where the crystallographic axes of the microcrystals are oriented at random, the perturbing interaction does not introduce a preferred direction in the radioactive source as a whole. In this case the directional correlation retains its essential form of Eq. (1), but each coefficient $A_{2k}{}^0$ is multiplied by an "attenuation" factor G_{2k} $(G_{2k}\leq 1)$:

$$W(\Theta)=1+\sum_{k=1}^{k_{\max}} A_{2k}{}^0 P_{2k}(\cos\Theta)$$
$$=1+\sum_{k=1}^{k_{\max}} A_{2k}{}^0 G_{2k} P_{2k}(\cos\Theta). \tag{3}$$

The attenuation coefficients $G_{2k}(t)$ are *periodic* functions of the time interval t during which the intermediate nuclear state is exposed to the static fields; e.g., $G_2(t)$ for the case of the electric quadrupole interaction involving an intermediate state of $I=5/2$ is represented by[7]

$$G_2(t)=[7+13\cos(\omega_e t)+10\cos(2\omega_e t)+5\cos(3\omega_e t)]/35. \tag{4}$$

In practice, a directional correlation is measured by means of a coincidence analyzer of finite resolving time τ_0. The time-integrated attenuation coefficient $\bar{G}_2$ is then easily obtained from the differential form Eq. (4), by integration:

$$\bar{G}_2=\int_0^{\tau_0} e^{-\lambda t}G_2(t)dt\Big/\int_0^{\tau_0} e^{-\lambda t}dt, \tag{5}$$

$$\bar{G}_2=[7+13J(\omega_e)+10J(2\omega_e)+5J(3\omega_e)]/35, \tag{6a}$$

where

$$J(n\omega_e)=\frac{1}{1+(n\omega_e\tau_N)^2}\times\frac{1-e^{-\lambda\tau_0}[\cos(n\omega_e\tau_0)-n\omega_e\tau_N\sin(n\omega_e\tau_0)]}{1-e^{-\lambda\tau_0}}. \tag{6}$$

For the static "isotropic" interaction the $\bar{G}_{2k}$'s will never vanish completely: a limiting or "hard core" correlation remains.[7] In fact, it can be shown that the irreducible minimum of $\bar{G}_{2k}$ for the integral correlation $(\tau_0\gg\tau_N)$ and for static interaction is

$$\bar{G}_{2k}(\min)=1/(4k+1). \tag{7}$$

2.3. Time-Dependent Interactions in Liquids

2.3. 1. Fluctuating Fields in a Liquid

The possibility of a perturbation of directional correlation phenomena by interactions of the nuclear

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[8] The symbols $A_{2k}{}^0$ are used to denote the coefficients of the unperturbed correlation; they correspond to the A_{2k} in references 3–5.

[9] R. M. Steffen and W. Zobel, Phys. Rev. **102**, 126 (1956) following paper.

moments with the *rapidly fluctuating fields* such as those present in a liquid due to the thermal motion of the molecules has first been pointed out by Pound and Abragam.[7] The mechanism involved in this interaction is similar to the relaxation mechanism which brings the nuclear spin system in nuclear magnetic resonance absorption back to thermal equilibrium with its environment.[10]

The quantitative analysis of this "relaxation" process in a liquid requires information as to how the local environment of a given nucleus changes in time. Any field at some point in the liquid, arising from a particular instantaneous configuration of the constituents of the liquid, must change continually in a random fashion. Any nucleus in the liquid is thus subjected to interacting fields which change rapidly in direction as well as in magnitude. In their model, Abragam and Pound[7] described the interaction as due to a field whose magnitude, on the average, is constant but whose direction fluctuates in a random manner.

2.3. 2. Time-Dependent Quadrupole Interaction

It is to be expected, and it has been corroborated by experiments, that the most effective time-dependent interaction mechanism in a liquid for the attenuation of a directional correlation is due to the interaction of the electric quadrupole moment Q in the nuclear intermediate state and the fluctuating electric gradients of average (mean square) magnitude $\langle(\partial E_z/\partial z')^2\rangle_{\text{Av}}$, where z' denotes the direction of largest field gradient in coordinates moving with the fluctuations.

The time-dependent quadrupole interaction is then characterized by parameters λ_{2k}, which also depend on the "correlation time" τ_c. The latter can be described as the time interval during which, on the average, a given orientation of the field axis z' changes appreciably. The correlation time τ_c is intimately related to the "characteristic" time τ_D in Debye's theory of dielectric dispersion;[11] that is, the time in which an assembly of molecules oriented by an external electric field loses its distribution with respect to a fixed direction by virtue of the Brownian motion, after the electric field has been removed. Following Debye's approach, treating the constituents of the liquid as rigid spheres of radius a in a viscous medium of viscosity η a crude approximation for τ_c can be constructed[10]:

$$\tau_c=\tfrac{1}{3}\tau_D=(4\pi a^3/3kT)\eta. \tag{8}$$

It should be kept in mind that this expression is necessarily the result of a much oversimplified picture. In particular, the viscosity η in Eq. (8), which is responsible for the "frictional" forces encountered in the rotational motion of the molecules, is not necessarily identical with the macroscopic viscosity of the liquid, which is measured on the basis of "friction" involving translatory motion of the molecules in the liquid. The presence of a time-dependent interaction in a liquid does not introduce a preferred direction at the radioactive source, and thus the perturbed directional correlation again can be represented by a function of the form of Eq. (3). The attenuation coefficients $G_2(t)$, however, are now *exponential* functions of t:[7]

$$G_{2k}(t)=e^{-\lambda_{2k}t}. \tag{9}$$

The interaction or relaxation parameters λ_{2k} have been calculated by Abragam and Pound.[7]

$$\lambda_{2k}=\frac{3}{80}\left(\frac{eQ}{\hbar}\right)^2\left\langle\left(\frac{\partial E_z}{\partial z'}\right)^2\right\rangle_{\text{Av}} \times\tau_c\frac{2k(2k+1)[4I(I+1)-2k(2k+1)-1]}{I^2(2I-1)^2}. \tag{10}$$

The time-integrated attenuation coefficients [compare Eq. (5)] for a coincidence resolving time τ_0 are given by the following expression:

$$\bar{G}_{2k}=\frac{1}{1+\lambda_{2k}\tau_N}\,\frac{1-e^{-(\lambda+\lambda_{2k})\tau_0}}{1-e^{-\lambda\tau_0}}. \tag{11}$$

If a delay $T(T>\tau_0)$ is incorporated into one channel of the coincidence analyzer, the time integrated attenuaattenuation coefficients are given by

$$\bar{G}_{2k}(T)=\int_{T-\tau_0}^{T+\tau_0}e^{-\lambda t}G_{2k}(t)dt\Big/\int_{T-\tau_0}^{T+\tau_0}e^{-\lambda t}dt, \tag{12}$$

$$\bar{G}_{2k}(T)=\frac{1}{1+\lambda_{2k}\tau_N}\,\frac{\sinh[(\lambda+\lambda_{2k})\tau_0]}{\sinh(\lambda\tau_0)}\,e^{-\lambda_{2k}T}. \tag{13}$$

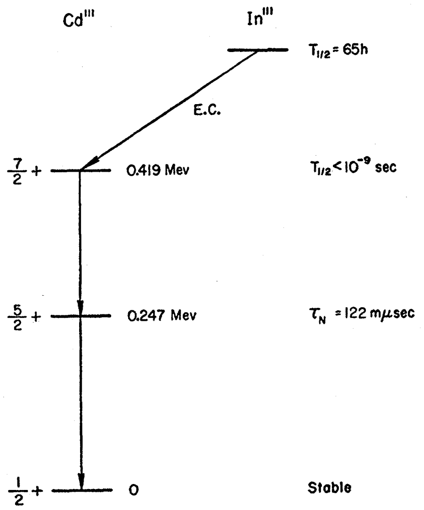

FIG. 1. The gamma-gamma cascade in Cd^{111} following the electron capture decay of In^{111}.

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[10] Bloembergen, Pound, and Purcell, Phys. Rev. 73, 679 (1948).

[11] P. Debye, *Polar Molecules* (Dover Publications, New York, 1945), Chap. V.

3. EXPERIMENTAL

3.1. Directional Correlation of the Cd^{111} Cascade

The unperturbed directional correlation of the Cd^{111} gamma-gamma cascade (Fig. 1) is of the form

$$W^0(\Theta)=1+A_2{}^0P_2(\cos\Theta)+A_4{}^0P_4(\cos\Theta). \qquad (14)$$

There was good, yet not conclusive, evidence that the values of $A_2{}^0$ and $A_4{}^0$ are close to -0.17 and 0, respectively. These values have been extracted from an analysis of direction correlation measurements on In^{111} sources of very different structure.[1] The directional correlation measurements, which will be described in this paper, allow an unambiguous, accurate determination of the Cd^{111} gamma-gamma correlation function:

$$W^0(\Theta)=1-(0.180\pm0.002)P_2(\cos\Theta)$$
$$+(0.002\pm0.003)P_4(\cos\Theta).$$

In the following discussion, the $P_4(\cos\Theta)$ term will be neglected.

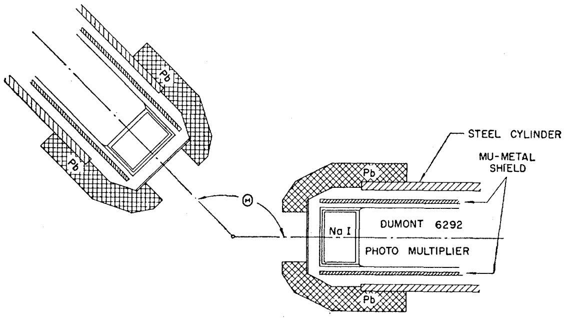

FIG. 2. Diagram of the detector arrangement.

The Cd^{111} cascade is ideally suited for the investigation of extranuclear effects on directional correlations. The intermediate lifetime, $\tau_N=1/\lambda=(122.3^{+1.8}_{-0.8})$ mμ sec,[12] is long enough to allow a strong influence on the directional correlation by external fields and yet is is short enough to perform coincidence measurements with high accuracy. Furthermore, the half-life ($T_{\frac{1}{2}}=65$ hr) of the parent In^{111} is of convenient magnitude and the decay of In^{111} is simple and well known.

3.2. Preparation of the In^{111} Sources

The In^{111} was produced by an (α,2n) reaction on Ag^{109} in the Purdue cyclotron. This method of preparing In^{111} has the advantage that no long-lived undesired radioactive indium isotopes (e.g., In^{114}), which might interfere with the correlation measurements, are present. In the chemical separation of the indium from the silver of the target, very small amounts of indium carrier (~0.2 mg) were used. Carrier-free coprecipitation of the $In(OH)_3$ with $Fe(OH)_3$ was used in some cases. The various indium compounds used in the investigation were prepared by dissolving the well washed $In(OH)_3$ precipitate in small amounts of acids or other chemicals. After evaporation to dryness the indium salts were redissolved in distilled water, ether, chloroform or other solvents and sealed into small Pyrex ampoules of 3-mm inside diameter. The height of the sources was about 10 mm. Metallic indium was obtained by electroplating the metal onto a thin indium wire from a formic acid solution. After purification by heating the metal in a hydrogen atmosphere the small indium metal cylinder (1 mm in diameter, 10 mm long) was vacuum sealed into a Pyrex tube of similar inside dimensions. Unless otherwise stated, the temperature of the sources was maintained constant at 25°C during the correlation measurements on solutions. The measurements on metallic indium sources were performed at source temperatures ranging from −190°C up to 695°C.

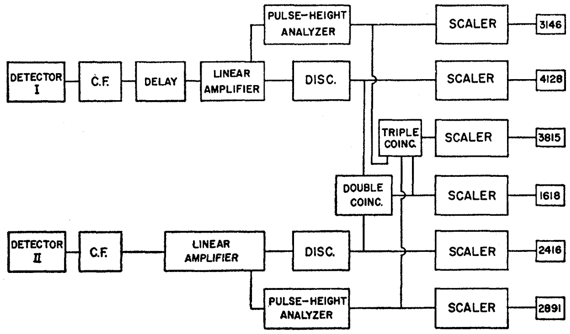

FIG. 3. Block diagram of the coincidence-spectrometer electronics.

3.3. Directional Correlation Apparatus

The essential features of the experimental arrangement are shown in Fig. 2. The detectors are NaI(Tl) cylindrical single crystals mounted on DuMont 6292 photomultiplier tubes. The energy resolution of the two detectors is between 8% and 9% (for the 661-kev gamma radiation of Cs^{137}). The directional correlation measurements were performed by means of a coincidence spectrometer which incorporated simultaneous differential and integral pulse height selection. The block diagram of the electronics is presented in Fig. 3. A more detailed description of the circuits will be published in the near future.[13] By inserting different lengths of RG7/U cable between cathode follower and linear amplifier one channel can be delayed with respect to the other. The over-all resolving time τ_0 of the equipment can be varied from 80 to 350 millimicroseconds. In most measurements presented here a resolving time of $\tau_0=(172\pm2)$ mμsec was used, except for the delayed correlation experiments, where a shorter resolving time, $\tau_0=(102\pm2)$ mμsec

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[12] P. C. Simms and R. M. Steffen, Bull. Am. Phys. Soc. Ser. II, **1**, 207 (1956); to be published in *The Physical Review.*

[13] H. Paul and R. M. Steffen (to be published).

was employed. The data accumulated in the integral pulse-height channels were used for control purposes only, unless otherwise stated. Drift effects of the equipment were found to be negligibly small as long as the temperature of the laboratory remained constant. Thus, special precautions were taken to keep the room temperature at a constant level.

Operation of the equipment is fully automatic. The angle between the axes of the detectors is changed at preset time intervals (15 min or 60 min) and the data are recorded photographically.

3.4. Experimental Procedure

Unless indicated otherwise, the coincidence counting rate was measured at eleven angular positions $\Theta = 90°$, $110°, 130°, 150°, 170°, 180°, 190°, 210°, 230°, 250°, 270°$. From these data the experimental correlation function $W'(\Theta)$ was obtained by a least-square fit to a function of the form $W'(\Theta) = A_0' + A_2' P_2(\cos\Theta)$, from which the actual correlation function $W(\Theta) = 1 + A_2 P_2(\cos\Theta)$ was computed by applying the corrections for the finite size of the detectors and for the finite extension of the source. The methods described by Frankel and Feingold[14,15] were employed for this purpose. It was verified experimentally that the expression for the local efficiency $\epsilon(\vartheta) = 1 - e^{-\mu x(\vartheta)}$ of the scintillation detectors as given by Rose[16] furnishes the proper correction only if differential pulse height analysis is used. With integral energy discrimination, degraded radiation scattered from the shields is accepted, and an experimentally determined efficiency function $\epsilon(\vartheta)$ must be used for the geometrical correction (compare reference 9).

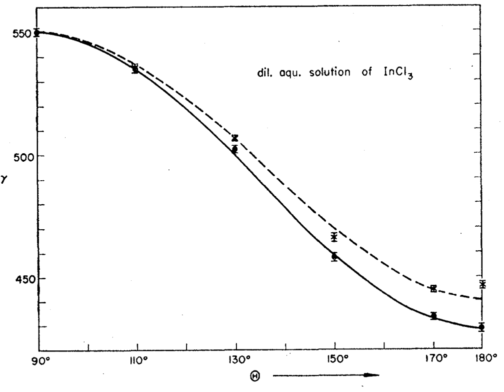

FIG. 4. The Cd^{111} gamma-gamma directional correlation displayed by an In^{111} source in the form of a dilute aqueous solution of $InCl_3$. The solid curve represents the measurements with differential energy selection. The dotted curve was obtained with integral energy discrimination.

4. MEASUREMENTS ON SOLUTIONS OF INDIUM SALTS

4.1. Directional Correlation Without Delay

The influence of the viscosity of the source on the undelayed directional correlation of the Cd^{111} gamma rays had been measured independently by the Purdue[17] and the Zurich group.[18] These investigations, however, were made without differential pulse-height selection in the two coincidence channels. It was found in the course of our measurements that the experimental anisotropy $A = [W(180°) - W(90°)]/W(90°)$ of the Cd^{111} directional correlation observed with differential pulse-height selection differed by as much as ten percent from the results obtained with integral discrimination where all gamma radiation above 70 kev was accepted.[19] This effect was attributed to scattering effects from the surroundings and in the source. Figure 4 shows the Cd^{111} directional correlation measured with an In^{111}

TABLE I. The Cd^{111} gamma-gamma directional correlation observed with In^{111} sources of different chemical composition. ($\tau_0 = 172$ mμ sec).

Form of the radioactive In^{111} source	Viscosity cgs units	A_2	$\bar{G}_2$
$InCl_3$, dilute aqueous solution	0.009	-0.168 ± 0.004	0.93 ± 0.03
$InCl_3$, dilute aqueous solution $+10^{21}Fe^{+++}$ ions per cm^3 added	0.009	-0.165 ± 0.004	0.92 ± 0.03
InI_3, dilute aqueous solution	0.009	-0.167 ± 0.004	0.93 ± 0.03
$In_2(SO_4)_3$, dilute aqueous solution	0.009	-0.165 ± 0.008	0.92 ± 0.05
$In(ClO_4)_3 \cdot 8H_2O$, dilute aqueous solution	0.009	-0.170 ± 0.005	0.94 ± 0.03
$In(NO_3)_3 \cdot 3H_2O$, dilute aqueous solution	0.009	-0.165 ± 0.007	0.92 ± 0.04
$In(NO_3)_3 \cdot 3H_2O$, dissolved in ethyl alcohol	0.011	-0.164 ± 0.008	0.91 ± 0.05
$In(ClO_4)_3 \cdot 8H_2O$, dissolved in ethyl alcohol	0.011	-0.168 ± 0.005	0.93 ± 0.03
InI_3, dissolved in xylene	0.007	-0.163 ± 0.007	0.90 ± 0.04
$In(C_9H_6ON)$, dissolved in chloroform	0.005	-0.158 ± 0.009	0.88 ± 0.06
InI_3, liquid salt (220°C)	?	-0.15 ± 0.01	0.83 ± 0.06
In, liquid metal (165°C)	0.009	-0.180 ± 0.02	1.00
$In(OH)_3$, gelatinous		-0.020 ± 0.006	0.11 ± 0.03
$In(OH)_3$, dry		-0.030 ± 0.006	0.16 ± 0.04
$InCl_3$, dried at 25°C		-0.021 ± 0.003	0.11 ± 0.03
$InCl_3$, vacuum dried at 150°C		-0.035 ± 0.003	0.19 ± 0.02
$InCl_3$, gas (1200°C)		-0.105 ± 0.008	0.58 ± 0.05
InI_3, polycrystalline		-0.031 ± 0.005	0.17 ± 0.03
In_2O_3, dry		-0.061 ± 0.004	0.34 ± 0.03
In metal, polycrystalline (25°C)		-0.059 ± 0.003^a	0.33 ± 0.015^a
In metal, polycrystalline (−190°C)		-0.055 ± 0.005^a	0.31 ± 0.03^a

[a] $\tau_0 = 295$ mμsec.

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[14] S. Frankel, Phys. Rev. **83**, 673 (1951).

[15] A. M. Feingold and S. Frankel, Phys. Rev. **97**, 1025 (1955).

[16] M. E. Rose, Phys. Rev. **91**, 610 (1953).

[17] P. B. Hemmig and R. M. Steffen, Phys. Rev. **92**, 832 (1953).

[18] Albers-Schönberg, Heer, Gimmi, and Novey, Helv. Phys. Acta **26**, 599 (1953).

[19] To give this comparison any significance, the possibility of disturbing coincidence counts due to Compton scattering from one counter to the other was eliminated in the case of integral discrimination by proper shielding of the scintillation detectors with lead.

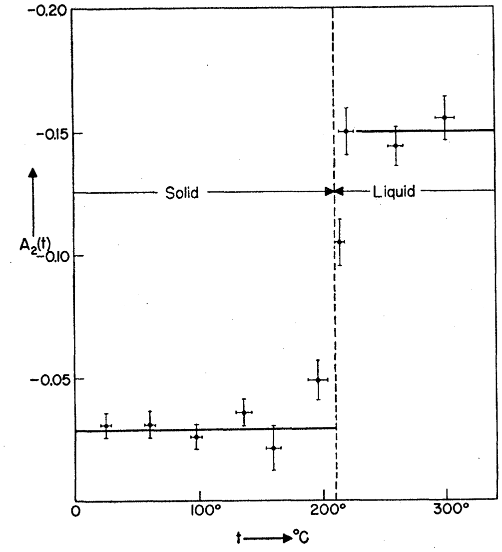

FIG. 5. The correlation factor $A_2(t)$ of the Cd^{111} gamma-gamma correlation displayed by sources of InI_3 as function of temperature.

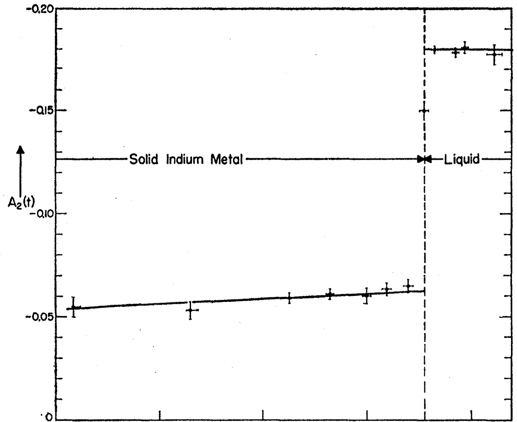

FIG. 6. The correlation factor $A_2(t)$ measured with indium metal sources as function of temperature ($\tau_0=295$ mμsec).

source in the form of a very dilute aqueous solution of $InCl_3$ using differential and integral pulse height selection (solid and dotted curve of Fig. 4, respectively). Table I summarizes the directional correlation measurements on liquid In^{111} sources of different types. For purposes of comparison, some results obtained with solid sources are included. The attenuation factors $\bar{G}_2$ were calculated on the basis of the "unperturbed" coefficient, $A_2^0=-0.180\pm0.002$, as determined later in this paper. It is interesting to note that of all In^{111} sources listed in this table only the gelatinous $In(OH)_3$ and the not too carefully dried $InCl_3$ exhibit an attenuation which is definitely below the hard core value, $G_2(\min)=0.2$. It is evident that the attenuation of the Cd^{111} correlation is small in all liquid sources of small viscosity, whatever the nature of the solvent or the solute.[1] As pointed out by Abragam and Pound[7] this is a consequence of the short correlation time τ_c in liquids of this kind [compare Eq. (10)]. For dilute aqueous solutions τ_c is of the order of 10^{-11} sec, thus for Cd^{111}: $(\tau_c/\tau_N)\simeq 10^{-4}$. The time-dependent quadrupole interaction is small if

$$\left[\left(\frac{eQ}{\hbar}\right)^2\left\langle\left(\frac{\partial E_z}{\partial z'}\right)^2\right\rangle_{\mathrm{Av}}\tau_N^2\right]\frac{\tau_c}{\tau_N}\ll 1.$$

The first factor, which measures, essentially, the effectiveness of the interaction in the case of a *stationary field* of magnitude $\langle(\partial E_z/\partial z')^2\rangle_{\mathrm{Av}}^{\frac{1}{2}}$ might be considerably larger than unity, resulting in a strong perturbation if the fields are static (solid sources) yet the small value of τ_c/τ_N, as is the case for the rapidly fluctuating fields in a nonviscous liquid, renders the interaction ineffective for the attenuation of a directional correlation. The argument still holds if the extranuclear fields are due to after-effects of the preceding electron-capture process in the electron shell of the Cd ion. The disturbed electron shell is then in a state of continuous fluctuation, which, *if fast enough*, causes the fields, produced by the electron distribution to average out to zero.

The difference between static interactions in solids and time-dependent interactions in liquid media is spectacularly evidenced by the sudden increase of the anisotropy of the Cd^{111} gamma corrrelation upon the change of the In^{111} source from the solid to the liquid phase. Figure 5 shows the factor A_2 of the Cd^{111} gamma correlation displayed by sources of InI_3 as a function of temperature. Whereas the polycrystalline solid sources exhibit a strongly perturbed directional correlation, which is, within our error limits, just compatible with the hard core attenuation, $G_2(\min)=0.2$, the perturbation becomes small upon reaching the liquid phase of InI_3.

A similar behavior is observed with In^{111} sources in the form of indium metal (Fig. 6). The sudden jump of the observed A_2 from -0.055 to -0.018 as soon as the melting point of indium is reached demonstrates again the small time-dependent interaction in the liquid phase.

The directional correlation observed with polycrystalline indium metal (e.g., at room temperature, $t=25$°C) corresponds to an attenuation factor $G_2=0.327\pm0.015$. With this value the static quadrupole coupling in the tetragonal indium lattice can be computed on the basis of Eq. (6):

$$\omega_e\tau_N=\frac{eQ}{\hbar}\frac{\partial E_z}{\partial z}\frac{3}{2I(2I-1)}\tau_N=1.68\pm0.10.$$

This coupling constant agrees well with those obtained from experiments on single crystals of indium.

The temperature dependence of A_2 observed with a solid polycrystalline indium source seems to indicate a tendency of the quadrupole interaction to become smaller with increasing temperature (Fig. 6). Such an effect could be related to the change of the axial ratio of the tetragonal lattice of metallic indium with temperature. According to observations of Graham, Moore, and Raynor[20] the c/a ratio of indium decreases from 1.082 at liquid air temperature to 1.066 at 135°C. Thus the electrostatic gradient, $\partial E_z/\partial z$, is expected to change by about 20% within this temperature range. The accompanying change of G_2 is then of the order of about 10% and would be consistent with our data. More accurate measurements, however, are needed before any conclusions as to the reality of this effect can be drawn. A more detailed investigation of this temperature effect on the static quadrupole interaction in indium metal is in progress.

The addition of paramagnetic ions (Fe^{+++}) in large concentrations ($10^{21}Fe^{+++}$ ions/cm^3) to aqueous solutions of $InCl_3$ did not result in any change of the attenuation. This fact is interpreted as an indication that the small attenuation observed in aqueous solutions is predominantly due to the time-dependent *quadrupole* interaction and not due to magnetic relaxation phenomena.

The correlation time τ_c of a liquid can be altered by changing the macroscopic viscosity η [compare Eq. (8)], and thus the attenuation factor $\bar{G}_2$ must depend strongly on η. Figure 7 shows the result of measurements on In^{111} sources of different viscosity. The sources used were dilute aqueous solutions of $InCl_3$ ($\lesssim 0.2$ mg $InCl_3$/ml) whose macroscopic viscosities were controlled by adding the proper amounts of waterfree glycerine. The viscosity at 25° was determined with the help of an Ostwald type viscosimeter. The experimental values represented in Fig. 7 agree fairly well with similar curves obtained by Hemmig and Steffen[17] and by the Zürich group,[18] yet the accuracy is considerably improved. The curve was computed on the basis of Eq. (11) and

$$\lambda_2 = c\eta, \qquad (15)$$

where c is a constant factor assumed to be independent of the viscosity. The expression represented in Eq. (15) follows directly from Eqs. (8) and (10). The best fit to the experimental points on the basis of Eq. (15) was obtained with the value, $c = 20$ g^{-1} cm^{-1}. However, there seems to exist a small, yet definite, discrepancy between the behavior of the experimental points and the shape of the calculated curve, a fact which casts some doubt on the validity of Eq. (15), at least as far as the time-dependent interactions in the water-glycerine mixtures are concerned.

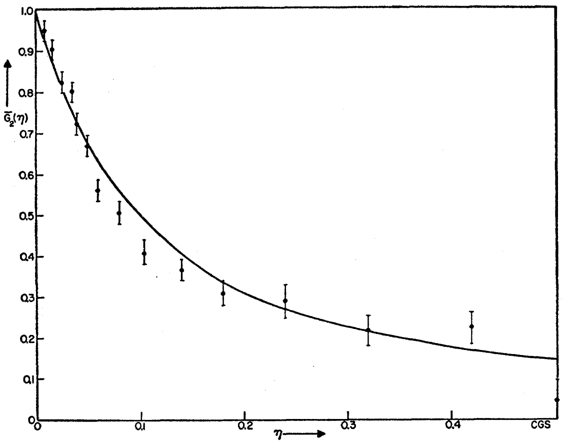

FIG. 7. The attenuation factor $\bar{G}_2$ observed with liquid In^{111} sources as function of the macroscopic viscoisity η.

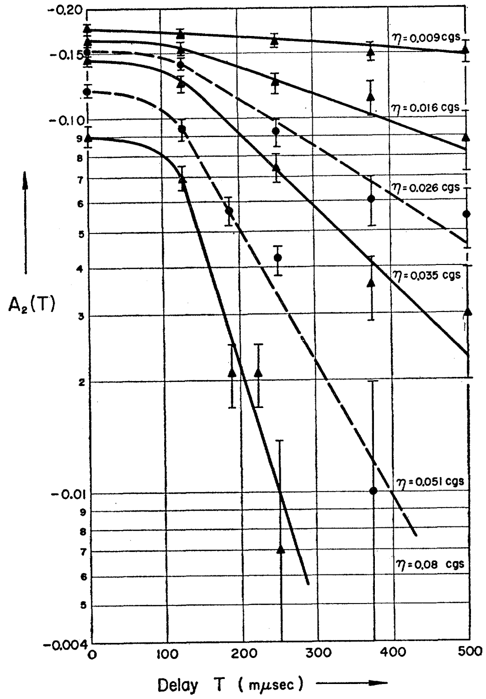

FIG. 8. Representative examples of measurements of the coefficient A_2 as function of delay T on In^{111} sources of different viscosities η.

4.2. Delayed Directional Correlation Measurements

The same sources on which the undelayed directional correlation was measured were used for the study of the correlation observed as a function of the delay time T.

Some of the experimental results for the coefficient $A_2 = A_2^0 G_2(T)$ of the Cd^{111} correlation function, which were obtained with sources of different viscosity, are plotted in Fig. 8. The experimental points follow, within the limits of error, an exponential law, an indication that the interaction mechanism in liquids is essentially the one proposed by Abragam and Pound[7] [compare Eq.

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[20] Graham, Moore, and Raynor, J. Inst. Metals 84, 86 (1955/56).

(13)]. The interaction parameters $\lambda_2(\eta)$ computed from the slope of the observed $A_2{}^0\bar{G}_2(T)$ curves are summarized in Table II. It is interesting to note that the value of λ_2 for a dilute aqueous solution is considerably smaller than that computed by Hemmig and Steffen,[17] who assumed the validity of Eq. (15). Nevertheless, the time-dependent interaction is not negligible as was reported earlier for sources of this kind.[21] The interaction parameter λ_2, as determined by the delayed correlation measurements, is plotted in Fig. 9 as a function of the viscosity η of the liquid sources. The curve fitted to the experimental points (solid line) is markedly different from the curve which represents the simple proportionality of Eq. (15) between λ_2 and η (dashed curve). The discrepancy is not surprising considering the simplifying assumptions made in obtaining this expression. Several reasons might be envisaged to explain why Eq. (8) is not too good an approximation:

(i) As mentioned in Sec. 2.3.2. the macroscopic viscosity of a liquid may not be a representative parameter to describe phenomena involving the mere *rotation* of molecules.

TABLE II. Interaction (relaxation) parameters λ_2 observed with liquid In^{111} sources of different viscosities $\eta(\tau_0 = 102\ m\mu\ sec)$.

η cgs	$\bar{A}_2(0)$	$\bar{G}_2(0)$	$\lambda_2\ sec^{-1}$
0.009 ± 0.001	-0.172 ± 0.003	0.955 ± 0.020	$0.56 \pm 0.12 \times 10^6$
0.016 ± 0.001	-0.163 ± 0.003	0.905 ± 0.020	$1.6 \pm 0.3 \times 10^6$
0.026 ± 0.001	-0.148 ± 0.004	0.822 ± 0.023	$3.0 \pm 0.6 \times 10^6$
0.035 ± 0.002	-0.145 ± 0.004	0.805 ± 0.025	$4.7 \pm 0.8 \times 10^6$
0.039 ± 0.002	-0.130 ± 0.005	0.722 ± 0.033	$7.4 \pm 0.8 \times 10^6$
0.051 ± 0.003	-0.120 ± 0.005	0.667 ± 0.033	$8.4 \times 1.6 \times 10^6$
0.060 ± 0.004	-0.101 ± 0.005	0.562 ± 0.035	$15.1 \pm 2.6 \times 10^6$
0.080 ± 0.006	-0.091 ± 0.005	0.505 ± 0.035	$16.2 \pm 2.5 \times 10^6$
0.105 ± 0.010	-0.075 ± 0.005	0.417 ± 0.030	$24.0 \pm 5.0 \times 10^6$

(ii) The molecular radius a of the molecules in the aqueous solution-glycerine mixtures was assumed to be constant. The assumption of an average molecular radius, however, according to

$$\bar{a} = (N_G a_g + N_W a_w)/(N_G + N_W), \tag{16}$$

where N_G and N_W are the number of glycerine and water-molecules of radius a_g and a_w, respectively, does not result in any better agreement with the experimental results.

(iii) Equation (15) implies that $\langle(\partial E_z/\partial z')^2\rangle_{Av}$ is independent of the glycerine concentration. Some justification for this assumption might be given.[22] Owing to the fact that the number of OH− groups per unit volume is approximately the same for glycerine and water, it is not improbable that the In^{+++} ions are surrounded by the same OH− configuration, independent of the glycerine concentration. To what extent this situation is realized in our case is not known.

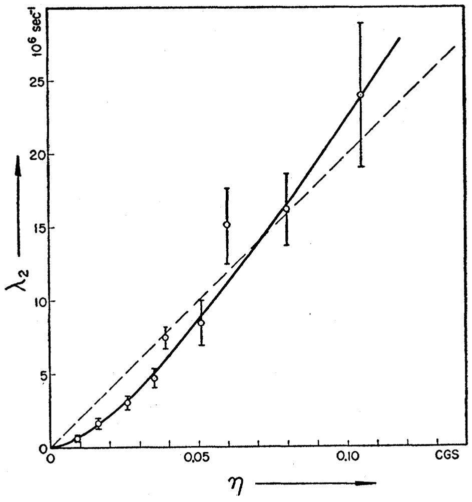

FIG. 9. The experimentally observed interaction parameters λ_2 as function of viscosity η. The dashed line represents the behavior of λ_2 according to Eq. (15).

(iv) A part of the attenuation effect might be due to after effects of the electron-capture decay which precedes the emission of the Cd^{111} gamma-gamma cascade. If, for a short time $\Delta t < \tau_c$, there is a coupling between the excited shell and the nucleus sufficiently strong to cause an appreciable influence on the correlation within Δt, then the condition for the applicability of Abragam and Pound's theory of time-dependent interactions is no longer satisfied. This is particularly true for viscous sources, where τ_c is large.

Since the interaction parameters λ_2 which are characteristic for the attenuation of the Cd^{111} directional correlation in the various viscous sources are known from the delay measurements, the true correlation of the Cd^{111} gamma rays can be computed from the un-delayed directional correlation displayed by these sources. The weighted average of the results obtained with nine sources of viscosities ranging from 0.009 cgs units to 0.11 cgs units suggest the following "true" directional correlation of the Cd^{111} gamma rays:

$$W^0(\Theta) = 1 - (0.181 \pm 0.003) P_2(\cos\Theta) \quad \text{(standard error).} \tag{17}$$

In these considerations, the $P_4(\cos\Theta)$ has been neglected entirely.

5. MEASUREMENTS ON LIQUID INDIUM METAL

5.1. Correlation without Delay

The possibility of electron-capture after-effects of the Cd atom can be reasonably excluded or, at least, can be greatly reduced by using metallic sources where

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[21] H. Albers-Schönberg and E. Heer, *Proceedings of the 1954 Glasgow Conference on Nuclear and Meson Physics* (Pergamon Press, London and New York, 1955).

[22] Albers-Schönberg, Heer, and Scherrer, Helv. Phys. Acta **27**, 637 (1954).

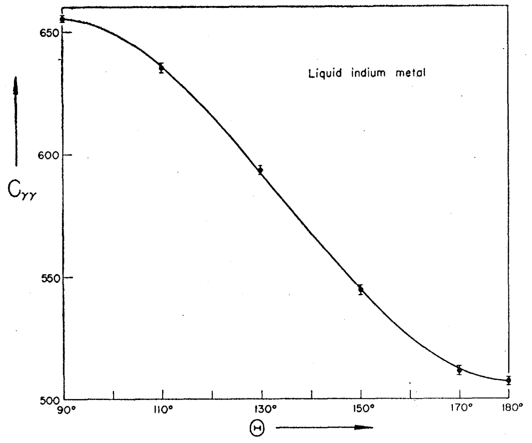

FIG. 10. The Cd^{111} directional correlation displayed by an In^{111} source in the form of liquid indium metal (temperature 225°C).

the recovery of the excited atom from preceding decay processes is fast enough to prevent an influence on the directional correlation.[23] Thus it seems desirable to study the time-dependent interaction (if any) in a liquid metal. In^{111} sources in the form of indium metal seemed to be most suited for experiments of this kind in view of the low melting point (157°C) of indium. Hence, the directional correlation as displayed by liquid indium metal sources was measured as a function of source temperature from 165°C to 695°C.

Figure 10 shows the result of a typical measurement at a source temperature of 225°C. The directional correlation displayed by sources of liquid indium metal is more pronounced than those exhibited by the solution sources, a fact which demonstrates again that the attenuation in the various solutions of indium salts, through very small, is not entirely negligible.[22]

The anisotropy for liquid indium metal sources as a function of temperature is presented in Fig. 11. The fact, that A is, within the limits of error, independent of the temperature, indicates that time-dependent interactions due to the Brownian motion in the liquid must be negligible in this source, since the correlation time τ_c changes by about a factor two over the temperature range considered. The latter statement follows from Eq. (8). It is also confirmed by measurements on the self-diffusion in liquid indium metal.[24]

Since the directional correlation exhibited by liquid indium sources seems to be unperturbed by extranuclear fields, a conclusion which is corroborated by the delay experiments discussed in the next section, the data obtained with these sources were used to arrive at an accurate representation of the Cd^{111} gamma-gamma correlation. A least square fit computation, this time including a $P_4(\cos\Theta)$ term, yields for the correlation function

$$W(\Theta)=1(-0.180\pm0.002)P_2(\cos\Theta)+(0.002\pm0.003)P_4(\cos\Theta). \quad (18)$$

5.2. Delayed Directional Correlation

In order to confirm that the function given above indeed corresponds to the undisturbed correlation, the delayed directional correlation displayed by In^{111} sources in the form of molten indium metal was measured as a function of the delay time T. The measurements were performed in the same fashion as for the aqueous solutions. The experimental value of $A_2(T)$ as a function of T is plotted in Fig. 12. The quadrupole interaction parameter, as computed from the curve of Fig. 12, is

$$\lambda_2<5\times10^4 \text{ sec}^{-1}, \quad (19)$$

or, in more significant terms,

$$\lambda_2\tau_N<0.006. \quad (20)$$

On the basis of this result the conclusion can be drawn that the directional correlation displayed by liquid indium metal sources is indeed, within our error limits, undisturbed by extranuclear fields and thus represents the true directional correlation of the Cd^{111} gamma rays. This absence of a disturbing interaction in the liquid indium metal is to be expected in view of the small correlation time in liquid indium metal [compare Eq. (10)]. From data on the self diffusion in liquid indium[24] τ_c can be estimated. At a temperature of 200°C, $\tau_c\cong10^{-12}$ sec.

6. UNPERTURBED DIRECTIONAL CORRELATION OF THE 172-KEV–247-KEV GAMMA CASCADE IN Cd^{111}

The determination of the "true" unperturbed directional correlation of a nuclear cascade which involves a lifetime of the intermediate state of 10^{-9} sec or longer requires special attention. In general, the

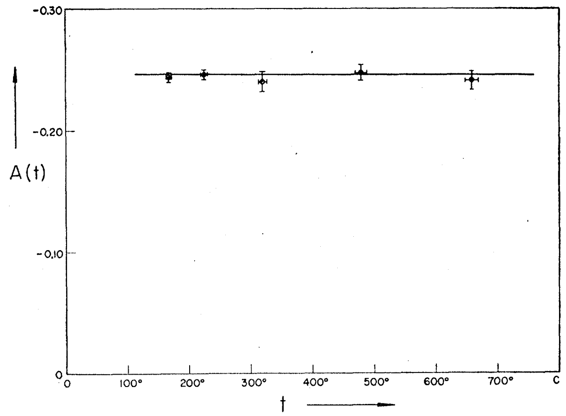

FIG. 11. The anisotropy $A=[W(180°)-W(90°)]/W(90°)$ of the Cd^{111} gamma correlation observed with liquid indium sources as function of temperature.

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[23] H. Frauenfelder, Phys. Rev. **82**, 549 (1951).

[24] G. Careri and A. Paoletti, Nuovo cimento **2**, 573 (1955).

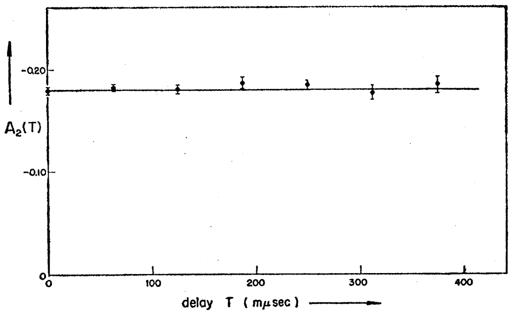

FIG. 12. The correlation coefficient A_2 as function of delay T, observed with a liquid indium metal source at a temperature of (210±30)°C.

directional correlation is attenuated due to the static or time-dependent magnetic and electric fields in the source and the interaction parameter must be well known and well controlled, before an extrapolation to the unperturbed correlation can be made. Under favorable circumstances, a strong magnetic field applied in the propagation direction of one of the nuclear radiations should cause a decoupling of the nuclear moments with the fields in the source and the unperturbed directional correlation should be displayed.[25] This effect, which is similar to the Back-Goudsmit effect (Paschen-Back effect of the hyperfine structure), requires strong magnetic fields and is applicable only if the interaction is mainly magnetic in character. Similarly, if the attenuation is due to a static (axially symmetric) quadrupole interaction with the crystalline fields of the source material, the unperturbed directional correlation may be observed by using as source a single crystal which can be orientated so that all electrostatic gradients acting on the radioactive nuclei are parallel.[1] In general no static quadrupole interaction should be expected in crystalline sources, whose crystal structure possesses cubic symmetry. The attenuating influence of extranuclear fields can, in theory at least, also be eliminated by observing the correlation by means of a coincidence analyzer which records the two cascade radiations within such a short resolving time τ_0, that the intermediate nuclear state is exposed to the external fields for a time too short to alter the directional correlation.[26] Again, in all these methods discussed, the after effects of decay processes, leading to the initial state of the casdcade, on the local environment of the nucleus in the intermediate state are difficult to consider in a quantitative way. This is particularly important if electron-capture or α decay is involved. In most cases the use of liquid metal sources might eliminate these atomic recovery effects, at least to some extent.

Probably the safest method to determine the true directional correlation is based upon the performance of adequate delayed correlation experiments from which the parameters which are characteristic for the interaction can be extracted. The undisturbed correlation can then be computed. However, not even this method is entirely free of objections. The possibility always remains that, due to a preceding decay process, a very strong interaction, large enough to affect the directional correlation, is present for a very short time Δt and then vanishes as the recovery process of the atom continues.

If this time interval Δt is considerably shorter than the resolving time τ_0 of the coincidence analyzer used in the delay measurements, the delayed directional correlation will not display any evidence of this effect and the directional correlation appears to be undisturbed. Furthermore, one would expect the electrons of an inner shell to be responsible for such a short-lived on-off interaction and thus to be independent of the local environment of an atom. Hence the attenuation effects probably would be the same for sources of quite different structure.

Dismissing the short-lived on-off interaction as very unlikely, we consider the correlation functions (17) and (18) extracted from the delay measurements on solutions and on liquid indium sources, respectively, as representative for the unperturbed directional correlation of the 172-kev−247-kev gamma cascade of In^{111}. This result agrees rather well with $W^0(\Theta)=1-(0.175\pm0.010)P_2(\cos\Theta)$ which was extracted from an analysis of correlation measurements on In^{111} sources of widely different structure (polycrystalline and single crystal sources).[1]

The directional correlation of the Cd^{111} gamma rays is characteristic for a cascade of a mixed $E2$-$M1$ transition with an $E2/M1$ intensity ratio of 0.021 ±0.002 ($\delta=+0.146$, in the notation of Biedenharn and Rose[3]), followed by a pure $E2$ transition involving states of angular momentum quantum number 7/2, 5/2, and 1/2.

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[25] G. Goertzel, Phys. Rev. **70**, 897 (1946).

[26] J. S. Fraser and J. C. Milton, Phys. Rev. **94**, 795 (1954).