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### Indications of the Interaction of Electric Field Gradients and Nuclear Electric Quadrupole Moments in Angular Correlation\*

ROLF M. STEFFEN

Department of Physics, Purdue University, Lafayette, Indiana

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THE influence of extranuclear fields on the angular correlation of successive nuclear radiation has been discussed by several authors.<sup>1</sup> It was generally assumed that the interaction between the nuclear magnetic dipole moment and magnetic fields, due to either the electron shell or the surrounding of the decaying atom, is mainly responsible for the reduction of the angular correlation. The dependence of this reduction on the source material was attributed to the difference in recovery time of the excited and ionized electron shell.

However, several directional correlations were reported recently, where the reduction of the anisotropy of the correlation could not be understood on the basis of the magnetic interaction alone, e.g.,  $\alpha$ - $\gamma$  correlation of  $\text{RdTh}^{228}$ ,<sup>2</sup>  $\gamma$ - $\gamma$  correlation of  $\text{Cd}^{111}$ ,<sup>3</sup> and of  $\text{Ta}^{181}$ .<sup>4</sup> Abragam and Pound<sup>5</sup> were able to show that the coupling between the nuclear electric quadrupole moment in the intermediate state and the  $\text{grad}E$  in axial crystals provides an explanation for the observed  $\alpha$ - $\gamma$  correlation of  $\text{RdTh}^{228}$ . It is well known that the  $\text{Cd}^{111}$   $\gamma$ -directional correlation depends strongly on the chemical state<sup>6</sup> and on the environment<sup>1</sup> of the decaying  $\text{In}^{111}$  atom. This was explained hitherto by magnetic interaction. However, magnetic decoupling experiments with a magnetic field of 7000 gauss applied in the direction of the propagation of one  $\gamma$ -ray showed no tendency to restore the maximum directional correlation (Table I). This field is probably too small for a complete decoupling of  $I$  and  $J$  (complete Paschen-Back effect of the hyperfine structure or Back-Goudsmit effect), but nevertheless it is large enough to cause a partial breaking of the  $(I, J)$  coupling (incomplete Back-Goudsmit effect). An increase in the anisotropy much larger than the experimental error of the measurements should have been observed if the magnetic coupling were the predominant cause for the attenuation of the correlation in the sources investigated. Similar results with even higher fields have been obtained by Heer *et al.*<sup>7</sup> This result seems to indicate that the attenuation of the  $\text{Cd}^{111}$  correlation in solid sources is due to the interaction between the crystalline  $\text{grad}E$  field and the electric quadrupole moment of the 0.247-Mev excited state of  $\text{Cd}^{111}$ , for which a rather large value of the quadrupole moment is expected from shell model considerations.

The largest anisotropy found for nonmetallic solids is exhibited by  $\text{In}_2\text{O}_3$  and  $\text{In}(\text{OH})_3$ . Both are cubic crystals with relatively small values of  $\text{grad}E$  at the position of the In, due to the high symmetry of their space groups:  $T_h$ <sup>7</sup> and  $T_h$ <sup>8</sup>, respectively. As a result of the preceding decay, however, these atoms may be displaced from the original lattice point and may be in regions of larger  $\text{grad}E$  values during a time comparable with the lifetime of the intermediate state.

TABLE I. Directional correlation of  $\text{Cd}^{111}$  with different sources and with application of a magnetic decoupling field.

Source	B gauss	Anisotropy: $A = \frac{W(\pi)}{W(\pi/2)} - 1$
Solid sources:		
$\text{InCl}_3$ , dry (20°C)	...	-0.012 $\pm$ 0.005
$\text{InCl}_3$ , dry (20°C)	7000	-0.015 $\pm$ 0.005
$\text{InCl}_3$ , dry (540°C)	...	-0.022 $\pm$ 0.006
$\text{InI}_3$ , dry (20°C)	...	-0.020 $\pm$ 0.006
$\text{InI}_3$ , dry (20°C)	7000	-0.019 $\pm$ 0.006
$\text{InI}_3$ , dry (200°C)	...	-0.021 $\pm$ 0.006
$\text{In}(\text{C}_6\text{H}_5\text{OH})_3$	...	-0.020 $\pm$ 0.006
$\text{In}_2\text{O}_3$	...	-0.045 $\pm$ 0.004
$\text{In}(\text{OH})_3$ , dry	...	-0.035 $\pm$ 0.006
$\text{In}(\text{OH})_3$ , dry	7000	-0.036 $\pm$ 0.007
In metal, electrodeposited (20°C)	...	-0.051 $\pm$ 0.005
In metal, electrodeposited (150°C)	...	-0.048 $\pm$ 0.006
In metal, electrodeposited (20°C)	7000	-0.056 $\pm$ 0.007
Liquid sources:		
$\text{InCl}_3$ , aqueous solution	...	-0.221 $\pm$ 0.005
$\text{InCl}_3$ , aqueous solution + $10^{21}$ $\text{Fe}^{+++}$ ions/cc	...	-0.215 $\pm$ 0.006
$\text{InI}_3$ , liquid salt (220°C)	...	-0.19 $\pm$ 0.02
$\text{In}(\text{C}_6\text{H}_5\text{ON})_3$ in $\text{CHCl}_3$ dissolved	...	-0.20 $\pm$ 0.01
In, liquid metal (180°C)	...	-0.20 $\pm$ 0.01

A further indication that the attenuation of the  $\text{Cd}^{111}$  correlation in the solid sources is caused by crystalline  $\text{grad}E$  fields stems from the fact that, without exception, liquid  $\text{In}^{111}$  sources of very different character give the maximum correlation (Table I). In the liquids used the character of the local configuration around a nucleus and thus the gradient of the electric field changes in a time much smaller than the lifetime of the intermediate nuclear state. Consequently the average perturbation on the nuclear  $m$  substates due to the quadrupole interaction is zero over the lifetime of the nuclear state.

During the progress of this work a convincing proof for the electric quadrupole interaction on the  $\text{Cd}^{111}$  correlation has been given by the beautiful experiments of the Swiss group<sup>7</sup> by using single crystals of indium as sources.

Attenuation effects very similar to those exhibited by the  $\text{Cd}^{111}$  correlation have recently been measured by McGowan<sup>4</sup> on the  $\text{Ta}^{181}$   $\gamma$ -cascade. Liquid sources of  $\text{Hf}^{181}$  also show a much more pronounced anisotropy than solid sources.<sup>4</sup> Moreover, there is evidence that the  $P_4(\cos\theta)$  term in the angular correlation function is less attenuated than is the  $P_2(\cos\theta)$  term, a result which is impossible with magnetic interaction alone.<sup>8</sup> Both effects, however, are explained easily if electric quadrupole interaction in the intermediate state of  $\text{Ta}^{181}$  is assumed. For this nucleus a very large electric quadrupole moment in the lower excited states is to be expected from shell model considerations, the ground state of  $\text{Ta}^{181}$  having one of the largest quadrupole moments known.

A more detailed account of this investigation will be submitted for publication in the near future.

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### Search for 1-Mev Gamma from $\text{N}^{16}$ Decay\*

F. BOEHM, D. C. PEASLEE, AND V. PEREZ-MENDEZ†

Columbia University, New York, New York

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IN the  $\beta$ -decay of  $\text{N}^{16}$  to the  $1^-$  (7.1-Mev) and  $3^-$  (6.1-Mev) excited states of  $\text{O}^{16}$ , respectively, a discrepancy exists between the branching ratio of about 1:1 determined from absorption measurements on the  $\beta$ -spectrum,<sup>1</sup> and the ratio of

about 1:12 from measurement of the associated  $\gamma$ -ray intensities.<sup>2</sup> If both measurements are to be reconciled, the most likely possibility is a strong  $E2$   $\gamma$ -ray transition from the 7.1-Mev to the 6.1-Mev level. The likelihood that this  $E2$  could give serious competition to the normal 7.1-Mev  $E1$  ground state transition is enhanced by the isotopic spin selection rule<sup>3</sup> forbidding  $E1$  transitions with  $\Delta T = N - Z = 0$ . In order to explain the quantitative discrepancy in this way, however, this selection rule would have to be much stronger<sup>4</sup> than suggested by simple estimates.

One check on this problem is to search for a strong 1-Mev  $\gamma$ -ray following  $N^{16}$  decay. The reaction  $O^{16}(n,p)N^{16}$  was used to produce  $N^{16}$  in the Columbia cyclotron, using fast neutrons from  $Be(d,n)$  on a water target. A flow system was installed to bring the water from a small chamber behind the Be cyclotron target to a measuring chamber. A 10-sec bombardment produced a very strong activity with the characteristic  $N^{16}$  half-life of 7 sec. After a waiting period of 5 sec to allow decay of any shorter activities, the  $\gamma$ -spectrum of  $N^{16}$  was measured with a scintillation counter consisting of a 1-inch NaI crystal. The pulse distribution from the crystal was photographed on a triggered oscilloscope.

A strong line was found in the 6-Mev region. In a weak exposure, a peak at 0.5 Mev is visible, owing to annihilation of positrons formed outside the crystal. Between these lines no  $\gamma$ -line can be seen. Assuming the scintillation to be caused by pair production for the 6-Mev line and photoelectric absorption for a hypothetical 1-Mev line, we find an upper intensity limit

$$I(1 \text{ Mev})/I(6 \text{ Mev}) \leq 0.05. \quad (1)$$

To account for the above-mentioned discrepancy in the branching ratios, the ratio (1) should be 10 times larger than observed, which is certainly ruled out by the present measurements. An error in the  $\beta$ -decay branching ratio seems the most likely explanation, in view of the inherent inaccuracy of absorption methods. It is of interest to note that if we assume approximate equality of the  $\beta$ -decay matrix elements, the branching ratio is determined by the ratio of  $(2J+1)f$ . In this case the ratio is

$$3f(3.2)/7f(4.2) \sim 1/7, \quad (2)$$

in order of magnitude agreement with the intensity ratio of 7-Mev to 6-Mev  $\gamma$ -rays.

This measurement does not provide any information on the failure of the isotopic spin rule forbidding electric dipole transitions. The result (1) implies that  $I(1 \text{ Mev})/I(7 \text{ Mev}) \leq 0.6$ . The ratio without restriction of the dipole transition is expected to be<sup>5-7</sup> of order  $10^{-6}$  or  $10^{-7}$ . Even if the isotopic spin selection rule reduces  $I(7 \text{ Mev})$  by a factor of  $10^3$ , an increase of sensitivity of at least  $10^3$  is expected to be necessary before the 1-Mev line could be detected.

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† Now at Project Whitney, Livermore, California.

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### Alpha-Particle Ionization in Pure Gases and the Average Energy to Make an Ion Pair

WILLIAM P. JESSE AND JOHN SADAUSKIS  
Argonne National Laboratory, Lemont, Illinois  
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FOR some time measurements have been in progress to determine the average energy required to make an ion pair by polonium alpha-particles in very carefully purified gases.

Since some of the values obtained are in marked contrast to those found in the literature, it is hoped that the publication of them will be of interest to others working in the field.

In the measurements a short collimating system directed the polonium alpha-particles along the axis of a long cylindrical ionization chamber. The effective path length was about 20 cm. The ions produced by each alpha-particle were collected and measured by a method already described.<sup>1,2</sup> From a knowledge of the capacity of the system and the change of potential of the system produced by the ionization from each alpha-particle, the absolute number of ion pairs produced may be determined. The average energy  $W$  to produce an ion pair is, of course, the energy of the polonium alpha-particle (5.298 Mev) divided by this number. A very small correction is made for the energy lost in the collimating system.

The capacity of the chamber was determined at intervals by comparison with a standard double-ended capacitance. During the progress of the work this capacitance was calibrated twice at the National Bureau of Standards with concordant results. The estimated error in the final values of  $W$ , of the order of one-half percent, includes, of course, a consideration of errors in such auxiliary measurements.

Since previous work<sup>2</sup> had shown the marked effect of minute gaseous impurities, the greatest precautions were taken to insure the purity of the gases used. These, in general, were introduced from breaker flasks into the chamber and were continuously circulated during the progress of the readings over a purification system by means of a small metal-bellows pump. The chamber was initially baked and pumped for more than twelve hours at a temperature above 200°C.

In Table I are shown the values of  $W$  obtained for the various gases used and for comparison very recent results of three other investigators. Two of these determinations were with polonium alpha-particles, and the third, that of Sharpe, was with  $Pu^{239}$ . Two values of  $W$  are given for each gas. First is given the absolute value of  $W$ , and then in parentheses is given the value relative to argon as a standard. In general, the agreement in the table is good, particularly between our own results and those of J. Sharpe.

In the case of helium there is a wide disagreement between the Argonne value and the other two values shown.<sup>3</sup> As has been discussed elsewhere,<sup>2</sup> this is no doubt due to impurities in the gas samples used, since in the other two determinations no extraordinary precautions seem to have been adopted to insure extreme gas purity. By reaction with the impurities the metastable atoms of helium are discharged with the production of additional ion pairs. For the same reason the Argonne value for neon is much higher than the older values found in the literature (about 29 ev/ion pair).

The agreement for argon in the table is good, particularly for three of the determinations. The effect of impurities is much less marked in argon, since the ionization potentials of the common

TABLE I. Values of  $W$ , in ev/ion pair. Values relative to argon are given in parentheses.

Gas	Argonne	Sharpe <sup>a</sup>	Valentine and Curran <sup>b</sup>	Haerberli, Huber, and Baldinger <sup>c</sup>
He	42.7 (1.617)		31.7 (1.22)	30.86 (1.176)
Ne	36.8 (1.394)			
A	26.4 (1.000)	26.3 (1.000)	25.9 (1.000)	26.25 (1.000)
Kr	24.1 (0.913)			
Xe	21.9 (0.830)			
H <sub>2</sub>	36.3 (1.375)		37.0 (1.43)	
CO <sub>2</sub>	34.5 (1.307)	34.2 (1.300)		33.5 (1.276)
Air	35.5 (1.345)	35.6 (1.354)	35.2 (1.36)	
O <sub>2</sub>	32.5 (1.231)	32.9 (1.251)	32.2 (1.24)	32.17 (1.226)
N <sub>2</sub>	36.6 (1.386)	36.4 (1.384)	36.0 (1.39)	36.3 (1.383)
CH <sub>4</sub>	29.2 (1.106)	29.1 (1.106)	29.0 (1.12)	
C <sub>2</sub> H <sub>2</sub>	27.5 (1.042)			
C <sub>2</sub> H <sub>6</sub>	26.6 (1.008)			
C <sub>3</sub> H <sub>4</sub>	28.0 (1.061)			

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