

mentum. Then  $\Delta K=3$ . In the present nuclides the spin sequence is  $1(\beta)2$ . The spin 2 may reasonably be interpreted as due to a collective excitation of the daughter<sup>20</sup> but the result is  $\Delta K=1$ ; hence  $K$  forbidden-

<sup>20</sup> A. Bohr and B. R. Mottelson, in *Beta- and Gamma-Ray Spectroscopy*, edited by K. Siegbahn (Interscience Publishers, Inc., New York, 1955).

ness does not seem to apply. The concept of “ $j$  forbiddenness” leads to fairly definite predictions only if the neutron undergoing decay and the proton into which it decays lie in the same major shell. For the present nuclides the neutron and proton lie in adjacent major shells, hence  $j$  forbiddenness does not yield a strong selection rule effect.

## Perturbation of Alpha Particle-Gamma Ray Angular Correlations in $\text{Am}^{241}$ , $\text{Am}^{243}$ , and $\text{Cm}^{243}\dagger$

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By utilization of the recoil properties associated with the alpha decay process, perturbations of alpha-gamma angular correlations in  $\text{Am}^{241}$ ,  $\text{Am}^{243}$ , and  $\text{Cm}^{243}$  were observed in a variety of media including vacuum, metals, Mylar, oxides and liquids. In the vacuum and Mylar environments the  $\text{Am}^{243}$  correlation is not only considerably more perturbed than in metallic environments but it exhibits an attenuation coefficient below the hard-core value for a static interaction. The perturbation in vacuum is attributed to a hyperfine structure interaction involving an electron shell excited in the alpha decay process. Several experiments designed to further explore the effect of alpha decay on the electron shell are suggested. It is pointed out that application of a strong magnetic field along the direction of one of the radiations may completely restore the correlation in vacuum. Improvement of the correlation in metals as compared to insulators may be related to the rapid recovery time of the electron shell in metallic media.

### INTRODUCTION

THE perturbation of alpha-gamma angular correlations has been reported in a number of even-even isotopes,<sup>1</sup> as well as in  $\text{Am}^{241}$ ,<sup>2,3</sup> and  $\text{Am}^{243}$ .<sup>4</sup> Interactions between the quadrupole moments of the intermediate nuclear states and electric field gradients of extra-atomic origin appeared responsible for attenuating the correlations. The present experiments were originally designed to eliminate these extra-atomic effects by observing angular correlations between alpha particles and the gamma rays from daughter nuclei recoiling into vacuum. After unexpectedly large perturbations were found to occur in vacuum, the experiments were extended to a variety of media including several whose influence on the angular correlation had not been previously explored.

### EXPERIMENTAL METHODS

The methods used to prepare and study thin  $\text{Am}^{241}$ ,  $\text{Am}^{243}$ , and  $\text{Cm}^{243}$  sources have been discussed in detail elsewhere.<sup>5</sup> Approximately 0.05  $\mu\text{g}$  of activity was freed from macroscopic impurities by repeated cation exchange and vaporized from a hot Ta filament onto an area  $\frac{7}{8}$  cm in diameter. The backing material consisted of thin metal or plastic films which were transparent to alpha particles but not to heavy recoils. The percentage of  $\text{Np}^{239}$  recoils escaping from the surface of the  $\text{Am}^{243}$  sources was determined by counting the beta activity collected over the source in vacuum. At least 45% of the recoils were found to escape into the vacuum; an equal percentage presumably were absorbed in the backing film, while less than 10% remained in the radioactive deposit.

The sources were centered in a cubical vacuum chamber at a 45° angle to the walls of the chamber. Gamma rays were detected by means of a fixed sodium iodide crystal which paralleled one face of the chamber and subtended a half-angle ranging from 11 to 19 deg at the source. Two zinc sulfide screens centered on adjacent faces of the chamber detected alpha particles at 90° and 180° to the observed gamma rays. Pulses from the two alpha detectors were successively counted and fed into a slow coincidence unit together with pulses from the

<sup>†</sup> Work done under the auspices of the U. S. Atomic Energy Commission.

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<sup>1</sup> J. Battey, L. Madansky, and F. Rasetti, *Phys. Rev.* **89**, 182 (1953); J. K. Beling, B. T. Feld, and I. Halpern, *ibid.* **84**, 155 (1951); J. C. D. Milton and J. S. Fraser, *ibid.* **95**, 628 (1954); G. M. Temmer and J. M. Wyckoff, *ibid.* **92**, 913 (1953); G. Valladas, J. Teillac, P. Falk-Vairant, and P. Benoist, *J. Phys. Radium* **16**, 123 (1955).

<sup>2</sup> J. S. Fraser and J. C. D. Milton, *Phys. Rev.* **94**, 795 (1954).

<sup>3</sup> V. E. Krohn, T. B. Novy, and S. Raboy, *Phys. Rev.* **105**, 234 (1957).

<sup>4</sup> F. Stephens, J. Hummel, F. Asaro, and I. Perlman, *Phys. Rev.* **98**, 261 (1955).

<sup>5</sup> E. Flamm, Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-9325, 1960 (unpublished).

gamma detector. The output of the coincidence unit gated a 50-channel pulse-height analyzer on which the gamma spectrum was displayed. After correction for accidental coincidences, the peaks of interest were integrated and normalized by the number of alpha particles gating the coincidence unit. The quantity thus determined was the anisotropy value,

$$A = [W(180^\circ)/W(90^\circ)] - 1,$$

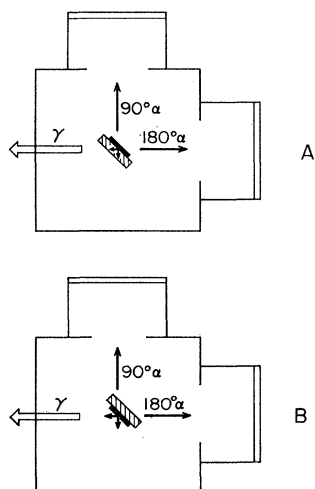
where  $W(\theta)$  is the relative probability that the propagation vectors of the alpha particle and the gamma ray form an angle  $\theta$ . The anisotropy value completely determines the angular correlation function for the cascades studied.

The frame on which the sources were mounted could be rotated through  $180^\circ$ , so that either the source or the backing plate faced the zinc sulfide screens. Gamma rays recorded in the coincidence spectrum are emitted by daughter nuclei recoiling in a direction opposite that of the observed alpha particles, i.e., into the backing plate if the source faces the zinc sulfide screens, into the vacuum or an absorber placed over the source if the backing plate faces the screens (Fig. 1). The recoil energies of the daughter nuclei correspond to initial velocities of nearly  $3 \times 10^7$  cm per sec and a range in heavy compounds of a few hundred atomic layers. Recoils escaping from the thin sources should therefore leave the radioactive deposit within  $10^{-13}$  sec of the alpha decay, an interval short compared with the lifetimes of the intermediate states in the alpha-gamma cascades. The angular correlations accordingly were expected to show perturbations characterizing the media into which the daughter nuclei recoil.

### RESULTS—Americium-243

A partial decay scheme<sup>6,7</sup> for  $\text{Am}^{243}$  is shown in Fig. 2. The angular correlation studied is that between the

FIG. 1. Diagram of vacuum chamber showing position of source (black) and backing plate (cross hatched) in relation to detectors. Long black arrows represent propagation vectors of detected alpha particles, white arrow represents propagation vector of detected gamma rays. Short black arrows indicate environment of recoil nuclei whose gamma rays contribute to the coincidence spectrum. A. Recoils into backing plate. B. Recoils into vacuum.



<sup>6</sup> F. Stephens, J. Hummel, F. Asaro, and I. Perlman, Phys. Rev. **98**, 261 (1955).

<sup>7</sup> John P. Unik, Ph.D. thesis, University of California Lawrence Radiation Laboratory Report UCRL-9105, 1960 (unpublished).

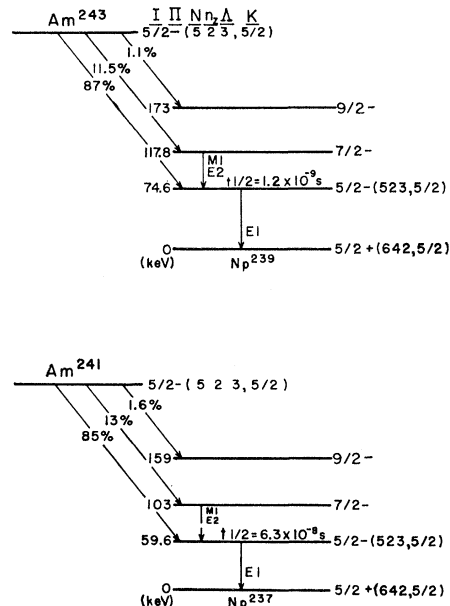


FIG. 2. Partial decay schemes of  $\text{Am}^{243}$  and  $\text{Am}^{241}$ . The quantum numbers  $N, n_z, \Lambda, K$  refer to Nilsson level assignments.

alpha particles populating the 75-keV state and the 75-keV gamma rays de-exciting this state. The unperturbed angular correlation can be calculated<sup>8</sup> if the multipole mixing ratios for the transitions are known. An estimate of the multipole mixing ratio for the alpha decay to the  $\frac{5}{2}-$  and  $\frac{7}{2}-$  states of  $\text{Np}^{239}$  may be made by the method described by Bohr, Fröman, and Mottelson.<sup>9</sup> These two states are members of a rotational band in which the odd nucleon occupies the same orbital as in the ground state of  $\text{Am}^{243}$ .<sup>10</sup> For alpha decay with no change in state of the odd nucleon, the multipole mixing ratio depends on geometrical factors involving only the multipole order and nuclear spins, and on hindrance factors for the various multipoles which may be estimated from the relative intensities of the alpha groups in neighboring even-even nuclei. Averaging the hindrance factors for the alpha decay of  $\text{Pu}^{242}$  and  $\text{Cm}^{244}$ ,<sup>11</sup> one obtains a value of  $\delta^2 = 0.22$ , for the ratio of  $L=2$  to  $L=0$  partial waves populating the  $\frac{5}{2}-$  level of  $\text{Np}^{239}$ . The calculated admixture of  $L=4$  is negligible. Previous alpha-gamma angular correlations have determined the sign of  $\delta$  to be positive.<sup>2-4</sup> The unperturbed correlation for the  $\frac{5}{2}- \rightarrow \frac{5}{2}- \rightarrow \frac{5}{2}-$  alpha-gamma cascade with an alpha mixing ratio of 0.22 and pure  $E1$  gamma radiation<sup>12</sup> is  $W(\theta) = 1 - 0.36P_2(\cos \theta)$ , and the corre-

<sup>8</sup> L. C. Biedenharn and M. E. Rose, Revs. Mod. Phys. **25**, 729 (1953).

<sup>9</sup> A. Bohr, R. P. Fröman, and B. R. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **1**, No. 8 (1959).

<sup>10</sup> B. R. Mottelson and S. G. Nilsson, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **1**, No. 8 (1959).

<sup>11</sup> Helen V. Michel, University of California Radiation Laboratory Report UCRL-9229, 1960 (unpublished).

<sup>12</sup> F. Asaro, F. S. Stephens, J. M. Hollander, and I. Perlman, Phys. Rev. **117**, 492 (1960).

TABLE I. Am<sup>243</sup>  $\alpha$ - $\gamma$  angular correlation results.

Experiment number	Source <sup>a</sup> position	Environment of daughter nucleus	Anisotropy value	$A_2$	$G_2$
1	A	oxide	$-0.20 \pm 0.01$	-0.14	0.39
2	A	aluminum	$-0.26 \pm 0.01$	-0.19	0.53
3	A	silver	$-0.28 \pm 0.01$	-0.21	0.58
4	A	iron	$-0.17 \pm 0.01$	-0.12	0.34
5	A	Mylar	$-0.04 \pm 0.02$	-0.03	0.08
6	B	vacuum	$-0.06 \pm 0.01$	-0.04	0.11
7	B	vacuum+metal	$-0.07 \pm 0.01$	-0.05	0.14
8	B	vaporized silver	$-0.25 \pm 0.02$	-0.18	0.50
9	B	air	$-0.10 \pm 0.02$	-0.07	0.19

<sup>a</sup> With the source in position A, the observed correlation corresponds to the daughter nuclei recoiling into the backing plate. With the source in position B, the observed correlation corresponds to the daughter nuclei recoiling away from the backing plate.

sponding anisotropy value is  $-0.46$ . Recent theoretical studies of the alpha decay of deformed nuclei suggest that a mixing ratio about 40% larger may be more accurate,<sup>13</sup> in which event the anisotropy value would decrease to  $-0.51$ .

The angular correlation observed in the experiments is a weighted average for the  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  double cascade and the  $\frac{5}{2} \rightarrow \frac{7}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  triple cascade with intermediate radiation unobserved. A relatively strong perturbation of the triple correlation may be expected owing to excitation of the electron shell associated with conversion of the unobserved radiation. The contribution of the triple cascade was, therefore, assumed to be isotropic.

A summary of the results obtained in Am<sup>243</sup> is given in Table I. Column 2 refers to Figs. 1A and 1B: Sources in position A face the alpha detectors: those in position B face the gamma detector. The entries in column 3 refer to the medium into which the daughter nuclei recoil. The attenuation factors  $G_2$  in the last column give the ratio between the coefficient of  $P_2$  calculated from the observed anisotropy and the unperturbed correlation coefficient of  $-0.36$  for an alpha mixing ratio of 0.22. Corrections have been made for the solid angles subtended by the detectors and for the contribution of the triple cascade.

Experiment 1 refers to a macroscopically impure sample considerably thicker than the recoil range. The remaining experiments were done in thin sources vaporized onto thin backing plates. Larger anisotropies were obtained when the recoils stopped in silver or aluminum than in iron or in the thick oxide sample. Mylar, a polyester of terephthalic acid and ethylene glycol, yielded almost no anisotropy.

A striking effect was the marked perturbation of the correlation in nuclei recoiling into vacuum (Table I, experiment 6). The correlation failed to improve when a silver or aluminum absorber was placed in loose contact with the source, although the recoils should have reached the metal in less than  $5 \times 10^{-10}$  sec (ex-

periment 7). When the silver absorber was vaporized onto the source, the anisotropies obtained in position B were comparable to those in position A (experiment 8). This experiment demonstrated that experimental errors related to the passage of alpha particles through the backing plates, such as scattering or loss of pulse height, were not responsible for the apparent attenuation of the correlation in vacuum. The dependence of the correlation on the medium receiving the recoils was most clearly seen in a source deposited on Mylar and covered with vaporized silver: This americium "sandwich" showed an anisotropy six times greater in position B than in position A. Variable anisotropies, ranging from  $-0.04 \pm 0.04$  to  $-0.17 \pm 0.04$ , were obtained when the daughter nuclei recoiled into air (experiment 9).

As first proposed by Goertzel,<sup>14</sup> angular correlations perturbed by interactions between the nucleus and atomic electrons in isolated ions may be restored by application of a strong magnetic field along the direction of one of the observed radiations. A decoupling experiment of this type was attempted in Am<sup>243</sup>. With the recoils escaping into a vacuum, a magnetic field of 7500 G was applied along the axis from the source to the gamma ray detector. Variations of the anisotropy value several times greater than the standard deviations were obtained, apparently owing to a defective arrangement for shielding the photo-multiplier tubes from the magnetic field. Although the results of the decoupling experiment are not reliable quantitatively, they indicated that an applied field of 7500 G is sufficient to restore part, but not all, of the Am<sup>243</sup> correlation.

### Americium—241

A partial decay scheme<sup>15,16</sup> for Am<sup>241</sup> is shown in Fig. 2. The pattern of the daughter energy levels and their alpha populations is very similar to that observed in Am<sup>243</sup> decay.

The calculated correlation for the  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  alpha-gamma cascade through the 63  $\mu$ sec state at 60 keV in Np<sup>237</sup> is approximately the same as that for the corresponding cascade in Am<sup>243</sup>. As shown in Table II,

TABLE II. Am<sup>241</sup>  $\alpha$ - $\gamma$  angular correlation results.

Experiment number	Source <sup>a</sup> position	Environment of daughter nucleus	Anisotropy value	$A_2$	$G_2$
1	A	aluminum	$-0.03 \pm 0.01$	-0.02	0.06
2	A	silver	$-0.04 \pm 0.01$	-0.03	0.08
3	A	Mylar	$-0.02 \pm 0.01$	-0.01	0.03
4	B	air	$-0.02 \pm 0.02$	-0.01	0.03
5	B	water	$0.00 \pm 0.03$	0.00	0.00
6	B	benzene	$-0.06 \pm 0.01$	-0.04	0.11

<sup>a</sup> With the source in position A, the observed correlation corresponds to the daughter nuclei recoiling into the backing plate. With the source in position B, the observed correlation corresponds to the daughter nuclei recoiling away from the backing plate.

<sup>14</sup> G. Goertzel, Phys. Rev. **70**, 897 (1946).

<sup>15</sup> J. K. Beling, T. O. Newton, and B. Rose, Phys. Rev. **87**, 670 (1952).

<sup>16</sup> F. Asaro and I. Perlman, Phys. Rev. **93**, 1423 (1953).

<sup>13</sup> R. R. Chasman and J. O. Rasmussen, Phys. Rev. **115**, 1260 (1959).

little or no anisotropy was obtained for the 60-keV gamma ray in any of the absorbers studied. Application of a magnetic field of 7500 G along the direction of the observed gamma rays failed to improve the correlation in Mylar.

**Curium—243**

A partial decay scheme<sup>17,18</sup> for Cm<sup>243</sup> is shown in Fig. 3. Four radiations from the 286-keV level of Pu<sup>239</sup> were observed in coincidence with Cm<sup>243</sup> alpha particles (see Fig. 4): Plutonium *K* x rays, the unresolved 210- and 228-keV gamma rays populating the  $\frac{7}{2}+$  and  $\frac{5}{2}+$  members of the ground-state rotational band, and the 278-keV gamma ray populating the  $\frac{3}{2}+$  level. The intensity ratio of the 210-, 228-, and 278-keV gamma rays is 0.32:1.0:1.1, and all three radiations are *M1* with less than 5% admixture of *E2*.<sup>17,18</sup> The  $L=2$  to  $L=0$  mixing ratio for the alpha waves populating the 286-keV state, estimated from hindrance factors<sup>11</sup> for the decay of Cm<sup>242</sup> and Cm<sup>244</sup>, is 0.20. The theoretical anisotropy values for the alpha-gamma cascades with an alpha mixing ratio of 0.20 and gamma mixing ratios up to the limit of 5% *E2* were calculated from the equations given by Biedenharn and Rose.<sup>8</sup> The calculated anisotropy values for the unresolved  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{7}{2}$  and  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  cascades with an intensity ratio of 1:3 range from +0.02 to -0.50, depending on the magnitudes and signs of  $\delta$  for the 210- and 228-keV gamma rays. The calculated anisotropy values for the  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{3}{2}$  cascade are positive for all gamma mixing ratios below 0.038 *E2*. As angular correlations involving *K* x rays are always isotropic, the *K* x-rays peak in the coincidence spectra serves as an internal check against experimental errors.

Table III presents a summary of the experiments on Cm<sup>243</sup>. Corrections have again been made for solid angle and for the presumably isotropic contribution of the triple cascade. The attenuation coefficients in the last column were calculated from the observed anisotropies

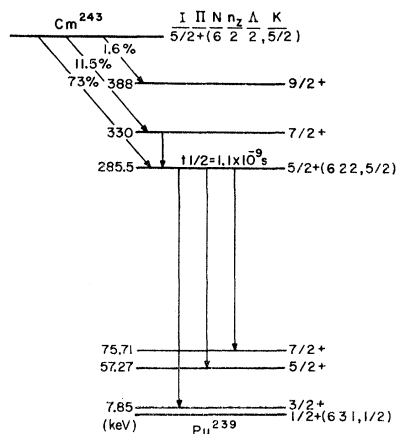


FIG. 3. Partial decay scheme of Cm<sup>243</sup> with Nilsson level assignments.

<sup>17</sup> G. T. Ewan, J. W. Knowles, and D. R. MacKenzie, Phys. Rev. **108**, 1308 (1957).

<sup>18</sup> G. T. Ewan, S. J. Geiger, R. L. Graham, and D. R. MacKenzie, Phys. Rev. **116**, 950 (1959).

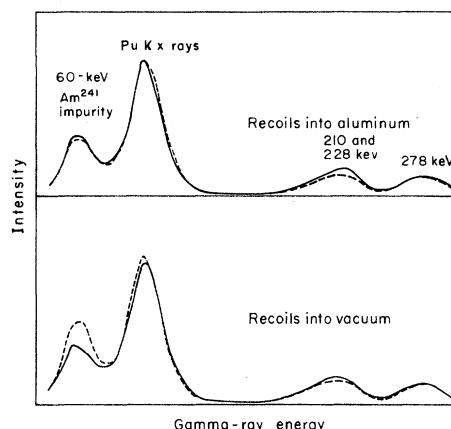


FIG. 4. Gamma ray coincidence spectra from Cm<sup>243</sup> source. Solid curve represents  $W(90^\circ)$ , broken curve  $W(180^\circ)$ .

of the combined 210- and 228-keV gamma rays. A range of  $G_2$  values is given for each experiment, corresponding to the range of unperturbed anisotropies consistent with gamma mixing ratios up to 5% *E2*. The upper limits of  $G_2$  are determined by the results of experiment 2, which establish that the unperturbed anisotropy for the combined  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{7}{2}$  and  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  cascades is at least -0.24. It should be noted that the attenuation coefficients apply to the  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{3}{2}$  correlation as well as the  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{5}{2}$  and  $\frac{5}{2} \rightarrow \frac{5}{2} \rightarrow \frac{7}{2}$  correlations since the intermediate state is the same in all three cascades.

Thick Cm<sub>2</sub>O<sub>3</sub> sources failed to yield a significant anisotropy. This is in contrast to the corresponding experiment on Am<sup>243</sup>, which showed a relatively large anisotropy in thick oxide sources. Thin Cm<sup>243</sup> sources, on the other hand, yielded negative anisotropies which varied with the environment of the daughter nuclei in the same manner as in Am<sup>243</sup>. The anisotropy of the combined 210- and 228-keV gamma rays was larger when the daughter nuclei recoiled into metals than when they recoiled into vacuum. A marked improvement of the correlation occurred when the recoils were prevented from escaping into vacuum by a layer of metal vaporized over the source. Variations of the anisotropy several times greater than the standard deviations were observed when the recoils escaped into air. Of the three Pu<sup>239</sup> peaks in the coincidence spectra, only the 210-228 keV peak showed these variations. The *K* x rays were consistently isotropic, and the small negative anisotropy of the 278-keV gamma ray varied within the relatively large statistical error throughout the experiments.

The apparent positive anisotropy of the 60-keV gamma ray of Np<sup>237</sup> seen in Fig. 4 represents an experimental error attributable to the long lifetime of the excited state. As indicated in Fig. 1B, the gamma rays recorded in the 180° coincidence spectrum come from recoils traveling toward the gamma detector, whereas the recoils contributing to the 90° spectrum travel on the average parallel to the gamma detector. Most of

TABLE III. Cm<sup>243</sup>  $\alpha$ - $\gamma$  angular correlation results.

Experiment number	Source position <sup>a</sup>	Environment of daughter nucleus	210-228 keV gamma anisotropy	$A_2$	278-keV gamma anisotropy	$A_2$	Approximate $G_2$ range
1	A	oxide	-0.01±0.03	-0.01	-0.1 ±0.02	-0.01	0.02-0.06
2	A	aluminum	-0.24±0.02	-0.17	-0.04±0.02	-0.03	0.42-1.0
3	A	silver	-0.19±0.02	-0.14	-0.01±0.02	-0.01	0.34-0.82
4	B	vacuum	-0.09±0.01	-0.06	-0.01±0.01	-0.01	0.15-0.35
5	B	gold	-0.19±0.02	-0.14	-0.05±0.04	-0.04	0.34-0.82
6	B	air	-0.15±0.02	-0.11	-0.04±0.02	-0.03	0.27-0.65

<sup>a</sup> With the source in position A, the observed correlation corresponds to the daughter nuclei recoiling into the backing plate. With the source in position B, the observed correlation corresponds to the daughter nuclei recoiling away from the backing plate.

the recoils escaping from thin Am<sup>241</sup> sources travel more than a centimeter in vacuum before emitting the 60-keV gamma ray. The solid angle subtended at the recoils by the gamma detector is, therefore, significantly larger when  $W(180^\circ)$  is recorded and the anisotropy value,  $[W(180^\circ)/W(90^\circ)]-1$ , appears to be positive. Solid angle errors of this type in the Cm<sup>243</sup> correlations would be readily detected as false anisotropies of the  $K$  x rays. No such error occurs in the Am<sup>243</sup> or Cm<sup>243</sup> correlations because the distance traveled by the recoils during the lifetime of the intermediate state is negligible compared to the source-to-detector distance.

The small negative anisotropy of the 278-keV gamma ray of Pu<sup>239</sup> is compatible only with  $E2$  admixtures of from 3 to 6%. These limits are consistent with those obtained from internal conversion coefficients.<sup>17</sup> The sign of  $\delta$  for the 278-keV gamma ray is positive according to the conventions of Biedenharn and Rose.<sup>8</sup>

### DISCUSSION

That perturbation of the angular correlation occurs in vacuum is not in itself surprising. In fact, attention was called to this possibility in Hamilton's classic paper setting forth the fundamental theory of the directional distribution of successive gamma rays.<sup>19</sup> Hamilton pointed out that an interaction between the nuclear magnetic moment and the magnetic fields arising in the electron shell of an isolated atom would perturb the correlation if the splitting of the hyperfine structure was comparable to the radiation width of the intermediate state. This conclusion was later confirmed by Goertzel,<sup>14</sup> in the first quantitative theoretical study of perturbed correlations, and again by Alder,<sup>20</sup> who reformulated the theory to obtain expressions for the attenuation coefficients. In view of the fundamental role played by free atoms in the development of the theory of perturbed angular correlations, it is of particular interest to consider whether interactions involving only the nucleus and the electron shell could account for the results of the present experiments.

The general condition<sup>14</sup> for a magnetic perturbation of the angular correlation is  $2\pi\Delta\nu\tau_N \gg 1$ , where  $\Delta\nu$  is the

over-all width of the hyperfine structure and  $\tau_N$  is the mean life of the intermediate state,  $1.7 \times 10^{-9}$  sec for the Am<sup>243</sup> cascade. A hyperfine structure splitting of the order of  $10^{-3}$  cm<sup>-1</sup> is, therefore, sufficient to result in perturbation of the Am<sup>243</sup> correlation. The magnetic moment of the ground state of Am<sup>243</sup> is 1.4 nm.<sup>21</sup> As the orbital of the odd proton does not change in the alpha decay, the intermediate level of the Am<sup>243</sup> cascade should also have a magnetic moment of approximately 1.4 nm. Prior to alpha particle emission, the electron shell consists of a closed radon core and five or six 5f electrons. Two processes associated with the alpha decay may alter this electron configuration: First, ionization may result from the sudden change in nuclear charge ("shakeoff effect")<sup>22</sup>; and second, inelastic collisions may occur during the recoil motion.<sup>23</sup> As discussed later, these processes may create holes in the radon core or they may result in loss or excitation of one or more 5f electrons. The overall width of the hyperfine structure in an ion with a magnetic moment of 1.4 nm, a closed radon core, and from two to five 5f electrons coupled according to Hund's rule is of the order of 0.1 cm<sup>-1</sup> for the most probable values of the total electronic angular momentum  $J$ .<sup>5</sup> Considerably wider hyperfine structure splittings would be expected if unpaired electrons were also present in the radon core or in the 7s shell. The condition for a magnetic perturbation in a free ion thus appears to be satisfied for the intermediate state of the Am<sup>243</sup> cascade for most of the electron configurations one might reasonably expect. The remarkable result of the experiments is not that the correlation was perturbed in vacuum, but that the attenuation coefficient for the Am<sup>243</sup> cascade was well below the "hard core" value of 0.20 for a static interaction.<sup>20</sup> According to present theory,<sup>24</sup> only a magnetic field or an electric field gradient which changes direction at the nucleus during the lifetime of the intermediate state can account for the observed attenuation coefficient of 0.11. Three processes which may give rise to such fluctuating fields will be considered:

<sup>21</sup> T. E. Manning, M. Fred, and F. S. Tomkins, Phys. Rev. **102**, 1108 (1956).

<sup>22</sup> A. Migdal, J. Phys. (U.S.S.R.) **4**, 449 (1941).

<sup>23</sup> N. Bohr, Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd. **18**, No. 8 (1948).

<sup>24</sup> A. Abragam and R. V. Pound, Phys. Rev. **92**, 943 (1953).

<sup>19</sup> D. R. Hamilton, Phys. Rev. **58**, 122 (1940).

<sup>20</sup> K. Alder, Helv. Phys. Acta **25**, 235 (1952).

(1) Partial reorientation of the intermediate state spins may occur before the recoils reach the surface of the radioactive deposit. A perturbation within the deposit could not account for attenuation coefficients lower than 0.5, the value obtained in metals, as the daughter nuclei traveled through the deposit whether they recoiled into vacuum or into metallic backing plates. Moreover, a comparison of the experiments in  $\text{Am}^{241}$  and  $\text{Am}^{243}$  indicates that the maximum attenuation coefficient of 0.5 represents at least partially perturbations occurring after the recoils leave the deposit. The electron shells and the extra-atomic environments of the  $\text{Np}^{237}$  and  $\text{Np}^{239}$  daughter nuclei should be similar when they recoil from comparable sources into the same absorber. Since the magnetic moments and quadrupole moments of the two intermediate states also are similar, an interaction occurring within the radioactive deposit would be expected to attenuate both correlations equally. The difference between the attenuation coefficients for the two isotopes recoiling into metallic environments thus appears to represent an interaction taking place in the metal, where the time available for spin reorientation is much greater in  $\text{Np}^{237}$  than in  $\text{Np}^{239}$  owing to the difference in half-life of the excited states. This interaction must have a decay constant of at least 100 per  $\mu\text{sec}$  to account for the almost complete destruction of the  $\text{Am}^{241}$  correlation, in which event it would also significantly attenuate the  $\text{Am}^{243}$  correlation. The experiments do not, however, exclude the possibility that small perturbations occur before the recoils reach the metal.

(2) A perturbation may occur at the boundary between the radioactive deposit and the vacuum. Large electric field gradients are encountered by the recoil nucleus as it traverses the region of decreasing field between the ionic deposit and a nonpolar medium. From simple estimates, however, it appears that the surface field gradient acts on the nucleus over too brief a period to affect the correlation. For a work function of 5 eV, a field falling to zero over atomic dimensions, and a nuclear quadrupole moment of 4 b,<sup>5</sup> the interaction frequency would be roughly 50 Mc/sec. At a velocity of  $10^7$  cm/sec, the recoils traverse the region of decreasing field in  $10^{-15}$  sec. The condition for an electric perturbation is  $\omega\tau \gg 1$ , where  $\omega$  is the interaction frequency and  $\tau$  is the length of time spent in the region of decreasing field. This condition is far from fulfilled and spin reorientation would not be expected.

(3) The electron shells of the recoil ions may undergo rapid transitions such as those associated with the Auger process following  $K$  capture or  $K$  or  $L$  shell conversion.<sup>25,26</sup> It should be noted that such a process would

account not only for the time-dependent character of the perturbation in vacuum but also for the improvement of the  $\text{Am}^{243}$  and  $\text{Cm}^{243}$  correlations in metals as compared with vacuum and nonmetallic crystals. The recovery time of the electron shell in metals is less than  $10^{-12}$  sec.<sup>27</sup> Perturbing fields associated with holes in the electron shell are, therefore, rapidly eliminated in a metallic environment. Conversely, recovery proceeds slowly in a plastic insulator such as Mylar, the environment which proved most unfavorable for the  $\text{Am}^{243}$  correlation. A similar relation between the attenuation coefficient and the conductivity of the environment of the intermediate state has been observed in the  $L$  conversion electron-gamma cascade in  $\text{Hg}^{197m}$ .<sup>28</sup> Partial restoration of the electron-gamma correlation in Mylar was achieved by application of a magnetic decoupling field, thus confirming the intra-atomic nature of the interaction.

Certain characteristics of the perturbing interaction in metallic environments may be deduced from a comparison of experiments 2 and 3 in  $\text{Am}^{243}$  (Table I) with experiments 1 and 2 in  $\text{Am}^{241}$  (Table II). The interaction in silver and aluminum must be time dependent, as the attenuation coefficient for the  $\text{Am}^{241}$  correlation is below the hard core. It must persist for longer than  $10^{-9}$  sec otherwise the attenuation of the  $\text{Am}^{241}$  and  $\text{Am}^{243}$  correlations would be the same. The time dependence of the interaction then cannot be related to de-excitation of the electron shell or to the recoil motion, both of which would be completed within  $10^{-11}$  sec of the alpha decay. Sources of a time-dependent interaction in silver and aluminum might be extra-atomic electric field gradients arising during readjustment of the lattice after penetration by the recoil nucleus or intra-atomic fields fluctuating with the rapid paramagnetic spin relaxation time of  $5f$  electrons.<sup>28</sup> The fact that stronger perturbations were observed in iron (Table I, experiment 4) than in silver or aluminum presumably represents a ferromagnetic effect.

Our interpretation of the present experiments may be summarized as follows: All the observed interactions are time dependent; that is, the direction of the perturbing field or field gradient at the nucleus changes within a period of the order of  $10^{-9}$  sec following the alpha decay. The perturbations in vacuum and at least part of the perturbations in insulators may be accounted for by interactions between the nucleus and an electron shell disturbed in the alpha decay process. The nature of the disturbance in the electron shell is not at all clear from present evidence. Of the two mechanisms mentioned earlier—inelastic collisions and shakeoff effect—neither is expected to produce ionization involving the innermost electron shells. The ionization probability

<sup>25</sup> H. Aeppli, A. S. Bishop, H. Frauenfelder, M. Walter, and W. Zunti, *Phys. Rev.* **82**, 550 (1951); H. H. Coburn and S. Frankel, *ibid.* **99**, 671 (1955); H. Frauenfelder, M. Walter, and W. Zunti, *ibid.* **77**, 557 (1950).

<sup>26</sup> B. G. Petterson, J. E. Thun, and T. R. Gerholm, *Nuclear Phys.* **24**, 223 (1961).

<sup>27</sup> S. Devons and L. J. B. Goldfarb, in *Encyclopedia of Physics*, edited by S. Flügge (Springer-Verlag, Berlin, 1957), Vol. 42, p. 517.

<sup>28</sup> W. Low, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press Inc., New York, 1960), Suppl. 2, p. 152.

associated with the change in nuclear charge has been shown experimentally to be less than 0.01% for the  $K$ ,  $L$ , and  $M$  shells of  $\text{Po}^{210}$ ,<sup>29</sup> in agreement with the results of calculations employing hydrogen-like wave functions.<sup>22</sup> For the outer shells of heavy atoms, neither experimental data nor suitable wave functions upon which to base calculations of the ionization probability are available. From semiclassical considerations, ionization of the neptunium and plutonium recoils would not occur at all in simple collision processes, as the orbital velocity of even the outermost electrons exceeds the recoil velocity.<sup>23</sup> Dense ionization nevertheless has been observed along the paths of alpha decay recoils,<sup>30</sup> but presumably this is attributable to inelastic processes involving only loosely bound electrons.

The magnitude of the perturbations observed in vacuum indicates that partially filled electron shells other than the  $5f$  shell are involved. Two experiments designed to determine if the radon core is disrupted in the alpha decay process may be suggested: (1) A determination of the charge on radon recoils from the alpha decay of radium isotopes; (2) alpha-gamma angular correlations in radium isotopes with the radon recoils escaping into vacuum. No perturbation of the radium correlations would be expected if the radon electron core remains intact in alpha decay.

A third experiment clearly called for by the results of the present study is an accurate determination of alpha-gamma angular correlations with the recoils escaping into vacuum as a function of the strength of a magnetic decoupling field. Two types of information may be derived from such an experiment. First, at sufficiently large applied fields, the correlation should be completely restored. The unperturbed correlation provides information on nuclear spins and on multipole

orders and multipole mixing ratios of the alpha particles and gamma rays. Second, information concerning the hyperfine structure may be extracted from an analysis of the variation of the correlation with applied field at intermediate field strengths.<sup>14</sup> If the state of the electron shell following alpha decay can be determined by other methods, the magnetic moment of the excited state can be deduced from the hyperfine structure. Conversely, if the magnetic moment of the excited state is known, the hyperfine structure provides information concerning the effect of alpha decay on the electron shell. It should be noted that Goertzel's treatment of the effect of a decoupling field on the perturbed correlation applies only if the electron shell is in a stationary state throughout the cascade. An extension of the theory to include electronic transitions accompanying the nuclear transitions is, therefore, required for an analysis of the variations of alpha-gamma correlations with applied magnetic field strength.

Finally, attention should be called to the possibility of studying radiation damage by means of alpha-gamma angular correlations. The attenuation of the gamma-gamma angular correlation in  $\text{Cd}^{111}$  produced by alpha-particle bombardment of  $\text{Ag}^{109}$  has recently been used to estimate the lattice vacancy concentration near the site of the product nuclei.<sup>31</sup> In the more complicated case of alpha-gamma angular correlations, attenuation may result from electromagnetic fields at the recoil nucleus during its flight as well as after it comes to rest. On the one hand, this renders the interpretation of the correlation more difficult. On the other hand, it suggests that such experiments might provide a means of exploring transient and highly localized effects associated with the passage of charged particles through matter.

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<sup>29</sup> M. Riou, *J. Phys. Radium* **16**, 583 (1955); W. C. Barber and R. H. Helm, *Phys. Rev.* **86**, 275 (1952); W. Rubinson and W. Bernstein, *ibid.* **86**, 545 (1952).

<sup>30</sup> B. S. Madsen, *Kgl. Danske Videnskab. Selskab, Mat.-Fys. Medd.* **23**, No. 8 (1945).

<sup>31</sup> G. W. Hinman, G. R. Hoy, and J. K. Lees, *Bull. Am. Phys. Soc.* **7**, 344 (1962).