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Flamm, Eileen.

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PERTURBATION OF ALPHA-GAMMA ANGULAR CORRELATIONS
IN TRANSURANIUM ISOTOPES

Eileen Flamm
(Ph. D. Thesis)
August 1960
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PERTURBATION OF ALPHA-GAMMA ANGULAR CORRELATIONS IN TRANSURANIUM ISOTOPES

Eileen Flamm

Lawrence Radiation Laboratory and Department of Chemistry
University of California, Berkeley, California
August 1960

ABSTRACT

The perturbation of alpha-gamma angular correlations has been studied in Am$^{241}$, Am$^{243}$ and Cm$^{243}$ by observing alpha particles in coincidence with gamma rays from daughter nuclei recoiling into vacuum, air, aluminum, silver, iron, Mylar, water and benzene. Attenuation coefficients below the hard core value for a static interaction were obtained in vacuum, indicating that fluctuating magnetic fields or electric field gradients are present at the daughter nuclei during the recoil motion. Significantly larger anisotropies were obtained in nonmagnetic metals than in iron or in insulators. The magnetic and electric moments of the intermediate nuclear states in the alpha-gamma cascades are estimated and the configuration of the electron shell following alpha decay is considered. Extra-atomic electric fields at the daughter nuclei during and after the recoil motion are discussed with reference to current theories of radiation damage and the chemical behavior of the actinides. The results and interpretations of previous alpha-gamma angular correlations are reviewed. From the observed correlations in Cm$^{243}$, a lower limit can be set for the E2/M1 mixing ratio $\delta^2$ of the 278 kilovolt gamma ray in Pu$^{239}$ and the sign of $\delta$ is determined.
INTRODUCTION

Perturbation of the angular correlation by extranuclear fields was first discussed in 1940 in Hamilton's classic paper formulating the theory of the directional distribution of successive gamma rays. As described in this paper, the link between the directions of the two radiations is the orientation of the nuclear spin in the intermediate state of the cascade. Hamilton pointed out that reorientation of the spin may occur before emission of the second radiation if the nuclear magnetic moment precesses about a strong magnetic field arising in the electron shell. This suggestion was developed in 1946 by Goertzel, who generalized the correlation function to include coupling between the nuclear spins and atomic electrons. From a comparison of the correlation functions obtained by Hamilton and Goertzel, it appeared that attenuation of the correlation would occur if the hyperfine structure was sufficiently wide, but a "hard core" anisotropy should remain regardless of the interaction energy. Alder in 1952, using group theoretical methods, further showed that the perturbation resulting from the action of a static field could be expressed in the Legendre polynomial expansion of the correlation function by attenuation coefficients depending only on the lifetime of the intermediate state, the interaction energy, and the angular momenta of the nucleus and electron shell. Despite these theoretical advances, attempts to explain the attenuation of alpha-gamma angular correlations proved unsuccessful until 1953, when Abragam and Pound extended the work of Goertzel and Alder to include time-dependent as well as static fields and electric as well as magnetic interactions between the nucleus and its environment. Electric fields at the recoil nucleus were shown to account for the perturbations observed in crystalline sources of Ra, Th, and Pu. More recently, delayed
correlations in dilute acid solutions of Am$^{241}$ were found to exhibit the exponential time dependence characteristic of electric interactions in nonviscous polar liquids.

I. ANGULAR CORRELATION EXPERIMENTS

Unfortunately, the interpretation of the perturbations observed in alpha-gamma angular correlations is complicated by changes in the electron shell connected with the alpha-decay process and by recoil of the daughter nucleus which disrupts the local structure in a liquid or solid source. On the other hand, the high recoil energy associated with alpha decay may prove useful in studying the perturbation process because it makes possible observations on nuclei in environments otherwise inaccessible for chemical reasons. Daughter nuclei in the heavy element region possess recoil energies of approximately 90 to 120 kilovolts, corresponding to initial velocities of 20 to 30 cm per μsec and a range in heavy material of about 40 μg per cm$^2$, or a few hundred atomic layers.$^{11,12}$ Recoils should therefore escape from a thin source within $10^{-13}$ second following alpha decay, a period short compared with the lifetimes of the intermediate states in alpha-gamma cascades. Angular correlations in thin sources might then be expected to show perturbations characteristic of the medium into which the daughter nuclei recoil, thus greatly increasing the variety of environments that can be explored. The experiments presented here were designed to examine this possibility by means of observations on nuclei recoiling into vacuum and into various absorbers.

I-A. Selection of Isotopes and Preparation of Sources

Sources considerably thinner than the range of the recoils were required, since most of the daughter nuclei travel obliquely through the source. A further limitation on source dimensions is the finite sourcesize correction, which becomes significant as the source height or width approaches the detector distance,$^{13}$ and is difficult to evaluate for the experimental arrangement employed. These restrictions limited the isotopes suitable for the experiments to those with relatively high specific activities and intense gamma rays in coincidence with primary alpha groups. Several otherwise suitable short-lived members of alpha-decay chains could
not be studied because gamma or x rays from lighter members of the chain coincided in energy with the gamma ray of interest. Separation of the members of such chains by alpha pulse-height analysis was not feasible as the alpha particles observed in the experiments pass at various angles through the backing plate, yielding a continuous energy spectrum.

Of the isotopes suitable for investigation, three were selected: Am$^{241}$, Am$^{243}$, and Cm$^{243}$. All three isotopes decay primarily to an excited state of the daughter which de-excites mainly by radiative transitions. The unperturbed correlation for the americium cascades is large and can be calculated to a good approximation. This constitutes an advantage because it makes possible a quantitative comparison of the experimental data with attenuation coefficients calculated for various magnetic and electric interactions. Three intense gamma rays, two of which were unresolved in the experiments, depopulate the intermediate state of the Cm$^{243}$ cascade. The recent work of Ewan and coworkers$^{14}$ has fixed the gamma mixing ratios within relatively narrow limits, but a wide range of anisotropies is nevertheless possible. A compensating feature of the curium cascades is the large K-conversion coefficient of the gamma rays of interest. The angular distribution of K x rays is always isotropic since the K shell is spherically symmetric; these radiations therefore serve as an internal check against experimental errors.

Thirty percent by activity of Cm$^{244}$, 4% of Pu$^{238}$, and 3% of Am$^{241}$ were present in the original Am$^{243}$ sample. A fraction of pure americium was obtained from the sample by elution from Dowex-50 cation-exchange resin with 0.28 N alpha-hydroxyisobutyric acid at a pH of 4.8.$^{15}$ The Am$^{241}$ used in the experiments was essentially free from other activities, while the Cm$^{243}$ contained 34% by activity of Am$^{241}$. Macroscopic impurities were removed by repeated adsorption on Dowex-50 cation-exchange resin at 87°C and elution with 6N hydrochloric acid prepared by bubbling the gas through conductivity water. Thin sources were then made by vaporizing approximately 0.05 µg or less of radioactive material from a tungsten or tantalum filament onto an area 7/8 cm in diameter. Quarter-mil aluminum, 0.1-mil silver, and 0.25-mil Mylar were used as backing materials. The source areas containing the radioactive deposit were indistinguishable from adjacent areas of the backing plate when viewed under a microscope. It may nevertheless be assumed that impurities amounting at least to several atomic layers were vaporized together with the activity.
The procedure used to determine the percentage of recoils escaping from the source was as follows: Alpha particles from each of four Am$^{243}$ sources were counted in a proportional counter of known efficiency. A collecting plate was then placed a few millimeters from each source and Np$^{239}$ recoils were collected in vacuum for several hours. The collecting plates were counted in a 2π proportional counter at the end of the collection period and at intervals for several days thereafter to verify the 2.3 day half life. Np$^{239}$ decays primarily to the 286- and 392-kilovolt states in Pu$^{239}$, which de-excite by highly converted cascade transitions. Assuming 100% counting efficiency (either a beta particle or a conversion electron counted for each beta decay), the initial counting rate multiplied by the mean life corresponds to the number of recoils collected. This number was corrected for the solid angle subtended at the source by the collecting plate and compared with the total number of alpha decays during the collection period. The results indicate that from 43 to 45% of all recoils escaped into the vacuum, and presumably an equal percentage into the backing plate. Essentially the same escape efficiencies were obtained when the recoils were collected in air in an electrostatic field. Since the four Am$^{243}$ sources yielded similar results and were prepared in the same manner as the Am$^{241}$ and Cm$^{243}$ sources, it was assumed that the recoil escape efficiencies of all the samples were comparable.

I-B. Equipment and Experimental Methods

In the majority of experiments a cubical vacuum chamber equipped with two zinc sulfide screens centered on adjacent faces was employed (Fig. 1). Collimators prevented passage of light between the two screens and defined the alpha-particle beams. Gamma rays were viewed by means of a fixed 1.5x1-inch or 3x3-inch sodium iodide crystal placed outside a thin aluminum window centered on a third face of the chamber. The half angles subtended at the source by the detectors ranged from 11 to 19 degrees. A frame fitted into a slot at the bottom of the chamber held the source at a 45° angle to the faces. The frame could be rotated through 180° 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;
Fig. 1. Diagram of vacuum chamber showing position of source (black) and backing plate (cross-hatched) in relation to detectors. Direction of gamma ray is indicated by white arrow, directions of alpha particles by long black arrows. The short black arrows indicate the paths of the recoils emitting the gamma rays recorded in the coincidence spectrum.

A. Source facing alpha detectors. Coincident gamma rays are emitted by recoils in the backing plate.

B. Source facing gamma detector. Alpha particles travel through the backing plate, coincident gamma rays are from daughter nuclei recoiling into vacuum.
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.

Light from the scintillators was transmitted through an optical coupling to RCA 6655A photomultiplier tubes and the output signals were suitably amplified, shaped, delayed, and fed into a coincidence unit with a resolving time of from 2 to 4 μ sec. The coincidence output pulse gated a 50-channel pulse-height analyzer on which the gamma spectrum was displayed. Before each experiment the noise level of the alpha-detection system was determined and discriminator settings were adjusted to prevent background pulses from gating the coincidence unit. Gamma rays in coincidence with gates from the two alpha detectors were then successively pulse-height analyzed. After correction for accidental coincidences, the peaks of interest were integrated and normalized by the number of alpha particles gating the coincidence unit. Corrections for the finite size of the gamma detectors were taken from the tables of Rose and West, while the corresponding correction for the alpha detectors was calculated from formulas in Reference 13. The quantity thus measured was the anisotropy value, \[ A = \frac{W(180°)}{W(90°)} - 1, \] where \( W(\theta) \) is the relative probability that the propagation vectors of the alpha particle and gamma ray form an angle \( \theta \). For correlation functions involving only the second power of cosine \( \theta \), the coefficient of \( P_2 \) in the Legendre polynomial expansion is obtained from the anisotropy value by the relation \( A_2 = A/(1.5 + 0.5 A) \).

In earlier experiments on \( ^{241} \text{Am} \) and \( ^{243} \text{Am} \), a beryllium-walled chamber equipped with a single zinc sulfide screen and a movable gamma-ray detector was employed. With this arrangement, singles spectra must be recorded at each position of the gamma detector to correct for differences in solid angle subtended at the source as the detector is moved. Accurate determination of the solid angles proved difficult because of the low activity of the sources and relatively high background radiation in the energy region of interest. Recoils collected on the walls of the chamber also contributed to the singles peaks, making it necessary either to subtract the chamber background or to stop
the recoils close to the source by covering it with thin absorber. Presumably because of these difficulties, the results of the earlier experiments were less precise than those obtained with the fixed detector arrangement.

The problem of dealing with background gamma rays does not arise in the experimental arrangement shown in Fig. 1 as the angle subtended by the gamma detector is constant. Corrections for decentering of the source are made in first order by dividing the number of alpha particles gating the coincidence unit into the number of counts under the coincidence peak. This ratio of coincidence counts to gates corresponds to the experimental value of $W(\theta)$ provided that all the gates represent alpha particles populating the intermediate state of the cascade. Since the anisotropy value depends only on the ratio $W(180^\circ)/W(90^\circ)$, it is not altered by the presence of alpha-active impurities if they constitute the same proportion of the total gate count for the two detector positions. The presence of Cm$^{244}$ in the Am$^{243}$ sources was found to introduce a significant error in one experiment during which the pressure in the chamber was high enough to reduce a large proportion of the pulses below the threshold of the alpha-detection system. A progressive increase in pressure occurred during the experiment, accompanied by a decrease in gate rate and a change in composition of the detected alpha particles in favor of the higher-energy Cm$^{244}$ alpha groups, which are not in coincidence with gamma rays. The ratio of coincidences to gates showed a corresponding decrease, resulting in a false anisotropy. The possibility of a similar effect was considered in experiments involving passage of alpha particles through backing plates, since a small proportion of the pulses fell below the detection threshold. In order to minimize such errors the following procedure was employed: The ratio of alpha-particle counting rates for the two detectors was determined with the source facing the zinc sulfide screens. In all cases these rates were equal to within a few percent, indicating that the efficiencies of the detectors were comparable and the sources were centered. The source was then rotated through 180 degrees and the discriminator settings adjusted above the noise levels to obtain the same ratio of gate rates. Under these conditions the Kx rays from impure samples of Cm$^{243}$ appeared isotropic in both source positions, and Am$^{243}$ sources containing Cm$^{244}$ impurities showed anisotropies for both positions consistent with those in pure Am$^{243}$ samples.
A further source of error noted during the experiments results from the long lifetime of the intermediate state in the $^{241}\text{Am}$ cascade. As shown in Fig. 1B, the gamma rays observed in the 180-degree coincidence spectrum come from recoils traveling toward the gamma detector whereas the recoils contributing to the 90-degree coincidence spectrum travel on the average parallel to the gamma detector. The half life of the 60-kilovolt state in $^{237}\text{Np}$ is $6.3 \times 10^{-8}$ second, during which the recoils travel more than a centimeter through the vacuum. Thus the source of the coincident gamma rays is significantly closer to the gamma detector when gates are obtained from the 180-degree alpha detector as compared to the 90-degree detector, and a false positive anisotropy is observed. No such error is expected in $^{243}\text{Am}$ or $^{243}\text{Cm}$ since the distance traveled by the recoils during the lifetimes of the intermediate states is negligible compared with the distance between the source and the gamma detector. This was demonstrated in $^{243}\text{Cm}$ by the isotropy of the K x rays.

**I-C. Americium-241 and Americium-243**

The decay schemes of $^{241}\text{Am}$ and $^{243}\text{Am}$ (Fig. 2) show the typical features of deformed odd-A nuclei. An explanation of the low-energy spectra in these nuclei in terms of collective motions of an axially symmetric core strongly coupled to the motion of the odd nucleon was proposed by Bohr and Mottelson. The single-particle orbitals in the strong coupling limit are designated according to Nilsson's level scheme by the asymptotic quantum numbers $(N, n_z, \Lambda, \Omega_p)$ and $K$, where $N$ is the number of nodes in the wave function, $n_z$ is the number of nodal planes perpendicular to the nuclear symmetry axis, $\Lambda$ is the component of orbital angular momentum along the symmetry axis, $\Omega_p$ is the projection of the total angular momentum of the particle along the symmetry axis, and $K$ represents the projection of the spin I along the symmetry axis. States differing only in the sign of $\Omega_p$ are degenerate. The lowest level of an odd-A nucleus is characterized by $K = \Omega$ of the odd nucleon, the remaining particles occupying states of the same $|\Omega|$ pairwise. Since alpha particles are formed most readily from paired nucleons, alpha decay between
Fig. 2. Partial decay schemes of $^{241}$Am and $^{243}$Am.
Experimental data from Reference 18 and 19.
states with the same asymptotic quantum numbers is favored over transitions involving a change in the orbital of the odd nucleon. Favored alpha decay is illustrated in \( \text{Am}^{241} \) and \( \text{Am}^{243} \), both of which show intense transitions from the \((523, 5/2)\) ground state to members of a rotational band based on the same intrinsic state in the daughter nucleus. The lowest member of the \((523, 5/2)\) band in the daughter is the intermediate state of the cascade observed in the experiments.

The selection rules for alpha decay allow transitions of multipole order 0, 2, and 4 between two states of spin \(5/2\) and the same parity. Ad-mixtures of the higher multipoles are responsible for the anisotropy of the cascade, since processes involving alpha particles with no angular momentum would be isotropic. The angular correlation is extremely sensitive to the mixing ratio because of interference between multipoles of different order (Fig. 3). As discussed by Bohr, Fröman, and Mottelson, an estimate of the mixing ratio for the americium alpha particles may be obtained by extrapolating experimental data from neighboring even-even nuclei. For transitions involving no change in intrinsic state, the relative intensity of alpha partial waves of multipolarities \(L\) and \(L'\) at the nuclear surface is given by

\[
\frac{I_L}{I_{L'}} = \frac{c_L (|II'K|II'K|^2}{c_{L'} (|II'K|II'1^K|^2),
\]

where \(I\) and \(I'\) are the spins of the initial and final states and the parentheses denote Clebsch-Gordan coefficients. The probability factor \(c_L\) for the \(2^L\)-pole alpha wave is normalized so that \(c_0 = 1\). Approximate values of \(c_L\) may be obtained by applying the above equation to the pure transitions populating members of the ground-state rotational band in even-even nuclei. The values of \(c_2\) from the alpha decay of \(\text{Pu}^{240}\) and \(\text{Cm}^{242}\) are 0.63 and 0.59, respectively. Assuming that the average of these values corresponds to the D-wave probability in \(\text{Am}^{241}\), a mixing ratio \(I_2/I_0\) of 0.22 is obtained. An almost identical mixing ratio is found for \(\text{Am}^{243}\) by averaging the \(c_2\) values of 0.71 for \(\text{Pu}^{242}\) and 0.52 for \(\text{Cm}^{244}\). Recent theoretical studies of the alpha decay of \(\text{U}^{233}\) by Chasman and Rasmussen indicate that mixing ratios about 40% larger than those
Fig. 3. Coefficient of \( P_2 \) for \( 5/2 \rightarrow 5/2 \) cascade with pure dipole gamma transition. The equation plotted is:

\[
A_2 = \frac{(-0.175[0.935\delta_a^2] + \cos(\sigma_0 - \sigma_2)(4.90\delta_a))}{(1 + \delta_a^2)}
\]

with \( \delta_a^2 = \left| \frac{(j_1 \parallel L \parallel j)}{(j_1 \parallel 0 \parallel j)} \right|^2 \) where \( (j_1 L j) \) is the reduced matrix element for the \( 2^L \) pole alpha partial wave between states of spin \( j_1 \) and \( j \). Equations 40, 67 and 78a of Reference 23 and the definition of the Coulomb phase shift difference \( (\sigma_1 - \sigma_2) \) in Reference 24 were used in the calculation.
predicted by Bohr, Frohman, and Mottelson may be more accurate. The admixture of $^4$-pole alpha partial wave populating the 5/2 minus states in Np$^{237}$ and Np$^{239}$ is expected to be negligible.

The admixture of 2$^+_4$-pole alpha partial wave populating the 5/2 minus states in Np$^{237}$ and Np$^{239}$ have been determined by means of conversion coefficients to be E1 with less than 1% M2. Admixtures of this magnitude would not alter the anisotropies plotted in Fig. 3. The ambiguity concerning the sign of $\beta$ has been resolved by previous alpha-gamma angular correlations in Am$^{241}$ and Am$^{243}$. Large negative anisotropies were obtained for the cascade through the 5/2 minus state, establishing that the S and D alpha partial waves are in phase. The angular-correlation function corresponding to a mixing ratio of 0.22 is then $W(\theta) = 1 - 0.36 P_2(\cos \theta)$, and the anisotropy value is -0.46.

If the mixing ratio is increased by 40% as suggested by Chasman and Rasmussen, $A_2$ decreases to -0.41 and the anisotropy value to -0.51. Before comparison with the calculated correlation the experimental data must be corrected for alpha decay to the 7/2 minus member of the rotational band. Highly converted M1-E2 radiation from the 7/2 state accounts for 13.5% of the population of the 5/2 state in Np$^{237}$ and 11.7% in Am$^{243}$. The angular correlation between all alpha groups and the E1 gamma ray from the 5/2 state is thus a weighted average for the 5/2 $\rightarrow$ 5/2 $\rightarrow$ 5/2 double cascade and the 5/2 $\rightarrow$ 7/2 $\rightarrow$ 5/2 triple cascade with intermediate radiation unobserved. If the triple cascade is assumed to be isotropic, the correction factors by which the experimental anisotropy values must be multiplied are 1.16 for Am$^{241}$ and 1.13 for Am$^{243}$. Correction factors about 2% larger are obtained if the triple cascade is assumed to contribute the unperturbed anisotropy value of +0.06 calculated for pure $L = 2$ alpha waves and an E2/M1 mixing ratio of 1.7 for the unobserved radiation. The larger correction factor would be appropriate only in the event that the perturbations of the double and triple correlations were comparable. Although the estimated lifetime of the 7/2 minus state is short compared to that of the 5/2 minus state, strong perturbations may be expected in the triple cascade owing to excitation of the electron shell associated with internal conversion of the unobserved radiation.
1. Results in Am$^{243}$

A summary of the results obtained in Am$^{243}$ 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; i.e., the backing plate in position A, the vacuum or absorber in contact with the source in position B. The attenuation factors $G_2$ in the last column give the ratio between the experimental coefficient of $P_2(\cos \theta)$ corrected for the presumably isotropic contribution of the triple cascade and the calculated coefficient of $-0.36$ for the $5/2 \rightarrow 5/2 \rightarrow 5/2$ double cascade.

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

Experiment 1 in Table I refers to an Am$^{243}$ source containing macroscopic impurities. The source was prepared by evaporating a dilute hydrochloric acid solution of the isotope onto a platinum plate and flaming the sample in air, a procedure expected to yield Am$_2$O$_3$ or AmO$_2$. Since the source thickness considerably exceeded the range of the recoils, the local environment
of the daughter nuclei presumably consisted largely of nonvolatile impurities deposited from the acid solution. The anisotropy of \(-0.20\) obtained under these conditions may be compared with the value of \(-0.15\) reported previously. 19a

Experiments 2 through 9 refer to \(^{243}\text{Am}\) sources free from macroscopic impurities vaporized onto thin backing materials. The coincidence rates in these sources varied from 3 to 6 counts per minute at half angles of 11 to 19 degrees. Anisotropy values for each source were alternately determined in positions A and B to a statistical accuracy of 5% and the procedure repeated until a 2% statistical error was obtained. Several sources were observed at intervals over periods ranging up to a year in order to determine if slow chemical or physical changes affected the results. No such changes were noted.

All the \(^{243}\text{Am}\) correlations showed considerable perturbation. The anisotropies obtained in silver and aluminum were significantly larger than in iron or in the thick oxide source. Mylar, a polyester of terephthalic acid and ethylene glycol, yielded almost no anisotropy.

A striking effect observed during the experiments was the marked attenuation of the correlation in nuclei recoiling into vacuum (Fig. 4). Thin sources on silver and aluminum as well as those on Mylar consistently showed anisotropies below the hard-core value for a static interaction when the recoils escaped into the vacuum from source position B. Part of the perturbation might be attributed to interactions occurring before the recoils left the source. This could not, however, account for an attenuation coefficient smaller than the largest value obtained in any absorber, \(G_2\) about 0.58, since the daughter nuclei travel through comparable source thicknesses whether they recoil into the vacuum or the backing plate. Most if not all of the perturbation thus appeared to result from interactions at the surface of the source or in isolated ions traveling through the vacuum. These possibilities were investigated further as follows:

a. An aluminum or silver plate was placed over the source and the anisotropy again determined in position B. Because of the fragility of the sources, pressure could not be applied to establish close contact between the plate and the radioactive deposit. The two surfaces appeared to be in contact over most of the active area and in any case were much less than 0.1 mm
Fig. 4. Coincidence spectra from thin Am\(^{243}\) source on aluminum backing in source positions A and B. Broken curve represents 180 degree spectrum, solid curve 90 degree spectrum. The spectra have been normalized to the same number of gates.
apart. At an average velocity of $10^7$ cm/sec the daughter nuclei should have reached the metal plate in less than the mean life of the excited state. The anisotropy nevertheless remained the same as in vacuum.

b. A layer of silver thick enough to stop the recoils was vaporized over the source. Observations in position B then yielded anisotropies comparable to those obtained when the recoils stopped in silver backing plates. The anisotropies in position A remained unchanged. This experiment established that the attenuation in vacuum did not result from errors related to the passage of the alpha particles through the backing plate, such as scattering or loss of pulse height. The dependence of the perturbation on the medium receiving the recoils was again clearly demonstrated in a source on Mylar over which a layer of silver was vaporized: This americium "sandwich" showed an anisotropy of -0.04 in position A and -0.25 in position B.

c. As discussed by Goertzel, Alder, and Biedenharn and Rose, angular correlations perturbed by magnetic interactions between the nucleus and atomic electrons in vacuum may be restored by the application of a strong magnetic field. A decoupling experiment of this type was attempted but yielded inconclusive results. The vacuum chamber was placed between the poles of an electromagnet with the field directed along the axis from the source to the gamma detector. Light from the scintillators was transmitted through 10-inch Lucite light pipes to photomultiplier tubes enclosed in concentric mu-metal and iron shields. Owing to light loss in the pipes, the alpha pulses were not well resolved from background noise. A reasonable coincidence rate could be maintained only by decreasing the detection threshold to admit the lower portion of the alpha spectrum together with a small background, which was subtracted from the gate count. Preliminary observations were made on an Am$^{241}$ source in position B with a Mylar absorber placed over the surface to stop the recoils. The correlation in this case was consistently isotropic to within $\frac{1}{4}$% in a field of 7500 gauss and also at zero field. This is in agreement with the results of Milton and Fraser, who observed no improvement of the Am$^{241}$ correlation in a solid source in a field of 7000 gauss.
Observations on $^{243}_{\text{Am}}$ were made in position B with the recoils escaping into vacuum. Variable anisotropies, ranging from $-0.02 \pm 0.03$ to $-0.17 \pm 0.03$ and averaging $-0.07$, were obtained with the field off. The anisotropies with the field at 7500 gauss were consistent within the standard deviations and averaged $-0.18 \pm 0.02$. These results were not considered significant, in view of the failure to obtain consistent data with the field off. Since no such problem was encountered in $^{241}_{\text{Am}}$, it appears probable that the variations arose from the background counts, which were negligible compared with the counting rate in $^{241}_{\text{Am}}$, but not with that in $^{243}_{\text{Am}}$. An anisotropy comparable to the unattenuated value of $-0.46$ should have been detectable despite the experimental error. Complete restoration of the correlation by a field of 7500 gauss thus appeared to be ruled out in this experiment.

d. A perturbation resulting from a magnetic interaction between the nucleus and atomic electrons would be altered by collisions with gas molecules owing to reorientation of the electron spins and possible changes in the electron configuration of the recoils. An attempt was therefore made to determine the anisotropy in nuclei recoiling into air. The zinc sulfide screens used in the previous experiments were replaced with smaller screens located $1.5$ cm from the source and subtending a half angle of $13$ degrees. Anisotropies were determined in source positions A and B with the chamber open to air. The results obtained for nuclei recoiling into backing plates were comparable to those of previous experiments, indicating that the smaller source-to-detector distance did not introduce significant errors. When the source was rotated to position B, variable anisotropies ranging from $-0.04 \pm 0.04$ to $-0.17 \pm 0.04$ and averaging $-0.10$ were obtained. Impurities adhering to the surface of the source or changes in composition or temperature of the air may have been responsible for the variations.

2. Results in $^{241}_{\text{Am}}$

Only integral correlations were done in $^{241}_{\text{Am}}$ (Table II). Experiments 1 and 2 show a strong interaction in aluminum and silver. Since $^{239}_{\text{Np}}$
recoils in these absorbers must undergo a similar interaction, the attenuation coefficients of 0.53 and 0.58 in Table I cannot be attributed entirely to perturbations occurring before the recoils leave the source.

Experiments 4, 5 and 6 in Am²⁴¹ were carried out in two sources, one vaporized onto Mylar and the other on Teflon. The plastic was cemented over a hole 1.5 cm in diameter in an aluminum disc which served as the lid of a thin aluminum container 2 mm deep and 2 cm in diameter. Before each experiment the disc was mounted vertically in the chamber with the plastic window in position B, and angular correlations were done in vacuum. Large positive anisotropies were observed, indicating that most of the recoils escaped into vacuum and traveled toward the 180-degree detector. The lid was then fastened on the aluminum container with the activity facing the inside of the container. This device was mounted in the chamber with the lid again in a vertical plane and the inactive side of the plastic facing alpha detectors 1.6 cm from the source. Angular correlations were done with the chamber open to air and daughter nuclei recoiling into the container. In Experiments 5 and 6, conductivity water or freshly distilled benzene was introduced into the container through a small hole until the liquid level was well above the top of the source.

<table>
<thead>
<tr>
<th>Experiment Number</th>
<th>Source position</th>
<th>Environment of daughter nucleus</th>
<th>Anisotropy value</th>
<th>$A_2$</th>
<th>$G_2$</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>A</td>
<td>aluminum</td>
<td>-0.03 ± 0.01</td>
<td>-0.02</td>
<td>0.06</td>
</tr>
<tr>
<td>2</td>
<td>A</td>
<td>silver</td>
<td>-0.04 ± 0.01</td>
<td>-0.03</td>
<td>0.08</td>
</tr>
<tr>
<td>3</td>
<td>A</td>
<td>Mylar</td>
<td>-0.02 ± 0.01</td>
<td>-0.01</td>
<td>0.03</td>
</tr>
<tr>
<td>4</td>
<td>B</td>
<td>air</td>
<td>-0.02 ± 0.02</td>
<td>-0.01</td>
<td>0.03</td>
</tr>
<tr>
<td>5</td>
<td>B</td>
<td>water</td>
<td>0.00 ± 0.03</td>
<td>0.00</td>
<td>0.00</td>
</tr>
<tr>
<td>6</td>
<td>B</td>
<td>benzene</td>
<td>-0.06 ± 0.01</td>
<td>-0.04</td>
<td>0.11</td>
</tr>
</tbody>
</table>
Periodic inspection showed that the liquid remained clear and in contact with the entire area of the source throughout the experiment. The correlation nevertheless failed to show the improvement generally observed in nonviscous liquids as compared to solids.

I-D. Curium-243

The intermediate state of the alpha-gamma cascades observed in Cm\(^{243}\) has been assigned a spin of 5/2 plus, an energy of 285.5 kilovolts, and a half life of \(1.1 \times 10^{-9}\) second (Fig. 5). Favored alpha decay populates a rotational band based on the 285.5-kilovolt state, indicating that the odd neutron occupies the same single-particle level as in the ground state of Cm\(^{243}\). The 285.5-kilovolt level is identified according to the Nilsson level scheme by the asymptotic quantum numbers (622, 5/2). The multipole mixing ratio of the alpha waves may be estimated from experimental data in neighboring even-even nuclei by the method described in connection with Am\(^{241}\) and Am\(^{243}\). Averaging the probability factors \(c_2\) of 0.59 and 0.52 from the alpha decay of Cm\(^{242}\) and Cm\(^{244}\), one obtains an estimated ratio of \(L = 2\) to \(L = 0\) partial waves of 0.20. The calculated admixture of \(L = 2\)-pole radiation is again negligible.

Decay of the 285.5-kilovolt level populates the members of a 1/2 plus rotational band based on the ground state of Pu\(^{239}\) to which the asymptotic quantum numbers (631, 1/2) have been assigned. Four radiations from the 285.5-kilovolt state were observed in coincidence with the Cm\(^{243}\) alpha particles: K x-rays at about 100 kilovolts, the unresolved 210- and 228-kilovolt gamma rays populating the 7/2 and 5/2 members of the rotational band, and the 278-kilovolt gamma ray populating the 3/2 state. The relative intensity of the three gamma rays \(^{30}\) as well as the K-shell and L-subshell conversion coefficients \(^{14}\) were determined by Ewen and co-workers from observations on the decay of Np\(^{239}\). According to these workers, the intensity ratio of the 210-, 228-, and 278-kilovolt gamma rays is 0.32:1:1.1, and all three radiations are M1 with less than 5% admixture of E2. As pointed out by Hollander, Smith, and Mihelich,\(^{31c}\) the occurrence of M1 radiation between the states (622, 5/2) and (631, 1/2) is forbidden by
Fig. 5. Partial decay scheme of Cm$^{243}$. Data from References 14, 30, 31.
a rigid selection rule requiring \( L \geq |K_\pi - K_\xi| \). This condition is satisfied and the gamma-ray intensities are best accounted for if the 285.5-kilovolt level contains a small admixture of the \((631, \frac{3}{2})\) state, which lies near the \((622, \frac{5}{2})\) state in the Nilsson level scheme.

As discussed by Bohr, Fröman, and Mottelson\(^{25a}\) and by Nilsson\(^{21}\), the reduced transition matrix elements for gamma rays from the same pure state populating different members of a single rotational band based on another pure state are identical. In that event the \(E2/ML\) mixing ratios for the gamma rays are proportional to an energy-dependent factor multiplied by the square of a ratio of Clebsch-Gordan coefficients which contain the entire spin dependence. This simple relationship may not apply to the 285.5-kilovolt level of \(^{239}\text{Pu}\), owing to the possibility of interference between \(E2\) radiations from the \((622, \frac{5}{2})\) and \((631, \frac{3}{2})\) components. Even if only a small admixture of the \((631, \frac{3}{2})\) state is present, its contribution to the \(E2\) transition probability cannot be assumed negligible compared with that of the \((622, \frac{5}{2})\) state for two reasons: First, the asymptotic quantum number selection rule \( \Delta n_z = 0 \) is violated by the \((622, \frac{5}{2}) \rightarrow (631, \frac{1}{2})\) transition but not by the \((631, \frac{3}{2}) \rightarrow (631, \frac{1}{2})\) transition; and secondly, a symmetrizing term in the wave function contributes an \(E2\) matrix element if \( L \geq K_\xi + K_\eta \), a condition satisfied only by the \((631, \frac{3}{2}) \rightarrow (631, \frac{1}{2})\) transition. The relative mixing ratios of the gamma rays from the 285.5-kilovolt state may therefore depend on the magnitudes and signs of the matrix elements and the amplitude of the \((631, \frac{3}{2})\) state as well as the Clebsch-Gordan coefficients. If the contribution of the \(3/2\) state to the \(E2\) transition probability is neglected, the relative mixing ratios predicted for the 210-, 228-, and 278-kilovolt gamma rays are 0.8:1:3.

The calculated anisotropy values for the three alpha-gamma cascades observed in the experiments are plotted as a function of the gamma mixing ratio in Fig. 6. In accordance with the preceding discussion,
Fig. 6. Anisotropy values for alpha-gamma cascades with $\delta_2 = 0.20$ plotted as a function of the gamma mixing ratio $\alpha$

$$\delta_2^2 = \frac{I(E2)}{I(M1)}$$
the alpha mixing ratio is assumed to be 0.20 and only gamma mixing ratios up to 6% E2 are considered. The calculated coefficient of P4 is less than 1% of A2 for all mixing ratios up to the limit set by Ewan and co-workers; a determination of the anisotropy value therefore also determines A2.

The anisotropy value for the 228-kilovolt gamma ray is negative and equal to or greater than 0.20| for all mixing ratios up to the limit, while the anisotropy value for the 210-kilovolt gamma ray has a maximum value of 0.066. Since the 228-kilovolt gamma ray is three times as intense as the 210-kilovolt radiation, a negative anisotropy may be expected for the unresolved peaks observed in the experiments unless both mixing ratios are near the maximum. The anisotropy for the 278-kilovolt gamma ray may be either positive, negative, or nonexistent, depending on the magnitude and sign of 8.

Table III presents a summary of the experiments on Cm243. The A2 and anisotropy values given in the table include corrections for solid angle, for accidental coincidences, and for the presumably isotropic contribution of the triple cascade through the 7/2 plus state. The column headings have the same significance as in Tables I and II, and were discussed in connection with Am241 and Am243. Each curium source contained 34% by activity of Am241. The sources were comparable in mass to the corresponding Am243 samples and yielded approximately the same coincidence rate for the 210-228-kilovolt peak. In all cases the K x rays observed simultaneously with the three gamma rays in the coincidence spectrum were isotropic.

**TABLE III**

<table>
<thead>
<tr>
<th>Experiment Number</th>
<th>Source position</th>
<th>Environment of daughter nucleus</th>
<th>210-228-kev anisotropy</th>
<th>278-kev gamma anisotropy A2</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>A</td>
<td>oxide</td>
<td>-0.01±.03</td>
<td>-0.01±.02</td>
</tr>
<tr>
<td>2</td>
<td>A</td>
<td>aluminum</td>
<td>-0.24±.02</td>
<td>-0.17±.02</td>
</tr>
<tr>
<td>3</td>
<td>A</td>
<td>silver</td>
<td>-0.19±.02</td>
<td>-0.14±.02</td>
</tr>
<tr>
<td>4</td>
<td>B</td>
<td>vacuum</td>
<td>-0.09±.01</td>
<td>-0.06±.01</td>
</tr>
<tr>
<td>5</td>
<td>B</td>
<td>gold</td>
<td>-0.19±.02</td>
<td>-0.14±.02</td>
</tr>
<tr>
<td>6</td>
<td>B</td>
<td>air</td>
<td>-0.15±.02</td>
<td>-0.11±.02</td>
</tr>
</tbody>
</table>
The source referred to in Experiment 1 was deposited from an impure hydrochloric acid solution and flamed to remove organic material. Macroscopic impurities thick enough to stop the recoils were present, but no chemical analysis was done to determine if these were similar to the impurities in the corresponding Am\(^{243}\) source. In any event the environment of the daughter nuclei probably consisted largely of oxides. The 210- and 228-kilovolt gamma rays as well as the 278-kilovolt gamma ray were isotropic in this source.

Thin curium sources on aluminum and silver backings also failed to show significant anisotropies for the 278-kilovolt gamma ray. A large anisotropy was observed in the 210-228-kilovolt peak when the daughter nuclei recoiled into metals, but the correlation significantly decreased when the recoils were allowed to escape into vacuum. As shown in Fig. 7, the K x rays and the 278-kilovolt peaks on either side of the 210-228 kilovolt peak exhibited no corresponding changes on rotation of the source. The false positive anisotropy discussed earlier appeared in the 60-kilovolt gamma ray of Np\(^{237}\) when the recoils escaped into vacuum. A similar effect could not be responsible for the decreased negative anisotropy of the 210-228-kilovolt peak, since the K x rays remained isotropic. It should be noted that the K x rays as well as the 210-1 228-, and 278-kilovolt gamma rays depopulate the 285.5-kilovolt state of Pu\(^{239}\), and therefore all four radiations have the same lifetime.

Experiments 1 through 4 in Cm\(^{243}\) are analogous to experiments 1, 2, 3, and 6 in Am\(^{243}\), and show qualitatively similar results: The anisotropies obtained from daughter nuclei recoiling into a metal are larger than in isolated ions traveling through a vacuum or in oxide deposits. Experiment 5 in Cm\(^{243}\) corresponds to Experiment 8 in Am\(^{243}\) and likewise shows a similar effect: The correlation improves when the recoils are prevented from escaping into vacuum by a layer of metal vaporized over the source. Again as in Am\(^{243}\), variable results were obtained when the recoils escaped into air. The anisotropies for the 210-228-kilovolt peak in this experiment ranged from -0.05±.03 to -0.18±.03 and averaged -0.15. Since these variations were observed in the 210-228-kilovolt peak while the K x rays and the 278-kilovolt peak remained isotropic, they appear to result from a variable perturbation rather than from experimental errors.

Certain conclusions may be drawn from a comparison of the Cm\(^{243}\) results with the calculated correlations in Fig. 6:
Fig. 7. Coincidence spectra at 90 degrees (solid curve) and 180 degrees (broken curve) from thin Cm$^{243}$ source on aluminum backing. The peaks, from left to right, are the 60-kilovolt gamma ray of Np$^{237}$ due to an Am$^{241}$ impurity; K x rays from the 285.5-kilovolt state in Pu$^{239}$; the unresolved 210-228-kilovolt gamma rays from the same state; and the 278-kilovolt gamma ray also from the 285.5-kilovolt state. The 90 and 180 degree curves have been normalized to the same number of gates.
a. The largest $A_2$ value possible for the combined 210- and 228-kilovolt gamma rays is $-0.41$, occurring if the two radiations have the maximum mixing ratio of 0.0525 set by Ewan and co-workers, and opposite signs of $\delta$. If the unperturbed correlation coefficient is $-0.41$, the attenuation coefficients for Pu$^{239}$ recoils in aluminum and silver would be 0.42 and 0.34. These minimum values are not much lower than the attenuation coefficients of 0.53 and 0.58 obtained for Np$^{239}$ recoils in the same absorbers, indicating that the interaction in metals is of the same order of magnitude for Cm$^{243}$ as for Am$^{243}$ or smaller. This is in contrast to the results in thick oxide sources, in which the attenuation coefficient is almost zero for Cm$^{243}$ and 0.39 for Am$^{243}$.

b. The anisotropy of the 278-kilovolt gamma ray would be positive for all mixing ratios lower than 0.03. Since the observed anisotropies are all slightly negative, the mixing ratio for the 278-kilovolt transition appears to be 0.03 or larger and the sign of $\delta$ is clearly positive.

c. Unfortunately, an accurate analysis of the shape of the 210-228-kilovolt peak could not be made because of instability of the recording equipment over the long counting period. In some instances, the peak appeared to shift to higher channels in the 90-degree spectrum as compared to the 180-degree spectrum, indicating that the anisotropy for the 228-kilovolt gamma ray is considerably more negative than that of the 210-kilovolt gamma ray.
II. DISCUSSION

A general formulation of the angular correlation problem including the influence of extranuclear fields has been provided by Abragam and Pound. As discussed by these authors, the angular distribution of the second radiation in a cascade depends on the relative populations of the various magnetic sublevels of the intermediate state. The population distribution among these sublevels in turn depends on the angle between the spins and the propagation vector of the first radiation. Thus the directions of the two radiations are correlated through the spin distribution in the intermediate state of the cascade. If transitions occur among the magnetic sublevels during the lifetime of the intermediate state, the anisotropic spin distribution established by the selection of the first radiation is gradually replaced with a Boltzmann distribution, which is essentially isotropic at ordinary temperatures. The "memory" of the direction of the first radiation is then partially or completely lost before emission of the second radiation and the angular correlation is correspondingly attenuated.

Transitions among the magnetic sublevels of the intermediate state may be induced by interactions between the nuclear magnetic moment and magnetic fields arising in the electron shell or between the nuclear quadrupole moment and the gradient of an electric field of extra-atomic origin. In either case the interaction may be static or time-dependent, depending on whether the direction of the field at the nucleus is constant or fluctuating during the lifetime of the intermediate state. Magnetic interactions are considered in Section II-A and electric interactions are discussed in Section II-B. Each section consists of a review of the pertinent theory and previous experimental data, followed by an estimate of the nuclear moments and extranuclear fields in the experiments. A summary of the discussion and some general observations concerning the interpretation of the experiments are given in Section II-C.
II-A. Magnetic Interactions


As mentioned previously, the theory of the perturbed angular correlation began with the comments by Hamilton and subsequent work by Goertzel and Alder on the magnetic interaction in isolated atoms. No experimental observations concerning the perturbation in free atoms have been reported during the 20 years since this problem was suggested, presumably because angular correlations are difficult to perform in a rarefied gas. With certain qualifications to be discussed later, the theory for free atoms should apply to the present experiments involving recoils in vacuum. A review of the principal results obtained by Goertzel and Alder is therefore in order.

The perturbing Hamiltonian operating on the wave function of the intermediate state is \( \mathbf{\alpha} \mathbf{I} \cdot \mathbf{J} \), where \( \mathbf{\alpha} = \mu / H(0) / IJ \). In this expression \( \mu \) is the magnetic moment of the intermediate nucleus, \( H(0) \) is the time-average magnetic field at the nucleus produced by the orbital electrons, \( I \) is the nuclear spin, and \( J \) is the angular momentum of the electron shell. The attenuation coefficient \( G_k \) in the integral correlation is

\[
G_k = \frac{1}{2F+1} \sum_{FF'} \left[ (2F+1)(2F'+1)w^2(IIFF'; KJ) \right] \left[ 1 + \left( \frac{\tau_N}{\tau_{FF'}} \right)^2 \right],
\]

where \( F \) is the vector sum of \( I \) and \( J \), and \( w(abcd;ef) \) is the Racah coefficient. The splitting between the hyperfine-structure levels, \( \omega_{FF'} \), is given in terms of \( \alpha \) for pure IS coupling by the Landé interval rule. \( \tau_N \) is the mean life of the intermediate state. The orientation of the nuclear spins with respect to the direction of the first radiation changes as \( I \) precesses about \( F \) with the Larmor frequency \( 2\pi \Delta \nu \), corresponding to the width of the hyperfine structure. If \( 2\pi \Delta \nu \tau_N \gg 1 \), the populations of the magnetic sublevels will be significantly altered during the lifetime of the intermediate state and the correlation will be attenuated.
A minimum or "hard core" correlation nevertheless remains regardless of the width of the hyperfine structure, as indicated by the presence of terms with $F=F'$ in the expression for $G_k$. The minimum $G^2$ values obtained as $\alpha$ approaches infinity are plotted as a function of $J$ in Fig. 8. It should be noted that the hard-core correlation approaches a lower limit of $1 + 0.20 A_Z^2 P_Z^2$, significantly greater than the value of $1 + 0.11 A_Z^2 P_Z^2$ obtained from $^{239}$Np recoils in vacuum. Apparently a magnetic interaction with an electron shell in a stationary state cannot account for the perturbation observed in this experiment.

2. **Time-Dependent Magnetic Interactions**

Perturbation of the angular correlation by time-dependent fields is generally discussed in connection with the quadrupole interaction in liquids, but certain features of the theory may apply to a variety of situations in which the environment of the intermediate state undergoes rapid change. The condition for a time-dependent interaction is $\tau_R \ll \tau_N$ where $\tau_R$ is the relaxation time, corresponding to the period during which the field at the nucleus acts in a given direction, and $\tau_N$ is the mean life of the intermediate state or the period during which the nucleus is exposed to the field. As in the case of a static interaction, transitions among the magnetic sublevels of the intermediate state are induced by the action of the fluctuating field. The transition probability per unit time, however, varies directly with the relaxation time $\tau_R$ rather than with the lifetime $\tau_N$ as in the static case. Rapidly fluctuating fields accordingly produce less perturbation of the correlation than static fields of comparable magnitude. Also in contrast to static fields, a fluctuating field may completely destroy the correlation if the interaction energy is large enough or the lifetime of the intermediate state sufficiently long. Time-dependent interactions may be distinguished experimentally from static interactions by the occurrence of attenuation coefficients below the hard-core value or by means of delayed correlations. The delayed correlation varies periodically in the presence of a static interaction but shows a characteristic exponential decay if the interaction is time-dependent, provided that $\omega \tau_R \ll 1$ where $\omega$ is the frequency of the perturbing interaction.
Fig. 8. Minimum values of the attenuation coefficient $G_2$ for a static magnetic interaction with intermediate state spin of 5/2 plotted as a function of the resultant electronic angular momentum $J$. 

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Fluctuating magnetic fields may be present at a nucleus owing to radiation-induced excitation of the electron shell or to spin-lattice coupling in a liquid or solid. Three examples of time-dependent magnetic perturbations associated with the emission of Auger electrons have been reported; these are the gamma-gamma correlation in Cd$^{111}$ following K capture by In$^{111}$, and the K-electron-gamma correlations in Hg$^{197}$ and in Ta$^{181}$. The Cd$^{111}$ correlation showed significantly less perturbation in polycrystalline metallic environments than in solid ionic crystals, presumably because vacancies in the electron shell were filled more rapidly in the metals. A similar improvement of the correlation in metals was noted in the experiments on Am$^{243}$ and Cm$^{243}$, suggesting that excited electronic states may be involved. Magnetic interactions have also been observed in several gadolinium isotopes in aqueous solution following Coulomb excitation, but not in neodymium or samarium under comparable conditions. Although the ground state of Gd$^{+3}$ has a much smaller hyperfine structure than the other rare earth ions, it is an S state and therefore exhibits relatively weak spin-lattice coupling and a long paramagnetic spin relaxation time. The neodymium and samarium correlations apparently escaped attenuation because the "averaging out" effect of the rapid spin relaxation more than compensated for the wide hyperfine structure. A similar explanation might apply to the experiments in which the Cm$^{243}$ correlation showed a stronger perturbation than the Am$^{243}$ correlation in oxide sources, although it appears improbable that the initial $^8S_{7/2}$ configuration of Cm$^{+3}$ would be maintained in plutonium following the alpha decay.

3. Magnetic Moments

The magnetic moment of a deformed odd-A nucleus is given by the expression

$$
\mu = \frac{\Omega K}{I+1} \left( s_n - s_N \right) + s_N I; \quad s_N = \frac{1}{n} \left[ s_N \langle s_z \rangle + s_L \langle l_z \rangle \right],
$$

where $\langle s_z \rangle$ and $\langle l_z \rangle$ are calculated from Nilsson wave functions for the state occupied by the odd nucleon. As discussed previously, the intermediate levels of the cascades observed in the experiments are rotational
ground states populated by favored alpha decay from the ground states of the parents. Since favored transitions involve no change in the wave function of the odd nucleon, $g_n$ as well as $\Omega$, $K$, and $I$ should be the same in the initial and intermediate states in each cascade. The gyromagnetic ratio $g_R$ is roughly equal to $Z/A$ and consequently is also essentially unchanged in favored alpha transitions. It follows that the magnetic moments of the intermediate states in the Am$^{241}$, Am$^{243}$, and Cm$^{243}$ cascades should be comparable to those of the ground states in the parent nuclei or in neighboring odd-A nuclei in which the unpaired particle occupies the same orbital. The ground-state magnetic moments of Am$^{241}$ and Am$^{243}$, determined from the hyperfine structure in Am(I) and Am(II), are $+1.4$ $\mu$m for both isotopes. A magnetic moment of $2.0\pm0.5$ $\mu$m was obtained for the 60-kilovolt state in Np$^{237}$ by means of an angular correlation in a magnetic field perpendicular to the plane of the detectors.

The odd neutron occupies the same orbital in the 285.5-kilovolt state of Pu$^{239}$ as in the ground state of Pu$^{241}$. A magnetic moment of 0.1 $\mu$m is calculated for the Pu$^{241}$ ground state from the ratio of the ground state moments in Pu$^{239}$ and Pu$^{241}$, together with the value of 0.02 $\mu$m for the Pu$^{239}$ ground state obtained from atomic beam measurements by assuming a pure $7F_1$ electronic configuration. Judd has recently recalculated the Pu$^{239}$ moment from the atomic beam data, taking into account admixtures in the $f^6$ ground state electronic wave function. The calculated mixture of $7F_1$ and $5D_1$ states yields a magnetic field at the nucleus only one-third as large as the field arising from a pure $F_1$ state, in which case the Pu$^{239}$ ground state moment would be increased to 0.06 $\mu$m and the Pu$^{241}$ moment to 0.3 $\mu$m. These estimates from experimental data may be compared with magnetic moments of 1.0 $\mu$m for the $(5/2, 5/2)$ ground state of Am$^{241}$ and Am$^{243}$, and 0.5 $\mu$m for the $(6/2, 5/2)$ ground state of Pu$^{241}$, calculated from Nilsson wave functions.


The sources used in the experiments are believed to consist of AmO$_2$, Am$_2$O$_3$, and Cm$_2$O$_3$. Considerable evidence has been presented that the ground-state configurations of the ions are similar to those of the corresponding rare earths; i.e., $(5f^5) \ 6H_5/2$ for Am$^{4+}$, $(5f^6) \ 7F_0$ for Am$^{3+}$, and $(5f^7) \ 8S_7/2$ for Cm$^{3+}$. Two processes associated with the alpha decay and subsequent recoil which may modify these initial configurations are considered.
a. Migdal Effect

The electronic binding energies of the daughter atoms in alpha decay are some 40 kilovolts lower than those of the parents. Transfer of all but a few hundred electron volts of this energy occurs adiabatically as the alpha particle passes through the inner electron shells with a velocity comparable to the electronic velocity. In the outer shells, the passage of the alpha particle is rapid compared with the electronic period; the electrons are thus confronted with a sudden decrease in nuclear charge, and ionization or excitation may occur if the radial wave functions of the parent atom differ greatly from those of the daughter.

The "shakeoff effect" in alpha decay was first treated theoretically by Migdal, who calculated ionization probabilities of $2.8 \times 10^{-6}$, $1.1 \times 10^{-4}$, and $1.4 \times 10^{-3}$ for the K, L, and M shells, respectively. Experimental verification of the Migdal effect and of the order of magnitude of the ionization probabilities predicted for the K and L shells was provided by studies of the x rays associated with the alpha decay of Po$^{210}$. Rough estimates of the ionization probabilities for the remaining shells may be made from Migdal's approximate formula, $P_1 = \frac{\Delta Z}{(Z-a)^2}$, where $\Delta Z = 2$ for alpha decay and $a$ is the screening constant for the shell under consideration. Using screening constants calculated from the Hartree self-consistent field for U$^{238}$ and from the americium spectra, one obtains single ionization probabilities of $4\%$ for the N shell, $6\%$ for the O shell, and $3\%$ for the P shell. Migdal's approximate formula gives ionization probabilities for the beta decay of Kr$^{85}$ only one-half to one-third the experimental values, indicating that the estimates are somewhat low but of the correct order of magnitude at least as far out as the N shell. Considerably less accuracy may be expected from the approximate formula for the O and P shells, since the deviation from hydrogen-like wave functions increases with the principal quantum number. Moreover, a marked change in the radial distribution of the 5f orbitals appears to occur in the region of americium; in that case the 6s and 6p orbitals must undergo compensating changes since they are screened by 5f electrons. The overlap between the wave functions of neptunium
and americium may therefore be considerably smaller than in hydrogenic atoms and the ionization probabilities correspondingly larger.

b. Inelastic Collisions

The maximum kinetic energy transfer between an electron and a 100-kilovolt heavy recoil is approximately 1 electron volt. Since this is well below the binding energy of outer electrons, neither the recoil nor the atoms of the absorber should be ionized in a simple collision process. More detailed theoretical analysis of the stopping of heavy particles likewise indicates that displacement of atoms rather than electron excitation is the primary mechanism of energy loss at velocities much smaller than the Bohr orbital velocity.\textsuperscript{49,50} Theoretical range-energy curves calculated on this assumption generally agree well with the experimental data for alpha-decay recoils as well as for slow recoils produced in nuclear reactions.\textsuperscript{12,51} It should be noted, however, that inelastic collisions may be relatively unimportant as a mechanism of energy loss but may nevertheless alter the electron shells of the recoils. Intense ionization has been observed along the paths of slow recoils in cloud chambers, although the energy loss per ion pair is several times that for alpha particles.\textsuperscript{52,53} Moreover, a better fit to the experimental data has in some instances been achieved by taking into account electronic stopping as well as atomic displacements in calculating range-energy curves for slow recoils.\textsuperscript{54} In any event, the maximum energy transfer to the electrons is large compared with the splitting between the electronic ground state and a number of excited levels observed in neptunium and plutonium spectra. The recoils would therefore be distributed among a variety of excited states even if ionization did not occur.

No systematic studies of the charge on alpha-decay recoils comparable to those on fission fragments have been reported. Essentially all the Np\textsuperscript{239} recoils in the present experiments were positively charged in air, as shown by the high collection efficiency in an electrostatic field. Larger charges might be expected in solid absorbers than in gases, for two reasons: First, an electron may be promoted to progres-
sively higher energy levels in successive collisions if the time between collisions is short compared with the radiative lifetime of the excited state.\textsuperscript{55} Ionization may then occur in condensed media after a series of collisions no one of which involves a large enough energy transfer to remove the electron from the ground state to the continuum. Secondly, autoionization may take place in a solid as a result of polarization of the surrounding medium by the moving ion.\textsuperscript{56} Since both of these processes become less probable as the charge on the recoil increases, it appears unlikely that all the outer electrons would be stripped off even in a dense solid.

To summarize the preceding discussion, the K, L, M, and N shells of the neptunium and plutonium recoils probably remain intact while the O and P shells may lose electrons by ionization or by excitation to higher bound states. The 5f shell should be partially filled and the 6d shell, which is almost coincident in energy with the 5f shell in this region of the periodic table, may also be partially occupied. The ground states of the singly ionized atoms would in addition have an unpaired 7s electron. Recent observations on transuranium elements by the method of atomic beams indicate that L-S coupling according to Hund's rule occurs within shells, whereas j-j coupling occurs between shells.\textsuperscript{57} The electronic structure of the recoils is therefore complicated by uncertainties concerning the coupling scheme as well as the number and configuration of the electrons. It is nevertheless interesting to estimate the effect on the angular correlation of a magnetic interaction involving Np\textsuperscript{239} recoils in vacuum for the most probable electron configurations.

The hyperfine-structure constant $\alpha$ may be calculated for the case of $n$ equivalent 5f electrons in the limit of pure L-S coupling assuming that Hund's rule is obeyed.\textsuperscript{38} Under these conditions, the magnetic interaction energy is

$$\hbar Q = 2 \mu_1 \mu_0 \sum_{l=0}^{j} \left[ \frac{j(j+1)+L(L+1)-S(S+1)}{2(j+1)} + \frac{2(2L-n^2) K}{n^2(2L-1)(2L-1)(2L+3)} \right]$$

$$K = L(L+1)(j(j+1)+S(S+1)-L(L+1)) \frac{3}{2j(j+1)} \frac{3}{4} \left[ (j(j+1)+L(L+1)-S(S+1)) \frac{3}{2j(j+1)} \frac{3}{4} \right]$$

where $L$, $S$, and $j$ are the quantum numbers of the total angular momentum, spin, and orbital angular momentum, respectively.
where $\mu_0$ is the Bohr magneton, $L$ is the total orbital angular momentum of the coupled electrons, $S$ is the total electron spin, and $l$ is the orbital angular momentum of the individual electrons. The value of $\langle r^{-3}\rangle_{5f^5}$, obtained from the self-consistent field for the uranium atom,$^{38}$ is $3.89/a_0^3$, where $a_0$ is the Bohr orbital radius. As discussed earlier, the estimated magnetic moment for the $5/2$ minus excited state of Np$^{239}$ is 1.4 $\mu_B$. Using these data in the formula for the interaction energy, one obtains values of $\bar{a}$ increasing from 274 Mc to 322 Mc for the $5f^n$ configurations as $n$ increases from 2 to 5. The corresponding values of $\Delta \nu$, the overall width of the hyperfine structure, vary from a minimum of 0.111 cm$^{-1}$ for $5f^5$ to a maximum of 0.232 cm$^{-1}$ for $5f^3$. The mean life of the intermediate state in the Am$^{243}_c$ cascade is $1.7 \times 10^{-9}$ second.

The condition $2\bar{a} \Delta \nu \tau_N > 1$ is therefore satisfied and transitions among the magnetic sublevels of the intermediate state may be expected. This is illustrated in Fig. 9, where the attenuation coefficient $\alpha$ for a static magnetic interaction with the electron shell in the $(5f^5)_{6}^{6}H_{7/2}$ ground state of Np$^{++}$ has been plotted as a function of $\alpha \tau_N$. The perturbation by the magnetic field of the electrons is large enough to destroy more than half of the Am$^{243}_c$ correlation at the estimated $\alpha \tau_N$ value of 0.55.

Similar results are obtained for other $5f^n$ configurations with $J > 2$. The hyperfine-structure constants for the recoil ions should actually be larger than the estimated values for the neutral atoms owing to decreased screening by the $5f$ electrons as the charge increases. Whether the addition of an unpaired 6d electron would increase or decrease the attenuation coefficient depends on the resultant $J$ for the configuration. The hyperfine-structure constant for a 7s electron, taken from the Am(II) spectrum, is $2 \times 10^4$ Mc. Any configuration involving an unpaired 7s electron should therefore reduce the correlation to the hard core.

Unpaired electrons in the inner shells would probably have a similar effect owing to the large values of $\langle r^{-3}\rangle$.

As indicated by the preceding estimates, a strong perturbation of the Am$^{243}_c$ correlation would be expected for most of the electron configurations one might reasonably postulate. The surprising feature of the angular correlation experiments is not that perturbations were ob-
Fig. 9. Attenuation coefficient $G_2$ for static magnetic interaction between a nucleus with spin 5/2 and an electron shell with resultant angular momentum of 5/2. The splitting between hyperfine levels is 

$$\omega_{FF'} = \frac{a}{2} \left[ F(F+1) - F'(F'+1) \right],$$

where $a$ is the hyperfine-structure constant.
served in vacuum, but that attenuation coefficients below the hard-core value were obtained in \( ^{239} \text{Np} \). One possibility is that partial spin re-orientation occurs in the source or at the surface, followed by a static interaction in vacuum. The alternative explanation appears to be a time-dependent interaction involving an electron shell that undergoes transitions in vacuum within \( 10^{-9} \) second of the alpha decay. The energy splitting between the \( 5f, 6d, \) and \( 7s \) orbitals in the region of neptunium is from 1 to 6 electron volts.\(^{59}\) Resonance radiation of this energy generally has a mean life longer than \( 10^{-9} \) second,\(^{60}\) indicating that interactions involving transitions only among the \( 5f, 6d, \) and \( 7s \) shells should be static rather than time-dependent over the lifetime of the intermediate state. Vacancies in the radon core presumably would have lifetimes shorter than \( 10^{-9} \) second, provided that electrons were available to fill the vacancies. This condition would be met in vacuum only if the inner electrons were lost and the outer electrons retained in the primary ionization process, while both the Migdal effect and inelastic collisions should preferentially remove the most loosely bound electrons. Of these two processes, the sudden change in nuclear charge appears to offer better prospects for perturbing inner shells, since it should make available about 70 electron volts of excitation energy.\(^{61}\) The attenuation coefficient for the \( ^{243} \text{Cm} \) correlation in vacuum is between 0.14 and 0.35, and is therefore probably above the hard-core value. This apparent difference between the behavior of neptunium and plutonium recoils in vacuum may be related to the difference in nuclear magnetic moments rather than to different electron configurations.

Vacancies in the electron shells of the recoils should be filled within \( 10^{-12} \) second in a metallic environment.\(^{62}\) The short electron recovery time may account for the improvement of the \( ^{243} \text{Am} \) and \( ^{243} \text{Cm} \) correlations in metals as compared with vacuum and nonmetallic absorbers. Conversely, recovery would be expected to proceed slowly in a plastic insulator such as Mylar, the environment which proved most unfavorable for the \( ^{243} \text{Am} \) correlation.

Neptunium and plutonium might be expected to show magnetic interactions even after the electron shell had de-ionized, owing to the
presence of unpaired 5f electrons in the ground state. The effect on
the angular correlation of a static magnetic interaction in a solid
depends on the anisotropy of the hyperfine structure and cannot be cal-
culated for alpha-gamma correlations because the positions of the
recoils in the lattice are not known. It is probable, however, that
the interaction in neptunium and plutonium would be time-dependent
rather than static in crystalline solids, since the paramagnetic spin
relaxation times may be extremely short. The perturbations observed
in Np$^{\text{237}}$ and Np$^{\text{239}}$ recoils in metallic environments indeed appear to be
time-dependent, as indicated by a comparison of the attenuation coeffi-
cients for the two isotopes. G$_2$ values of about 0.55 were obtained for
Np$^{\text{239}}$ recoils in metals, while G$_2$ was about 0.07 for Np$^{\text{237}}$ recoils under
comparable conditions. The nuclear moments of the intermediate states
in the two cascades are similar, and the electron shells and extra-
atomic fields should also be similar in the same absorber. The differ-
ence between the attenuation coefficients may therefore be related to
the difference between the lifetimes of the intermediate states. A
reasonable conclusion is that a time-dependent interaction occurs in
both cases, with a frequency large enough to destroy about half of the
correlation in a few millimicroseconds and most of the remaining half
during the next 100 millimicroseconds. The time-dependent character of
the perturbation cannot be related to de-excitation of the electron
shell or to the recoil motion, since both of these processes should be
completed in considerably less than 10$^{\text{-11}}$ second.

Fig. 10 shows the integrated attenuation coefficient G$_2$ for a
time-dependent electric or magnetic interaction with an intermediate-
state spin of 5/2 plotted against the product of the decay constant $\lambda$
and the mean life $\tau_N$. For a time-dependent magnetic interaction in a
liquid or solid with short spin-relaxation time, $\lambda$ is proportional to
$\omega^2 \tau_R S(S+1)$, where $\omega$ is the frequency corresponding to the magnetic
interaction energy $\Delta_A \omega I \cdot S$, $S$ is the resultant electron spin, and $\tau_R$
is the relaxation time. This interaction refers to ions displaying
"spin only" paramagnetism; presumably, however, the shape of the theo-
retical curve would be similar if the orbital contribution to the elec-
tronic magnetic moment were only partially quenched, as appears to be
Fig. 10. Experimental attenuation coefficients for $^{239}\text{Np}$ (open circles) and $^{237}\text{Np}$ (black dots) recoils in metal absorbers plotted along theoretical curve for time-dependent interaction.
the case for the actinides. Several comments may be made concerning
the experimental $Q_2$ values for $^{237}\text{Np}$ and $^{239}\text{Np}$ recoils in metals plotted
on the theoretical curve.

a. One would expect that the $\lambda$ values for $^{237}\text{Np}$ and $^{239}\text{Np}$ recoils
in the same absorber would be equal if the perturbation in both isotopes
resulted entirely from a time-dependent interaction in the ground state
of the electron shell after the recoils came to rest. The $\lambda$ values
 Corresponding to the experimental attenuation coefficients actually
appear to be about three times as large for $^{239}\text{Np}$ as for $^{237}\text{Np}$ in both
silver and aluminum. This suggests that the average interaction energy
may be larger during the first $10^{-9}$ second following alpha decay than
during the next $10^{-7}$ second, indicating a strong initial perturbation
associated with electron excitation or with extra-atomic fields at the
recoiling nucleus. A similar suggestion has been made by Heer and
Novey on the basis of a comparison between the delayed correlation in
a solid $^{241}\text{Am}$ source and integral correlations in even-even alpha
emitters with intermediate-state lifetimes of the order of $6 \times 10^{-10}$
second. It should be noted, however, that the attenuation coefficients
plotted for $^{237}\text{Np}$ in Fig. 10 represent anisotropies of 3 to 4%, which
are difficult to measure accurately. Relatively small experimental
errors in the $^{241}\text{Am}$ correlation may be responsible for the apparent
discrepancy between the $\lambda$ values for the two isotopes.

b. If the initial perturbation in $^{237}\text{Np}$ is neglected, the $\lambda$ values
for that isotope must represent an interaction of constant magnitude
occurring after the recoils come to rest and reach a stable electronic
state. $^{239}\text{Np}$ recoils presumably undergo a similar interaction after
stopping in silver and aluminum, since their electron shells and extra-
atomic environment should be similar to those of $^{237}\text{Np}$. The attenuation
coefficients of about 0.55 for $^{239}\text{Np}$ in silver and aluminum thus can be
attributed only partially to interactions in the moving ion and presumably
even less to perturbations occurring before the recoils leave the thin
sources.

c. If the magnetic interaction frequency is about $300 \text{ Me}$, as esti-
mated for $5^n$ configurations with $n$ from 2 to 5, a spin relaxation time
of the order of $10^{-10}$ second would be required to account for the observed
perturbations in metals. A relaxation time of $10^{-12}$ second or less was estimated for neodymium and samarium ions in aqueous solution from the absence of observable perturbations following Coulomb excitation.

d. The $\lambda$ value for $\text{Np}^{239}$ recoils is about twice as large in iron as in aluminum or silver. One might suspect that this discrepancy is related to the ferromagnetism of iron, although the nature of the interaction is not apparent.

II-B. Electric Quadrupole Interactions

1. Static Electric Interactions

The effect on the angular correlation of a static electric field depends on the symmetry as well as the gradient of the field at the intermediate nucleus. Randomly oriented fields of low symmetry might be expected at recoil nuclei in crystals because the ions come to rest at interstitial sites where the chemical bonding and crystal structure may differ from those of the normal compound. Alpha-gamma angular correlations in a series of even-even nuclei ranging from $\text{Ra}^{224}$ to $\text{Pu}^{238}$ nevertheless show perturbations apparently consistent with the action of a static electric field of axial symmetry. For this special case, the interaction may be visualized as a precession of the nuclear quadrupole moment about the axis of the field gradient with a fundamental precession frequency

$$\omega_0 = 0.15(eQ/A)(\partial E/\partial z)$$

for an intermediate-state spin of $5/2$, where $Q$ is the nuclear quadrupole moment and $\partial E/\partial z$ is the gradient of the field. The attenuation coefficient $G_2$ again exhibits a hard-core value which varies with the symmetry of the field but is never less than 0.20 provided that the microcrystals are randomly oriented. Static interaction frequencies ranging from 1100 to 2500 Mc would be required to produce the perturbations observed in $\text{Np}^{239}$ recoils in metals and in the thick oxide source.

2. Time-Dependent Electric Interactions

The local electric field at a nucleus in a nonviscous liquid is continually changing in a random manner owing to Brownian motion. Time-dependent interactions characterized by the correlation time of the liquid
might therefore be expected, and these have been observed in a number of aqueous solutions as well as in molten metals.\textsuperscript{6b} The decay constant \( \lambda \) for the time-dependent quadrupole interaction is proportional to \( \langle \omega_0^2 \rangle \tau_R \), where the relaxation time \( \tau_R \) corresponds to the correlation time of water, \( 10^{-11} \) second, for ions in dilute aqueous solutions. A series of delayed correlations in dilute acid solutions of Am\textsuperscript{241} has been reported by Krohn, Novey, and Raboy.\textsuperscript{10} Anisotropies decaying exponentially with a decay constant of 13.5 per microsecond in perchloric and hydrochloric acid solutions and 21 per microsecond in sulfuric acid were obtained in this study. In each instance the correlation extrapolated to zero delay time corresponded to an \( A_2 \) value of \(-0.36\), in agreement with the calculations for a mixing ratio \( \xi_{\alpha} \sim 0.22 \). Solutions of Am\textsuperscript{241} in dilute acetic acid failed to show the characteristic exponential decay, presumably because of complexing by the acetate ion which introduces a large field gradient at the neptunium nucleus. Two conclusions of interest in connection with the present study may be drawn from these experiments:

a. The calculated correlation coefficient \( A_2 = -0.36 \) depends on estimates of the phase shifts and mixing ratio of S and D alpha partial waves. Fortunately, the correlation function is relatively insensitive to variations of these parameters within the range of probable values indicated by recent experimental and theoretical work.\textsuperscript{25a, 25b} It is nevertheless significant that the delayed correlations in liquid sources provide an experimental lower limit of \( A_2 = -0.36 \) for the unperturbed Am\textsuperscript{241} correlation and presumably also for the Am\textsuperscript{243} correlation, which should involve approximately the same mixing ratio and phase shift. The attenuation coefficients in Tables I and II are therefore upper limits, and the apparent occurrence of values below the hard core cannot be attributed to errors in the calculated correlation.

b. Complexing of neptunium ion by perchlorate and chloride in the dilute acid solutions was assumed to be negligible. One might then expect that uncomplexed neptunium ions in pure water would also yield a decay constant of 13.5/\( \mu\text{sec} \). An even smaller decay constant might be predicted for the interaction in benzene, since the correlation time is
shorter than that of water and the medium is nonpolar. The gamma rays from Np$^{237}$ recoils in water and in benzene, however, were almost isotropic, indicating that the $\lambda$ values exceeded 80 per microsecond. Complexing of the neptunium in these liquids may account for the strong interaction.

A second type of time-dependent quadrupole interaction of concern in the recoil experiments involves field gradients through which the nucleus passes during its flight. In particular, the interpretation of the experiments in thin sources is complicated by the possibility that such interactions occur before the recoils escape from the deposit. As discussed previously, the experiments involving Np$^{237}$ and Np$^{239}$ recoils in metals indicate that considerably less than half of the correlation is lost before the recoils reach the surface although there may be a relatively strong initial perturbation associated with the alpha decay or the recoil motion. An interaction taking place during the flight of the recoil would not be detected in delayed correlations because the resolving time of available coincidence circuits is much longer than the time of flight of the ions. The delayed correlations in Am$^{241}$ nevertheless suggest that little or no initial perturbation occurs in aqueous solutions, since the anisotropies obtained appear to be close to the unattenuated value. A correlation equal or close to the theoretical value has also been reported for the 0-1-0 cascade in metallic Th$^{230}$; unfortunately, the lifetime of the intermediate state in this instance is not known. The perturbations of the 0-2-0 correlations in even-even nuclei were originally attributed to static electric fields, in which case the interactions must take place after the recoils come to rest. This interpretation, however, was based largely on the observation that each isotope yielded values of $G_2$ and $G_4$ corresponding to a single value of $\omega_{QN}$ for a static electric interaction. The attenuation coefficients for the even-even isotopes actually are equally consistent with either a time-dependent or a static electric field and exclude only magnetic fields arising in the electron shell. Further evidence of electric interactions was obtained from a comparison of the 0-2-0 and 0-4-2 cascades in a metallic Th$^{230}$ source; the values of $\omega_{QN}$ calculated from the attenuation coefficients for the two cascades are in the ratio predicted by Bohr and Mottelson$^{20}$ for the 2-plus and 4-plus
members of a rotational band. Perturbation of alpha-gamma angular correlations by extra-atomic electric fields thus has been demonstrated for a number of nuclei but it is not yet clear whether the interactions occur during or after the recoil.

3. Quadrupole Moments

In contrast to the magnetic moment, the quadrupole moment of a deformed nucleus is primarily a collective property and varies relatively little with the addition of one or two particles in the region far from closed shells. The intermediate states in the Am$^{241}$, Am$^{243}$, and Cm$^{243}$ cascades should therefore have quadrupole moments comparable to one another and to the ground-state moments of neighboring nuclei. Intrinsic quadrupole moments of 9.0 to 10.3 barns have been reported from Coulomb excitation of U$^{235}$, U$^{238}$, Np$^{237}$, and Pu$^{239}$. These values correspond to an average spectroscopic quadrupole moment of about 3.6 barns for nuclei with $I = K = 5/2$, as in the intermediate states of the americium and curium cascades. A quadrupole moment of 4.9 barns was calculated from the hyperfine structure of Am$^{241}$ and Am$^{243}$. The experimental data may be compared with a recent calculation by Mottelson and Nilsson in which the nuclear deformations were estimated by minimizing the total energy of odd-A nuclei in a deformed harmonic oscillator potential. The calculated deformations correspond to spectroscopic quadrupole moments of 4.0 to 4.5 barns for the rotational ground states in Np$^{237}$, Np$^{239}$, and Pu$^{239}$.

4. Extra-Atomic Electric Fields

Abnormally large electric field gradients may be present at a recoil nucleus both during its flight through an absorber and after it stops at an interstitial lattice site. Since the quadrupole moments of the recoil nuclei also are large, perturbation of the correlation by electric interactions might be expected in all the absorbers studied. The possibility of coupling to extra-atomic fields can be eliminated only in vacuum, and even here a quadrupole interaction may occur between the nucleus and field gradients arising in the excited electron shell. One conclusion to be drawn from the experiments is that a static electric interaction cannot account for the observed perturbations. There appears to be either a fluctuating
electric field gradient, a fluctuating magnetic field, or perhaps electric and magnetic fields acting simultaneously or successively. In the absence of more specific information, some suggestions may be made concerning the extra-atomic fields to be expected under the conditions of the various experiments.

(a) The order of magnitude of the field gradients encountered by a recoiling nucleus may be estimated from the average rate of energy loss, \( W/R = ZeE \), where \( W \) is the work required to stop the recoil or about 100 kilovolts, \( Ze \) is the nuclear charge, \( E \) is the average electric field opposing the recoil motion, and \( R \) is the recoil range or a few hundred angstroms. The collision diameter \( b = \frac{2ZZ'}{\mu v^2} \), where \( Z' \) is the atomic number of the absorber atoms, \( \mu \) is the reduced mass of the system of colliding particles, and \( v \) is the velocity. In an absorber consisting mainly of oxygen, the collision diameter is about 3.5 angstroms and the average field gradient \( q = \frac{W}{ZeRb} \) is of the order of \( 5 \times 10^{13} \) esu. An interaction of frequency \( \frac{1}{\hbar}(eQq) \) occurs between the nuclear quadrupole moment and the field gradient during the time of flight of the recoil; this corresponds to \( \omega_q \sim 100 \text{ Mc} \) for a quadrupole moment of 4 barns. In general, the minimum interaction frequency detectable in an angular correlation experiment is given by the condition \( \omega_T > 10^{-2} \), applying to a randomly oriented static magnetic field or electric field gradient. The field gradient at a recoiling nucleus presumably fluctuates rapidly in direction, in which case the minimum detectable frequency would be considerably larger than that for a static field. The estimated interaction of 100 Mc occurring over the time of flight of the recoils corresponds to \( \omega_T < 10^{-3} \). Thus it appears that field gradients several orders of magnitude larger than the estimated value would be required to perturb the correlation during the flight of the recoils. The possibility nevertheless exists that abnormally large field gradients may be present at a recoiling
nucleus owing to distortion of the electron cloud or other processes associated with the details of the motion. It is interesting to note that alpha-gamma angular correlations, at least in principle, provide a means of studying such processes through the dependence of the attenuation coefficient on extra-atomic fields.

(b) Large field gradients may be encountered by the recoil nucleus as it enters a vacuum or a nonpolar medium, owing to the rapid change in electric field strength across the surface of the oxide source. In that event, the perturbations of the Am$^{241}$ correlation observed in vacuum, air, and Mylar may be related to extra-atomic fields arising at the oxide surface rather than magnetic fields arising in the electron shell. A similar explanation would apply to the attenuation of the Cm$^{243}$ correlation in vacuum and air, but not to the disappearance of the anisotropy in oxide crystals. A rough estimate of the surface field gradient may be obtained by equating the work function to the average force on an electron multiplied by the distance over which the field falls to zero. For a work function of 5 electron volts, a field extending over atomic dimensions, and a recoil velocity of $10^7$ cm per second, the average field gradient would be of the order of $10^{14}$ esu, while the interval during which the gradient interacts with the nuclear quadrupole moment would be $10^{-15}$ second. These estimates correspond to $\omega \tau \sim 10^{-7}$, an action several orders of magnitude smaller than the minimum detectable limit of $10^{-2}$. It should be noted that the estimated average gradients at the recoil nucleus within an oxide absorber and in the region of decreasing field adjacent to the surface of the oxide in vacuum are comparable. The experiments in Am$^{241}$ and Am$^{243}$ indicate that little or no perturbation occurs as the ions traverse the thin oxide sources. No perturbation would then be expected as the recoils cross the surface into a vacuum or nonpolar medium unless the gradients adjacent to the surface showed considerably larger fluctuations from the average value than those within the oxide.
(c) According to current theories of radiation damage in crystals, a heavy particle displaces atoms or ions until its kinetic energy drops below the displacement energy, estimated at about 25 electron volts for metals. The remaining energy is then transferred to the lattice in the form of vibrations, producing a "temperature spike" or "hot spot" at the end of the path which cools rapidly as heat diffuses to the surroundings. Atomic rearrangements with relatively low activation energies occur in the hot spot, followed by reordering of the lattice as the temperature falls. Fluctuating rather than static extra-atomic fields might therefore be present even after the recoil energy is spent, owing to migration of neighboring atoms or of the recoil itself.

(d) All three metals included in the experiments as well as the oxides normally exhibit cubic structures, in which case the field gradient vanishes at regular lattice sites. The recoils, however, presumably occupy interstitial positions at which the field gradients may be abnormally large. Angular correlations involving the gamma-gamma cascade following K capture by In\(^{111}\) showed considerably less attenuation in silver than in metals with lower symmetry, even when the indium was produced in the silver lattice by an \((\alpha,2n)\) reaction. Diffusion of the indium recoils into regular lattice positions after the nuclear reaction was credited with the improved correlation in this instance. It appears improbable that the recoils from alpha decay would achieve regular lattice positions in times comparable to the lifetime of the intermediate state.

(e) The possibility of large field gradients at the nucleus generated by electrons in covalent bonds must be considered in neptunium and plutonium. Neptunium ions would be expected to hydrolyze in water, probably to NpOH\(^{+++}\). The asymmetrical distribution of the electrons in the Np-O bond may account for the larger perturbation of the Am\(^{241}\) correlation in pure water than in dilute acid solutions. Both Mylar and benzene are relatively resistant to radiation damage owing to the resonance stability of the benzene ring. Reactions between the neptunium recoils and impurities dissolved in these absorbers rather than free-radical reactions might therefore be expected. In particular, reactions with oxygen may yield a linear molecule containing...
Np-O bonds similar to those in NpO$_2^{++}$. As discussed by Eisenstein and Pryce,$^{68}$ four of the electrons in neptunyl ion occupy two highly directional hybrid orbitals composed of the 5f, 6d, and 7s orbitals of neptunium and the sp$_g$ orbital of oxygen; the resulting cylinder of charge along the O-Np-O axis gives rise to an axially symmetric field gradient large enough to account for the quadrupole coupling constant $\Delta \nu_q = 1.2 \times 10^4$ Mc. A time-dependent interaction of this magnitude could be responsible for the perturbation of the Am$^{241}$ correlation in benzene.

**II-C. Summary**

The experiments described in this report demonstrate that alpha-gamma angular correlations are perturbed in vacuum as well as in solids, liquids, and gases. The perturbation of the correlation in vacuum may result from an interaction between the nucleus and an electron shell which undergoes transitions within $10^{-9}$ second after alpha decay. Whether the electronic changes are associated with the sudden decrease in nuclear charge or with inelastic collisions in the source could not be determined, although the former possibility appears more consistent with current theory. Rapid recovery of the electron shell or rapid paramagnetic spin relaxation may be credited with the improvement of the correlation in metals as compared with oxides, plastics, and vacuum. Abnormally large electric field gradients encountered by the recoils within the radioactive films or adjacent to the surface may contribute to the perturbation, but simple estimates indicate that these gradients act over too short an interval and fluctuate too rapidly to affect the correlation. The perturbations in solids and liquids may be attributed either to electric field gradients of extra-atomic origin or to electric or magnetic fields generated by the atomic electrons. In any case the fields in solids as well as in liquids appear to be time-dependent rather than static.

The 278-kilovolt transition between the 5/2 plus and 3/2 plus states of Pu$^{239}$ consists of at least 3% E2 with the M1 and E2 components in phase; i.e., $\delta_r = (3/2 || 5/2) \geq +0.03$. A large negative anisotropy was demonstrated for the 228-kilovolt gamma ray in solid metals, in agreement with calculations for the 5/2-5/2-5/2 alpha-gamma cascade in the decay of Cm$^{243}$.
with S and D alpha waves in phase and $\beta_0^2 \leq 0.05$. The $\text{Cm}^{243}$ correlation was completely destroyed in oxide crystals. These observations suggest that alpha-gamma angular correlations designed to study nuclear properties should be performed in nonmagnetic metals rather than in insulators.

Much of the ambiguity encountered in interpreting the experiments is related to the $5f$ electrons of neptunium and plutonium. Large magnetic fields generated by the unfilled $5f$ shell may be expected even in the electronic ground states, while large electric field gradients also are probable owing to the tendency of $5f$ electrons to form asymmetric covalent bonds. These problems are characteristic of the actinides and would not arise in the alpha decay of thorium or lighter elements. In particular, the observation of radon or radium recoils in vacuum should indicate whether or not the radon core is affected by alpha decay. An additional advantage of such experiments in even-even thorium and radium isotopes is the possibility of distinguishing between electric and magnetic interactions by comparing the two attenuation coefficients for the 0-2-0 alpha-gamma cascade.

A second complicating factor in the experiments is the recoil motion, which alters the electron shells as well as the extra-atomic environment of the daughter nuclei. Since many of the effects of atomic collisions are transient and highly localized, they may not be demonstrated directly by the methods generally employed in radiation-damage studies. On the one hand this introduces unknowns into the interpretation of alpha-gamma angular correlations. On the other hand it suggests that such experiments, performed under properly controlled conditions, might be a useful supplement to the techniques presently used to explore radiation effects.
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