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Perturbation of α–γ angular correlation in a polycrystalline ^{241}Am source

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Abstract. Perturbation studies of the α–γ angular correlation for the (5486α) (59·6γ) ($\frac{5}{2}^- - \frac{5}{2}^- - \frac{5}{2}^+$) cascade have been carried out in a polycrystalline ^{241}Am sample using the time-differential technique. The exponential trend of the angular correlation coefficients $A_2(\bar{t})$ as a function of the mean time delay $\bar{t}$ shows a strong time-dependent behaviour of the interaction phenomena. This is consistent with the theory of Abragam and Pound for polycrystalline sources. The value of the relaxation constant λ_2, characteristic of the time-dependent perturbation, is found to be $\lambda_2 = (1{\cdot}81 \pm 0{\cdot}17) \times 10^9\ \mathrm{s}^{-1}$.

The time-integrated perturbation coefficient obtained is $\bar{G}_2(\bar{t} = 2{\cdot}4\ \mathrm{ns}) = 0{\cdot}19 \pm 0{\cdot}01$. Using the experimental λ_2 value, the spin-relaxation time τ_s for the 5f electron shell is calculated as $\tau_s = (1{\cdot}56 \pm 0{\cdot}63) \times 10^{-11}$ s. The validity of the condition $\omega_s \tau_s \ll 1$ in the present case strongly suggests that the time-dependent character of the angular correlation is carried primarily by magnetic interactions between the unfilled 5f electron shell of ^{237}Np and the magnetic moment of the nucleus and, secondarily, by electron excitation of the Np atom.

1. Introduction

The α–γ directional-correlation method plays an important role in nuclear structure studies due to the sensitivity of the correlation pattern to phase and amplitude mixtures of alpha waves with different angular momenta. However, due to large recoil energy (~ 100 keV) of the α decay, the angular correlation is often perturbed. This may lead to either attenuation or, in some cases, a complete smearing out of the angular correlation. The time-dependent interaction is manifested quantitatively by an attenuation factor

$$G_k(t) = \mathrm{e}^{-\lambda_k t} \tag{1}$$

of the angular correlation function

$$W(\theta, t) = \sum_k G_k(t) A_k P_k(\cos\theta) \tag{2}$$

where the symbols have their usual meaning. Most of the angular correlation measurements involving α particles, reported in the literature, have been performed to determine the attenuation coefficients both in odd and even mass heavy isotopes mainly by using integral-correlation techniques. Such experiments usually yield an attenuated correlation which is evidence of an interaction, but is insensitive to the details of time dependence of the interaction.

The present work describes a study on the interaction mechanisms in a polycrystalline americium source. The time-differential PAC technique has been employed for measuring the (5486α) (59·6γ) correlation in the decay of ^{241}Am. The study of this cascade is interesting because of the long half-life of the 59·6 keV level in ^{237}Np as well as large anisotropy of this cascade. As a prelude to the above mentioned studies the half-life of the 59·6 keV level was measured by the authors and is reported elsewhere (Garg *et al* 1971).

Similar measurements on the cascade were performed by Fraser and Milton (1954) in a solid source of ^{241}Am; by Krohn *et al* (1957) (using viscosity method) and recently by Günther and Parsignault (1967) in a liquid source. Flamm and Asaro (1963) performed integral correlation measurements in the decay of ^{241}Am, ^{243}Am and ^{243}Cm and investigated the perturbation behaviour as a function of the material into which the daughter nuclei recoil. The results of the integral correlation exhibit a strong attenuation for all material backings. Similar measurements (Orre *et al* 1970, 1971, Hesselink and Kampen 1971, Ansaldo *et al* 1969, 1970) have also been recently carried out on ^{226}Ra, ^{226}Th, ^{230}U, ^{241}Am and ^{249}Cf.

In this paper we report on (i) the time-dependent behaviour of the interaction, (ii) the time-integrated attenuation coefficients and (iii) the spin-relaxation time of the 5f electron shell in neptunium.

2. Experimental details

2.1. Source

^{241}Am was obtained from the Radiochemical Centre, Amersham, UK. The source was prepared by drying up Am $(NO_3)_3$ onto a 0·5 mm thick plane perspex backing. The thickness of the source was estimated to be less than 1 μm; it is presumed that the recoils stop within the source as their range in americium is of the order of 100 Å.

2.2. Apparatus and measurements

A schematic diagram of the experimental set-up and other relevant details are given elsewhere (Garg *et al* 1971). The two-detector (α particles in a Si surface barrier detector and γ rays in movable NaI (Tl)) time-differential coincidence system with fast leading-edge pickoff timing and a time-to-pulse height converter (TPHC) with a time span of 800 ns were used. The FWHM of the system was $\simeq 7{\cdot}5$ ns. The data were recorded on a Nuclear Data 2048-channel analyser. The differential nonlinearity of the system was found to be less than 3% over the entire 800 ns range and no correction was applied to this effect.

The time calibration of the system obtained by the pair–point method was 384 ± 3 ps per channel.

The α side was set to accept all α particles of energy greater than 5 MeV. The γ channel accepted an energy range of 50 to 70 keV. The α–γ delayed coincidence time spectra were recorded over 20 ks intervals at relative detector positions of 90°, 135° and 180°. A total of over 20 000 counts was collected at each angle at the coincidence peak.

2.3. Analysis of data

The finite solid angle correction for the gamma ray detector was made in the usual way; no correction was, however, applied for the α detector as the geometry was less than 1%.

The prompt resolution curve obtained with the system by recording coincidences between the 4·620 MeV α particles from ^{230}Th and the 68 keV γ rays closely resembles a gaussian shape. The measured angular correlation coefficients at each delay can then be expressed as (Bodenstedt *et al* 1961),

$$A_2(t_v) = \frac{\int_0^\infty e^{-\lambda t} A_2(t) \exp[-\pi\{(t-t_v)/2\tau\}^2]\, dt}{\int_0^\infty e^{-\lambda t} \exp[-\pi\{(t-t_v)/2\tau\}^2]\, dt} \tag{3}$$

where t_v is the delay from the centre of the prompt curve. The strong exponential trend of the experimental $A_2(t_v)$ against t_v curve leads us to assume that the coefficients follow a time-dependent behaviour of the form

$$A_2(t) = A_2(0)\, e^{-\lambda_2 t}. \tag{4}$$

Equation (3) can then be evaluated for different t_v delays.

Since the system has a finite time resolution, it follows that an event which occurs at time $t \neq t_v$ may be recorded in the channel corresponding to t_v. The effect of the finite time resolution was, therefore, taken into account by plotting the experimental $A_2(t_v)$ coefficients as a function of the mean time delay $\bar{t}$, which is obtained from the prompt resolution curve fitted to a gaussian shape,

$$\bar{t} = \frac{\int_0^\infty t\, e^{-\lambda t} \exp[-\pi\{(t-t_v)/2\tau\}^2]\, dt}{\int_0^\infty e^{-\lambda t} \exp[-\pi\{(t-t_v)/2\tau\}^2]\, dt} \tag{5}$$

The observed angular correlation function calculated at each delay t_v is a weighted average of (5486α) (59·6γ) double cascade and the interfering (5443α) (43·4γ) (59·6γ) triple cascade with intermediate radiation unobserved. A correction for the interference of the triple cascade was applied on the basis that it is isotropic and the feeding to the 59·6 keV level is 14% through the 43·4 keV M1+E2 strongly internally converted γ ray transition. The experimental $A_2(t_v)$ values were thus multiplied by a factor of 1·16.

The time-integrated expansion coefficient $\bar{G}_2(\bar{t})A_2(0)$ was obtained by numerical integration of the data at the three angles over a time span of $\bar{t}$ = 2·4 ns (ie t_v = 15 ns), subtracting the chance counts and normalizing the coincidence counts by the α and γ channel rates. The time-integrated attenuation coefficient $\bar{G}_2(\bar{t})$ was then obtained from the relation

$$\bar{G}_2(\bar{t}) = A_2(\bar{t})/A_2(0). \tag{6}$$

3. Results

The partial decay scheme of ^{241}Am is shown in figure 1. The γ ray transitions depopulating the $\frac{5}{2}^-$ intermediate state in ^{237}Np have been determined from internal conversion data to be E1 (Yamazaki *et al* 1966, Asaro *et al* 1960).

The $A_2(\bar{t})$ against $\bar{t}$ curve, deduced from the experimental $A_2(t_v)$ against t_v curve, as described in § 2.3, is shown in figure 2. The shape of the curve below $\bar{t} \simeq 0{\cdot}8$ ns is uncertain due to finite time resolution of the system. It is, however, noteworthy that up to a mean time delay of 0·8 ns practically the unperturbed anisotropy persists, whereas within the next nanosecond the $A_2(\bar{t})$ value gets attenuated roughly by a factor of ten. As suggested by Bodenstedt (1972 private communication), this may be attributed to a static interaction with a strong frequency distribution dominating at short initial delays.

A least squares fit of the curve beyond 0·8 ns to the function in equation (3) yields the value of the relaxation constant λ_2 which seems to indicate the influence of a time-dependent interaction at longer delays and this brings about almost complete isotropy

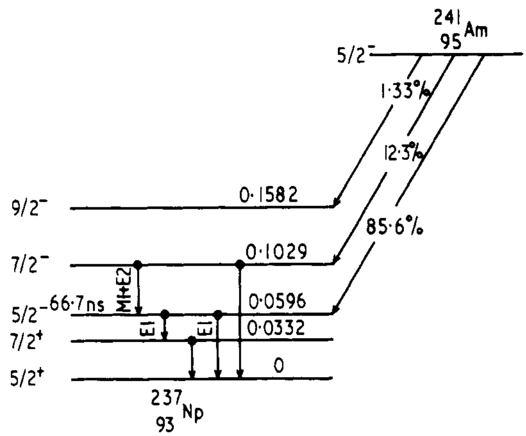

Figure 1. Partial decay scheme of ^{241}Am.

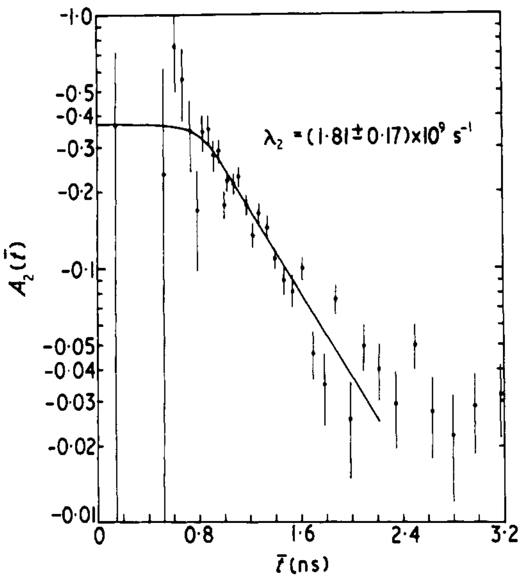

Figure 2. Plot of $A_2(\bar{t})$ against $\bar{t}$, the mean delay time.

of the perturbation at very long delays. This can be seen from the plot of figure 3 where the long tail of the attenuation lies well distributed about the isotropic value.

The present value of λ_2 along with the results of other authors is shown in table 1. It can be seen that the value of λ_2 is larger by a factor of about 50 times than that reported by Günther and Parsignault (1967) and Krohn *et al* (1957). For a $\frac{5}{2}^{-}$–$\frac{5}{2}^{-}$–$\frac{5}{2}^{+}$ (5276α) (74·6γ) cascade in ^{243}Am, Hutchinson (1967) reports $\lambda_2 = 1{\cdot}0 \times 10^8\ s^{-1}$ using a liquid source. This $\alpha\gamma$ cascade, being identical to the one studied here, should have electronic shell configuration and extra-atomic environments similar to that of ^{237}Np. The parameter λ_2 is characteristic of the physical and chemical environments of the source. Since perturbation interactions are highly diluted in liquid sources due to rapid disappearance of magnetic electronic states (Abragam and Pound 1953), as a result of

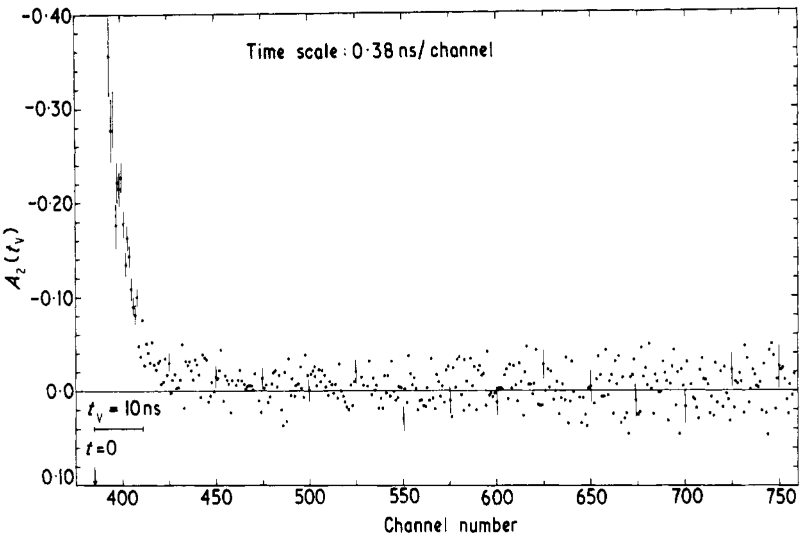

Figure 3. Experimental $A_2(t_v)$ against t_v curve (a few initial points are not shown).

Table 1. Values of relaxation constant λ_2 in ^{241}Am

Reference	Source form	λ_2 (s^{-1})
Fraser and Milton (1954)	Solid	12.6×10^7
Krohn, Novey and Raboy (1957)	Liquid film	$(1.3–2.1) \times 10^7$
Günther and Parsignault (1967)	Liquid	$(3.4 \pm 0.17) \times 10^7$
Present work	Solid polycrystalline	$(1.81 \pm 0.17) \times 10^9$

brownian motion, the present value of λ_2 obtained for a polycrystalline source of ^{241}Am is not unreasonable.

In order to obtain the time-integrated attenuation coefficient $\bar{G}_2(\bar{t})$, the unperturbed coefficient $A_2(0)$ should be known. Because of strong initial perturbations, the $A_2(0)$ value cannot, however, be determined with sufficient accuracy from the present measurement. Using liquid sources of varying normalities, Krohn *et al* (1957) obtained $A_2(0) = -0.36 \pm 0.02$ for the (5486α) (59·6γ) cascade in ^{241}Am. The theoretical value on the basis of Bohr, Fröman and Mottelson theory (Bohr *et al* 1955) is −0·36. Applying the Chaseman and Rasmussen (1959) ^{233}U correction, which arises from the interaction of the α particle with the quadrupole moment of the nucleus, the $A_2(0)$ value becomes −0·405. With a decoupling experiment on the aforesaid identical cascade in ^{243}Am, Falk *et al* (1967) reported experimental $A_2(0) = -0.404 \pm 0.010$ and, using a liquid source, Hutchinson (1967) obtained $A_2(0) = -0.41 \pm 0.02$. Accepting the value of $A_2(0) = -0.405$ and using our value of $\bar{G}_2(\bar{t})A_2(0) = -0.0768 \pm 0.0023$ we get $\bar{G}_2(\bar{t}) = 0.19 \pm 0.01$.

4. Discussion

Our measurements show a strong time-dependent attenuation of the type $G_2(t) = e^{-\lambda_2 t}$ as predicted by the theory of Abragam and Pound (1953) for polycrystalline sources. As is obvious from figure 2, the angular correlation coefficients or the correlation anisotropy decays to an almost isotropic value ($\simeq -0.02$) in $\bar{t} \sim 2.5$ ns. Using α–γ IMPAC technique, Ansaldo *et al* (1970) have shown that the experimental anisotropy in copper for the 59·6 keV state in ^{237}Np decays to a value below the hard core value in about 2 ns. Günther and Parsignault (1967) have also observed that the electron shell recovers itself after α decay in a time of about 2 ns. The strong initial perturbation may, therefore, be attributed to electron excitation (or even ionization of the daughter atom due to the Migdal (1941) effect), or to extra-atomic fields at the recoiling nucleus. The 5f electron shell of the Np ion would produce a strong magnetic field at the nucleus and give rise to a magnetic hyperfine interaction. Because of the long half-life of the intermediate state this interaction would result in a perturbation of the angular correlation, due to the coupling between the nuclear magnetic moment and the HF field. Such perturbations give rise to a time-dependent paramagnetic interaction. One might also expect the perturbation from the triple cascade owing to the excitation of the electron shell associated with the internal conversion of the unobserved radiations even though the lifetime of the 103 keV ($\frac{7}{2}^-$) state in ^{237}Np is very short $T_{1/2} = (8 \pm 4) \times 10^{-11}$ s (Wolfson and Dixon 1965) as compared to that of the 59·6 keV state. However, the radiation damage caused to the lattice structure by the α recoil may also give rise to a time-dependent perturbation. In this context, the magnetic relaxation phenomena (Aharoni 1969, Orre *et al* 1971) might play an important role in the interaction mechanism.

Flamm and Asaro (1963) and Hutchinson (1967) have also predicted a strong, initial short term interaction in the α–γ correlation of ^{243}Am. The time-dependent character of the perturbation cannot, however, be related to the de-excitation of the electron shell or to the recoil motion, since both of these processes should be completed in considerably less than 10^{-11} s.

We now make an approximate estimation of the spin-relaxation time. The ground state electron configuration for Np, which has a paramagnetic electronic structure, is probabily either $5f^4 6d 7s^2$ or $5f^5 7s^2$; the unpaired 5f electrons could give rise to perturbing magnetic fields which interact with the magnetic moment of the nucleus in the intermediate state. No reliable calculations of this internal field $B_i(0)_{5f}$ are available because of its strong dependence on the departures of the 5f wavefunctions from Russell–Saunders coupling (Wybourne 1965).

Considering the effect of the large spin–orbit coupling of the 5f shell, Günther and Parsignault (1967) define the paramagnetic correction factor for Np^{6+} ions as

$$\beta = 1 - \frac{g_J \mu_B (J+1)}{3kt} B_i(0)_{5f}. \tag{5}$$

From their value of $\beta = 1.68 \pm 0.11$ and $g_{5/2} = 0.762 \pm 0.060$, one gets

$$B_i(0)_{5f} = -(1.84 \pm 0.33) \times 10^6 \text{ G}.$$

This agrees well with the value of approximately 3×10^6 G in $NpAl_2$ and $NpCl_4$ observed by Stone and Pillinger (1968) in the Mössbauer spectra of the 59·6 keV γ ray in ^{237}Np.

For time-dependent magnetic interactions the theory of Abragam and Pound (1953) gives

$$\lambda_2 = a'\{1 - (2I+1)W(I12I/II)\} \tag{6}$$

where

$$a' = \tfrac{2}{3}\tau_s \omega_s^2 I(I+1)J(J+1) \tag{7}$$

$$\omega_s^2 = \frac{g^2 B_i^2(0)_{5f}\mu_2^2}{\hbar^2 J(J+1)} \tag{8}$$

τ_s is the spin-relaxation time of the 5f electron shell and ω_s is the magnetic interaction frequency. Using our experimental λ_2 value, we get for $I = \frac{5}{2}$, $a' = (4 \cdot 11 \pm 0 \cdot 38) \times 10^9 \, s^{-1}$ and $\tau_s = (1 \cdot 56 \pm 0 \cdot 63) \times 10^{-11}$ s at room temperature. An extremely short value of τ_s confirms the predictions of Low (1960) for the actinides.

Eisenstein and Pryce (1960) have interpreted the resonance spectrum in NpF_6 in terms of a magnetic interaction of the electrons with the Np nucleus. Their calculations give $\omega_s = 4 \cdot 47 \times 10^9 \, s^{-1}$. For a time-dependent magnetic interaction the condition $\omega_s \tau_s \simeq 0 \cdot 07 \ll 1$ is thus fulfilled. It is, therefore, more probable that the interaction in Np would be time dependent, rather than static, in crystalline fields.

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