Time-differential perturbed angular correlations using ^{161}Dy

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The 49–25.6-keV γ-γ cascade in ^{161}Dy has been used for making time-differential perturbed-angular-correlation measurements. For ^{161}Dy in Gd_2O_3 the internal fields are so large that only the hard-core correlation is seen. For ^{161}Dy in gadolinium metal above the Curie temperature the correlation function decays exponentially due to the presence of rapidly varying time-dependent fields. The half-lives of the 25.6- and 74.6-keV levels have been measured and found to be 30.0 ± 0.5 and 3.16 ± 0.05 nsec, respectively.

I. INTRODUCTION

The introduction of high-gain low-noise photomultiplier tubes such as the RCA 8575 has made it possible to make time-differential perturbed-angular-correlation (PAC) measurements with γ rays of much lower energy than before.[1-3] We report here the results of our attempt to make PAC measurements using the 49–25.6-keV γ cascade in ^{161}Dy using Dy in Gd_2O_3 and in Gd metal as sources.

II. THEORY

The relevant γ transitions are shown in Fig. 1. ^{161}Dy is produced by neutron irradiation of ^{160}Gd. The resulting ^{161}Gd decays to ^{161}Tb which has a convenient half-life of 6.9 days. The ^{161}Tb then decays by β emission, mainly populating the 74.6-keV level in ^{161}Dy. This level then is the starting point of the 49- and 25.6-keV cascade. The intermediate level has a half-life of 30 nsec. The spins and multipolarities involved give a theoretical correlation function

$$W(\theta) = 1 - 0.16 P_2(\cos\theta) . \quad (1)$$

Thus, with adequate time resolution PAC measurements should be possible over at least a narrow range of electric quadrupole or Larmor precession frequencies.

III. EXPERIMENTAL

A standard fast-slow coincidence arrangement was used. The detector for the 49-keV γ rays was an RCA 8575 coupled to a 5×25.4 mm NaI crystal. For the 25.6-keV γ rays, a 1×25.4 mm crystal on the RCA 8850 was used. The fast discriminators were of the constant fraction timing type. The time resolution was measured directly in the following way: The 25.6-keV γ detector was shielded from the ^{161}Dy source and positioned to detect radiation emitted from the face of the 49-keV γ detector. Prompt coincidences were then accumulated from 74.6-keV γ's for which the 28-keV iodine K x ray escaped, and the escape x ray itself. The prompt peak resulting from detection of these $74.6 - 28 \simeq 47$-keV events in coincidence with 28-keV x rays had a full width at half maximum (FWHM) of 5 nsec. Unfortunately, these escape x rays interfered with the PAC measurements, but they were filtered out satisfactorily by placing a thin metal shield in front of the 49-keV γ detector to absorb them. Another difficulty arose from the fact that the k-shell binding energy for ^{161}Dy is 53.8 keV. As a result, the 132–74.6-keV transition is almost totally coverted with concomitant emission of x rays of ~44 keV. The resulting pulse height spectrum taken with a germanium detector is shown in Fig. 2. The x-ray and 49-keV γ-ray peaks are, of course, not resolved by the NaI crystals. This would not be a serious difficulty

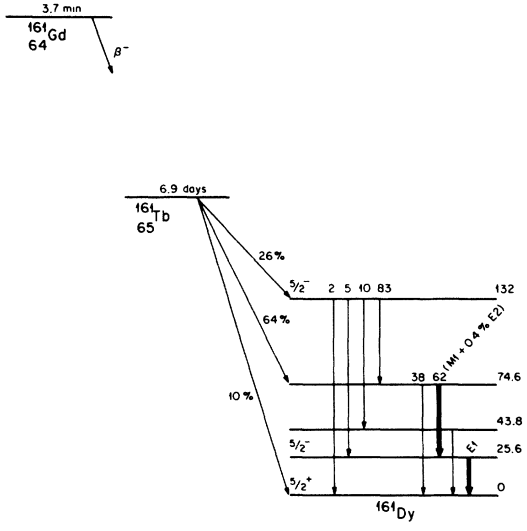

FIG. 1. Levels in ^{161}Dy following the decay of ^{161}Tb.

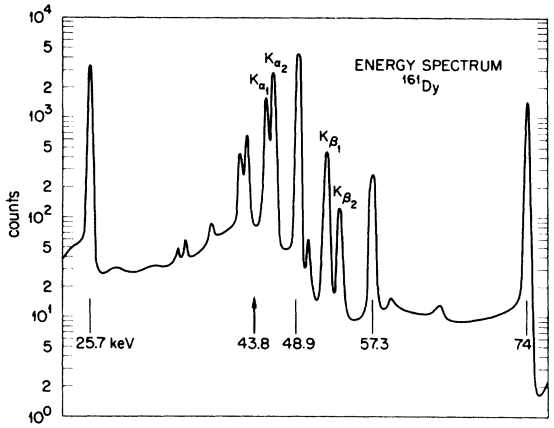

FIG. 2. Pulse-height spectrum of ^{161}Dy following the decay of ^{161}Tb taken with a germanium detector.

except that most of the x rays signal the formation of the 74.6-keV level so that they are in delayed coincidence with the 25.6-keV γ rays. By carefully placing the window of the single channel analyzer on the upper half of the 49-keV peak, however, we were able to eliminate most of these unwanted coincidences.

IV. LIFETIME MEASUREMENTS

As a preliminary to the PAC measurments the lifetimes of the 25.6- and 74.6-keV levels were measured. The 74.6-keV level lifetime was obtained by setting both single channel analyzers on the 49-keV peaks. In this way, an x ray signaled the formation of the level and a 49-keV γ ray signaled its decay. Both detectors served to "start" and "stop" the time interval measurement with the resulting spectrum shown in Fig. 3. The lifetime measured from the slope of this curve is 3.16 $\pm$ 0.05 nsec. The lifetime of the 25.6-keV level obtained from 49–25.6-keV coincidences was found to be 30.0 $\pm$ 0.5 nsec. Both measurements are in

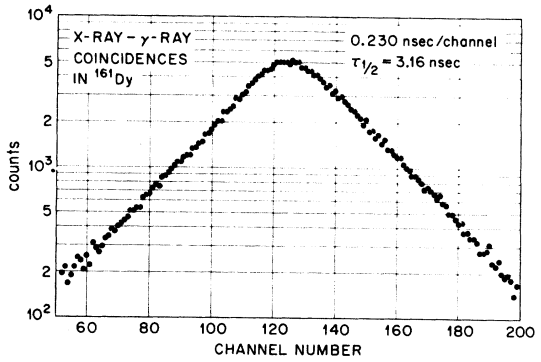

FIG. 3. Decay curves for 49-keV γ rays in coincidence with K x rays from ^{161}Dy.

agreement within experimental accuracy with the most recent previous measurments.[4-7]

V. PAC MEASUREMENTS

A. ^{161}Dy in gadolinium metal

Gadolinium metal has a Curie temperature (T_C) of 293 °K. Mössbauer measurements of ^{161}Dy in gadolinium metal near $T = 0$ °K reveal a magnetic hyperfine field at the ^{161}Dy nucleus of 7×10^6 Oe.[8] Perturbed-angular-correlation measurements have also been made using ^{111}Cd^m as a dilute impurity in Gd metal.[9] At room temperature the spectrum was produced by an axially symmetric electric field gradient with $e^2qQ \sim 25$ MHz. This fairly small value is characterisitic of results found for ^{111}Cd^m at substitutional sites in hcp lattices in which the c/a ratio does not deviate by a large amount from the ideal value of 1.633. Measurements have also been made using ^{181}Ta as a probe.[10] In this case a fairly large asymmetry parameter ($\eta = 0.3$) was found. However, in this case the spectrum was rapidly damped, indicating that a substantial number of Ta ions were not at regular substitutional sites.

On the basis of these results, using the value $g = 0.234$ for the 25.6-keV level in ^{161}Dy, we would expect near $T = 0$ a Larmor precession frequency $\omega_L \sim 9\times10^9$ rad/sec giving a period $\tau \sim 0.7$ nsec for the oscillations in a PAC spectrum. The amplitude of these oscillations would clearly be attenuated due to the finite time resolution of our apparatus so that we would expect to see only the "hard-core" value of the correlation function. Above T_C, however, it might be possible to observe oscillations in the correlation function resulting from the expected axially symmetric electric field gradient.

A thin gadolinium foil enriched to ~99% ^{160}Gd was irradiated and used for the source. Data were taken with the same source at 301 °K, which is slightly above the Curie temperature, and near liquid-nitrogen temperature. As expected, at the low temperature only the time-independent hard-core anisotropy of the correlation function was observed.

At the higher temperature a slowly varying time-dependent anisotropy was observed. The data were reduced to the form

$$\tfrac{1}{2}A_{22}G_{22}(t) = \frac{N(\pi, t) - N(\tfrac{1}{2}\pi, t)}{N(\pi, t) + 2N(\tfrac{1}{2}\pi, t) - 3C}\ , \qquad (2)$$

where $N(\theta, t)$ is the number of counts collected at the angle θ as a function of time and C is a background constant. The results are shown in Fig. 4. Nonlinear least-squares fits were made to the two sets of data; the results are discussed below.

a. Low-temperature spectrum. The measured

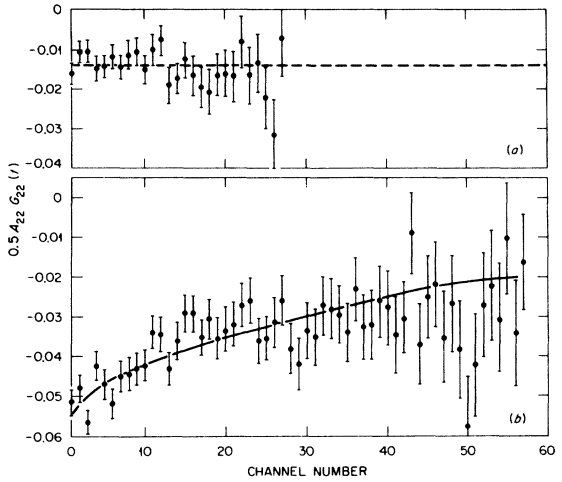

FIG. 4. PAC spectra of ^{161}Dy in gadolinium metal at (a) $T = 90\,^{\circ}$K and (b) $T = 301\,^{\circ}$K. The time scale in (b) is 1.4 nsec/channel.

hard-core correlation was found to be

$$\tfrac{1}{2}A_{22}G_{22}(t) = -0.014 \pm 0.001\ ,$$

where the error reflects only the statistical uncertainty. This is exactly the value expected for $A_{22} = -0.16$ after correcting for solid angle effects. This indicates that the measured asymmetry is not distorted by source thickness effects or by the unresolved K x rays seen by the detectors.

b. High-temperature spectrum. The high-temperature data cannot be fitted using the function resulting from a static electric quadrupole or magnetic hyperfine interaction even if a wide distribution of frequencies is assumed. The shape of the spectrum appears instead to result from some kind of relaxation process. The effects of such processes on PAC spectra have been discussed by a number of authors.[11-13] Several different models for describing the relaxation process have been used, all of which predict an exponential decay of $G_{22}(t)$ for high relaxation rates.

We have, therefore, fitted the spectrum with a function of the form

$$\tfrac{1}{2}A_{22}^{\text{eff}}\, e^{-\lambda_2 t}\ ,$$

with the resulting values

$$A_{22}^{\text{eff}} = -0.1 \pm 0.01\ ,$$

$$\lambda_2 = (1.2 \pm 0.1) \times 10^7\ \text{sec}^{-1}\ .$$

The effective value of A_{22} is only 60% of the theoretical value. Since excellent agreement between measured and calculated hard-core values were found at the low temperature this low value indicates that a large number of the gadolinium ions are at sites where very large quadrupole interactions are present with the resulting rapid decays being attenuated by the finite time resolution of our apparatus.

If the PAC spectrum is presumed to result from an electric quadrupole interaction modified by spin-lattice relaxation, then the decay constant is proportional to the product $(e^2qQ)^2$ and "the spin-correlation time"[11] τ_c. Unfortunately, these two factors can be separated only if measurements can be made at low temperatures where the relaxation effects have vanished.

B. ^{161}Dy in Gd_2O_3

Several Mössbauer measurements using ^{161}Dy as a source in Gd_2O_3 have been reported.[14-16] Interpretation of the results has been hampered by the lack of an absorber with a narrow unsplit absorption line.

Mørup and Trumpy[16] interpreted their results as being due to a unique electric quadrupole splitting with $e^2qQ = 485$ MHz at room temperature. On the basis of this result, assuming an axially symmetric electric field gradient, the fundamental period in the time spectrum obtained in a PAC measurement would be

$$\tau = 2\pi/\omega_0 \sim 14\ \text{nsec}.$$

This should be observable with our apparatus, even when considerable allowance is made for attenuation due to finite time resolution and a fairly wide distribution of electric field gradients. Surprisingly, however, at room temperature only the hard-core correlation was observed as in the case of the Dy and Gd metal at low temperature.

VI. CONCLUSION

We have observed time-differential perturbed angular correlations using the 49-26-keV cascade in ^{161}Dy. For Dy in Gd_2O_3 only the hard-core correlation was observed but for Dy in Gd metal above the Curie temperature a large time-dependent anisotropy was seen. The anisotropy decays exponentially with a time constant

$$\lambda_2 = (1.2 \pm 0.1) \times 10^7\ \text{sec}^{-1},$$

indicating the presence of a rapidly fluctuating hyperfine field. The constraints of time resolution and intermediate-state lifetime combined with the large hyperfine fields in Dy limit the application of PAC to this isotope. Further measurements are in progress using the mixed fluoride $DyGdF_6$ which has a much narrower Mössbauer emission line than Dy in Gd_2O_3 and using Dy in Gd metal just below T_C where the Larmor precession frequency is small enough to be observable.

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