THE
PHYSICAL REVIEW

A journal of experimental and theoretical physics established by E. L. Nichols in 1893

SECOND SERIES, Vol. 138, No. 4B 24 MAY 1965

Perturbed Directional Correlation in Tantalum-181†

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(Received 6 January 1965)

The effect of a static quadrupole interaction on the 133-482-keV gamma-gamma angular correlation in Ta^{181} has been studied by the delayed-coincidence method. Using a source of Hf^{181} in polycrystalline hafnium metal, the anisotropy was measured as a function of the time between the formation and decay of the 482-keV state with a fast-slow coincidence spectrometer. The anisotropy as a function of time clearly revealed fluctuations arising from the reorientation of the nuclear spin due to coupling of the quadrupole moment of the 482-keV state with the electric-field gradients of the crystals. The interaction strength evaluated on the basis of the theory for axially symmetric gradients was $\omega_0=299\pm8$ Mc/sec. Evidence for the presence of a rhombic (nonaxially-symmetric) electric interaction was observed in the departure of the results from the predictions for the axially symmetric case. The results are, however, in agreement with the theory for rhombic interactions if the electric field gradient in the hafnium crystals is assumed to have an asymmetry parameter $\eta=0.30\pm0.03$, and in addition, if a normal frequency distribution of full width at half-maximum equal to 10% is used to describe the variation of the field gradient from site to site. Further, using the evidence from Coulomb excitation for the strong deformation of the tantalum nucleus, we determined the quadrupole moment of the 482-keV state to be $Q=(2.53\pm0.10)\times10^{-24}$ cm^2. This allows the calculation of the field-gradient magnitudes in metallic hafnium, which yields $V_{zz}=\pm(5.18\pm0.27)\times10^{17}$ V/cm^2, $V_{yy}=\mp(3.37\pm0.19)\times10^{17}$ V/cm^2, and $V_{xx}=\mp(1.81\pm0.25)\times10^{17}$ V/cm^2. Incidentally, the half-life of the the 482-keV state was remeasured to be 10.4 ± 0.2 nsec, consistent with prior determinations.

INTRODUCTION

THE determination of electric and magnetic moments of excited states of nuclei has not proceeded as swiftly as other aspects of the spectroscopy of such states, principally because of the experimental difficulties involved. Investigations of the interaction of the static nuclear electric-quadrupole moment with its environment has been particularly difficult because of the impossibility of providing sufficiently strong electric-field gradients in the laboratory. In a few cases, the crystalline environment is suitable for such studies; for excited nuclear states, the perturbations of the nuclear alignment due to such interactions may be detected by means of angular-correlation experiments and Mössbauer studies. Theories have been worked out for most cases of interest.

For ordinary gamma-gamma directional-correlation experiments, the information to be gained is restricted to the multipole order of the radiation in the decay of the excited nuclear states, and to the spins of the states. If the resolving time of the apparatus is long compared to the mean life of the intermediate state, the experiment is an integral experiment. Integral experiments usually yield an attenuated correlation which is evidence of an interaction, but the details of the interaction are inevitably lost. Differential experiments, in which events are accepted on a time scale comparable to or shorter than the mean life of the intermediate state, are capable of distinguishing between electric and magnetic interactions. Measurements of the strengths of such interactions from the perturbation of the angular correlation lead to direct information concerning the nuclear moments, provided the properties of the nuclear environment are known. As yet, only a few differential experiments on the static electric-quadrupole interaction have been done. This paper reports a differential angular-correlation measurement on the 133–482-keV gamma-gamma cascade in Ta^{181}. Preliminary results

† This work was performed with support from the U. S. Atomic Energy Commission.
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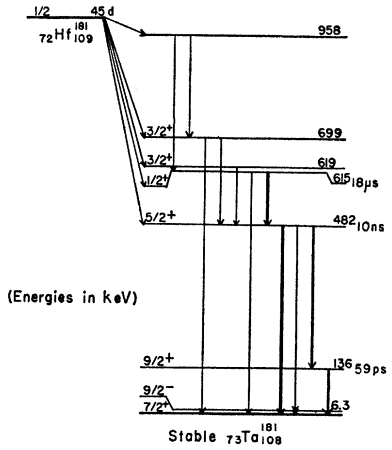

FIG. 1. Decay scheme of Hf^{181}.

have been previously reported.[1] A partial decay scheme is shown in Fig. 1.

EXPERIMENTAL METHOD

The differential correlation was measured with a two-counter scintillation spectrometer using $1\frac{1}{2}$-in. by 1-in. NaI(Tl) crystals coupled to 6810-A photomultiplier tubes. A fast-slow arrangement allowed energy discrimination and fast time-to-height conversion concurrently with a resolution of about 4.0 nsec. The source consisted of polycrystalline natural Hf metal, sized between U. S. Series 140 and 200 mesh screens, which had been irradiated in a reactor to produce the desired Hf^{181}. The source-to-counter distance was 5 cm, and the source strength was such as to produce a true-to-accidental ratio of 80 at the peak of the delayed-coincidence curve. Data were taken at 180 and 90° in alternate runs of 2-h duration. No serious drift problems were encountered over a total running time of the order of one thousand hours.

As a check on the apparatus, the lifetime of the intermediate state in Ta^{181} at 482 keV was measured by the usual delayed-coincidence technique, using 133–482-keV gamma pairs from a liquid source. The resulting half-life of 10.4 ± 0.2 nsec is in agreement with previous measurements of this well-established lifetime. As a further check, the anisotropy was measured using a liquid source by dissolving the polycrystalline metal in hydrofluoric acid. Our results agree with those of a similar experiment by McGowan.[2]

RESULTS AND DISCUSSION

The differential anisotropy of the 133–482-keV gamma-gamma cascade was measured with the results shown in Fig. 2. Finite-time resolution was taken into account by correcting the data for accidentals and by folding the prompt resolution curve into the theoretical curve. Finite-space resolution in the correlation has been accounted for by modifying the theoretical curve in the usual way to correct for the nonzero solid angle accepted by the detectors. Because of the suggestion of periodic behavior in the results, the data were first analyzed in terms of the theory of Abragam and Pound[3] for an axially symmetric field gradient as the nuclear environment. This yields a quadrupole precession period of 21.0 ± 0.5 nsec which corresponds to an interaction strength $\omega_0=3eQV_{zz}/2I(2I-1)\hbar=299\pm8$ Mc/sec. The obvious divergence of the experimental data from this simple theory is most likely due to two causes: (a) a nonaxially symmetric electric field gradient, and (b) a frequency spectrum resulting from a nonunique value of the gradient from nucleus to nucleus. These possibilities can result from nuclear-recoil effects and imperfect crystal structure. We first attempted to take account of the possibility of a nonaxially symmetric field gradient by the inclusion of the asymmetry parameter $\eta=(V_{xx}-V_{yy})/V_{zz}$ into the theory in the way suggested by Matthias, Schneider, and Steffen.[4] The best fit was obtained for the value $\eta=0.30\pm0.03$ as shown in Fig. 3, although the agreement still was not completely satisfactory.

We next included the effect of a variation in field gradient from site to site. Following a further suggestion of Matthias, Schneider, and Steffen,[4] we used a frequency-averaged attenuation coefficient

$$\bar{g}(\omega_0 t)=\int g(\omega t)P(\omega-\omega_0)d\omega \Big/ \int P(\omega-\omega_0)d\omega,$$

choosing a normal frequency distribution

$$P(\omega)d\omega=(1/(2\pi)^{1/2}\sigma)\exp[-(\omega-\omega_0)^2/2\sigma^2]d\omega.$$

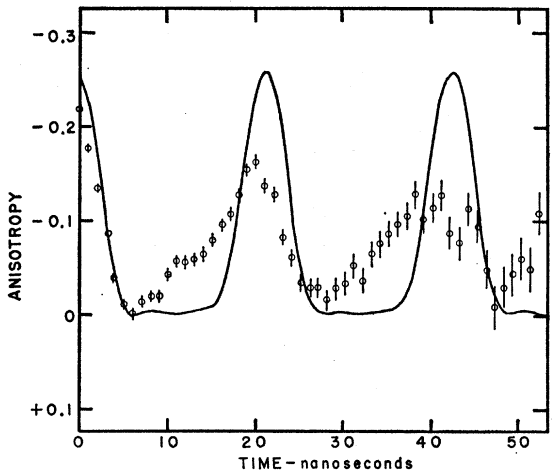

FIG. 2. The anisotropy of the 133–482-keV gamma-gamma cascade obtained with a polycrystalline source. The solid curve is the theory of Abragam and Pound, assuming a static interaction with an axially symmetric electric-field gradient.

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[2] F. K. McGowan, Phys. Rev. 93, 471 (1954).
[3] A. Abragam and R. V. Pound, Phys. Rev. 92, 943 (1953).
[4] E. Matthias, W. Schneider, and R. M. Steffen, Phys. Letters 4, 41 (1963).

With σ chosen so that the full width at half-maximum of the distribution is 10%, we get the very satisfactory fit shown in Fig. 4. A search for simultaneous best values of $\delta=\sigma/\omega_0$, ω_0 and η did not alter the values quoted above.

Although the requirement of a frequency distribution is reasonable, the reason for a nonaxially symmetric field gradient as the nuclear environment is unknown, since hafnium metal has a hexagonal close-packed (hcp) structure at temperatures below 1760°C. A recent experiment[5] with single-crystal sources emphasizes the dilemma which arises from this unexpected effect, since no nonaxially symmetric gradient is required to fit the data. It may be, however, that the particular angles used in the single-crystal experiment are such that the anisotropy would be insensitive to such effects.

Evidence from spectroscopy and from Coulomb excitation experiments indicates that Ta^{181} belongs to the class of highly deformed nuclei with energy levels which result from the excitation of collective rotations. In such a case, the spins of the rotational levels are given by $I=K, K+1, K+2, \cdots$, where K is the projection of $\mathbf{I}$ on the symmetry axis of the nucleus. The quadrupole moment Q of any of these states in odd-A nuclei is simply related[6] to the "intrinsic" quadrupole moment Q_0 of the nuclear shape in the ground state,

$$Q=Q_0\frac{3K^2-I(I+1)}{(I+1)(2I+3)}$$

and Q_0, in turn, can be determined from Coulomb-excitation reduced-transition probabilities between any pair of such a rotational band,

$$B(E2;I_i\rightarrow I_f)=(5/16\pi)e^2Q_0^2(I_i2K0|I_i2I_f0)^2.$$

Using the best available Coulomb excitation results[7] for the 301-keV transition in the $K=\frac{7}{2}$ band of Ta^{181} and assuming the 482-keV level has the parameters $I=\frac{5}{2}$, $K=\frac{5}{2}$, we obtain for the quadrupole moment of this level, $Q=(2.53\pm0.10)\times10^{-24}$ cm^2. Having both ω_0 and Q for this state, we calculate the absolute value $|V_{zz}|=2I(2I-1)\hbar\omega_0/3eQ$, and from the definition of η and the fact that $\nabla^2V=0$, we determine

$$V_{zz}=\pm(5.18\pm0.27)\times10^{17}\ \text{V/cm}^2,$$
$$V_{yy}=\pm(3.37\pm0.19)\times10^{17}\ \text{V/cm}^2,$$
$$V_{xx}=\pm(1.81\pm0.25)\times10^{17}\ \text{V/cm}^2,$$

to be the electric-field gradients in hafnium metal. A similar calculation has recently been carried out for hafnium ammonium hexafluoride.[8]

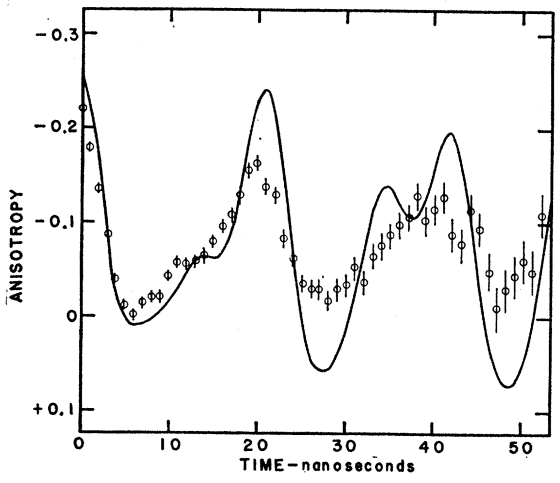

FIG. 3. Comparison of the measured anisotropy with a rhombic interaction using an asymmetry parameter $\eta=0.30$.

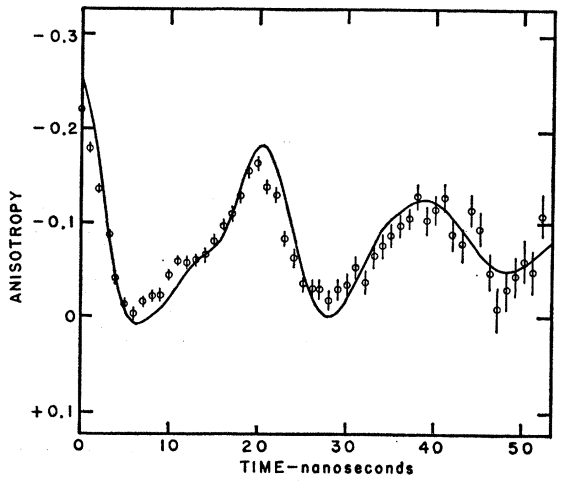

FIG. 4. Comparison of the measured anisotropy with a rhombic interaction including a normal frequency distribution having a full width at half-maximum of 10%.

ACKNOWLEDGMENTS

The authors wish to express their gratitude for the inestimable aid gained from discussions with Professor Rolf M. Steffen of Purdue University. In addition, we wish to thank Dr. R. M. Steffen and Dr. W. Schneider for certain computer programs from which the theoretical curves were calculated. Thanks are also due to Dr. E. A. Yunker for his continuing encouragement.

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