

Angular Distribution of Gamma Rays from Coulomb Excitation of Pt and W†

W. I. GOLDBURG* AND R. M. WILLIAMSON
Duke University, Durham, North Carolina

(Received January 30, 1957)

Data on the unresolved 330- and 358-keV gamma rays from thin natural platinum bombarded by 2- to 4-MeV protons show that the anisotropy of the angular distribution agrees closely with first order Coulomb excitation theory. The thick-target anisotropy of the 114-keV gamma rays from natural tungsten was found to be much more isotropic than the theory predicts. This result can be explained by assuming an extranuclear interaction between the excited tungsten nuclei and electric field gradients within the tungsten lattice.

INTRODUCTION

THE original semiclassical theory of Coulomb excitation¹ has been shown experimentally to be inadequate in its description of the incident energy dependence² and the angular distribution³ of gamma rays. More refined theoretical considerations, which give better agreement with observation, have since been published.⁴⁻⁶ We wish to describe measurements of the incident energy dependence of the anisotropy of the unresolved 330- and 358-keV gamma rays from a thin platinum target Coulomb-excited by protons of 2 to 4 MeV. Complete angular distributions of these gamma rays have been reported by Stelson and McGowan,³ who used thick platinum targets. The major experimental problem in gamma-ray measurements is the determination of the background components in crystal scintillator spectra. Therefore, it is worthwhile to report angular distribution data which were obtained with considerably different experimental conditions than those of Stelson and McGowan. Also, our thin-target data may be compared directly with theory, but this is not a large advantage because thick-target corrections are small.^{3,4}

We have also remeasured, with more precision, the thick-target anisotropy of the 114-keV line in natural tungsten.² This gamma ray is the sum of contributions of the many isotopes of natural tungsten. Most of the photopeak intensity comes from the 0-2-0 transition in the three even-even isotopes.^{7,8} Our data show an attenuation of the anisotropy, which appears to be a result of the interaction between the quadrupole moment of tungsten nuclei and the electric field gradients existing within the tungsten lattice.

† This work was supported by the U. S. Atomic Energy Commission.

* Present address: Department of Physics, Carnegie Institute of Technology, Pittsburgh, Pennsylvania.

¹ K. Alder and A. Winther, *Phys. Rev.* **91**, 1578 (1953).

² W. I. Goldberg and R. M. Williamson, *Phys. Rev.* **95**, 767 (1954).

³ F. K. McGowan and P. H. Stelson, *Phys. Rev.* **99**, 127 (1955).

⁴ K. Alder and A. Winther, *Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd.* **29**, No. 19 (1955), also CERN Report T/KA-AW-4, October, 1955 (unpublished).

⁵ Biedenham, Goldstein, McHale, and Thaler, *Phys. Rev.* **101**, 662 (1956).

⁶ Breit, Ebel, and Russell, *Phys. Rev.* **101**, 1504 (1956).

⁷ McClelland, Mark, and Goodman, *Phys. Rev.* **97**, 1191 (1955).

⁸ McClelland, Mark, and Goodman, *Phys. Rev.* **93**, 904 (1954).

The proton beam from a Van de Graaff accelerator was collimated to 1° and the diameter of the target spot was $\frac{3}{8}$ in. The last collimating slit was followed by an antiscattering slit. The target chamber consisted of a 2-in. diameter, 3-in. deep cylindrical brass chamber with $\frac{1}{32}$ -in. walls. The chamber was lined with 0.002-in. Ta and was insulated and electrostatically shielded from the collimators. Targets were supported by a thin brass ring. This ring was free to rotate about an axis perpendicular to the beam. The 0.0001-in. Pt foils, backed by 0.005-in. W discs, were mounted on the ring. When turned 45° with respect to the incident beam, these Pt foils had thicknesses which produced 310-keV and 220-keV energy loss for 2-MeV and 4-MeV protons, respectively. The target assembly and the scattering chamber were electrically connected for purposes of current integration. The gamma counter was a 1.5-in. diameter, 0.5-in. thick NaI(Tl) crystal mounted 2 in. from the axis of the chamber and centered to within 0.010 in. relative to the axis of the chamber. Appropriate absorbers were placed in front of the detector, and the pulse-height spectrum was recorded by a 10-channel analyzer.

PROCEDURE

The procedure for the platinum measurements was to record at each proton energy the gamma-ray intensity at counter angles of 270°, 0°, and 90° and target angles of plus and minus 45°. These angles are measured relative to the incident beam. When the settings were such that the gamma rays did not pass through the 0.0005-in. tungsten backing, an extra 0.007 in. of tantalum was placed in front of the detector to provide the equivalent absorption (this absorption was about 5% for 350-keV gammas). When the tungsten backing was turned toward the incident beam the observed radiation in the 350-keV region consisted of bremsstrahlung, slit background, and machine background. It was this spectrum from the tungsten backing alone that was actually used in making the background subtraction. This procedure is valid if (a) the proton beam is sufficiently steady that a target measurement and a corresponding background measurement take about the same time, and (b) the bremsstrahlung produced by the thin platinum foil plus backing is about equal to that produced by the beam striking the

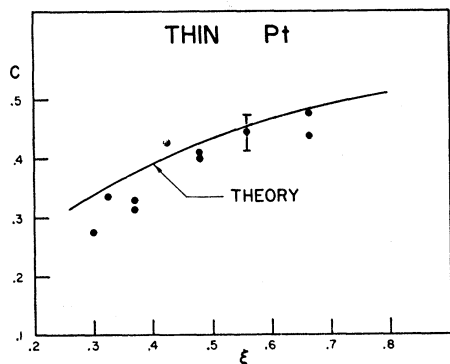


FIG. 1. The measured anisotropy of the unresolved 330- and 358-keV gamma rays from proton bombardment of thin natural platinum. The solid line corresponds to the first order theory of Alder and Winther.⁴ The theoretical curve has been corrected for the solid angle subtended by the counter and for finite target thickness. The energy dependent parameter ξ is defined as $\xi = \eta_f - \eta_i$, where $\eta_{f,i} = Z_1 Z_2 e^2 / \hbar v_{f,i}$. Here $Z_1 = 1$, $Z_2 = 78$, and the subscripts on v refer to the average proton velocities in the target before and after excitation.

backing alone. Both conditions were fulfilled, the latter by virtue of the fact that the atomic numbers of platinum and tungsten are about the same, resulting in their cross sections for bremsstrahlung production being approximately equal. In this experiment the areas under the bremsstrahlung spectrum were 8% to 20% of the corresponding photopeak areas. Gross changes were made in the proton beam position and therefore in the target spot illumination. These changes indicated that the uncertainties due to beam position, current integration, and background from collimators were less than 2% for the normal stability experienced during runs at any one proton energy.

The experimental procedure for the tungsten angular distribution measurements was the same as described above. The tungsten gamma-ray spectrum shapes were analyzed by comparison with known line shapes of x-rays and Ta gamma rays.² The determination of the background below the photopeak was somewhat less certain for these thick-target measurements than for the platinum data.

RESULTS

The anisotropy C of the unresolved 330- and 358-keV gamma rays from thin platinum is compared with the theory in Fig. 1. C is defined as the ratio of the gamma-ray yield at 0° minus that at 90° divided by the yield at 90° . The incident energy-dependent parameter ξ is defined in the caption of Fig. 1. In Fig. 1 ξ corresponds to the mean energy of the proton in the target. The theoretical curve corresponds to quadrupole excitation of a target of spin zero to a state of spin two.⁴ The calculation is completely quantum mechanical but is carried to first order only. The theoretical curve has been adjusted to take into account the finite, though small, thickness of the target as well as the solid angle

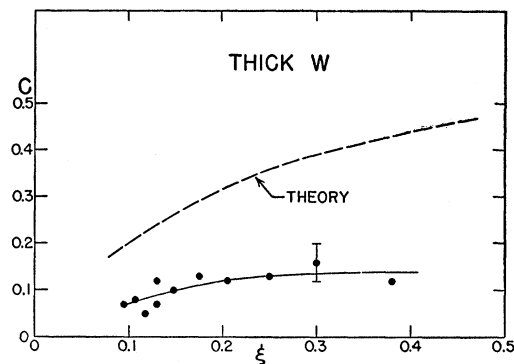


FIG. 2. Anisotropy of 114-keV (average energy) gamma ray from proton bombardment of thick tungsten. The dashed line corresponds to the (thick-target) theory of Alder and Winther.⁴ The solid line represents the maximum attenuation of the theoretical anisotropy that can result from interaction of the tungsten nuclei with the electric field gradients within the tungsten lattice. This curve corresponds to the assumption of axial symmetry of the field gradients at the positions of the excited nuclei.

subtended by the NaI detector.⁹ The solid-angle correction to the anisotropy amounts to about 4%; the thickness of the target results in about a 10% increase of the anisotropy over the zero-target-thickness value.

Figure 2 shows the observed anisotropy of the 114-keV tungsten line as a function of ξ . It is seen that the experimental points fall far below the upper dotted curve, which is a plot of the exact theory⁴ corrected for the thickness of the target.

The discrepancy between the theory and the data can be the result of (a) the "reorientation effect"¹⁰ and (b) the interaction of extranuclear fields with the tungsten nuclei. The reorientation effect refers to the alteration of the target nucleus orientation brought about by the electric field of the impinging projectile. The magnitude of this effect has been calculated by Breit *et al.*¹⁰ and in the case of 2-4 MeV protons on tungsten is estimated to be a correction to the anisotropy of the order of several percent. This correction is not large enough to account for the attenuation actually observed.

The attenuation of the anisotropy can be explained as the result of the interaction of the quadrupole moment of the nucleus with the interstitial field gradients within the tungsten lattice (there is a corresponding interaction between the magnetic moment of the nuclei with the crystalline magnetic fields but this is a much smaller effect in metals¹¹). A possible description of the excitation process is as follows: The Coulomb-excited nucleus, on being struck by an incident proton, recoils from its regular lattice position to perhaps an interstitial position where the field gradient is nonzero (since the tungsten crystal has cubic symmetry the field gradient

⁹ M. E. Rose, Phys. Rev. **91**, 610 (1953).

¹⁰ Breit, Gluckstern, and Russell, Phys. Rev. **103**, 727 (1956).

¹¹ R. M. Steffen, *Advances in Physics* (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 293.

is zero at regular lattice positions, provided there is little effect from strains or imperfections in the lattice¹². For impinging protons in the energy range 2–4 Mev the tungsten nucleus recoils several nuclear diameters and comes to rest in a time of the order of 10^{-12} sec.¹² The nucleus remains at the interstitial position in an excited state, finally returning to its ground state in $\sim 10^{-9}$ sec, the lifetime of the nuclear level(s) under consideration.¹³ Because this lifetime is so long, the extranuclear fields are capable of appreciably altering the angular distribution of the emitted gamma ray.¹¹ It is also to be expected that the impinging projectile causes many of the excited nuclei to vibrate about their equilibrium positions rather than recoil through the lattice.¹² These vibrational frequencies are of the order of 10^{12} cps; the amplitudes of vibration are of the order of an atomic diameter. These excited, vibrating nuclei would be expected to move into regions of large field gradient many times during the lifetime of the state, thereby experiencing an interaction which would contribute to the attenuation of the anisotropy.

Quantitatively, the condition that the angular distribution be appreciably attenuated is that the precession period of the excited nucleus in a quadrupole field be comparable to the lifetime of the excited state.

The precession frequencies of a quadrupole nucleus of moment Q in an inhomogeneous axially symmetric field (e.g., around the z axis) for the $0-2-0$ transitions with which we are dealing are given by¹¹

$$f_n = n \frac{e^2 Q}{8h} \left(\frac{1}{e} \frac{\partial E_z}{\partial z} \right) \text{ cps, } n=1, 3, 4.$$

The quadrupole moment is in units of cm^2 . The factor in parenthesis is the field gradient expressed in units of cm^{-3} . As is customary, we shall henceforth abbreviate this term as q .

The attenuation of the anisotropy of the gamma ray is determined by f_n and the lifetime of the excited state. The observed anisotropy of the 114-kev line is consistent with a "hard core" correlation.¹¹ It is the hard-core anisotropy (corrected for the effectively infinite thickness of the target) which is plotted as a function of ξ in Fig. 2 (solid line). This curve corresponds to the case where (a) the sample is polycrystalline and (b) the field gradients at the sites of the decaying nuclei are axially symmetric. The dotted line in Fig. 2 represents the theory of Alder and Winther⁴ for nuclei in a field-free environment. The excellent agreement between the data and the hard-core theory may be fortuitous in that condition (b) above may not be fulfilled.

In the absence of detailed knowledge of the potential distribution within the tungsten lattice, one of course cannot calculate the positions into which the Coulomb-excited nuclei recoil. Nonetheless one might empirically

define an "average quadrupole interaction frequency" corresponding to the observed transitions^{11,14}:

$$\langle f_0 \rangle = \frac{1}{8} (e^2 Q/h) \langle q \rangle \text{ cps.}$$

This equation may be regarded as defining the "mean field gradient" $\langle q \rangle$ in terms of the quadrupole moment of the tungsten nuclei and the observed interaction energy. Our measurements give a lower limit on $\langle f_0 \rangle$ of 10^{+9} cps. Using known Coulomb excitation data^{4,7,8} in conjunction with the collective model of nuclear excitation, one can estimate the quadrupole moment of the tungsten nucleus in the excited state(s) under consideration to be $1.0 \times 10^{-24} \text{ cm}^2$. We then find $\langle q \rangle \geq 2 \times 10^{26} \text{ cm}^{-3}$. This figure does not take into account polarization effects,¹⁵ which probably enhance Q , thus causing the above estimate for the lower bound of the mean field gradient to be too large.

SUMMARY

The thin-platinum anisotropy measurements described above are in good agreement with the first-order quantum-mechanical calculations of the angular distribution of Coulomb-excited gamma rays. There is some indication from these data (in agreement with the experimental results of Stelson and McGowan⁹) that the theoretical anisotropy is slightly larger than is actually observed.

Recent semiclassical calculations by Breit *et al.*¹⁰ contain a second-order "reorientation" term, which reduces the first order anisotropy by several percent. This correction was not applied in calculating the theoretical curve of Fig. 1 because of the limitations of semiclassical approach² and because the correction term contains an unknown factor, the expectation of the square of the radius of the nucleus in the excited state.

The tungsten anisotropy measurements establish the fact that for long-lived nuclei it is possible to observe the effect of the interaction of Coulomb-excited nuclei with extranuclear fields. Additional experiments are required to determine whether the extranuclear fields at the sites of the decaying nuclei result from the excited nuclei recoiling into the lattice or whether the interaction is a result of the vibration of these nuclei on being struck by the impinging particle. By measuring the anisotropy of long-lived nuclei in various environments, by using different types of bombarding particles, and by making measurements at various temperatures, one could perhaps resolve the above question. As has been illustrated with the calculation in the previous section, it is also possible that Coulomb-excited nuclei can be used as a probe to measure field parameters in solids.¹⁴

The authors are grateful to Professor E. Merzbacher, Professor R. Smoluchowski, and Professor W. Kohn for valuable discussions.

¹⁴ N. Bloembergen and T. J. Rowland, *Acta Metallurgica* **1**, 731 (1953).

¹⁵ R. M. Sternheimer and H. M. Foley, *Phys. Rev.* **92**, 1460 (1953).

¹² F. Seitz and J. S. Koehler, *Solid State Physics* (Academic Press, Inc., New York, 1956), Vol. 2, p. 305.

¹³ A. W. Sunyar, *Phys. Rev.* **95**, 626 (1954).