Coulomb Excitation of Ta, W, and Au†

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By using protons as bombarding particles, a study has been made of the yields and angular distributions of Coulomb-excited gamma rays in tantalum (137, 166, 303 kev), tungsten (114 kev), and gold (277, 195, 545 kev). Coincidence experiments show that the 277-kev and the previously unreported 545-kev transition go to the ground state and that the Ta 166-kev line represents a cascade from 303 kev. Thin-target yield data for the 277-kev gamma gives an absolute cross section which is in approximate agreement with the theory while the absolute cross section for the 545-kev transition is seven times too large. [*Note added in proof.*—On correcting an arithmetical error the absolute cross section for the 545-kev transition is also found to be in approximate agreement with the theory.] In both cases the experimental cross sections increase more rapidly than the theory. Thick-target yields from the five strongest gamma rays are greater than the theory by factors ranging from 6 to 100 percent between proton energies of 2 and 4 Mev. Agreement with the theory becomes worse with increasing level energy and decreasing proton energy. On the basis of the theory the angular distribution data permit an unambiguous spin assignment of 7/2 to the 545-kev level and give agreement with the previously established spins for the 277 (Au), 137, 303 (Ta), and 114-kev (W) levels. The ratio of $E2$ to $M1$ radiation is $7_{-5}^{+13}/100$ for the 277-kev radiation (Au) and less than 5/100 for the 137-kev radiation (Ta).

INTRODUCTION

THE present investigation was undertaken to examine the quantitative predictions of present theories concerning the yields[1] and angular distributions[2] of gammas resulting from Coulomb excitation of nuclei by incident charged particles. We also wish to compare the observed energy levels with the predictions of the collective model.[3] Since the initial experimental work done by Huus and Zupančič[4] and McClelland and Goodman,[5] a search for Coulomb excited gamma rays has been carried out in many elements using both alpha particles[6,7] and protons. Thick target yield studies[4,6] have been made on Ta and the angular distribution of Ta lines has been studied.[8] We have surveyed spectroscopically pure targets of Ta, W, and Au bombarded by 4-Mev protons, for gammas having energies different than the K and L x-rays. Coincidence measurements were made in order to check decay schemes. The thin target yields of two gold gammas and thick target yields of gammas from all targets are compared with each other and with theory. Angular distributions of the five strongest gammas are reported and compared with theory.

EXPERIMENTAL ARRANGEMENT

The proton source for these experiments was the Duke 4-Mev Van de Graaff accelerator.[9] The proton energy resolution and accuracy were both better than 0.05 percent after 25° deflection in a magnet equipped with a proton-moment fluxmeter. The target and detector area was shielded from the machine by a concrete block wall. Figure 1 shows the setup for the thin-target yield measurements. Since Rutherford scattering of the proton beam by the target foil was considerable, a large-diameter collecting chamber lined with lead was used. (We have verified Goodman's report[5] that protons excite no levels in lead.) Thin Ta lining in the tubing ahead of the target improved the background. When thick targets were mounted at D (Fig. 1), the collector (E) was removed. The target insulation and current integrator[10] input were periodically checked with a megger. The lead diaphragm (A) in Fig. 1 was made larger than the defined beam; it served to stop collimating slit-scattered particles and to absorb gammas. Current integration appeared reproducible to ±1 percent when the beam current was held constant to 10 percent.

The cylindrical scattering chamber used in the angu-

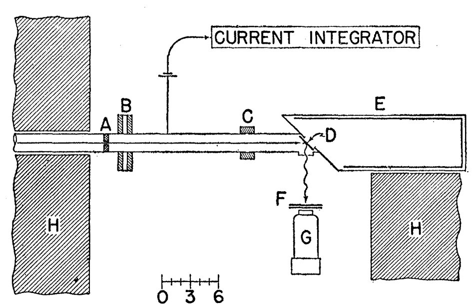

FIG. 1. Thin-target assembly showing: A—lead diaphragm; B—insulated flanges; C—coupling joint; D—thin foil; E—lead-lined beam collector; F—absorbers; G—scintillation counter; H—concrete shielding.

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

[1] K. A. Ter-Martirosyan, J. Exptl. Theoret. Phys. (U.S.S.R.) 22, 284 (1952).
[2] K. Alder and A. Winther, Phys. Rev. **91**, 1578 (1953).
[3] A. Bohr and B. R. Mottelson, Kgl. Danske Videnskab. Selskab. Mat.-fys Medd. **27**, No. 16 (1953).
[4] T. Huus and Č. Zupančič, Kgl. Danske Videnskab. Selskab, Mat.-fys Medd. **28**, No. 1 (1953).
[5] C. L. McClelland and C. Goodman, Phys. Rev. **91**, 760 (1953).
[6] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **93**, 351 (1954).
[7] N. P. Heydenburg and G. M. Temmer, Phys. Rev. **93**, 906 (1954).
[8] Class, Cook, and Eisinger, Phys. Rev. **94**, 747 (1954).
[9] Designed and built by the High Voltage Engineering Corporation.
[10] R. C. Mobley (to be published).

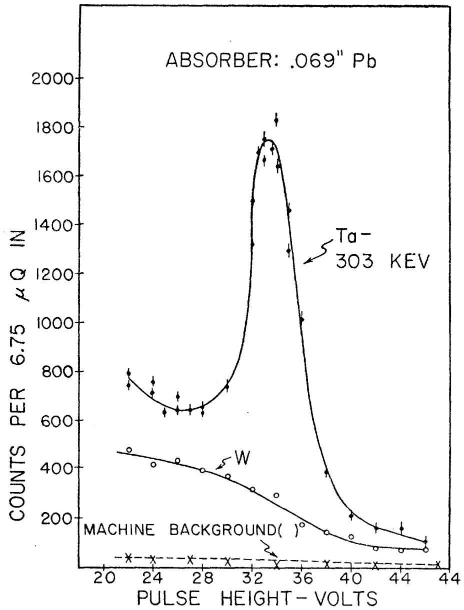

FIG. 2. Pulse-height spectrum from the 1.5-in.×0.5-in. NaI crystal showing the thick Ta 303-kev photopeak, the thick W spectra, and machine background. The proton energy is 4 Mev and the counter distance is 2 in.

lar distribution measurements was 5.25 in. in diameter, 6.0 in. deep, and had 0.12-in. aluminum walls. It was connected at point (*C*) in Fig. 1. The target could be rotated about the axis of symmetry. The chamber was lined with 0.020-in. cadmium in order to prevent the scattered protons from producing gammas in the aluminum walls. It was necessary to use thick (0.004-in.) targets for all angular distribution measurements. An attempt to use thin foils failed because of the size of the background produced when the proton beam struck the chamber wall.

A NaI crystal counter 1.5 in. in diameter and 0.5 in. thick was placed at 4 in. to 6 in. from the target in all angular distribution measurements. The DuMont 6292 photomultiplier tube was magnetically shielded in order that the tube gain remain the same as the direction of the counter axis was changed. The x-ray region of gamma energies was surveyed using a thin NaI crystal mounted directly on top of the phototube; a $\frac{1}{8}$-in. diaphragm placed ahead of the crystal limited the photocathode area involved. (These techniques will be described in a forthcoming paper on *L* x-ray production by H. W. Lewis and E. Bernstein.) The gamma energy region above 600 kev was surveyed using a 2-in.×2-in. NaI crystal counter. The 1.5-in.×0.5-in. crystal was used for the intermediate region. Phototube pulses were fed throgh a cathode follower into an A-1 linear amplifier and then put into a single-channel differential pulse-height analyzer. Care was taken at all times not to exceed maximum equipment counting rates. Gamma-energy calibrations were made relative to the Pb *K* line and 191, 277, 364, 511, and 662-kev gammas from In^{114*}, Hg^{203}, I^{131}, Na^{22}, and Cs^{137}. The coincidence circuit was similar to that published by Burke and Risser.[11] It has pulse-height discrimination in both channels, a resolving time of 0.1 microsecond, and recorded gross and accidental counts simultaneously.

PROCEDURE AND DISCUSSION OF ERRORS

Our ability to locate the background below the NaI scintillator photopeaks was the main factor limiting our determination of relative gamma intensities. We approached the problem two ways. Figure 2 shows the Ta 303-kev gamma photopeak along with the W yield in that region as well as the machine background. The Pb yield (not shown) was found to be very much the same as the W yield. These targets were spectroscopically pure. Curves taken with added absorber in front of the scintillator indicated a hard background, presumably due to gammas from vacuum system impurities, about three times machine background in each target. The remainder of the W and Pb yield is consistent with calculated bremsstrahlung yield from the incident protons.[12] Since Pb and W have no gammas in or above this region, they should be a measure of the Ta background.[4] The shape of the corrected Ta curve is in excellent agreement with the spectrum of 277-kev gammas from a Hg^{203} source placed at the position of the Ta target. The gold 277-kev gamma (Fig. 3) also submitted nicely to this comparison after the gold 545-kev Compton distribution was subtracted from the data. Data taken with varied absorber thicknesses and counter distances indicated that the background could be located within 25 percent. Gamma peaks varied from four to ten times background. Statistical errors were limited to 2 percent. Absolute cross sections quoted are based on calculated scintillator efficiency and are therefore approximate.

Full-proton-energy yield curves were taken on all strong lines in order to verify their nonresonant behavior and similarity to the theoretical yields. Then several repeated yield ratios were taken at the selected

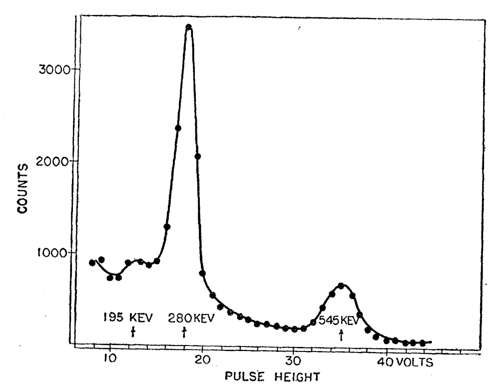

FIG. 3. Gammas from thick gold bombarded by 3.86-Mev protons. 0.5-in.×1.5-in. NaI crystal at 5 in. with 0.069-in. Pb absorber added.

[11] W. H. Burke and J. R. Risser, Phys. Rev. 87, 294 (1952).
[12] Č. Zupančič and T. Huus, Phys. Rev. 94, 205 (1954).

proton energies. The proton beam current was held constant within 10 percent for all data. In all cases these ratios agreed with each other within the ±3-percent counting statistics.

The symmetry of the angular distribution setup was checked by observing the Au 545-kev gamma at angles of 0°, 45°, 90°, 135°, 225°, 270°, and 315°. The 0.004-in. target foil was thin for this radiation. Since the rather small anisotropies observed showed nothing other than $\cos^2\theta$ dependence, repeated points were then taken at 270°, 0°, and 90° counter angles. The target angle was set so that gammas always traversed $\sqrt{2}$ times the target thickness. Runs were taken at both 45° target angles when the counter angle was 0° in order to check the target setting.

The effect of Compton scattering of the observed gamma rays both in the target itself and in the material between it and the detector was considered. Placing a 0.010-in. Ta sheet, which was thicker than any of the targets used, behind the gold target had no effect on the anisotropy of the 277-kev line. The change of the Compton cross section, as a function of gamma-ray energy, is not sufficiently great to require our repeating this experiment for all the lines studied. By varying the amount of scattering material near the detector we likewise showed that Compton scattering produced in the chamber wall and in the absorbers was negligible.

RESULTS

A. Observed Energy Levels

Only previously observed 137, 166, and 303-kev gammas were found in pure Ta. There were 1.7±0.1 times as many 166 as 303-kev gammas—in agreement with previous data.[8] Data using both 0.25-in. Cu and 0.069-in. Pb absorbers were analyzed in order to establish this figure. We also established the coincidence of the 137- and 166-kev gammas, but were unable to use this data as a check on the 303-kev level branching

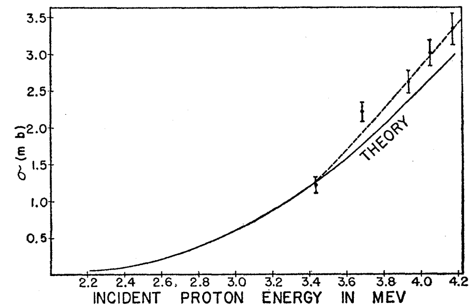

FIG. 5. Yield of 545-kev gammas from 0.0001-in. Au foil mounted at 45° to the incident proton beam. The theoretical cross section has been slightly modified to allow comparison with the 300-kev target thickness. The measured absolute cross section has a possible error of +50 percent. *Note added in proof.*—Cross-section scale should be multiplied by 1.0×10^{-1}.

ratio. No levels other than the 114 kev, the average of gammas from four isotopes,[13] were observed in W.

Previously reported gammas[7,14] of 195±10 and 277 ±5 kev were observed in gold. A new gamma at 545±8 kev is shown in Fig. 3. (This gamma has also been observed by C. M. Class, private communication.) The 195-kev gamma is better defined in data at lower proton energies. The most recent beta decay data[14] give levels at 77, 268, and 279 kev which involve excitation spin changes equal to or less than two. We compared our 277-kev photopeak shape very carefully (at 2- and 4-Mev proton energies) with the 277-kev Hg^{203} gamma and find no evidence of a 268-kev gamma. We also looked for 277–268 kev coincidences and found that the 545-kev level decays by cascade radiation to these levels less than 10 percent of the time. No 545–277 kev or 545–191 kev coincidences were observed. 545–77 kev coincidences could not be checked because of the dominant Au K x-rays. Thus it appears that Au has a 545-kev level decaying directly to the ground state.

Weak high-energy gammas of 840, 1000, 1360, and 1720 kev were observed in each of the three targets and are assumed to be from surface impurities from the vacuum system. These peaks were 2 to 10 percent of the Ta 303-kev peak and were about 10 times machine background. A yield curve run on the 1720-kev/gamma showed thin target resonances. Further, we observed both the K x-ray escape peaks and Ag K x-rays in commercial purity gold and platinum. Pure gold later showed no Ag K x-ray. The $1/Z^{12}$ cross-section dependence of K x-ray production greatly magnifies light element impurities when the low-gamma-energy region is being surveyed.

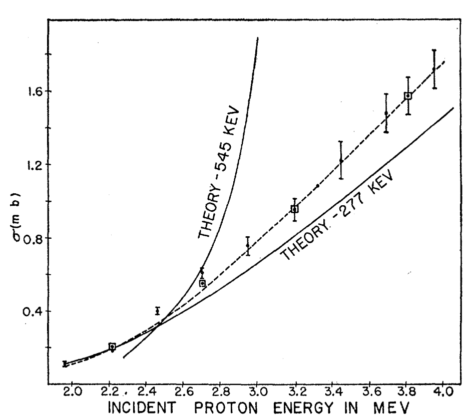

FIG. 4. Yield of 277-kev gammas from 0.0001-in. Au foil mounted at 45° to the incident proton beam. The theoretical cross section has been slightly modified to allow comparison with the 300-kev target. The measured absolute cross section has a possible error of +50 percent. Theoretical curves are drawn for several assumed level energies.

B. Yields

Figures 4 and 5 show the energy dependences of the thin target yields of the 277- and 545-kev gold gammas compared with the theoretical excitation of levels of those energies. A comparison of the magnitudes of

[13] McClelland, Mark, and Goodman, Phys. Rev. **93**, 904 (1954).
[14] J. W. Mihelich and A. de-Shalit, Phys. Rev. **91**, 78 (1953).

TABLE I. Theoretical and experimental thick-target yield ratios at 4.0, 2.5, 2.25, and 2.0-Mev proton energy for the five strongest gammas in W, Ta, and Au. The experimental error is given in parentheses.

Gamma energy (kev)	$I(4.0)/I(2.0)$ Theory	$I(4.0)/I(2.0)$ Exp.	$I(4.0)/I(2.25)$ Theory	$I(4.0)/I(2.25)$ Exp.	$I(4.0)/I(2.5)$ Theory	$I(4.0)/I(2.5)$ Exp.
545					40	65 (6)
303	46	97 (12)	20.8	33 (3)	10.7	12.7(0.7)
277	46	79 (6)	20.8	28 (2)	10.7	12.9(0.7)
137	14	13.8(0.8)	···	···	5.2	5.5(0.3)
114	11.2	13.4(0.8)	7.1	7.8(0.5)	5.2	5.1(0.3)

measured cross sections with the theory for the excitation of rotational levels[3] is necessarily approximate because the quadrupole moment of gold has not been measured and our scintillators were not accurately calibrated. We have tried using $Q=0.6\times10^{-24}$ cm²—obtained by extrapolation from the curve of experimentally measured quadrupole moments.[15] The 277-kev cross section is within a factor of two of the calculated cross section, while the 545-kev cross section is eight times the calculated cross section at a proton energy of 4 Mev.* The size of the 277-kev gamma yield curve at low energies indicates that it is not a transition from a level of much higher energy.

Table I lists thick target yield ratios. The experimental error quoted is the sum of the background uncertainties at the two energies being compared.‡

C. Angular Distributions

The third column of Table II contains the results of our distribution measurements. We have tabulated the anisotropy (C) at various energies for the five gamma rays studied. C is defined as

$$C=[I(E_p,0°)-I(E_p,90°)]/I(E_p,90°).$$

$I(E_p,\theta)$ is the gamma-ray photopeak intensity, corrected for background, at a proton energy E_p and at an angle θ with respect to the proton beam.

The strong energy dependence of the Coulomb excitation cross sections makes the comparison of the experimental thick-target angular distributions and the calculated thin-target values meaningful. With the exception of tungsten, the data support the predicted slow energy dependence of the distribution.[2] Both of these factors, *viz.*, the strong energy dependence of the yield and the weak energy dependence of the anisotropy, make comparison of the magnitude of the thick-target angular distribution and the theoretical thin-target distribution valid. The intensity of the 545-kev gamma was insufficient to measure its angular distribution below 4 Mev.

[15] W. C. Gordy, Phys. Rev. **76**, 139 (1949).
* See abstract, note added in proof.
‡ *Note added in proof.*—Alder and Winther have recently improved the theory by taking into account the energy loss of the incident particle. With this correction the calculated relative cross section is brought into agreement with our measurements for both the 277- and 545-kev gold lines over the full range of proton energies studied. We are indebted to A. Bohr and B. Mottelson for the communication of this information.

Only the anisotropies are quoted because all angular distributions measured were indistinguishable from $\cos^2\theta$ distributions. The theoretical values of the anisotropy which best fit the observed distributions appear in the column headed "C_2+C_4." The angular distribution function, whose derivation by Alder and Winther is based on the assumption of pure electric quadrupole excitation, is given by

$$W(\theta)=\frac{8-4B_2a_2+3B_4a_4}{8}\left(1+\left[\frac{12B_2a_2-30B_4a_4}{8-4B_2a_2+3B_4a_4}\right]\times\cos^2\theta+\left[\frac{35B_4a_4}{8-4B_2a_2+3B_4a_4}\right]\cos^4\theta\right).$$

The coefficients of $\cos^2\theta$ and $\cos^4\theta$ are denoted by us as C_2 and C_4, respectively. It is readily seen that the quantity, C_2+C_4 is the theoretical counterpart of the experimental parameter C. B_2 and B_4 are angular correlation coefficients and have been calculated by Biedenharn and Rose.[16] They depend only on the multipolarity of excitation and emission and on the spins of the states involved. The coefficients, a_2 and a_4,[17] the only energy-dependent parameters in the distribution function, are functions of the nuclear matrix elements and are given in reference 2. The last two columns of Table II list the spin of the excited state and the type and multipolarity of radiation that is implied by our choice of C_2+C_4.

The choice of spin and multipolarity from our data is usually unambiguous. This is seen in Table III, which shows values of C_2+C_4 for various assumed spins and multipolarities. The calculations were all made assuming a proton bombarding energy of 4.0 Mev.

The errors quoted in Table II are ±3 percent background uncertainty and ±1 percent statistics for all gammas except those from tungsten. Here, the 0.004-in. target apparently caused more absorption difficulties than in the other cases. Both Ta gamma distributions agree with previous data[8] and support the level assignments of 9/2 (137 kev) and 11/2 (303 kev) made by

TABLE II. Measured anisotropy (C) and calculated (C_2+C_4) are given at different proton energies, E_p(Mev). The most probable gamma transition is listed.

Gamma	E_p	C	C_2+C_4	Transition
Ta, 137 kev	2.7	0.00±0.04	−0.005	9/2(M1)7/2
	3.8	0.00±0.04	−0.005	
Ta, 303 kev	2.7	+0.14±0.04	+0.18	11/2(E2)7/2
	3.8	+0.16±0.04	+0.17	
Au, 277 kev	2.7	−0.17±0.04	−0.06	5/2(M1)3/2
	3.8	−0.14±0.04	−0.06	
	4.1	−0.15±0.04	−0.06	
Au, 545 kev	4.0	+0.25±0.04	+0.23	7/2(E2)3/2
W, 114 kev	2.0	+0.21±0.07	+0.36	2(E2)0
	2.8	+0.18±0.07	+0.31	
	3.6	+0.10±0.07	+0.27	
	4.2	+0.11±0.07	+0.21	

[16] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25**, 729 (1953).
[17] a_4 is a negative number. C. M. Class (private communication).

Huus and Zupančič.[4] A calculation of mixed $M1$ and $E2$ radiation from the 137-kev level indicates that $E2$ is either more than 90 percent or less than 5 percent of the total radiation. Internal conversion electron data[18] support the latter assumption.

The Au 545-kev gamma anisotropy clearly demands the assumption of a $7/2(E2)3/2$ transition. The Au 277-kev gamma distribution may be fitted by mixed $E2$ and $M1$ radiation using a phase shift of 180° for the interference term.§ The intensity ratio of $E2$ to $M1$ is either $(7_{-5}{}^{+13})\div 100$ or $(3_{-1.5}{}^{+2})\div 1$. Beta-decay data support the former assumption.

The tungsten gammas are expected to be predominantly $2(E2)0$ transition in the even isotopes. W^{183} (14 percent abundant) has a ground-state spin of $\frac{1}{2}$ and an observed gamma at 103 kev.[13] The only possible transitions in W^{183} which could decrease the observed anisotropies are $M1$. Further, the internal conversion coefficient for $M1$ is large compared to the $E2$ expected from the even isotopes. The 102-kev conversion electron peak observed by Huus and Bjerregaard[18] is comparable to peaks from the other even isotopes and should, therefore, be mainly due to the 102-kev level in W^{182}.[13] We, therefore, do not think that the low experimental anisotropy can be explained by the radiation from W^{183}. However, if we assume that the intrinsic quadrupole moment (Q_0) of tungsten[4] is 7×10^{-24} cm^2, the lifetime of rotational levels in W is about 10^{-9} sec.[19] We might therefore expect some decrease in anisotropy due to interaction between the quadrupole moment of the nucleus and atomic electric-field gradients.[20] Further study of this possibility is planned.

It was hoped that the W gammas would afford a particularly sensitive check on angular distribution theory, since it had a relatively large anisotropy and rapidly changing energy-dependent coefficients, a_2 and a_4. The order of magnitude of the predicted anisotropy change with energy is observed but the possible effect discussed above may also depend on the recoil velocity of the tungsten nucleus.

CONCLUSIONS

The determination of scintillator line shapes with artificial sources appears the most direct approach to the problem of background. Detection of high-energy gammas is limited by target surface impurities rather than machine background when no special cold baffles are employed near the target. Very pure targets are particularly important when the x-ray energy region is surveyed. Gamma-gamma coincidence counting rates in Ta showed that angular correlations demand better than 0.1-microsecond resolving time.

TABLE III. Calculated values of the anisotropy C_2+C_4 for various transitions using a proton energy of 4 Mev. Except for the 137-kev Ta line, transitions for which B_2 and B_4 equal zero are not listed. Because of the small change in C_2+C_4 with gamma energy, we have not listed separately the transitions possible for each gamma from one nucleus.

Target	Ground state	Transition	C_2+C_4
Ta^{181}	7/2	$7/2(M1)7/2$	−0.09
	7/2	$7/2(E2)7/2$	+0.06
	7/2	$9/2(M1)7/2$	−0.005
	7/2	$9/2(E2)7/2$	+0.015
	7/2	$11/2(E2)7/2$	+0.17
Au^{197}	3/2	$5/2(M1)3/2$	−0.07
	3/2	$5/2(E2)3/2$	+0.04
	3/2	$7/2(M1)5/2$	−0.15
	3/2	$7/2(E2)5/2$	+0.03
	3/2	$7/2(E2)3/2$	+0.23
W^{even}	0	$2(E2)0$	+0.38
W^{183}	1/2	$3/2(M1)1/2$	−0.20
	1/2	$3/2(E2)1/2$	+0.20
	1/2	$5/2(E2)1/2$	+0.24

The identification of the two Ta^{181} levels as pure rotational levels[4] is further supported by our angular distribution and coincidence data. The gold 545-kev level does not fit pure rotational level spacing when related to any of the lower states. Its energy is 2 times that of the 277-kev level, while the calculated figure is 2.5. However, the vibrational distortions discussed by Bohr and Mottelson[3] would tend to lower the energy of the upper level.

The Au 277-kev gamma thin target yield curve departs from theory by about 20 percent between 2- and 4-Mev proton energy. It will be noted that the discrepancy between theory and experiment is much greater for the thick target yield ratio between 2 and 4 Mev. The data also suggest that this gamma is from a level not more than 50 percent above 277 kev. This indicates that the experimental yield is much too low below 2 Mev and rises slightly too rapidly between 2 and 4 Mev. It is also seen (Table I) that the theory is most in error for higher level excitation. This is reasonable in light of the neglect of the change in energy of the incident proton in present theory.[2] The yield ratio errors do not seem to be systematic with excitation spin change. This point needs further data despite the fact that magnetic dipole excitation by incident particles moving slowly, compared to velocities within the nucleus, is expected to be small.‡

We have verified that there is no rapid change in angular distribution of gammas with incident proton energy. The magnitude of the anisotropies is in good agreement for all levels except those in tungsten. Further study of $2(E2)0$ transitions is planned.

We are very grateful for the aid and advice given us by our colleagues, H. W. Newson, H. W. Lewis, S. A. Cox, and E. M. Bernstein, and E. Merzbacher of the University of North Carolina.

[18] T. Huus and J. H. Bjerregaard, Phys. Rev. **92**, 1579 (1953).

§ For this particular nucleus a phase shift of 180° is predicted by the theory, assuming the transition is rotational. A. Bohr and B. Mottelson (private communication).

[19] A. W. Sunyar, Bull. Am. Phys. Soc. **29**, No. 4, 38 (1954).

[20] Alder, Albers-Schonberg, Heer, and Novey, Helv. Phys. Acta **26**, 761 (1953).