The nucleus of $Mn^{55}$ is one of the exceptions to Meyer's rules for nuclear spin.[23] This nucleus is thought to have 5 protons in $f_{7/2}$ orbits outside of a closed shell which are coupled to a total spin 5/2. The observed sign and the exceptionally large quadrupole moment of this nucleus is consistent with the expected value of such a state. In fact, for all possible spins consistent with Pauli's exclusion principle which one can expect from a $(f_{7/2})^5$ configuration, the state of total spin 5/2 gives the largest positive quadrupole moment. It is remarkable that all nuclei which are exceptions to the normal shell structure rules for spin have a ground state with an exceptionally large positive quadrupole moment,[24] as is found here for $Mn^{55}$.

The authors wish to thank Dr. G. Silvey for preparing samples of $ReO_3Cl$, Dr. A. V. Grosse for his interest in the preparation of the Mn compounds, and Mr. J. Lotspeich for his construction of the wave guide and the gas-handling system used in this experiment. Thanks are also due various personnel of the Columbia Radiation Laboratory.

We should like to take this opportunity to express our appreciation to Professor C. H. Townes for his guidance throughout the course of this research.

---

[23] M. G. Meyer, Phys. Rev. 78, 16 (1950).

[24] S. A. Moszkowski and C. H. Townes, Phys. Rev. 93, 306 (1954).

---

PHYSICAL REVIEW VOLUME 96, NUMBER 3 NOVEMBER 1, 1954

# Electric Excitation of $Au^{197}$†

C. F. Cook,* C. M. Class, and J. T. Eisinger‡
*The Rice Institute, Houston, Texas*

(Received July 30, 1954)

The gamma radiation emitted from $Au^{197}$ following the electric excitation of the nucleus by protons of from 2 to 5 Mev has been studied. The angular distributions and absolute yields of the two intense gamma rays of the spectrum at 279 and 555 kev have been measured. The angular distributions of these two gamma rays were found to agree well with those calculated for $5/2\rightarrow3/2$ $(M1+E2)$ and $7/2\rightarrow3/2$ $(E2)$ transitions, respectively, the excitation being $E2$ in both cases. The yield from the 279-kev state was observed to increase with beam energy in the manner predicted by theory for electric quadrupole excitation. On the basis of the Bohr-Mottelson unified model of the nucleus the results indicate a larger than expected cross section for the formation of the 555-kev state. Values for the quadrupole moment and magnetic moment of the ground state of gold calculated from the cross section for the formation of the 279-kev state agree with spectroscopic values.

## INTRODUCTION

The low-lying states of $Au^{197}$ have been accessible heretofore through the beta decay of $Hg^{197}$ and $Pt^{197}$, and through the decay from the metastable state excited by the inelastic scattering of neutrons.[1,2] On the basis of the $Hg^{197}$-$Au^{197}$ decay, Mihelich and de-Shalit have proposed a level scheme for gold having excited states at 77, 268, 279, and 409 kev, as shown in Fig. 1, which is compatible with the observed gamma ray and internal conversion electron spectra. The results from inelastic neutron scattering indicate that the 409-kev state is metastable. Spin assignments of $1/2^+$ or $3/2^+$ have been made to both the 77- and 268-kev levels. Both values are consistent with the available data, although the $1/2^+$ assignment is preferred for the 77-kev state. Some evidence also supports the prediction of the shell model that the 279-kev and 409-kev levels are $d_{5/2}$ and $h_{11/2}$ states, respectively.

Recent work has shown that the low-energy states of gold can also be reached by electric (or Coulomb) excitation of the nucleus. Using a beam of 3-Mev alpha particles Heydenburg and Temmer[3] have observed gamma rays emitted subsequent to excitation having energies of 77, 191, and 279 kev, which can be identified as due to transitions between the known levels listed. The 191- and 279-kev gamma rays have also been found with proton bombardment,[4,5] and in addition, a gamma ray of 555 kev has been reported which is now assigned to a new level of that energy.[6]

Further study of the gamma radiation due to the Coulomb excitation of gold has been undertaken in the

---

† Supported in part by the U. S. Atomic Energy Commission. A preliminary account of some of this work was given at the Austin, Texas and Washington, D. C. meetings of the American Physical Society.

* Now at the University of Florida, Gainesville, Florida.

‡ Now at the Bell Telephone Laboratories, Murray Hill, New Jersey.

[1] J. W. Mihelich and A. de-Shalit, Phys. Rev. 91, 78 (1953).

[2] A. A. Ebel and Clark Goodman, Phys. Rev. 93, 197 (1954); Martin, Diven, and Taschak, Phys. Rev. 93, 199 (1954).

[3] N. P. Heydenburg and G. M. Temmer, Phys. Rev. 93, 906 (1954).

[4] W. I. Goldburg and R. M. Williamson, Phys. Rev. 94, 747 (1954).

[5] Class, Cook, and Eisinger, Phys. Rev. 94, 809 (1954).

[6] Goldburg, Cox and Williamson, Phys. Rev. 95, 628 (1954).

work reported here. As has been pointed out,[7,8] this method of excitation is particularly suitable for studying the collective properties of the nucleus, which are described by the unified model of Bohr and Mottelson.[8,9] According to the model, nuclei in the region of mass values removed from closed shell configurations are subject to large deformations which result in a spectrum of low-lying "rotational" states. Because of these deformations large quadrupole matrix elements are associated with the rotational states which cause them to be especially susceptible to electric excitation by the $E2$ component of the radiation field of the incident particle. States due to single particle excitations may, of course, appear in addition to those associated with the collective motion.

The rotational states for an even-odd nucleus have spins which follow the sequence $I=I_0, I_0+1, \cdots$, and are at energies

$$E_I=(\hbar^2/2\mathcal{I})[I(I+1)-I_0(I_0+1)],$$

where $\mathcal{I}$ is the effective moment of inertia (proportional to the square of the nuclear deformation). A similar formula applies for even-even nuclei. Departures from this purely rotational spectrum may occur as a result of centrifugal distortion and perturbations due to neighboring vibrational states. These effects are expected to be small for nuclei far removed from closed-shell configurations, and this is amply confirmed by experiment.[9–11]

In the case of gold, which is close to the magic nuclide Pb, the pure rotational model predicts 12:5 for the ratio of the energies of the second to first rotational states, while the 555- and 279-kev levels which are presumably rotational in character, are in the ratio of 2:1. This departure can be understood in a semi-quantitative way as due to the effects mentioned above. However, other features associated with rotational levels, such as an appropriate sequence of spin values and suitable $E2$ transition probabilities should be retained by these states. Experiments pertaining to these points are reported on in this paper. By measuring the angular distribution of the gamma radiation from these levels with respect to the beam, information relating to their spins was obtained. The observed functional dependence of the yield of these radiations on the energy of the bombarding particle allowed a check on certain of the assumptions in the theory of the excitation process to be made; and finally, the measurement of the absolute yield permitted a comparison of the experimental values of the reduced transition probabilities with those predicted by the unified model.

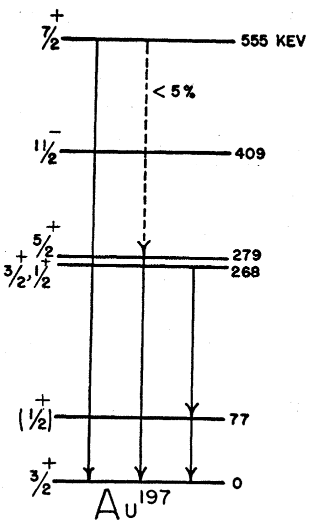

FIG. 1. Energy level diagram of $Au^{197}$. The arrows indicate transitions observed in Coulomb excitation. Spin assignments are taken from reference 1 and the present work.

## APPARATUS AND PROCEDURE

Targets of gold, 99.95 percent pure,[12] were bombarded with protons accelerated by the Rice Institute 6.0-Mev Van de Graaff generator. After deflection through a 90° analyzing magnet, the resolution of the beam was ±10 kev, and the absolute energy was known to ±10 kev. The proton current incident on the target was measured with a current integrator of the type described by Watt.[13]

For the yield measurements, a thin gold target was used in order to reduce the inaccuracies of the stopping power calculation. The target was a gold foil 0.00011 inch thick (about 300 kev for 2-Mev protons) mounted at 45° to the beam. For the angular distribution measurements, it was possible to use a thick target (0.005 inch) since the distributions are not very sensitive to the beam energy.

Background radiation presented a serious problem throughout the experiment but was especially important with the thin target used for the yield measurements. In this case, unless the beam was absorbed on emerging from the target in such a way as to produce relatively little background radiation, the gamma rays under examination were obscured. To reduce this background the target chamber was constructed as shown in Fig. 2, so that the beam was absorbed in lead. Lead was chosen because no level is excited by protons in the energy range up to 5 Mev. By stopping the beam two feet beyond the target-counter position, it was possible to

---

[7] T. Huus and Č. Zupančič, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 28, No. 1 (1953).

[8] A. Bohr and B. Mottelson, Kgl. Danske Videnskab. Selskab, Mat.-fys. Medd. 27, No. 16 (1953); hereafter referred to as B-M.

[9] A. Bohr, *Rotational States of Atomic Nuclei* (Ejnar Munksgaards Forlag, Copenhagen, Denmark, 1954).

[10] G. M. Temmer and N. P. Heydenburg, Phys. Rev. 94, 1399 (1954).

[11] F. Asaro and I. Perlman, Phys. Rev. 91, 763 (1953).

[12] Based on a spectroscopic analysis. The targets were prepared from gold foil obtained from Baker & Company, 113 Astor Street, Newark 5, New Jersey.

[13] B. E. Watt, Rev. Sci. Instr. 17, 334 (1946).

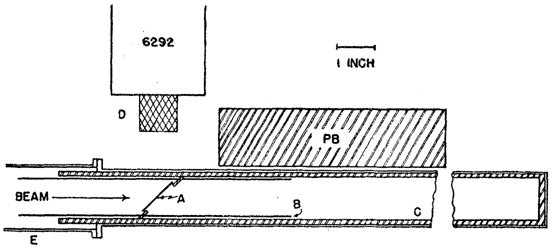

FIG. 2. The target chamber used for the absolute yield measurements. A: a 0.0001-inch thick gold target; B: 0.004-inch tin foil; C: lead lining; D: a 1-inch×1-inch NaI crystal; E: beam tube.

take advantage of the inverse square effect to reduce the intensity of the radiation at the counter. Further shielding was accomplished with a lead block placed so as to intercept much of the radiation produced as the beam was absorbed. To reduce the bremsstrahlung[14] due to scattered protons in the vicinity of the counter, the inner surfaces of the chamber were lined with 0.004-inch thick tin foil. The target chamber was located 20 feet from the analyzing magnet and at this distance the accelerator background was negligible. These measures reduced the background at $E_p<4.0$ Mev to about 15 percent of the total intensity of the photopeaks of the 279- and 555-kev lines. With increasing beam energy, the background rose to about 40 percent at $E_p=5.2$ Mev and at higher energies it prevented significant data being obtained.

The target chamber for the angular distribution measurements was a cylinder of copper 2 inches in diameter and 6 inches deep, which was mounted with its axis transverse to the beam tube. The target and a quartz plate used to position the beam were mounted on a rod free to rotate and slide along the axis of the chamber, and a divided head was provided so that the target could be accurately positioned from outside the chamber at any desired angle to the beam. The gamma-ray counter was mounted on an aluminum platform which rotated about an axis carefully aligned to coincide with the axis of the chamber. Background in these measurements was chiefly due to bremsstrahlung resulting from the scattering of the beam in the thick gold target. However, the improvement in yield due to the greater target thickness caused the problem to assume less importance.

The gamma rays were detected with a conventional NaI(TlI) scintillation spectrometer which had a resolution of 10 percent for 660-kev radiation. Standard electronic circuits were used for amplification, pulse-height analysis, and recording. The spectrometer was calibrated with gamma rays from $Hg^{203}$, $I^{131}$, $Sn^{113}$, and $Cs^{137}$ sources.

The principal problem encountered in analyzing the data was in estimating the amount of background to be subtracted from the photopeak of the gamma rays being measured. This subtraction was guided by data on the shape of pure gamma-ray lines. The spectra of $Hg^{203}$ and $Cs^{137}$ radiations were taken with the sources mounted in the target positions of both chambers and by making use of the peak to valley ratio, resolution, and the location of the upper edge of the photopeak of the pure lines the magnitude of the background was estimated. Subtraction of the background resulted in line shapes for the gold gamma rays which agreed well with the pure lines from radioactive sources. Such measurements also showed that scattering material external to the target chamber had a negligible effect on the line shape. In the case of the angular distribution measurements, sources mounted in the target position made it possible to check the symmetry of the target and counter assemblies.

To make absolute yield measurements, it was necessary to determine the efficiency of the counter assembly. This was done by placing a standardized $I^{131}$ source[15] in the target position under conditions identical to those under which the yield data were taken. Appropriate corrections were made to take into account the difference in the energies of the gamma rays from $I^{131}$ and gold.

## RESULTS AND DISCUSSION

### 1. Spectrum

Figure 3 shows the spectrum of radiation obtained from a thick target of gold when bombarded with 3.0-Mev protons with a 1-mm thick gold absorber used to attenuate the target x-rays. Gamma rays having energies of 191, 279, and 555 kev can be identified. A known gamma ray of 77 kev was obscured in these measurements due to the x-rays from $Au^{197}$, while

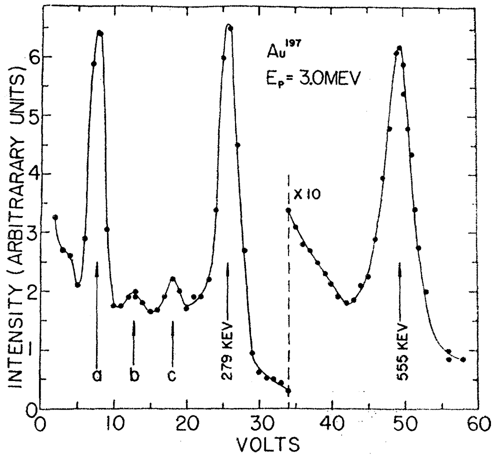

FIG. 3. The pulse-height distribution of gamma radiation from a thick gold target bombarded with 3.0-Mev protons obtained with a NaI crystal using a 1-mm gold absorber. Peaks *a*, *b*, and *c* correspond to the gold *K* x-ray, the Compton edge of the 279-kev gamma ray, and a 191-kev gamma ray.

---

[14] Č. Zupančič and T. Huus, Phys. Rev. **94**, 205 (1954).

[15] The source was calibrated by the National Bureau of Standards and was made available to us by Miss F. Garrett of the Radio-isotope unit of the V. A. Hospital, Houston, Texas.

possible gamma rays of 276 or 287 kev corresponding to transitions between the 555- and 279- or 268-kev levels could not have been observed with the available resolution. However, a search for gamma-gamma coincidences in this region showed that less than 5 percent of the 279-kev radiation resulted from a cascade from the 555-kev level. (This result was also obtained by Williamson.[6]) No evidence was found for a 478-kev gamma ray corresponding to a transition between the 555- and 77-kev levels. Since the 555-kev level is almost certainly a spin 7/2 state, as will be shown below, the absence of this gamma ray would seem to favor the choice of a spin assignment of 1/2 for the 77-kev level out of the two possibilities of 1/2, 3/2 allowed on the basis of the beta decay measurements.[1] The transitions observed with Coulomb excitation are indicated in Fig. 1. Other transitions in either excitation or decay are presumably negligible because of the large spin changes involved.

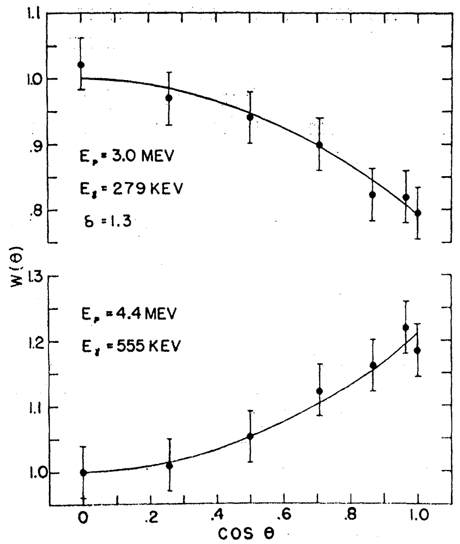

FIG. 4. The angular distributions of the 279-kev and 555-kev gamma rays from gold measured between 0 and 90 degrees to the beam. The curves were obtained from least square fits to the data.

## 2. Angular Distributions

It has been shown by Alder and Winther[16] that information on the spins of electrically excited states can be obtained by measuring the angular distribution of the subsequent gamma radiation with respect to the beam. Figure 4 shows the experimental angular distributions obtained for the 279- and 555-kev radiations at beam energies of 3.0 and 4.4 Mev, respectively. The counter subtended a solid angle of 0.13 steradian and a 1-mm thick gold absorber was again used to reduce the x-ray intensity. The data were corrected for absorption in the target (5 mils gold), background and the finite angle subtended by the counters. Errors shown in Fig. 4 are statistical, but systematic errors due to uncertainties in background subtraction could change the asymmetries by about 3 percent.

The angular distribution data were analyzed under the assumption that the excitation of the state in question occurs by an electric quadrupole transition, which is in keeping with the Bohr-Mottelson theory, as discussed above. Transitions from the excited state may, of course, go by either $E2$ or $M1$ radiation or by a mixture of these. For $Au^{197}$, which has a ground state spin of 3/2, the only states accessible under this assumption which would result in anisotropic distributions are those having spins 5/2 and 7/2. The angular distribution corresponding to the 7/2→3/2, $E2$ transition has been calculated at the appropriate beam energy using the following expression given by Alder and Winther[16]:

$$W(\theta)=1+a_2(\xi)B_2P_2(\cos\theta)+a_4(\xi)B_4P_4(\cos\theta), \quad (1)$$

where the $a_k(\xi)$ are energy dependent factors which enter because of the electric character of the excitation process, the $B_k$ are the gamma-gamma correlation coefficients tabulated by Biedenharn and Rose,[17] and the $P_k$ are the Legendre polynomials.[18] Expression (1) when evaluated for $E_\gamma=555$ kev and cast in a form easily compared with experiment gives

$$W(\theta)=1+0.248\cos^2\theta-0.014\cos^4\theta\approx 1+0.23\cos^2\theta.$$

This distribution is to be compared with $W(\theta)=1+0.21\cos^2\theta$ obtained by a least squares fit to the data for the 555-kev radiation shown in Fig. 4. The agreement between the calculated and observed distributions appears adequate to assign a spin 7/2 to the level.

The observed negative anisotropy of the 279-kev gamma-ray angular distribution obviously excludes the possibility of a 7/2→3/2, $E2$ transition. Indeed, a 5/2→3/2, $E2$ transition is also excluded since this leads to a small positive coefficient for the $\cos^2\theta$ term, and although a 5/2→3/2, $M1$ transition gives a negative coefficient, the anisotropy amounts to only 7 percent. However, by considering a mixture of $M1$ and $E2$ radiations, it is possible to obtain an anisotropy compatible with the experimental results for a 5/2→3/2 transition. The angular distribution for this case is given by

$$W(\theta)=1+\frac{(0.0365-0.362\delta-0.0715\delta^2)}{1+\delta^2}a_2(\xi)P_2(\cos\theta)+\frac{0.49}{1+\delta^2}a_4(\xi)P_4(\cos\theta), \quad (2)$$

---

[16] K. Alder and A. Winther, Phys. Rev. **91**, 1578 (1953).

[17] L. C. Biedenharn and M. E. Rose, Revs. Modern Phys. **25** 729 (1953).

[18] The error in the coefficients made in neglecting the effect of the thick target is small because of the form of the yield curve and the slow variation of $a_k(\xi)$ with energy.

where $\delta^2$ is the ratio of the intensities of $M1$ and $E2$ transitions and can be treated as a parameter to give the best fit to the data.

The experimental distribution is $W(\theta)=1-0.21\cos^2\theta$, and the best agreement between it and theory is achieved by assuming $\delta^2=1.7$. The theoretical distribution is then

$$W(\theta)=1-0.174\cos^2\theta-0.014\cos^4\theta\approx1-0.19\cos^2\theta,$$

and the negative coefficient describing the asymmetry is a maximum. This value of $\delta^2$ which corresponds to about 63 percent $M1$ transitions, when taken together with the theoretical $K$-conversion coefficients[19] $\alpha_K{}^2=0.08(E2)$ and $\beta_K{}^1=0.50(M1)$ gives a total conversion coefficient of about 0.35 as compared to ~0.3 measured by Mihelich and de-Shalit.[1] It should be noted, however, that the calculated distribution is not strongly dependent upon $\delta^2$. Values of $\delta^2$ from 1 to 3 would lead to anisotropies within the range of experimental error.

### 3. Yield Measurements

The cross sections measured for the emission of gamma rays from the 279- and 555-kev levels are shown in Figs. 5 and 6, and for comparison the theoretical yield-curve for excitation by $E2$ transitions is also shown, normalized to the experimental data at about 4 Mev. The data were corrected for the finite thickness of the target and the angular distribution of the radiation. The errors indicated are due to some arbitrariness in the subtraction of background. Systematic errors in the absolute calibration of the counter contribute an uncertainty of about 20 percent to the scale of the cross sections.

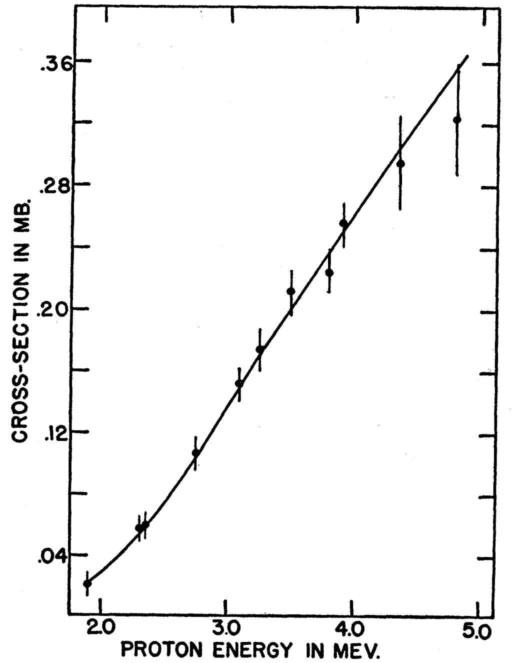

FIG. 5. The cross section in millibarns from the 279-kev gamma radiation as a function of the energy of the incident protons. The theoretical curve which is shown is normalized to the data at 4.0 Mev.

The theoretical yields have been calculated from the cross-section formula of Alder and Winther,[16]

$$\sigma=\frac{2\pi^2}{25}\frac{1}{Z_2{}^2e^2}\left(\frac{Mv}{\hbar}\right)^2B(E2)g_2(\xi), \qquad (3)$$

where $Z_2e$ is the charge of the target nucleus, $v$ is the relative velocity, $M$ is the reduced mass, and $B(E2)$ is the reduced electric quadrupole transition probability. The definition of the parameter $\xi$ of reference 16 has been replaced here by the more rigorous definition,

$$\xi=\frac{Z_1Z_2e^2}{\hbar}\left(\frac{1}{v_f}-\frac{1}{v_i}\right),$$

in order to improve the agreement with the experimental data in the low-energy region. $Z_1$ is the charge of the incident particle and $v_{i,f}$ are the relative velocities before and after the excitation. For excitation energies which are small compared to the bombarding energy this definition approaches that given by Alder and Winther. The factor $g_2(\xi)$ involves integrals over the orbit of the impinging particle which are evaluated numerically under the assumption that the path of the particle is a classical trajectory. This implies that the energy lost in the excitation process must be small in comparison to the kinetic energy of the interacting particle.

The agreement observed between the theoretical yield and the experimental measurements for the 279-kev transition supports the assumption of $E2$ excitation and also indicates sufficient accuracy in the evaluation of the trajectory factor. For the 555-kev transition, detectable yields occur when the excitation energy is as much as 20 percent of the energy of the impinging proton, so that a deviation from the theoretical yield might be expected at low bombarding energies. This is, indeed, observed as seen in Fig. 6 for the range of $E_p<3.5$ Mev. That this discrepancy could be due to multipole excitation other than $E2$ is not believed likely in view of the results on the angular distribution of this radiation.

To get the total cross section for the excitation of the 279-kev state, the probability of decay by internal conversion must be taken into account. From the experimentally determined $K$-conversion coefficient of ~0.3 and a $K/L$ ratio of 6,[1] a total cross section of 0.35 mb for the excitation of this level at $E_p=4.0$ Mev was obtained. The total cross section for the excitation of the 555-kev state is 0.26 mb at the same proton energy, the correction for internal conversion and decay by cascades being assumed negligible.

---

[19] Rose, Goertzel, Spinrad, Harr, and Strong, Phys. Rev. 83, 79 (1951).

## 4. Discussion

From the experimental total cross sections for the excitation of the 279- and 555-kev states and the theoretical cross section formula (3), the reduced transition probabilities $B(E2)$ were calculated. If the states are assumed to have rotational character, the $B(E2)$ are given by the unified model as

$$B(E2)=\frac{15}{16\pi}e^2Q_0^2\frac{I_0}{(I_0+1)(I_0+2)},\quad I_0\rightarrow I_0+1,\quad (4)$$

and

$$B(E2)=\frac{15}{8\pi}e^2Q_0^2\frac{1}{(2I_0+3)(I_0+2)},\quad I_0\rightarrow I_0+2,\quad (5)$$

where $Q_0$ and $I_0$ are the intrinsic nuclear quadrupole moment and spin of the ground state. By using spins of 5/2 and 7/2 for the 279- and 555-kev states, respectively, and the ground state spin of 3/2, $Q_0$ has been calculated from the above formulas. From the data for the 279-kev transition, $Q_0=1.9\times10^{-24}$ cm$^2$ has been obtained while that from the 555-kev transition yields $Q_0=3.0\times10^{-24}$ cm$^2$. This $Q_0$ is related to the spectroscopically measured quadrupole moment $Q$ by

$$Q=[I_0/(I_0+1)][(2I_0-1)/(2I_0+3)]Q_0,$$

so that the two cross section measurements yield values for $Q$ of $0.4\times10^{-24}$ and $0.6\times10^{-24}$ cm$^2$, respectively. Recent measurements[20] of the hyperfine structure of Au give $Q=(0.56\pm0.1)\times10^{-24}$ cm$^2$.

Quadrupole moments of other nuclei derived in a similar manner from the collective model matrix elements have been found to be systematically smaller than those obtained from spectroscopic measurements (see B-M, VIId, iii). In view of this, the larger value of $Q$ deduced from the 555-kev transition data suggests that the cross section for the excitation of this state is greater than would be expected from the theory. That a difficulty exists here is also indicated by the ratio of the reduced transition probabilities for the 279- and 555-kev states which is independent of $Q_0$. From formulas (4) and (5), $B(279):B(555)$ is expected to be 9:5 while the experimental ratio is of the order of 1:2. A similar comparison for the first two levels of Ta$^{181}$ gave excellent agreement between experiment and theory.[9]

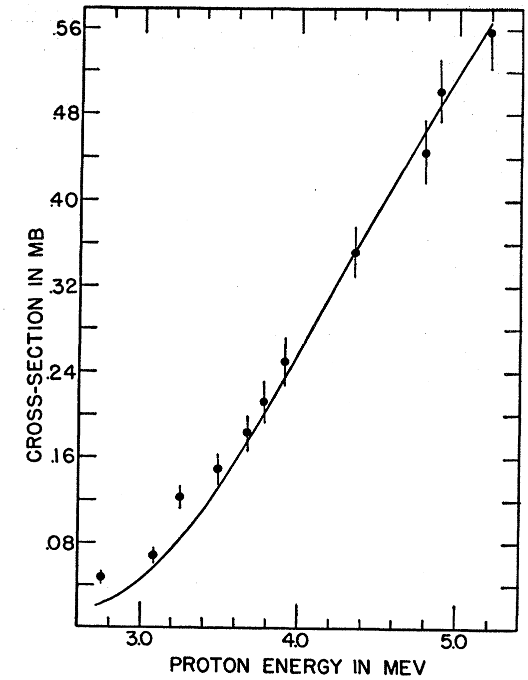

FIG. 6. The cross section in millibarns for the 555-kev gamma radiation as a function of the energy of the incident protons. The theoretical curve which is shown is normalized to the data at 4.2 Mev.

By making use of the $Q_0$ and $M1$ to $E2$ ratio determined for the 279-kev transition, the ground-state magnetic moment of gold was calculated (see B-M, IVb and VII 18, 20) to be ~0.13 nm as compared to 0.13±0.01 nm as measured by the atomic beam method.[21] These data also enable the intensity of the cascade transition from the 555-kev level through the 279-kev level to be calculated and compared with the intensity of the cross-over transition (see B-M, VII 18, 19, 20). Seven percent of the decays were computed to occur by the cascade route whereas the coincidence measurements gave an upper limit of about 5 percent.

We wish to acknowledge several helpful comments by Dr. T. Huus of the Institute for Theoretical Physics, Copenhagen, Denmark.

---

[20] W. von Siemens, Ann. Physik **13**, 158 (1953).

[21] G. Wessel and H. Lew, Phys. Rev. **92**, 641 (1953).