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EXCITATION OF NUCLEAR ROTATIONAL STATES BY THE ELECTRIC FIELD OF IMPINGING PARTICLES

BY

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1. Introduction.

When heavy nuclei are bombarded with charged particles with an energy appreciably below the Coulomb barrier the short range nuclear forces cannot operate. The collisions may, however, still give rise to nuclear excitations produced by the electric field of the impinging particles. Such reactions are especially simple to interpret, and the cross-sections can be expressed in terms of the same nuclear properties that determine the transition probabilities for electromagnetic radiative processes.*†§

It has been suggested^{**} that the method of Coulomb excitation should provide a powerful tool for the study of nuclear collective properties. According to the nuclear model which describes the dynamics of the nucleus in terms of the coupled motion of individual particles and surface oscillations^{***}, lowlying excited states of collective type are expected for nuclei possessing large deformations. These states exhibit a rotational spectrum, and should reveal themselves by their especially large cross-sections for Coulomb excitation.

In order to test some of these predictions we have undertaken an investigation of the Coulomb excitation effects produced

[†] Nuclear excitations with cross-sections too large to be explained by barrier penetration have been observed on a number of occasions, and the possibility of attributing the effects to Coulomb excitation has been discussed. Cf., e. g., BARNES and ARADINE (1939); RISSER, LARK-HOROWITZ and SMITH (1940).

§ While the present work was being prepared for publication, we have learned that γ -rays resulting from Coulomb excitation of heavy nuclei have recently been observed by C. L. MCCLELLAND and C. GOODMAN. We are indebted to Professor GOODMAN for sending us a manuscript of their work in advance of publication.

** A. BOHR and B. MOTTELSON. (Cf., e.g., Report of the International Physics Conference, Copenhagen, June 1952).

*** A. BOHR (1952); BOHR and MOTTELSON (1953); the latter paper will be referred to in the following as B.-M.

^{*} The mechanism of nuclear excitation by the Coulomb field of bombarding particles has been discussed by several authors: WEISSKOPF (1938); RAMSEY (1951); MULLIN and GUTH (1951); HUBY and NEWNS (1951); BREIT, HULL, and GLUCK-STERN (1952); TER-MARTIROSYAN (1952); BOHR and MOTTELSON (1953).

by the protons from a 2 MeV electrostatic accelerator^{*}. We here report the results obtained by the bombardment of tantalum and tungsten.

In § 2, a brief summary is given of some of the relevant aspects of the theory of Coulomb excitation. A description of the nuclear rotational spectrum appears in § 3. The experimental arrangement is described in § 4. In § 5, the excitation crosssections for the first excited state in Ta¹⁸¹ are given and compared with the theory of Coulomb excitation; the nuclear data obtained are discussed in relation to the theory of rotational states. The excitation of the second rotational state in Ta is described in § 6. In § 7 the Coulomb excitation of the first excited states of the even-A isotopes of W is reported. The yields of the characteristic K X-rays, which are also excited by the protons, are discussed in § 8. A survey of main conclusions is contained in § 9.

2. Theory of Coulomb Excitation**.

When the energy of the bombarding particles is low enough to exclude penetration through the Coulomb barrier, the parameter

$$\varkappa = 2 \, \frac{Z_1 Z_2 e^2}{\hbar v} \tag{1}$$

is large compared to unity and the trajectory may be described by means of classical mechanics (N. BOHR, 1948). In expression (1), the charge numbers of the projectile and the target nucleus are denoted by Z_1 and Z_2 , respectively, while v is the relative velocity.

^{*} In previous experiments performed with the electrostatic accelerator of the California Institute of Technology (DAY and HUUS, 1952), a strong γ -ray had been observed to arise from the Ta target backing. The present experiments were undertaken after the recognition that this γ -ray of 137 keV resulted from Coulomb excitation.

^{**} The present formulation is based on the work of TER-MARTIROSYAN (1952). Cf. also B.-M., Appendix VI, who, in connection with a review of this work, have especially discussed the relationship between Coulomb excitation and electromagnetic transitions, and the applications to the study of nuclear collective properties. We here follow the presentation given in the latter reference.

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The effect of the projectile on the target nucleus can then be described in terms of the time varying electric potential, given by

$$V(t) = \sum_{p=1}^{Z_2} \frac{Z_1 e^2}{\left| \vec{r}_p - \vec{r}(t) \right|},$$
(2)

where \vec{r}_p are the coordinates of the protons in the target nucleus, and where $\vec{r}(t)$ gives the classical trajectory of the projectile considered as a point charge. Since the probability for exciting the nucleus in any single collision is small, the excitation process may be treated by quantum-mechanical perturbation theory.

The collective nuclear excitations are produced by the electric quadrupole component of (2) and, for the total cross-section for excitation of a given level, one obtains

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

where *m* is the reduced mass. The quantity $B_e(2)$ is a constant containing the nuclear matrix element; this quantity also determines the electric quadrupole (*E*2) decay probability for the inverse transition. Theoretical values of *B* for rotational excitations are quoted in the following paragraph. The last factor $g_2(\xi)$ can be expressed in terms of integrals over the trajectories of the bombarding particles and depends on the parameter

$$\xi = \frac{\Delta E}{2E} \frac{Z_1 Z_2 e^2}{\hbar v},\tag{4}$$

where E is the bombarding energy and ΔE the nuclear excitation energy. The parameter ξ thus represents the ratio of the collision time to the nuclear period; for small ξ , the function g_2 approaches the constant value 1.13 while, for large ξ , the collisions become adiabatic, resulting in an exponentially decreasing g_2 . The function $g_2(\xi)$ has been evaluated numerically (cf. forthcoming publication by A. WINTHER, whose results for g_2 are also given in B.-M., Appendix VI).

The angular distribution of the emitted radiation can also be expressed in terms of integrals over the trajectories, which have been calculated numerically (cf. forthcoming publication by K. ALDER and A. WINTHER).

3. Nuclear Rotational States.

The coupled particle-surface model predicts the occurrence of low-lying collective excitations for the strongly deformed nuclei encountered in regions removed from closed shells (B.-M., Chapter VI). These states exhibit a spectrum of rotational character, and also reveal themselves by their very large E2 transition probabilities.

In even-even nuclei, the spectrum is given by

$$E = \frac{\hbar^2}{2\Im} I(I+1) \qquad I = 0, 2, 4 \cdots$$
even parity
(5)

where the effective moment of inertia \Im is proportional to the square of the nuclear deformation, and is expected to vary slowly with the atomic number A. States of this type have recently been identified by the regularity of the spectrum, the systematic dependence of the energies on A, and the lifetimes which are often more than a hundred times shorter than expected for single-particle transitions (BOHR and MOTTELSON, 1952, 1953a, 1953b; cf. also FORD, 1953 and ASARO and PERLMAN, 1953).

In odd-A nuclei, the rotational spectrum is given by

$$E = \frac{\hbar^2}{2\Im} (I(I+1) - I_0(I_0+1)) \qquad I = I_0, I_0+1, I_0+2 \cdots \qquad (6)$$

same parity as ground state

where I_0 is the ground state spin^{*}. Since \Im is expected in general to vary slowly with A, the rotational excitation energies in odd-A nuclei can be related to those in even-even nuclei.

Due to the very large E2 transition probabilities for these rotational states, the method of Coulomb excitation is especially suited for their identification. The excitation cross-section (3) depends on the reduced transition probability $B_e(2)$ which for the rotational excitations $I_0 \rightarrow I_0 + 1$ and $I_0 \rightarrow I_0 + 2$ is given by (B.-M., § VII c.ii and Appendix VI)

* An additional term in the rotational energy, resulting in a less regular spectrum, occurs when the angular momentum of the particles along the nuclear axis equals $1/2 \hbar$ (cf. B.-M., § VIc.iii).

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$$B_e(2) = \frac{15}{16\pi} e^2 Q_0^2 \frac{I_0}{(I_0+1)(I_0+2)} \quad I_0 \to I_0 + 1$$
(7)

and

$$B_e(2) = \frac{15}{8\pi} e^2 Q_0^2 \frac{1}{(2I_0+3)(I_0+2)} \quad I_0 \to I_0+2.$$
 (8)

The latter formula also refers to the excitation of the (2+) first excited state in even-even nuclei.

The transition probabilities (7) and (8) are expressed in terms of the intrinsic nuclear quadrupole moment Q_0 , measured with respect to the nuclear axis (B.-M., Chapter V). The spectroscopically measured quadrupole moment Q is related to Q_0 by

$$Q = \frac{I_0}{I_0 + 1} \frac{2I_0 - 1}{2I_0 + 3} Q_0.$$
(9)

Thus, the measurement of the excitation cross-sections, just as of the corresponding E2 decay probability, provides a measure of the nuclear deformation which can be directly compared with the spectroscopic data (cf. B.-M., Table XXVII).

The rotational character of the collective excitation spectrum represents a limiting situation realized when the zero-point oscillations of the nuclear surface are negligible compared with the total deformation. These zero-point oscillations give rise to deviations from the expressions given in this section. In the region of closed shells, where the deformations are small, an entirely different spectrum results (B.-M., § VIc.i and ii).

4. Experimental Arrangement.

The 2 MeV electrostatic generator was used to produce a separated beam of protons, which passed through the system of stops shown in Fig. 1. The guard ring (G in the figure) was kept at a negative voltage of 100 volts in order to avoid the influence of secondary electrons on the current measurements.

The target holder was mounted in a horizontal tube in such a way that a number of different targets could be inserted into

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the beam and set at an arbitrary angle. An aluminum target holder and target tube were employed to make the absorption small. Aluminum, however, radiates strongly when bombarded



Fig. 1. Experimental arrangement.

with protons, and the target tube was, therefore, lined with a 0.1 mm tin foil in order to reduce the effect from the scattered protons. Tin is good for this purpose because its closed proton shell structure implies a low cross-section for Coulomb excitation; moreover, its nuclear charge is great enough that capture radia-

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tion is unimportant and small enough that the characteristic X-radiation does not interfere with the present experiments.

A 10 cm thick layer of lead, $50 \text{ cm} \times 50 \text{ cm}$, was placed above the counter in order to shield it from the hard X-rays from the acceleration tube. These X-rays would produce a background of soft, secondary radiation. The counter was further mounted in a 5 mm lead tube to absorb the scattered radiation. On the top of this tube various absorbers could be placed for the purpose of filtering the radiation from the target. The distance between the counter and the target tube could be fixed by means of two lucite rods when measuring angular distributions.

The γ -rays were detected by a cylindrical NaI (Tl) crystal, 4 cm in diameter and 3 cm high. The crystal was mounted on a RCA 5819 photo multiplier tube by means of a glass cap which was filled with Nujol mineral oil and covered on the outside with a layer of MgO.

The pulse spectrum was measured with a single-channel analyzer. The linearity of the equipment was checked by a relay-pulser connected to the preamplifier, and energy calibrations were made by means of the annihilation radiation from Na²² and by means of the characteristic K X-rays emitted by the bombarded targets.

5. Excitation of First State in Ta.

For a detailed study of the Coulomb excitation process, Ta is a particularly suited target, since it is known to have a large nuclear quadrupole moment, and occurs in a region of the elements where the trends of the rotational states seem well established. Moreover, Ta possesses only a single isotope ($_{73}$ Ta¹⁸¹).

A piece of Ta metal of 0.1 mm thickness (i. e. thick to protons of 2 MeV) was used as a target. The γ -ray spectrum shown in Fig. 2a was obtained with 1.75 MeV protons; the measurements were made at an angle of 80° with respect to the beam. The strong peak is due to the KX-radiation from the Ta atoms, which results mainly from the ejection of K-electrons by the bombarding particles (cf. CHADWICK, 1913; BOTHE and FRÄNZ, 1928; HENNEBERG, 1933 LIVINGSTON, GENEVESE and KONO-PINSKI, 1937), and partly also from the internal conversion of the nuclear excitation. The weak peak on the low energy side corresponds to the K-radiation from the Sn-lining of the target tube.



Fig. 2. Pulse spectra measured at a proton energy of 1.75 MeV. The Ta- C_1 peak is due to Coulomb excitation of the first excited state of Ta, and the K-peaks to the characteristic X-radiation from the atoms. Figs. a, b, and c show the spectra which are obtained for tantalum when various absorbers are used; Fig. d shows the spectrum for lead with a 1.5 mm Cu absorber.

The high energy peak (C_1 in the figure) is due to the Coulomb excitation of the Ta-nuclei. The relative intensity of the C_1 -peak with respect to the Ta K-peak can be increased by the insertion of absorbers, since the absorption coefficients are strongly energy dependent in this region. This is illustrated in Figs. 2 a, b and c. It was convenient, for most of the quantitative measurements, to use an absorber of 1.5 mm Cu, which makes the two peaks about equally strong.

The nuclear origin of the C_1 -peak was checked by measurements on Pb (Fig. 2*d*), which has a similar X-ray spectrum to that of Ta, but whose closed-shell nuclear structure implies high excitation energies and correspondingly small cross-sections for Coulomb excitation under the present conditions.

The cross-section for formation of a compound nucleus in the bombardment of Ta by 1.75 MeV protons is expected to be of the order of 10^{-38} cm² (cf., e.g., BLATT and WEISSKOPF, 1952, p. 352), which is many orders of magnitude too small to explain the observed yield. It is therefore strongly indicated that the excitations result from the influence of the electric field of the protons.

The energy of the C_1 -peak was found to be 135 ± 5 keV from the pulse size. An independent determination of the energy was made by absorption measurements which yielded 140 ± 7 keV, and also confirmed the monochromatic character of the radiation. As an average, we adopt the value 137 ± 5 keV for the energy of the C_1 -line. This value is in good agreement with the value of 136 keV found for the first excited level of $_{73}$ Ta¹⁸¹ in other experiments (cf. GOLDHABER and HILL, 1952).

The angular distribution of the C_1 -radiation was measured at bombarding energies of 1.25 MeV and 1.75 MeV in the region from 0° to 80°. When corrections were made for the absorption in the target material, approximately isotropic distributions were found, in agreement with theoretical expectations for the transitions in question.

The thick target yield for the C_1 -radiation was measured at 80° with respect to the beam for bombarding energies from 1 to 2.2 MeV. The total yield of γ -quanta per proton, obtained by assuming isotropic distribution, is shown in Fig. 3. The solid curve represents the theoretical energy dependence for E2 Coulomb excitation obtained from (3), using the numerically calculated g_2 function. Since the energy loss ΔE in the nuclear excitation process is not quite negligible compared with the bombarding energies used, the theoretical curve has been calculated for an effective energy equal to the incident energy minus $1/2 \Delta E$. The stopping power for Ta is taken from the semi-empirical relation given by LINDHARD and SCHARFF (1952).

The close agreement between the energy dependence of the experimental yield of the C_1 -radiation and that given by the theory confirms the interpretation of the observed effects in terms of Coulomb excitation by the electric quadrupole field of the protons.

For a bombarding energy of 2 MeV, a cross-section for γ -emission of approximately 0.5 millibarns (mb) was obtained. In order to derive the total excitation cross-section, it is necessary to take into account the de-excitation by internal conversion. The decay following the E2 excitation is expected to be E2 or M1, both of which yield total conversion coefficients of about 2 (Rose et al., 1951; GOLDHABER and SUNYAR, 1951), which is also consistent with the experimentally measured electron yield (FAN,



Fig. 3. Total yields for the Ta-K, Ta- C_1 , and Ta- C_2 peaks. The theoretical curves are adjusted to fit the measured yields at a proton energy of about 1.8 MeV.

1952). Employing the value 2 for the total conversion coefficient, an excitation cross-section of approximately 1.6 mb is found for protons of 2 MeV.

From the measured cross-section, one derives from (3) the reduced transition probability $B_e(2)$, and the value obtained is more than a hundred times that expected for the excitation of a single particle in the nucleus (cf. B.-M. § VII b.i for estimates of $B_e(2)$ for single-particle transitions). Thus, the observed large cross-section directly indicates a collective excitation and suggests a rotational interpretation.

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The rotational spectrum (6) for an odd-A nucleus depends on the ground state spin, which for $_{73}$ Ta¹⁸¹ is known to be 7/2 (cf. MACK, 1950). The first rotational excitation should thus have I = 9/2 and an energy of $9 \hbar^2/2$ \Im which is 3/2 times the rotational energy expected for the first excited (2+) state in an even-even nucleus with the same value of \Im (cf. (5)). The energy of 93 keV for the first excited state in $_{72}$ Hf¹⁸⁰ (cf. Scharff-Gold-HABER, 1953) thus implies an energy of about 140 keV for the 9/2 state in Ta, in good agreement with the 137 keV γ -ray produced by Coulomb excitation^{*}.

Assuming the rotational interpretation of the 137 keV state, one derives from the empirically determined $B_e(2)$, by means of (7), the intrinsic nuclear quadrupole moment $Q_0 \simeq 7 \times 10^{-24}$ cm². This may be compared with the value $Q_0 \simeq 14 \times 10^{-24}$ cm². derived by means of (9) from the quadrupole moment Q determined from the atomic hyperfine structure (BROWN and TOM-BOULIAN, 1952). Another estimate of deformations for nuclei in this region may be obtained from measured lifetimes of rotational states in even-even nuclei, which yield $Q_0 \simeq 8 \times 10^{-24}$ cm² (cf. B.-M., Table XXVII). The three estimates of Q_0 are of the same order of magnitude; it does not seem excluded that the differences may be attributed to experimental uncertainties.

6. Excitation of Second Rotational State in Ta.

The rotational interpretation of the first excited state in Ta implies the existence of a second rotational state with spin 11/2, and an energy of 20/9 times that of the first state. Since this state may also be excited in an E2 transition of collective type, it is expected to have an appreciable cross-section for Coulomb excitation. However, the yield of the higher energy γ -ray is expected to be considerably smaller than that for the 137 keV γ -line for several reasons: partly, the value of $B_e(2)$ for the former transition is about four times smaller than for the latter (cf. (7) and

* A level scheme for Ta^{181} has been given (GOLDHABER and HILL, 1952) according to which the first excited state of 136 keV is assigned a spin of 7/2. However, it does not seem inconsistent with the data to identify this level with the one observed in the Coulomb excitation process, and to assign it a spin of 9/2.

(8)); partly, the higher value of ξ implies smaller values of $g_2(\xi)$; finally, the de-excitation of the higher state may proceed either by a cascade through the 137 keV state or by a cross-over transition to the ground state, of which only the latter could be detected with the available resolution.



Fig. 4. Pulse spectra for tantalum and tungsten, measured at a proton energy of 2.1 MeV and with a 5.7 mm Sn-absorber. In the insert, the difference between the yields for Ta and W is plotted in the region of the Ta- C_2 line, which corresponds to the second excited state in Ta. No other γ -lines were found in Ta.

The importance of this level as a test of the rotational spectrum led us to undertake a careful search for higher energy γ -rays. In order to determine the background radiation, measurements were also made with targets of the neighbouring element W, since, in the relevant energy range, W is not expected to give rise to appreciable γ -radiation resulting from Coulomb excitation (cf. the following paragraph).

The comparison between the γ -spectra obtained with Ta and W targets, for a proton energy of 2.1 MeV, is shown in Fig. 4 and clearly reveals a higher energy line in Ta (the Ta- C_2 peak).

In order to exclude the possibility of effects arising from the coincidence of two C_1 -quanta or from the distortion of the back-

ground due to absorbers, the spectrum was measured with a number of different strong absorbers. The interpretation of the yield from W-targets as representing the background for higher energies was further supported by the observation that the Ta and W yields in all cases coincided at a point somewhat above the Ta- C_1 peak.

Subtracting the background from the observed Ta-spectrum, the Ta- C_2 peak shown in the insert to Fig. 4 was obtained. The energy of the Ta- C_2 line was determined to be 300 ± 10 keV, which corresponds to a ratio of 2.2 for the C_2 and C_1 energies. This ratio is in very good agreement with the ratio 20/9 predicted for the rotational spectrum.

The observed yield for the C_2 -line, which is plotted in Fig. 3, also agrees with the theoretical energy-dependence for E2 Coulomb excitation, shown by the solid line. The cross-section for the emission of the 300 keV γ -ray was found to be about 0.02 mb for a proton energy of 2 MeV.

From (3) and (8), assuming the value of Q_0 deduced from the excitation cross-section of the 137 keV line, one calculates a cross-section of 0.10 mb for the excitation of the 300 keV level. The conversion coefficient for the 300 keV γ -line is expected to be only of the order 0.05—0.10, since this transition is of E2 type. The comparison between the excitation cross-section and the γ -yield therefore indicates that about 20 % of the de-excitations take place via the cross-over transition to the ground state.

The relatively large probability for cross-over transitions is characteristic of the rotational spectrum, arising from the strong enhancement of the E2 radiation (cf. B.-M. § VIIc.ii).

From the branching ratio one can obtain an estimate of the M1 transition probability for the $\Delta I = 1$ radiative transition within the rotational family. This transition probability can also be estimated from the static magnetic moment of the ground state (B.-M. § VIIc.ii). The value obtained in this way is somewhat smaller than that indicated by the branching ratio, but the estimate is very sensitive to the value of the magnetic moment. A precision measurement of this moment would thus be of interest.

7. Coulomb Excitation of W.

In even-even nuclei with large deformations, the first excited (2+) states should show up strongly in the Coulomb excitation process. As a first example, $_{74}W$ was selected since it consists predominantly of even isotopes, and since the excitation energies in this region indicated that it would be possible to resolve the nuclear radiation from the *K* X-rays.

The W-spectrum is shown in Fig. 5 and clearly exhibits the well separated peaks resulting from the X-radiation (W-K) and the Coulomb excitation $(W-C_1)$. The $W-C_1$ peak was observed



Fig. 5. Pulse spectrum for tungsten, measured at a proton energy of 1.75 MeV and with a 1.5 mm Cu absorber.

to be composite, presumably arising from the different isotopes, since its position and shape could be altered by the use of absorbers.

As an average energy of the $W-C_1$ lines, we obtained about 115 keV, which is consistent with the known excitation energies of 102 keV for W^{180} and 123 keV for W^{186} (cf. SCHARFF-GOLD-HABER, 1953).

The cross-section for γ -emission at a proton energy of 2 MeV is found to be about 1.5 mb, assuming the theoretical energy dependence (3). Using an average total internal conversion

coefficient of 2 for these E2 transitions, one obtains from (8) a deformation of $Q_0 \simeq 7 \times 10^{-24}$ cm². which is just of the same magnitude as that observed in the excitation of the neighbouring element Ta. This value of Q_0 is also in agreement with that derived from the measured lifetimes of excited states of eveneven nuclei in this region.

8. Yield of KX-rays.

As a by-product of these investigations, we obtained the yield of X-rays resulting from the ejection of K-electrons by protons on Ta. The results are shown in Fig. 3. The X-ray yield has been corrected for internal conversion of the Ta- C_1 line, which amounts to about 20 % of the total X-ray yield. The solid line represents the theoretical energy dependence (HENNEBERG, 1933), which is seen to agree rather well with the measurements. The absolute yield is found to be about a factor of two smaller than predicted by the theory, but it is not yet clear to what extent this discrepancy is to be attributed to experimental uncertainties or to the approximations involved in the theory. A comparison of the Ta-K and Pb-K yields for a proton energy of 1.75 MeV was found to be in good agreement with the theoretical Z-dependence.

9. Conclusions.

One may summarize as the main conclusions of these investigations:

1) The feasibility of nuclear excitation by the electric field of bombarding particles has been confirmed.

2) Over the energy region investigated the theoretical energy dependence of the cross-sections has been verified.

3) The first two rotational levels in Ta have been found at the predicted energies.

4) The large cross-sections for excitation of the levels in Ta and W confirm the collective character of the excitation.

5) The nuclear deformations deduced from the measured Dan. Mat. Fys. Medd. 28, no: 1. 2

cross-sections are of the same magnitude as those derived from spectroscopic evidence and measured lifetimes.

The use of the electric field of charged particles to excite nuclear levels provides the possibility of studying the spectra of a wide variety of nuclei. Preliminary experiments have also exhibited the effect in other elements, and further investigations of the Coulomb excitation processes are in progress.

This work has been performed at the Institute for Theoretical Physics, University of Copenhagen. We wish to thank Professor NIELS BOHR for his continued interest in the experiments and for the good working facilities provided at the Institute. We are also very grateful for the many stimulating discussions on the subject, which we have had with A. BOHR and B. MOTTELSON, in close co-operation with whom the theoretical parts of this paper have been written. To A. WINTHER and K. ALDER we are indebted for making results of their calculations available to us in advance of publication. Our thanks are further due B. MADSEN for developing the electronic equipment used in the investigations, and J. BJERREGAARD for aid in the experiments.

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