MULTIPLE COULOMB EXCITATION IN Th^{232} AND U^{238}

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Electromagnetic excitation or "Coulomb excitation" of a nucleus by the electric field of a passing ion occurs most readily in nuclei having low-lying states with the character of collective excitations of the ground-state configurations.[1] Electric quadrupole excitations are by far the most important. "Multiple Coulomb excitation" refers to excitation of higher states through successive quadrupole excitation steps. In an earlier paper,[2] the double Coulomb excitation of even-even tungsten isotopes by O^{16} ions was reported (excitation of the 4+ member of the ground-state rotational band through the 2+ state).

The probability of multiple Coulomb excitation should increase rather rapidly with increasing projectile charge, and Coulomb excitation in general is most favorable where the rotational level spacings are smallest. With these factors in mind, search was made for higher than second-order Coulomb excitation in Th^{232} and U^{238} by irradiating with Ne^{20}, S^{32}, and A^{40} ions produced by the Berkeley heavy-ion linear accelerator (Hilac). These particular target elements also have the virtue of being monoisotopic. (The uranium used was isotopically depleted in U^{235} and U^{234}.) In this paper evidence is presented for excitation up to sixth order in U^{238} and fifth order in Th^{232}.

Because of low beam intensities (1-5 micro-microamperes) it was necessary to employ thick targets of thorium and uranium metal (2-4 mils). The gamma rays resulting from Coulomb excitation were detected with a 3 in. $\times$ 3 in. NaI(Tl) crystal assembly mounted behind the target and recorded with a 100-channel pulse-height discriminator. In order to enhance the events from multiple Coulomb excitation and to reduce the background radiation, the gamma-ray detector was operated in fast coincidence (3×10^{-8} sec) with pulses produced by the back-scattered heavy ions in an argon scintillation chamber. These gating pulses were cut off below about 10 Mev.

The gamma-ray spectra obtained with A^{40} irradiation of U^{238} and Th^{232} are shown in Figs. 1(a) and 1(b). The gamma-ray energies observed are listed in columns 2 and 5 of Table I except that the energies for the 2+ → 0+ transitions were taken from other experiments. The 4+ → 2+ transition in U^{238} (103 kev) is somewhat uncertain both in energy and intensity because uranium K x-rays lie at 94 to 98 kev and accurate resolution was not possible. In Figs. 1(a) and 1(b) the small peaks which appear beyond energies of 311 kev and 273 kev can be reasonably well explained both in energy and intensity, as due to coincident arrival at the detector of lower energy photons. Such pile-up peaks, occasioned by the high acceptance angle of the detector, would make a negligible contribution to the gamma rays listed in Table I.

The interpretation of these data in the form of level schemes is shown in Fig. 2. The evidence is largely indirect since it would be experimentally difficult to determine all of the gamma-gamma coincidences in the de-excitation sequences. However, as a partial check, it was established that in U^{238} the 214-kev (8+ → 6+)

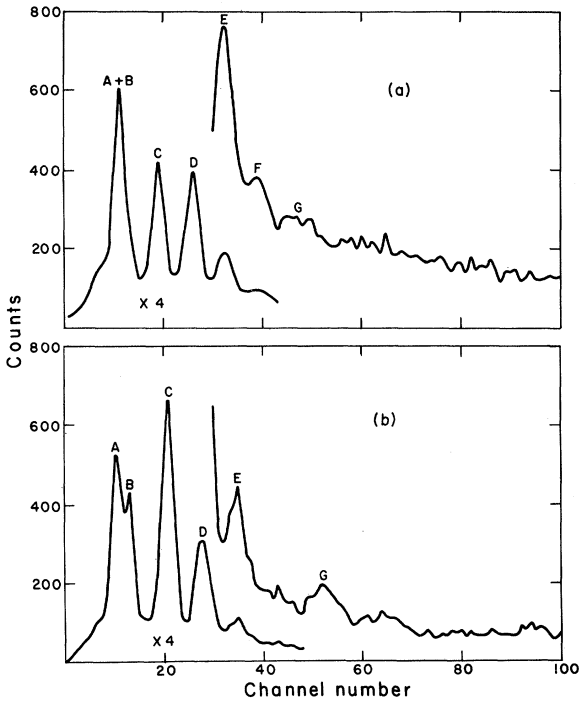

FIG. 1. De-excitation gamma-ray spectrum of (a) U^{238}, (b) Th^{232} irradiated with 190-Mev A^{40} ions. A, B, C, D, E, and F correspond to K x-rays, 4+ → 2+, 6+ → 4+, 8+ → 6+, 10+ → 8+, 12+ → 10+ transitions, respectively, and G is a pile-up peak (see text).

Table I. Transition energies between rotational states produced by multiple Coulomb excitation.

Transition	U^{238} transition energies (kev)			Th^{232} transition energies (kev)			
	Exper.	Calc. for $A=7.45$	Calc. for $A=7.46$, $B=-2.7\times10^{-3}$	Exper.	Calc. for $A=8.33$	Calc. for $A=8.38$, $B=-8.5\times10^{-3}$	Calc. for $A=8.34$, $B=-1.2\times10^{-2}$, $C=4\times10^{-5}$
2+→0+	(44.7 ± 0.2)[a]	44.7	44.7	(49.75 ± 0.25)[b]	50	50	50
4+→2+	103 ± 2	104	103	113 ± 2	117	114	113
6+→4+	161 ± 2	164	162	170 ± 3	183	173	170
8+→6+	214 ± 3	224	215	222 ± 4	250	222	221
10+→8+	264 ± 4	283	265	273 ± 5	317	259	272
12+→10+	311 ± 5	343	310				

[a]Taken from J. O. Newton, reference 5.
[b]This value was measured by the present authors from U^{236} α decay using a xenon-filled proportional counter.

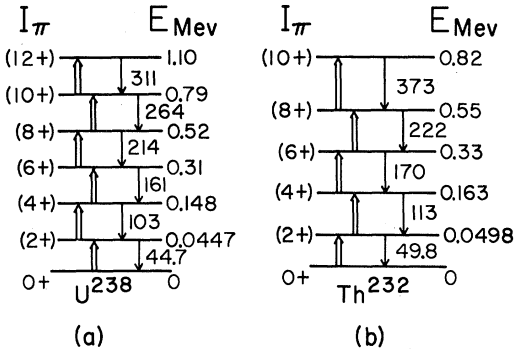

FIG. 2. Rotational energy level schemes proposed for (a) U^{238} and (b) Th^{232}.

gamma ray and part of the 103-kev (4+→2+) intensity are in coincidence with the 161-kev (6+→4+) transition. The principal pieces of evidence are the following: (1) The energies of the gamma rays are in agreement with expectations for excitation of rotational bands, (2) the yields of the photons for different bombarding ions and energies would be difficult to explain on any basis other than multiple Coulomb excitation of rotational bands, and (3) no other low-lying states are known in this region of the periodic system which would not at the same time give abundant crossover transitions of easily discernible energies.

It is now well known that levels of a rotational band of an even-even nucleus, such as those in Fig. 2, have energies given approximately by $E_I=AI(I+1)$, where E_I is the energy of the state with spin I and I is restricted to even integers.[3]

The empirically determined constant A is related to the effective moment of inertia, $\mathcal{I}$, by $A=\hbar^2/2\mathcal{I}$. The first excited states (2+) have been established by other Coulomb-excitation work,[4,5] and the energies obtained have been used to determine A. With this value of A the energies of other states may be calculated. The corresponding energy spacings of the rotational bands are listed in columns 3 and 6 of Table I. It is not surprising that agreement with the experimental results is increasingly poor for the higher states since it is expected (and found in other cases) that higher order terms should be added to the expression for E_I to account for rotational-vibration interactions. The second term $BI^2(I+1)^2$ contains another constant, B, which can be related to vibrational energies. Columns 4 and 7 of Table I show that this second term is sufficient to bring into agreement the U^{238} data; but for Th^{232} a third term, $CI^3(I+1)^3$, is required (column 8).

Table II gives a comparison of these constants for several nuclei in the heavy-element region.

Table II. Rotational constants in the heavy elements.

Nuclide	A (kev)	$-B$ (kev)	C (kev)
Pu^{240}	7.17	3.9×10^{-3}	
Pu^{238}	7.37	3.6×10^{-3}	
U^{238}	7.46	2.7×10^{-3}	$<2\times10^{-6}$
U^{234}	7.29	6.9×10^{-3}	$(\sim3\times10^{-5})$
Th^{232}	8.34	1.2×10^{-2}	4×10^{-5}
Ra^{226}	11.74	8×10^{-2}	$(\sim9\times10^{-4})$

As is well known, the value of A is rather constant for elements above thorium and increases rather sharply for lighter elements approaching the closed shells at Pb^{208}. Indeed, below radium the rotational picture is no longer a proper description of lowest-energy collective modes of motion. It is apparent that if accurate values for B and C are to be obtained and only a few members of a rotational band can be measured, great precision in the energy determination is required. However, if (as in the present study) a large number of states are discernible, high precision is not required. The values of C shown for Ra^{226} and U^{234} are only estimates which are entered to give some comparison with the more accurate values obtained in the present experiments.

Cross sections for Coulomb excitation have proved to be of great value because they are directly related to $E2$ transition probabilities and provide information for nuclear models. Such information on multiple Coulomb excitation would also be of interest as a guide in extending Coulomb excitation theory. Unfortunately, meaningful cross sections cannot be obtained from the present experiments because (1) it was necessary to use thick targets and the inherent difficulties in obtaining an excitation function are complicated by uncertainties in the dE/dx relation for argon ions; (2) only a portion of the excitations were observed, those for which the heavy ion was scattered through angles between 90° and 160° and emerged from the target with enough energy to enter the gas scintillation counter with greater than 5 to 10 Mev. Nevertheless some relative yields have been calculated for two different A^{40} energies (see Table III) and these will be explained presently.

There has been no published theoretical treatment applicable to high-order multiple excitation, but Alder and Winther[6] are now considering the problem. Their first results seem to be in excellent agreement with our observations in that qualitatively the predicted trends are being followed.

In columns 2 and 3 of Table III we have summarized the relative independent yields for excitation of the indicated levels in Th^{232} at two different A^{40} ion bombarding energies. These values were obtained from the integrated photopeaks corrected for absorption in the target and backing plate and for the counting efficiency of the NaI crystal. Correction was made for internal conversion (assuming $E2$ transitions), and allowance was made for the cascade from higher levels.

Table III. Relative yields from A^{40} on Th^{232}.

E_γ	Parent level	Direct yield of parent level $E(A^{40}) = 158$ Mev	$E(A^{40}) = 190$ Mev
113	4+	100	100
170	6+	46	88
222	8+	10	26
273	10+	0.7	5.9

The yield for the 4+ level has been normalized to 100 for each energy. It is seen that each successive level has an appreciably steeper excitation function so that at the higher bombarding energy the 10+ level is about eight times more intense relative to the 4+ than at the lower bombarding energy. This is explained in a natural way according to the scheme in Fig. 2, since here each higher state results from a higher order multiple excitation. Other explanations of this behavior are not so easy. If, for example, the gamma rays come from ordinary single $E2$ excitations from the ground state, then we would expect the 273-kev peak to increase only about 10% more than the 113-kev peak over this range of bombarding energy. To obtain the much greater variation in yield indicated in Table III, one would have to assume either a much higher (and different) energy of excitation for each transition, or excitations of higher multipole orders. Neither of these possibilities is very likely. The latter—the direct excitation of the rotational band members by $E4$, $E6$, etc., excitations instead of by the multiple process—was shown to be negligible in the case of double excitation in the tungsten isotopes using O^{16},[2] and should be even less important with a particle of higher charge. The yields seem entirely reasonable for the experimental conditions used (thick targets and back-scattered projectile ions). Had we used thin targets, the results of Winther and Alder would suggest that at the higher bombarding energy the primary population of the 6+ and perhaps the 8+ states would have been higher than for the 4+ state.[6] Such reversals in yield would be interesting to observe, but the difficulties associated with thin targets in our apparatus are rather severe. Data similar to those in Table III have been obtained for both U^{238} and Th^{232} with different bombarding ions. In all cases these data are consistent with the schemes in Fig. 2 insofar as it is possible to determine at the

present time.

We are grateful to Dr. J. O. Newton who participated in many of the early phases of this work. We are also greatly indebted to Dr. E. Hubbard and the operating crews of the Hilac for the indispensable help they have given us in carrying out these experiments.

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CAPTURE IN (K^-, p) ATOMS

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Day, Snow, and Sucher[1] have concluded that the reactions of K^- mesons at rest in liquid hydrogen almost invariably result from the interaction of the (K^-, p) system in states of zero relative orbital angular momentum. This conclusion results from a calculation, based on a suggestion by Madansky, that indicates that collisions of the protons in the liquid hydrogen with the (K^-, p) atom induce in the atom a strong Stark effect which mixes S-wave orbital states into states of higher orbital angular momentum. The very strong (K^-, p) S-wave absorption then depopulates these states before radiative transitions leading to the $2P$ state can occur. Previously, it had been generally believed that capture from the $2P$ state might be more important than radiative $2P \rightarrow 1S$ transitions, and that the (K^-, p) capture process might proceed predominantly through P-state capture. If it can be established that this view is incorrect and that capture almost invariably results from the S states, the parity of the K meson might be determined,[2] and other important conclusions might be reached, by a study of the $K^- - p$ and $K^- - d$ reactions for stopped K^- mesons. It is the purpose of this note to point out corrections to the specific calculations of Day et al. which appear to vitiate largely their specific conclusions that P-state capture cannot be important.

Day et al. consider the (K^-, p) atom moving essentially with thermal velocity through the liquid hydrogen. Frequent collisions with protons in the liquid result in the atom being subjected to intense electric fields which polarize the atom, or induce Stark transitions, for a period of time of the order of $2a_0/V$, where a_0 is the proton Bohr radius and V is the velocity of the atom. Dealing with the $n = 6$ level, largely for the sake of definiteness, they calculate the Stark transition rate due to an electric field, $E = e/a_0^2$, of a proton at a distance of one Bohr radius. These transition rates are large, and Day et al. then conclude that the mixing is such that P states are completely depopulated in a collision while the other angular momentum states are rearranged into a $2l + 1$ statistical population. They do not consider immediate capture from states of higher orbital angular momentum presumably because the collision time is not assuredly long compared to the time required to establish a complete mixing of states. Using these approximations, they follow an assembly of atoms through their history and conclude that, from statistically weighted $n = 6$ states, 1.4% eventually reach the $2P$ level by radiative transitions while 98.6% are captured in S states. From states of higher n even fewer reach the $2P$ level.

The collisions considered by Day et al. are collisions of the second kind with a negligible energy transfer but with the transfer, from the (K^-, p) atom to the system of the atom and the colliding proton, of one or more units of angular momentum. The DeBroglie wavelength characteristic of an average collision is about 2π A while the effective collision radius a_0 used by Day et al. is a Bohr radius or about $\frac{1}{2}$ A. Since the ratio a_0/λ is then about $\frac{1}{2}$, classically collisions with $l \geq 1$ are forbidden. An estimate of the correction to