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DETERMINATION OF THE MULTIPOLARITY OF THE 0.3-sec Ta^{182} ISOMERIC TRANSITION BY ITS L X-RAY PATTERN ALONE*

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We have determined the multipolarity of the short-lived isomeric transition in Ta^{182} to be $M2$; we have measured the half-life and transition energy; we have observed population of the state from 16-min Ta^{182m}; and we make putative spin and parity assignments. Determination of the multipolarity was by a new method which should have applicability to other cases of low-energy highly converted transitions in nuclides with $Z > 60$.

A 0.33-sec isomer produced in neutron irradiation of Ta was discovered by Campbell and Good[1] in 1949 and additional details were reported in 1950 and 1951 by Goodrich and Campbell[2] and by Kahn.[3] The only radiations observed consisted of 8- to 11-keV photons, presumably L x rays from a low-energy, highly converted nuclear transition. No further experimental work on the isomer has been published, and questions about it such as its multipolarity, transition energy, isotopic assignment, and location in a nuclear level scheme have long remained unanswered.[4] Thermal-neutron-capture gamma data[5] and (d,p) reaction data[6] do not provide any direct evidence for the character or even the existence of the isomer. However, on the theoretical side, Bizzarri, Nunberg, and Prosperi[7] in a detailed treatment of the level structure of Ta^{182} predict a low-lying 5^+ level to be the bottom level in the decay of the well-known[8] 16-min Ta^{182m}.

To clarify this situation, we have reinvestigated the 0.33-sec activity, using a pulsed TRIGA reactor (peak flux $>10^{15}$ n/cm^2 sec), a fast pneumatic rabbit (transit time ~50 msec), and NaI and Ge(Li) detectors with 10-mil Be windows. The Ge(Li) detector[9] has a cooled field-effect transistor preamplifier; the full width at half-maximum at 14.4 keV is 550 eV. The overall system is briefly described elsewhere.[10] The samples used were 99.988% natural Ta vacuum evaporated onto $\frac{1}{2}$-mil Mylar foil; sample thickness was 0.15 μ, and the polyethylene rabbit capsule had a 1.2-mm wall. We have studied the photon spectrum, the half-life, and the relationship of the 0.33-sec activity to the 16-min isomer.

The only short-lived radiations seen in the present experiment were Ta L x rays, which were clearly resolved into L_α, L_β, and L_γ components; see Fig. 1. Intensity ratios (after wall, window, and sample absorption corrections) were $\beta/\alpha = 1.005 \pm 0.018$ and $\gamma/\alpha = 0.193 \pm 0.006$. From these observations alone, we are able to deduce that the isomeric transition has $M2$ multipolarity. Because the method employed to obtain this result is new, it is explained in some detail below. The half-life was found to be 0.283 ± 0.003 sec.

Experimental evidence that the short-lived level is fed by the 16-min isomer was obtained by counting delayed L x rays that follow K x rays arising from internal conversions in the 16-min decay. Samples irradiated to a weak level of 16-min activity were placed between two $\frac{1}{32}$-in. NaI Be-window detectors in close contact; a K x ray in either detector triggered a multiscaler (20 msec/channel) in which L x rays were counted. The resulting L x-ray half-life was 0.4 ± 0.2 sec. (The statistical accuracy is poor here because very low counting rates were required to reduce chance counts to an acceptable level.)

Combination of our results with the known decay properties of the 16-min isomer[8] and the reasonable assumption that the 283-msec isomer decays directly to the 3^- ground state leads to the level scheme shown in Fig. 2. These experimen-

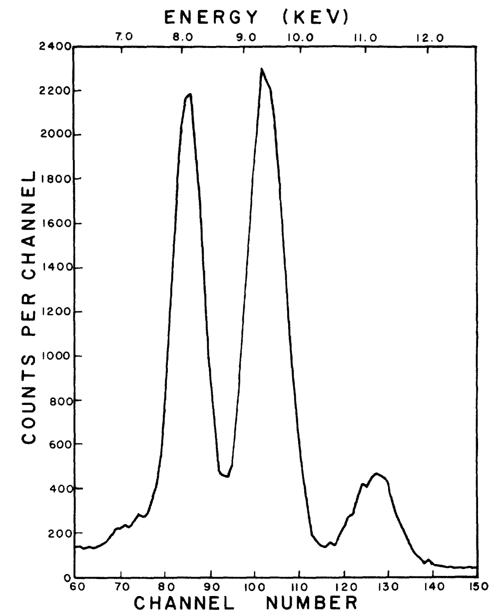

FIG. 1. The L x-ray spectrum of the short-lived Ta^{182} isomer. The curve is a sum of data from nine irradiations, each counted for 1.5 sec with a Be-window Ge(Li) detector. Corrections for sample self-absorption, window absorption, capsule wall absorption, and variation with energy of detector efficiency have not been applied in this plot. No other photons of intensity greater than ~0.5 % of the total L x-ray intensity were observed in an energy range between L-shell and K-shell binding energies.

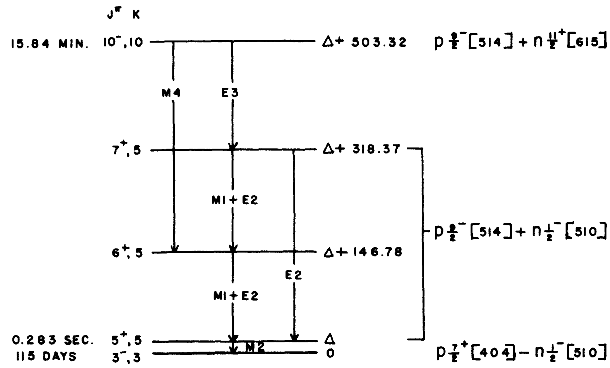

FIG. 2. Level structure of the two isomers in Ta^{182}. Present results establish the $M2$ multipolarity of the 0.283-sec transition and its position in the decay scheme. The other information shown is taken from references listed in footnote 8. The energy Δ is not known precisely; upper and lower limits are discussed in the text. The Nilsson assignments are those of Bizzarri, Nunberg, and Prosperi (Ref. 7).

tal results thus confirm the spins and parities of these levels as predicted by Bizzarri, Nunberg, and Prosperi.[7] The Nilsson assignments given by them are also shown in Fig. 2. The excitation energy Δ has not been measured but must lie between the L_1 subshell binding energy (11.68 keV) and an upper limit of ~35 keV which is set by the lack of low-energy conversion electrons in an experiment[7] with an electron-energy lower cutoff of 25 keV. Additional limits on the value of Δ are discussed below.

Determination of the multipolarity of highly converted low-energy (less than K-shell binding energy) transitions has always been difficult. In some cases, L-subshell conversion ratios have been used, but if (as in the present case) the transition energy is close to L-shell binding energies, conversion-electron intensities become extremely hard to measure accurately because of energy loss and scattering of low-energy electrons. The new approach developed and utilized in the present experiment is based upon using the relative intensities in the L x-ray pattern as clues to the L-subshell conversion coefficient ratios, and hence to the multipolarity. Observation of electrons is thus not necessary, and though low-energy photons are strongly absorbed, experimental or theoretical absorption corrections are readily applied.

Calculated internal conversion coefficients give the relative numbers N_1, N_2, N_3 of vacancies produced in the respective L subshells as a function of Z, transition energy, and multipolarity; in this work, the tables of Hager and Seltzer[11] were used. These vacancies can be filled by radiative transitions from higher shells, or by radiationless (Auger-type) transitions. Radiationless transitions are usefully divided into two classes, depending upon the initial state of the electron filling the vacancy. If it is in a higher subshell of the atomic shell containing the vacancy, it is called a Coster-Kronig transition; if in a higher (major) shell, an Auger transition. For example, an L_1 vacancy can be filled by a radiative transition from the M or higher shells, by an L_2 or L_3 electron in a Coster-Kronig transition, or by an M or higher shell electron in an Auger transition. The relative probabilities of these alternatives are designated by ω_1, f_{12}, f_{13}, and a_1 respectively; their sum is equal to 1. Similar parameters (ω_2, f_{23}, a_2, ω_3, and a_3) apply to the L_2 and L_3 subshells. The Coster-Kronig yields f_{ij} are limited to $i<j$ by energy considerations. Because of Coster-Kronig transitions, a vacancy initially in the L_1 shell may result in L x-radiation charac-

teristic of (radiatively) filling an L_2 or L_3 vacancy. The full relationships for the number X_i of L_i-subshell x rays produced by N_1, N_2, N_3 initial vacancies are

$$X_1 = N_1\omega_1,$$

$$X_2 = (N_2 + N_1 f_{12})\omega_2,$$

$$X_3 = [N_3 + N_2 f_{23} + N_1(f_{13} + f_{12}f_{23})]\omega_3.$$

(A more complete treatment of these topics may be found in Fink et al.[12] and references listed therein, especially Listengarten.[13])

However, the fluorescence yields ω_i as conventionally defined include all L x rays filling an L_1 subshell vacancy. There are in fact 44 different L x-ray wavelengths listed for Ta in Bearden[14]; under moderate resolution such as ours these fall into the three main groups called L_α, L_β, and L_γ. For our purposes it is necessary to introduce new parameters which we call "partial fluorescence yields" designated by $\omega_{i\alpha}$, $\omega_{i\beta}$, and $\omega_{i\gamma}$, corresponding, respectively, to the production of L_α, L_β, and L_γ x rays by radiative filling of an L_i vacancy; their sum is ω_i.[15]

Application of these relationships to a multipolarity determination involves calculating curves of intensity ratios β/α and γ/α for various multipolarities as a function of transition energy for the relevant Z. (Ratios are sufficient since absolute L x-ray intensities are not determined.) A major difficulty is encountered in these calculations: For most Z not all of the six parameters ω_i and f_{ij} have been experimentally determined (see Ref. 12) and theoretical estimates are not reliable. Fortunately, in the case of Ta good experimental values exist (due largely to the work[16] of Crasemann's group) for ω_2, ω_3, f_{23}, and also for an equation relating ω_1 and f_{12}. The parameter f_{13} has a relatively weak influence on the L x-ray pattern. Using Listengarten's work[13] as a rough guide to reasonable ranges for the parameters, families of β/α and γ/α curves were plotted for various values of ω_1; this was done for $E1, E2, E3, E4$ and $M1, M2, M3, M4$ transitions for energies between L- and K-shell binding energies. Representative curves are shown in Fig. 3. A check on the overall method was carried out by measuring in a coincidence arrangement the β/α and γ/α ratios (1.21 ± 0.05 and 0.226 ± 0.019, respectively) for the known $E3$ transition in the 16-min isomer. Very good agreement was found with calculated values[17] which ranged from 1.16 to 1.20 for β/α, and 0.212 to 0.226 for γ/α, depending on the values used for ω_1 and f_{12}.

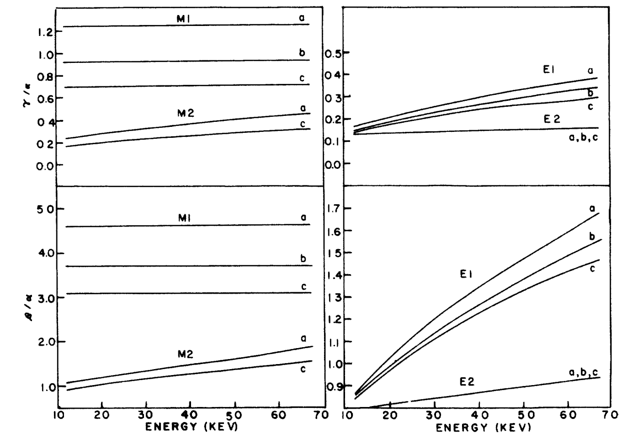

FIG. 3. Calculated β/α and γ/α ratios for Ta. Only $E1$, $E2$, $M1$, and $M2$ multipolarities are shown. Curves with label a are for $\omega_1 = 0.15, f_{12} = 0.17$; with b, $\omega_1 = 0.10, f_{12} = 0.33$; and with c, $\omega_1 = 0.05, f_{12} = 0.50$. The value assumed for f_{13} was 0.10. See text for details.

For the short-lived isomer itself, comparison of experimental ratios with the calculated curves immediately eliminated all multipolarities except $M2$, $E1$, and mixed $M1 + E2$. The latter two were eliminated on the basis of the absence of (unconverted) gammas in the measured spectrum. The calculated ratios of gamma intensity to total L x-ray intensity vary (depending on assumed transition energy Δ) from 0.25 to 2 ($E1$), 0.016 to 0.13 ($M1$), and 3×10^{-5} to 7×10^{-4} ($M2$); in the experimental spectrum any peak with ratio greater than 0.005 would have been clearly visible. The further possibility that Δ lies between L-subshell binding energies was investigated for all multipolarities but in no case was it possible to reproduce the experimental values. Corrections to the experimental values for variation of detector efficiency with energy (including Ge K-x-ray escape) were not made; however, careful estimates showed that such corrections could not affect the conclusions reached.

Interestingly, the multipolarity was determined without observing either gammas or conversion electrons, nor was it necessary to know Δ precisely. In fact, were the fluorescence parameters well known, Δ could have been determined, albeit indirectly, within 2 keV. Even with the present knowledge, we can conclude that Δ lies between 11.68 and ~18 keV.

This new technique for multipolarity determination should be applicable to other cases of low-energy highly converted transitions. With present instrumental resolution, the L x-ray pattern should be adequately resolved for $Z > 60$. The general lack of fluorescence parameters can be remedied in part by using known nuclear transitions to determine the (Z-dependent) transformation matrix between a vacancy pattern N_1, N_2, N_3 and the resulting x-ray pattern $L_\alpha, L_\beta, L_\gamma$. Work along these lines is underway at this laboratory.

A more detailed description of the technique and of its application to Ta will be submitted for publication elsewhere.

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