Δj selection rule does not seem to be responsible for the large relative $\int B_{ij}$ component. The absence of any indication of the $\int B_{ij}$ component in most first-forbidden β transitions with $\Delta I=0$, is possibly due to the large contributions of the $\lambda=0$ matrix elements ($\int i\gamma_5$, $\int \boldsymbol{\sigma}\cdot\mathbf{r}$).

The considerations presented above are obviously of a qualitative nature only. Even though a large amount of configuration mixing *within* major shells is undoubtedly present in the case of Sb^{124}, this does not change the qualitative aspect of the conclusion that the reduction of the $\lambda=1$ matrix elements is due to Δj selection rule effects. On the other hand, the fact that a large reduction of the $\lambda=1$ matrix elements has been established, shows that little or no configuration mixing *outside* major shells is present.

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L to K Ratios in the Electron Capture Decay of W^{181} and Ta^{179}†

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The L to K ratios in the electron capture decay of the isotopes Ta^{179} and W^{181} have been redetermined. Thin (0.060 in.) NaI crystals with thin (0.002 in.) Be windows were used to detect the L and K x rays. The partial fluorescence yields of the L subshells of Hf and Ta were also measured by determining the coincidence rates between the L and the K x rays emitted by the sources, It is necessary to know the fluorescence yields if the L to K ratios are to be determined from the measured intensities of the L and the K x rays. The L to K ratio of Ta^{179} was found to be 0.63 ± 0.06, which implies a total decay energy of approximately 115 ± 5 kev for this isotope. This energy is consistent with the observation that no gamma rays accompany the decay of Ta^{179} since the first excited level of Hf^{179} has an energy (122 kev) exceeding the total decay energy of Ta^{179}. The L to K ratio of W^{181} was found to be 0.23 ± 0.05, from which a decay energy of approximately 260 kev is computed. This result is in agreement with the fact that two weak gamma rays are emitted by the source, one at 137 kev and the other at 152 kev. These gamma rays correspond to excited levels in Ta^{181}. These gamma rays are in coincidence with the L x rays emitted by the source but not with the K x rays, which means that the total decay energy of W^{181} must exceed 166 kev. The L to K capture ratios reported here are not in good agreement with previously reported values.

INTRODUCTION

WHENEVER a nucleus is unstable against positron emission, but does not have the decay energy necessary to create the positron, it decays by capturing an orbital electron and emitting a neutrino. The most prominent electromagnetic radiations observed in an electron-capture decay are usually the characteristic x rays of the daughter nucleus. These may be either the K, L, or higher-shell x rays, depending upon which electron is captured in the decay. The relative number of vacancies created in the various atomic sub shells by the decay process depends on the nature of the β decay and on the total energy of the transition.

If the decay energy is very much larger than the electron binding energy in the various sub shells, then, for allowed and first-forbidden transitions, the ratios of K to L to M capture depend only on the electron densities in these shells at the nucleus. On the other hand, if the decay energy is comparable to the binding energy in the K shell, then the L to K capture ratio is a function of the decay energy, the dependence being such that the emission of high-energy neutrinos is favored.[1] Since no other way exists to determine the decay energy for electron capture processes, it is of some importance to measure the L to K capture ratios as accurately as possible.

It is difficult to determine the relative number of L and K vacancies from the relative intensities of the L and K x rays emitted by the source. In the case of K capture, most of the K x rays emitted also create an L-shell vacancy, and the L x rays emitted by the filling of these vacancies must somehow be distinguished from L x rays which follow L-electron capture. In addition every K or L vacancy created does not always result in the emission of the corresponding x ray. The energy of the transition can be dissipated by the emission of one of the outer electrons of the atom. (Electrons resulting from such processes are called Auger electrons.) For the isotopes studied here, this effect was not too important for K electrons since only about 7% of the

† Work done under the auspices of the U. S. Atomic Energy Commission.

[1] R. E. Marshak, Phys. Rev. **61**, 431 (1942).

K vacancies lead to the emission of an Auger electron. However over 70% of the L vacancies decay by emitting electrons, and it is therefore very important to know the ratio of electrons to x rays emitted by L vacancies since only the x rays are counted. The usual way in which Auger transitions are taken into account is to define a quantity called the "fluorescence yield" which is the ratio of the number of x rays emitted to the number of vacancies created in a given shell. Part of the object of the present experiment is to measure the fluorescence yields of the various atomic shells which take part in the electron-capture decay.

Two isotopes which decay by pure electron capture were studied, W^{181} and Ta^{179}. Both of these have long enough half-lives so that very detailed experiments can be performed. Previous work on W^{181} is somewhat contradictory. Bisi *et al.*[2] have measured the L to K ratio and find a number ($C_L/C_K=1.54\pm0.1$) which implies a decay energy of 92 kev. On the other hand, Cork *et al.*[3] have observed internal conversion electrons emitted by a W^{181} source corresponding to energy levels in Ta^{181} at 137 and 152 kev. The observed intensity of these lines is about 0.2% of the total number of decays. Debrunner *et al.*[4] have also observed these gamma rays with a scintillation spectrometer. These workers have also shown that the gamma rays are in coincidence with L x rays emitted by the W^{181} source, indicating that the gamma rays are not due to impurities. If these gamma rays correspond to levels in Ta^{181} at 137 and 152 kev, then the decay energy must exceed 164 kev and the L to K ratio must be considerably smaller than the value reported in reference 2. The results reported in the present work confirm the existence of the high-energy gamma rays and the value obtained for the L to K capture ratio is consistent with the higher decay energy required by the existence of the gamma rays. The present work therefore resolves the contradictory situation with regard to W^{181}.

In the case of Ta^{179}, the decay is simpler since no gamma rays are emitted by the source. The measured L to K ratio is sufficiently large so that the decay energy computed from this result does not exceed the energy of the first known level in Hf^{179}. However the L to K ratio reported here is not as large as the value (1.4 ± 0.4).[5]

EXPERIMENTAL METHODS

Scintillation counters were used in all the measurements reported here. There are two important features which a scintillation crystal sensitive to low-energy x rays must possess. The first is that the light output per ion pair must be as large as possible so that pulses due to low-energy quanta can be distinguished from phototube noise. This requirement means that NaI(Tl) crystals must be used. The second requirement is that absorption of the low-energy quanta in the window of the crystal must be minimized. (CsI crystals, which are not hygroscopic, could be operated without any windows, however their light output is not sufficiently large for the purposes of this experiment.) The most successful crystals were made as follows. A large piece of sodium iodide was taken, and thin ($\approx$0.060 in.) slices were cleaved from it using the usual techniques. A slice was then placed into a brass holder with a glass face on one side. Optical contact with the glass face was made with standard Dow-Corning silicone grease. A 0.002-in.-thick beryllium window was then cemented to the top of the holder in such a way that the container was air-tight. The glass face was attached to the photomultiplier with mineral oil when the crystal was used. It was found that it was not necessary to polish the faces of the crystal if it had good cleaveage surfaces. In fact better resolution was usually obtained with unpolished crystals. No levigated alumina was used as a light reflector, since a layer of this material on the surface of the crystal would introduce a large, unknown absorption factor for the low-energy x rays. The pulse-height spectra obtained with the W^{181} and Ta^{179} sources are shown in Fig. 1. Three peaks are clearly visible in each spectrum, one at about 60 kev corresponding to the K

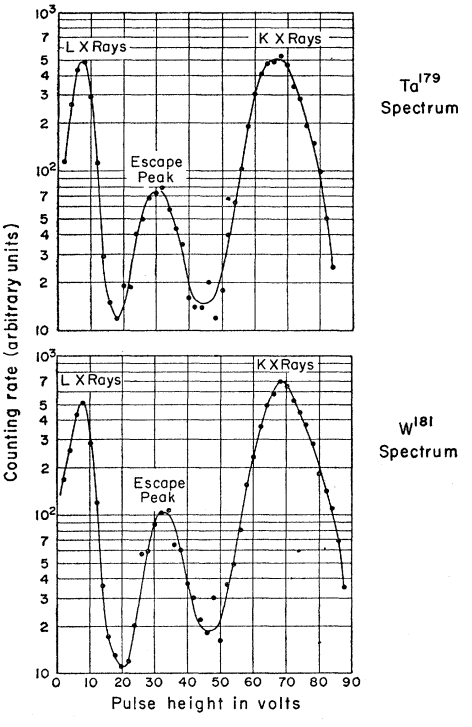

FIG. 1. Pulse-height spectra of Ta^{179} and W^{181} sources. Note that the K x-ray peak of the W^{181} spectrum is larger relative to the L x-ray peak than the K x-ray peak in the Ta^{179} spectrum. This difference is reflected in the values of N_L/N_K listed in Table I.

[2] A. Bisi, S. Terrani, and L. Zappa, Nuovo cimento **1**, 651 (1955).
[3] J. M. Cork, W. H. Nester, J. M. LeBlanc, and M. K. Brice, Phys. Rev. **92**, 119 (1953).
[4] von P. Debrunner, E. Heer, W. Kündig, R. Rüetschi, and T. Lindquist, Helv, Phys. Acta **29**, 432, 463 (1956).
[5] A. Bisi, L. Zappa, and E. Zimmer, Nuovo cimento **4**, 307 (1956).

x ray, one at about 30 kev corresponding to the escape peak of the high-energy x rays, and finally a peak at about 9 kev corresponding to the L x rays emitted by the source. The resolution obtained for the 59-kev Am^{241} gamma-ray peak with this system was 16%. The rather prominent escape peak is observed because the crystals were thin. Thin crystals give lower backgrounds and also give a slightly larger light output for the very low-energy x rays.

The preparation of the sources used for these experiments required considerable care. The W^{181} source was made by bombarding Ta^{181} with 7-Mev protons in the LRL Livermore 90-inch cyclotron. The target was made of four 0.00025-in. Ta foils. These foils were then dissolved and the W^{181} was chemically separated using carrier-free methods so that the number of heavy atoms in the source was reduced to a minimum. The tungsten was prepared in a water solution as an ammonium salt. Some drops of this solution were then placed on a 0.001-in. Mylar backing and evaporated. Another 0.001-in. Mylar piece was then glued to the top of the source to make the seal. The Ta^{179} source was made by bombarding hafnium metal with 24-Mev deuterons at the 60-inch Crocker Laboratory cyclotron. The Ta was again separated chemically by carrier-free methods and the sources were prepared as described above. Since many short-lived activities due to Ta isotopes result from this bombardment, old ($\approx$2 years) sources were used for this work. (The half-life of Ta^{179} is about 660 days.)

Standard electronic equipment was used throughout the experiment. RCA 6810A photomultiplier tubes were employed and the tube base circuits were modified so that the resistances between the cathode, the focusing grid, and the first two dynodes could be varied to adjust the tubes for maximum resolution. For the coincidence measurements on the L and K x rays, circuits with a resolving time of about 1 μsec were used.

EXPERIMENTAL RESULTS

In order to interpret the results obtained in the measurements to be described, certain relations between the observed counting rates and the actual L and K vacancies created by electron capture must be derived. These relations involve the fluorescence yields of the L and K shells. The object of some of the experiments described in this paper is to make new measurements of the fluorescence yields of the L and K subshells of these materials.

Let D be the number of disintegrations per second of the source, C_K the fraction of K-shell vacancies produced by the source, so that the L to K ratio is C_L/C_K. If Ω_K, E_K, and A_K are the geometrical factor, efficiency, and transmission for the K x-ray counter and Ω_L, E_L, and A_L are similar quantities for the L x-ray counter, then the number of K counts per second is

$$N_K=(C_KD)(\Omega_KA_KE_K)\omega_K, \quad (1)$$

where ω_K is the fluorescence yield of the K shell. The expression for the number of L x rays observed is complicated by the fact that the L x rays arise from two sources; x-ray emission after L capture which is proportional to (C_LD), and L x-ray emission following K x-ray emission which is proportional to (C_KD). The number of L counts observed is therefore,

$$N_L=(C_LD)(\Omega_LA_{LL}E_{LL})\omega_{LL}+(C_KD)(\Omega_LA_{KL}E_{KL})\omega_{KL}n_{KL}. \quad (2)$$

In this formula, A_{LL}, E_{LL}, and ω_{LL} are the transmission, efficiency, and fluorescence yields for L x rays following L capture, and A_{KL}, E_{KL}, and ω_{KL} are similar quantities for L x rays following K capture. The reason for distinguishing between the two species of L x rays is that the L-capture process creates vacancies mainly in the $L_{\rm I}(2s_{\frac{1}{2}})$ shell and hence leads to transitions into this level, whereas K capture creates vacancies mostly in the $L_{\rm II}(1p_{\frac{1}{2}})$ and $L_{\rm III}(1p_{\frac{3}{2}})$ shells. Since the energies of the resulting transitions are slightly different (8 to 10 kev), the transmissions, efficiencies, and fluorescence yields will also differ. These differences must be taken into account if accurate results are desired. The quantity n_{KL} in Eq. (2) is equal to the number of L vacancies created per K vacancy. This quantity is defined in detail in the review paper by Robinson and Fink.[6] For the present computations, values given in that review article were used.

The L to K capture ratio, C_L/C_K, can now be expressed in terms of the quantities defined in Eqs. (1) and (2). If the ratio of the number of L counts to K counts (N_L/N_K) is measured, then the L to K capture ratio is given by

$$\frac{C_L}{C_K}=\left(\frac{N_L}{N_K}\right)\left(\frac{\Omega_K}{\Omega_L}\right)\left(\frac{A_K}{A_{LL}}\right)\left(\frac{E_K}{E_{LL}}\right)\left(\frac{\omega_K}{\omega_{LL}}\right)-n_{KL}\left(\frac{A_{KL}}{A_{LL}}\right)\left(\frac{E_{KL}}{E_{LL}}\right)\left(\frac{\omega_{KL}}{\omega_{LL}}\right). \quad (3)$$

In order to compute C_L/C_K from N_L/N_K, all the quantities in the parentheses on the right-hand side of the equation must be evaluated. Since the same counter is used to detect both the K and L x rays, $\Omega_K=\Omega_L$ if the distance between the source and the counters is large enough so that edge effects can be neglected. For the beryllium-window crystals used here, the ratios of the A's and the E's are quite close to unity. The most important correction which must be applied is the absorption in air of the L x rays, which appears in (A_K/A_{LL}) and this is only of the order of 5% for the geometries used in the experiment. However the L-shell fluorescence yields are not well known and it is therefore necessary to perform another experiment to determine them.

[6] B. L. Robinson and R. W. Fink, Revs. Modern Phys. **32**, 117 (1960).

TABLE I. Summary of results.

Isotope	N_L/N_K	n_{KL}	a	ω_{KL}	ω_{LL}	C_L/C_K	Decay energy (kev)
Ta^{179}	0.375±0.005	0.83	0.79	0.24±0.01	0.27±0.02	0.63±0.06	115±5
W^{181}	0.312±0.005	0.83	0.79	0.28±0.01	0.32±0.02	0.23±0.05	260_{-60}^{+160}

The partial fluorescence yield, ω_{KL}', can be obtained by measuring the coincidence rate between the L and the K x rays. This counting rate is given by

$$N_C = N_K'(a\omega_{KL}'A_{KL}E_{KL}\Omega_L), \tag{4}$$

where N_K' is the observed K x-ray counting rate in the K x-ray counter, ω_{KL}' is the fluorescence yield of L vacancies produced in filling K vacancies only when K fluorescence occurs, and the expression in the parenthesis is the probability that the L counter will detect an x ray. Solution of Eq. (4) for ω_{KL}' gives

$$\omega_{KL}' = \left(\frac{N_C}{N_K'}\right)\frac{1}{aA_{KL}E_{KL}\Omega_L}. \tag{5}$$

The factor a must be included in Eq. (5) because the K x-ray counter cannot resolve the K_α from the K_β x rays. The number a is therefore the fraction of the observed K x-ray counts due to K_α x rays. To compute ω_{KL}', the values of a given in reference 6 were used. Since ω_{KL}' and ω_{KL} can be shown to differ by only a few percent, ω_{KL}' has been used for ω_{KL} in Eq. (3).

The arguments made in the last paragraph are true only if there is no angular correlation between the L and the K x rays emitted following K capture. The assumption of isotropy is implicit in Eq. (4). Several experiments were performed with both sources to measure the coincidence rate at 180° (back-to-back) and 90°. The coincidence rate in these two positions was found to be the same to within 5%. This result is not surprising since a great many atomic levels are involved in the transitions, and the counters are not able to resolve the individual K and L x-ray fine structure in the spectrum. Estimates of the expected distribution show that it should be nearly isotropic.

The decay of Ta^{179} proceeds directly to the ground state of Hf^{179}. No radiations other than the Hf K and L x rays were observed from this source. The transition is most probably an allowed transition since the ground state of Hf^{179} has the spin $9/2^+$ and the ground state of Ta^{179} probably has a spin $\frac{7}{2}^+$. The decay energy can be computed from the L to K capture ratio using the formula given in reference 1. The decay energy obtained in this manner is consistent with the fact that the first-excited level in Hf^{179} is at 122 kev, an energy exceeding the computed decay energy. The results are summarized in Table I.

In the case of W^{181}, the decay is more complex since it proceeds to the ground state about 99.7% of the time. In the remainder of the transitions two energy levels in Ta^{181} at 137 and 152 kev are populated. A weak composite peak corresponding to these transitions is observed in the scintillation pulse-height spectrum. The 152-kev gamma rays is about twice as intense as the 137-kev line. Both gamma rays are in coincidence with the L x rays emitted by the source but are not in coincidence with the K x rays. The two gamma rays are also not in coincidence with each other. The decay scheme for this isotope, given in reference 4, is therefore confirmed and the intensities of the gamma-ray lines relative to the x rays quoted by these authors are found to be correct. The transition to the ground state of Ta^{181} is a first-forbidden transition since the spin of Ta^{181} is $\frac{7}{2}^+$ and the probable spin of W^{181} is $\frac{7}{2}^-$. The decay to the 137-kev level also proceeds by a first-forbidden transition since the spin of the 137-kev level is $9/2^+$. The decay energy for this isotope as obtained form the L to K ratio is computed using the formula for allowed transitions. This procedure is justified by the fact that the energy-dependent terms in the first-forbidden transition probabilities for the conditions of this experiment are unimportant.[7,8] It follows therefore that the energy dependence of the L to K capture ratio for these transitions also is due only to the phase space factor which means that the formula of reference 1 is applicable. The decay energy computed from the L to K ratio using this method is sufficiently large (260 kev) to explain the existence of the observed gamma rays. The electron wave functions used to compute the decay energies are tabulated in reference 8. The large errors quoted in the decay energy of W^{181} are characteristic of small (≈ 0.2) values of C_L/C_K, since the decay energy in this region is a very sensitive function of C_L/C_K. It should also be pointed out that the decay energy of W^{181} is probably not quite as large as the value given in Table I, since K capture to the excited levels would then be possible. Since no K capture is observed (i.e., no K x rays are observed in coincidence with the gamma rays), the decay energy must be less than 210 kev, which is within the limits of error of the quoted energy. The above argument is true only if K capture to the excited levels is not forbidden for other reasons.

The circumstance that W^{181} can also decay to excited levels in Ta^{181} by pure L-shell capture, makes it possible to measure the partial fluorescence yield ω_{LL} by a method similar to that used to determine ω_{KL}. The coincidence rate between the gamma rays and the L x rays

[7] H. M. Mahmoud and E. J. Konopinski, Phys. Rev. 88, 1266 (1952).
[8] H. Brysk and M. E. Rose, Revs. Modern Phys. 30, 1169 (1958).

is given by

$$N_C = N_\gamma b(\omega_{LL} A_{LL} E_{LL} \Omega_L). \quad (6)$$

This formula is identical in form to Eq. (4). The quantity b is the fraction of L capture to $L+M+N$ capture and has the value 0.735 according to best estimates of electron densities.[9] N_γ is the gamma-ray counting rate in the gamma counter. Since the transition to the 137-kev level is also a first-forbidden transition the value of ω_{LL} computed from Eq. (6) can be used in calculating C_L/C_K for the ground-state transition in Eq. (3).

It should also be pointed out that the method outlined for the measurements of ω_{KL} and ω_{LL} has the additional advantage that some of the efficiency and transmission factors in Eq. (3) are cancelled. It is therefore not necessary to compute them.

The experimental results are summarized in Table I. In order to show how the final values of C_L/C_K were obtained, all the important numbers appearing in Eqs. (3) and (5) are included. The measured ratios of N_L/N_K are shown in the first column. The next columns show the quantities n_{KL} and a. The measured fluorescence yields, ω_{KL} and ω_{LL}, are shown in the next two columns. Finally the values of C_L/C_K and the decay energies are listed in the last two columns. Since no measurement of the partial fluorescence yield, ω_{LL}, could be made in the case of Ta^{179}, it was assumed that the values of ω_{LL} for the two isotopes studied are in the same ratio as the values of ω_{KL}. This assumption is not unreasonable since both ω_{LL} and ω_{KL} probably have the same Z dependence.

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The authors are indebted to Dr. Manfred Lindner and K. V. Marsh for the chemical separation of W^{181} from the Ta targets. Thanks are also due to LeRoy Erickson and the 90-inch cyclotron crew for their help during the bombardment.

[9] A. H. Wapstra, G. J. Nijgh, and R. van Lieshout, *Nuclear Spectroscopy Tables* (North-Holland Publishing Company, Amsterdam, 1959).