Decay of ^{193m}Ir via an electron bridge

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Nuclear decay via an electron bridge was observed for the first time. The process was obtained by decay of the 80.27 keV ($T_{1/2} = 10$ d) isomer level in ^{193m}Ir. The internal conversion coefficient on the K shell was measured. It is shown that the probability of decay of the nucleus via an electron bridge reaches 21% of that via the γ channel.

INTRODUCTION

A theory of higher-approximation effects was developed in Refs. 1 and 2 for γ radiation and internal conversion in atomic nuclei. According to these papers, third-order effects in the fine-structure constant (second-order effects constitute internal conversion) should be substantial if the internal conversion coefficients (ICC) are very large. In particular, there exists a certain probability that the next step of a conversion electron after absorbing a γ photon is to return to its initial place, emitting at the same time a real γ photon of energy equal to the initial one. According to Ref. 3, the ratio of the probability $\lambda_\gamma^{(3)}$ of this effect to the probability $\lambda_\gamma^{(1)}$ of the "bare" nucleus is

$$\lambda_\gamma^{(3)}/\lambda_\gamma^{(1)} \approx \Big(\sum_{\varkappa\varkappa'} \alpha_{\varkappa\varkappa'}\delta_{\varkappa\varkappa'}\Big)^2,$$

where $\alpha_{\varkappa\varkappa'}$ are the partial ICC for shells with numbers $\varkappa\varkappa'$; $\delta_{\varkappa\varkappa'} = \mathrm{Re}R_{\varkappa\varkappa'}/\mathrm{Im}R_{\varkappa\varkappa'}$; $R_{\varkappa\varkappa'}$ is the radial integral of the conversion matrix element. The Im $R_{\varkappa\varkappa'}$ and Re $R_{\varkappa\varkappa'}$ values calculated in the ICC tables are always intermediate. Estimates made using the programs of Ref. 4 yield a probability $\lambda_\gamma^{(3)} \sim 10^{-5}\lambda_e^{(2)}$, where $\lambda_e^{(2)}$ is the probability of nuclear decay with emission of a conversion electron. It follows also from the expression above that the probability $\lambda_\gamma^{(3)}$ is always higher for transitions from high electron shells. The authors of Ref. 3 called this effect an electron bridge (EB). This process has not yet been observed in experiment. We report here an attempt to assess the role of such effects in decay of radioactive nuclei.

We chose for the study the ^{193}Ir nucleus. It has an isomer (see Fig. 1) whose probability of decay via a γ channel is $4.6\cdot 10^{-3}$%. The theoretical ICC for this transition with $E_\gamma = 80.27$ keV are $\alpha_K = 112$ and $\alpha = 22\,240$, on the K shell and the total one. By definition $\alpha_K = \lambda_K^{(2)}/\lambda_\gamma^{(1)}$, and if a γ transition through an EB exists, experiment should yield

$$\alpha_K^{\exp} = \lambda_K^{(2)}/\lambda_\gamma,$$

where $\lambda_\gamma = \lambda_\gamma^{(1)} + \lambda_\gamma^{(3)}$; the value of $\alpha_K^{\exp}$ should decrease compared with theoretical one.

EXPERIMENT

In our experiment α_K was determined by measuring the intensity ratio of the KX x rays of iridium to the intensity of the γ transition. It is known that $I_{KX} = \lambda_K^{(2)}\omega_K$, where ω_K is the fluoresence yield. It follows hence that α_K can be determined by measuring I_{KX}/λ_γ. Measurement of the x-ray spectrum requires that the investigated source contain no other radioactive atoms that decay into iridium isotopes. In addition, since the measurements are performed in the low-energy region, it is more convenient to operate with thin sources, to avoid problems with self-absorption. A higher degree of radiochemical purity is also necessary, since the probability of decay via the γ channel is low (10^{-5}). The ^{193m}Ir isomer was therefore built up by irradiating ^{192}Os (99% enriched) by thermal neutrons, via the chain

$$^{192}\mathrm{Os} \xrightarrow{(n,\gamma)} {}^{193}\mathrm{Os} \xrightarrow[T_{1/2}=30\ \text{ч}]{\beta^- 0{,}3\%} {}^{193m}\mathrm{Ir}.$$

After a week the irradiated sample revealed γ activities of ^{193m}Ir and ^{191}Os (the latter was produced via the isotope ^{190}Os and its concentration was 0.86%) in a ratio 1:10^6. The radioactive Ir atoms were separated by chromatography. The active ^{193m}Ir was deposited on the walls of a quartz tube of 5 mm diameter. Radiochemical treatment equalized the γ activities of the ^{193m}Ir and ^{191}Os.

The γ spectra of the obtained source was measured with a γ spectrometer having a pure-germanium detector of 5 cm^3 volume and resolution 0.45 keV on a ^{241}Am γ line of energy 59.6 keV. This energy resolution enabled us to separate the $K\alpha$ and $K\beta$ rays both from each other and from impurities of neighboring nuclei. By way of example, the figure shows one of the spectra typical of the described measurement. To determine the relative intensities one must know the dependence of the spectrometer γ-photon recording probability on the energy—the efficiency curve. We used for this purpose a ^{192}Ta source produced for the measurement of conversion electrons. It was placed in a similar quartz tube. The obtained efficiency curve enabled us to determine the corrections for the registration efficiency with accuracy 1% for the $K\alpha$ rays and 0.5% for the $K\beta$ rays and the γ photons. Some deterioration of the accuracy for the $K\alpha$ transitions is due to the inflection point on the efficiency curve at $E_\gamma = 0.65$ keV.

Altogether we performed eight measurement runs in three geometries. The source was placed at a distance 2, 4, or 6 cm from the detector. The measurement results are listed in the table. The value of α_K was determined from the equation

$$\alpha_K = I_{KX} f_\gamma / \omega_K I_\gamma f_{KX},$$

where f_γ is the relative efficiency of the spectrometer and f_{KX} is the relative intensity of the KX rays. Table I lists only the errors due to the processing method and to the statistics. Values of the accuracies of ω_K and f_{KX} will be given below, with an aim at refining them in the future. Detailed information on the accuracy of contemporary data for ω_K and f_{KX} can be found in Refs. 5–7.

Analysis of experimental data and of contemporary

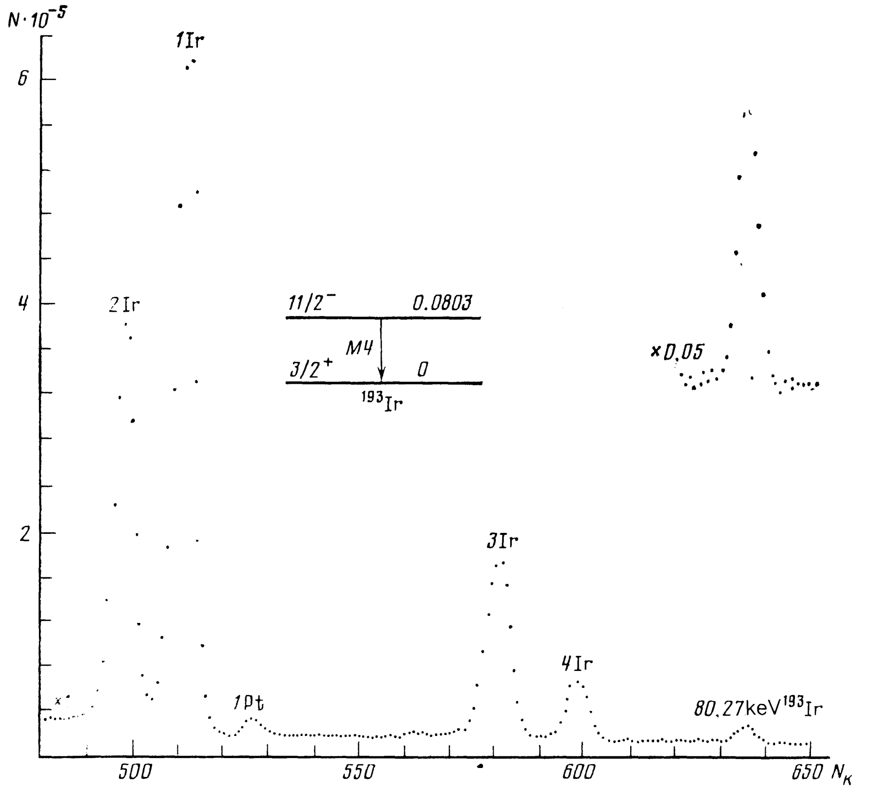

FIG. 1. Fragment of spectrum and of decay scheme of ^{193m}Ir($T_{\frac{1}{2}} = 10d$): 1,2,3,4-$K_{\alpha I}$, $K_{\alpha II}$, $K_{\beta I}$ and $K_{\beta II}$ lines of the x-ray spectra.

theoretical calculations leads to the conclusion that in heavy nuclei the accuracy is $\leqslant 1\%$ for the $K\alpha$ group, $\leqslant 2\%$ for $K\beta'$, and $\leqslant 5\%$ for $K\beta'$. With these taken into account, we obtained $\alpha^{\rm exp} = 92.6 \pm 0.9$ as against the tabulated $\alpha_K = 112.1$ (Ref. 9). So large a difference can be attributed to some processes that decrease the probability $\lambda_K{}^{(2)}$ or to an increase of the probability λ_γ. The conversion K transition takes place near a threshold region with energy $E_K = 4.17$ keV. It is known that an important role can be played in this region by effects such as the choice of the exchange potential or the role of the vacancy produced in the internal-conversion process.[10] These questions were examined in detail and it was concluded that the vacancies must be taken into account,[11] whereas there was no meeting of mind concerning the exchange potential. We have therefore compared the experimental α_K with the tabulated theoretical values,[9] with account taken of the role of the vacancy. These tables, furthermore, were calculated using an exchange potential that leads to ICC values lower than in other tables. In general, α_K varies from 112 to 117 if the exchange potential is varied. Understandably, this only increases the difference between the theoretical and experimental values. The last and most important process that can decrease $\lambda_c{}^{(2)}$ is the so-called internuclear conversion.[12] However, the penetration parameters that characterize this conversion can be determined from ICC on other shells. Therefore, by measuring the ratio $L_{\rm I}:L_{\rm II}:L_{\rm III}$ (L_i is the probability of internal conversion on the i-subshell) we can estimate the role of this process. The ICC ratios on almost all shells were measured in Ref. 13 accurate to 0.1–0.4%, starting with L_1, and it follows unequivocally from these measurements that if intranuclear conversion does indeed take place, it can only increase α_K by 1–2%. It remains for us only to assume that the observed decrease of α_K is connected with the probability λ_γ, and the latter must be increased in order to decrease α_K, i.e., this is a process of the electron-bridge type.[14]

TABLE I. Internal conversion coefficients on the K shell for 80.27 keV transitions in ^{193m}Ir.

Distance to source, cm	$\alpha_K^{(1)}$	$\alpha_K^{(2)}$	$\alpha_K^{\rm Theory}$
2	93,1±1,0	91,9±0,7	112,1
4	93,0±1,0	92,3±1,0	112,1
6	92,0±1,0	92,3±0,7	112,1

Note. $\alpha_K^{(1)}$ and $\alpha_K^{(2)}$ were obtained from the $K\alpha$ and $K\beta$ x-ray spectra.

It follows from our data that $\lambda_\gamma{}^{(3)} = \left(9.5 \begin{array}{c} +0.4 \\ -1.6 \end{array}\right) \cdot 10^{-6} \lambda_e{}^{(2)}$. The errors of $\lambda_\gamma^{(3)}$ must include also the systematic decrease (by 1.5–2%) from theory observed in Refs. 15 and 16, of the experimental values of λ_K and for M4 transitions.

We see that $\lambda_\gamma^{(3)}$ is close to the estimates obtained in the Introduction. It follows from the value of $\lambda_\gamma^{(3)}$ that in the decay of ^{193m}Ir decay 21% of the γ-channel intensity is via the EB. The value obtained indicates that third-order effects can be appreciable in certain situations. It is therefore, of course, necessary to attempt a direct measurement of such processes. One aspect to study is the angular distribution of $\gamma^{(\theta)}$ from the decay of ^{193m}Ir and of oriented nuclei. Allowance must be made here for the possiblity that the multipolarity of the γ photons due to the EB is not necessarily $M4$. This, naturally can be easily observed in experiments with oriented nuclei. Our calculations of this process for the L electron yielded $\lambda_\gamma^{(3)}(L_3) \approx 0.18\lambda_\gamma^{(1)}$. This theoretical value is practically the same as the experimental. It is obvious, however, that $\lambda_\gamma^{(3)}$ will be substantially larger if account is taken of the contributions of the higher shells, which can be qualitatively estimated by using the calculations for this effect in the decay of ^{235m}U (Ref. 17). Estimates show that $\lambda_\gamma^{(3)}(M,N,...) \sim (0.1-0.2)\lambda_\gamma^{(1)}$. We see that the theoretical calculations lead to overestimates. This may be due to the choice of Coulomb wave functions. We point out in conclusion the possibility of verifying the accuracy of the radiative corrections in quantum electrodynamics[14,18] with the aid of such processes.

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