Correlation effects in the internal conversion of γ rays

M. D. Bondar'kov, V. A. Zheltonozhskiĭ, A. G. Zelinskiĭ, L. V. Sadovnikov
and M. A. Ukhin

Institute of Nuclear Studies, Ukrainian National Academy of Sciences, 252650 Kiev, Ukraine

(Submitted 12 December 1995)
Zh. Éksp. Teor. Fiz. **110**, 443–449 (August 1996)

This paper reports accurate measurements of the internal conversion coefficients of γ rays at the K shell in ^{187}Re and ^{234}Pa. A value of $\alpha_K=0.765(8)$ is obtained for the ^{187}Re transition with energy $E_\gamma=72$ keV, and a value of $\alpha_K=0.212(16)$ is obtained for the ^{234}Pa transition with energy $E_\gamma=112.8$ keV. These values differ from the theoretical values by (5–10)%. Based on the analysis carried out here, the substantial difference of the experimental values of α_K from the theoretical values is treated as a manifestation of correlation effects in the internal conversion of γ rays. © *1996 American Institute of Physics.* [S1063-7761(96)00708-1]

1. INTRODUCTION

As is well known,[1] the interaction between the atomic electrons belonging to the ion residue and the emerging electron has a substantial effect on the photoionization cross section. The direct interaction disturbs the independent motion of the electrons in the atom and causes them to be correlated. The manifestations of direct interaction are called correlation effects. They are especially large in the near-threshold region.

Similar phenomena occur in the process of internal conversion of γ rays when the nuclear transition energies are close to the binding energy of the given shell.

However, unlike the photoelectric effect, correlation effects in the internal conversion of γ rays have not been experimentally studied.

The role of such effects has been considered theoretically in Ref. 2, which showed that, for $E_e=E_\gamma-E_i<2$ keV, where E_γ is the energy of the γ quantum and E_i is the binding energy of the ith shell, the internal-conversion probability can vary by a factor of 2.

We have investigated correlation effects in the internal conversion of γ rays in the nuclei ^{187}Re and ^{234}Pa. These nuclei exhibit transitions with energies of $E_\gamma=72$ keV in ^{187}Re, $\Delta E=E_\gamma-E_K=0.3$ keV and $E_\gamma=112.8$ keV in ^{234}Pa, $\Delta E_e=0.2$ keV, where E_K is the binding energy at the K shell. In both cases, these are $E1$ transitions, and therefore the effect is expected to be maximal.

2. EXPERIMENTAL TECHNIQUE AND RESULTS OF THE MEASUREMENTS

When the kinetic energies of the conversion electrons are very small, the absolute values of the internal conversion coefficients can be measured only by comparing the intensity of the characteristic radiation (I_{K_x}) that accompanies the conversion with the intensity I_γ of the γ radiation. For internal conversion at the K shell, the internal conversion coefficient α_K will be determined from

$$\alpha_K=I_{K_x}/I_\gamma\omega_K, \tag{1}$$

where ω_K is the fluorescence yield.

To determine α_K for the indicated γ transitions, the decay of ^{187}W to the ^{187}Re levels and of ^{234}Th to the ^{234}Pa levels was studied (Fig. 1).

The measurements were made on $\gamma\gamma$ coincidence spectrometers with Ge and NaI(Tl) detectors and on a Si(Li) semiconductor detector. The coincidence spectrometer had a resolving time of $\tau=24$ ns. The Ge detector had a beryllium input window, the detector volume was 12 cm^3, and the resolution was 450 eV at the γ line with an energy of $E=60$ keV (in what follows, we shall write $\gamma60$) of ^{241}Am; the Si(Li) detector had a resolution of 450 eV at $\gamma72$. Measurements were made of the separate γ and K_x spectra and of the $\gamma\gamma$ and $K_x\gamma$ coincidence spectra.

In ^{187}Re, the relative intensity I_{K_x}/I_γ of the transition with energy 72 keV was determined from the coincidence spectra (see Fig. 1a). Here we used the fact that $\gamma72$ decays from a state with $\tau=560$ ns, while the resolving time of the apparatus was $\tau=24$ ns. In this case, $\gamma134$ coincides only with $\gamma72$, $\gamma551$, and the characteristic radiation that corresponds to them; $\gamma551$ has $E1$ multipolarity. Its contribution to the characteristic radiation is less than 0.5%.

Using Eq. (1), we see from the $K_x\gamma$ coincidence-data that $\alpha_K(72)=0.765\pm0.008$, where α_K corresponds to the energy of the $E=72$ keV transition; in what follows we shall denote this as $\alpha_K(72)$.

To determine the $M2$-multipole contribution, measurements were made of the total internal conversion coefficient α. To do this, the relative intensities of the γ transitions that populate and depopulate the state with $E=206$ keV (see Fig. 1a) were carefully measured. It was found that $I_{\gamma479}=100$, $I_{\gamma72}=50.8\pm0.5$, and $I_{\gamma206}=0.65\pm0.02$. Using these data, tabulated values of the internal conversion coefficients of $\gamma206$ and $\gamma479$, and the expression

$$I_{\gamma72}[1+\alpha(72)]+I_{\gamma206}[1+\alpha(206)]=I_{\gamma479}[1+\alpha(479)] \tag{2}$$

we determined the total internal conversion coefficient $\alpha(72)=0.960\pm0.015$. Note that this value is independent of the choice of tables of internal conversion coefficients, since the contribution to the value of $\alpha(72)$ associated with these coefficients corresponding to the 206- and 479-keV transitions is less than 5%. Also, as is well known, the accuracy of

1063-7761/96/080236-04$10.00

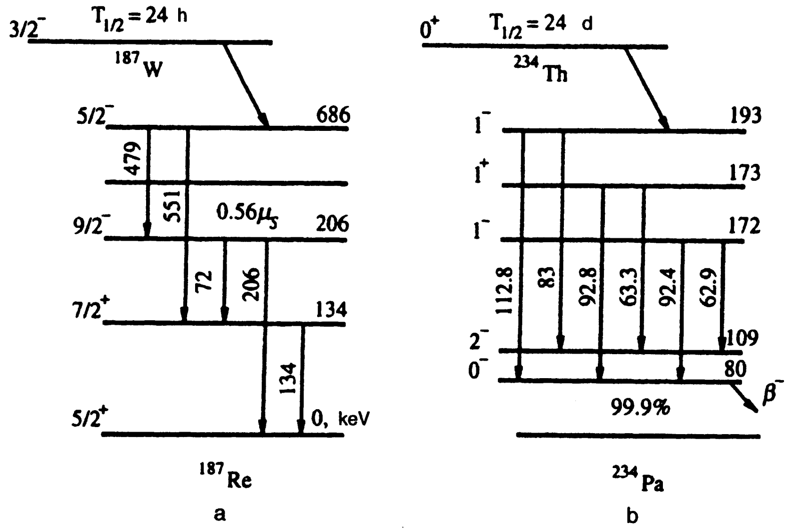

FIG. 1. Decay diagram of ^{187}W (a) and ^{234}Th (b).

modern tables of internal conversion coefficients for γ transitions remote from threshold values is better than 1%.[3]

The $\alpha_K(112.8)$ measurements of ^{234}Pa were made with a natural uranium source, an equilibrium product of which is ^{234}Pa (see Fig. 1b). The decay of ^{238}U occurs along the squence $^{238}\mathrm{U}\xrightarrow{\alpha}{}^{234}\mathrm{Th}\xrightarrow{\beta}{}^{234}\mathrm{Pa}\rightarrow$. The characteristic K_x radiation of ^{234}Pa is mainly caused by internal conversion of γ112.8, since $E_K = 112.6$ keV.

In the K_x spectra of Pa, there is also a component due to the decay $^{235}\mathrm{U}\xrightarrow{\alpha}{}^{231}\mathrm{Th}\xrightarrow{\beta}{}^{231}\mathrm{Pa}\rightarrow$.

We therefore carried out two series of measurements. The decay of the activity of the target made from natural uranium and 100 mg/cm^2 thick was measured on a Ge spectrometer for a time for which the statistical measurement error of the $K_{\alpha \mathrm{I}}$ line of Pa became $\leqslant 1\%$. The characteristic K_x and γ spectra are shown in Fig. 2. The ^{234}Pa activity in the spectrum is identified from γ92, and that of ^{231}Pa from the $K_{\alpha \mathrm{II}}$ line of Th.

Measurements were also carried out on a target made from enriched ^{235}U ($>70\%$). The relative value of $I_{K_{\alpha \mathrm{I}}}(\mathrm{Pa})/I_{K_{\alpha \mathrm{II}}}(\mathrm{Th})$ was determined with high accuracy from these measurements. The resulting value was allowed for in $I_{K_{\alpha \mathrm{I}}}(\mathrm{Pa})$ from the natural uranium, taking into account that $I_{K_\alpha}(\mathrm{Th})$ in the target made from natural uranium was caused only by the α decay of ^{235}U.[4]

As a result of these measurements, we found $\alpha_K = 0.212(16)$. It was assumed in determining this value that the K_α line of ^{234}Pa is associated with internal conversion only for γ112.8. This conclusion is based on an analysis of the γ spectra from our measurements and Refs. 4 and 5. The entire complex of these data shows that all the γ quanta except for γ112.8 have an energy less than the binding energy of the K electrons.

References 4 and 5 indicated the existence of γ184, with an intensity of about 0.012%. In our measurements, this value is $<0.007\%$. An analysis of the level diagram of Pa

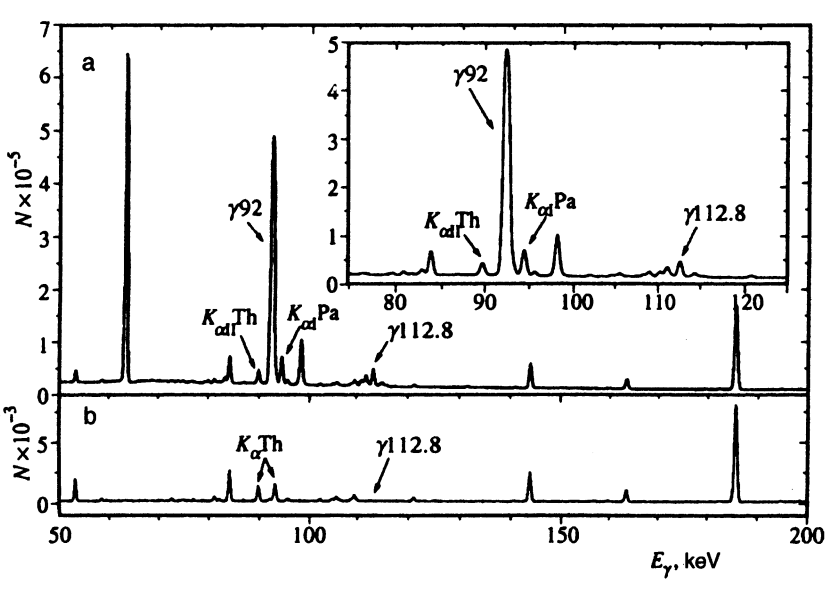

FIG. 2. K_x and γ spectra of natural uranium (a) and ^{235}U (b).

shows that this is most likely an $E3$ transition. The contribution of this transition to I_{K_x} is $<5\%$. We allowed for it when determining $\alpha_K(112.8)$.

To analyze the data on the internal conversion coefficient, the energies of the γ quanta must be known with high accuracy. At the beginning of our studies, it was known that $E_\gamma(\mathrm{Pa})=112.8(3)$, while the data on $E_\gamma(\mathrm{Re})$ were contradictory [$E_\gamma=71.963(4)$ (Ref. 6) and $E_\gamma=72.004(2)$ (Ref. 7)]. We therefore made special measurements of the energies of the γ quanta studied here.

The measurements were made on a semiconductor spectrometer with a Si(Li) detector that had an energy resolution of 450 eV at $\gamma72$, and the energy dependence of the spectrometer's efficiency curve in the 60–100-keV region was linear. For measurements on semiconductor spectrometers the choice of standards, the differential nonlinearity of the spectrometer, and the processing method have great significance. Modern processing methods make it possible to determine the position of a line with a relative accuracy of $(0.05\pm0.01)\%$. It follows from this that, to determine the $\gamma72$ energy with an accuracy of (1–2) eV requires that one of the γ standards be spaced no further than (2–3) keV from it. For such accuracies, the positions of the standards and of the γ line of interest cannot be measured separately, since, even though the differential and integral nonlinearities in our spectrometer were $<0.1\%$, methodological errors associated with some variation of the external conditions cannot be eliminated. Therefore, a mixed source was prepared, with approximately equal activities of ^{169}Yb [$T_{1/2}=32$ d, $E_\gamma=(63.12080\pm0.00019)$ keV], ^{170}Tm [$T_{1/2}=130$ d, $E_\gamma=(84.3049\pm0.0014)$ keV], ^{187}W [$T_{1/2}=24$ h, $E_\gamma=72$ keV], and ^{199}Au [$T_{1/2}=3.2$ d, $E_{K_{\alpha\mathrm{II}}}=68.893$ keV, $E_{K_{\alpha\mathrm{I}}}=70.818$ keV].

The target was prepared by sputter-coating the corresponding isotopes or elements onto an aluminum substrate; the target thickness was 50 μg/cm^2. This made it possible to eliminate distortions of the line shape due to self-absorption.

The target was irradiated at the reactor of the Institute of Nuclear Studies, Ukrainian National Academy of Sciences, for 3 d and, after a day's delay, ten series of measurements were made. In the first series, the duration of the measurements was from 5 to 10 h; in last series, it was 1–2 d. The measurements were designed so that the statistical accuracy for all the γ lines was no less than 0.01% in all the series.

The processing was done by the method of splines.[5] To do this, we chose one of the standards $\gamma63$ or $\gamma84$ as the tabulated line and inserted it into the (K_αHg + K_βRe + $\gamma72$) spectrum at $\gamma63$ or $\gamma84$. Unfortunately, there is no γ line spaced 1–2 keV from $\gamma72$ and measured with an accuracy of $\leqslant 1$ eV, and so we used the K_α lines of Hg as standards, which resulted in certain features during the processing. This is because the natural width of the K_α line is 50 eV (the calculation was done neglecting the width of the L holes). Since the width of the instrumental line is 450 eV, the shape of the x-ray lines begins to be affected by the natural width of the K hole. Because the K_x lines are described by a Poisson distribution, while the instrumental line is gaussian, "tails" appear in K lines measured on semiconductor spectrometers. Much attention was devoted to the role of such tails, and they were allowed for when a precise measurement was made of the internal conversion coefficients at the K shell.[6] In particular, it was shown[6] that it is necessary when processing K_x spectra to introduce false lines, displaced from the maximum by 2–2.5 times the width of the given line. The same procedure was used in this paper. As a result of the measurements that were carried out, it was found that the energy is $E_\gamma=(72.001\pm0.002)$ eV, with the scatter of the values in all the series within the indicated limits, but errors of 2 eV are associated with the necessity of using the tabulated values of the energies of the K_α line of Hg, while they differ in different tables by 1 eV.[4,5]

TABLE I.

	E_γ, keV	$E_\gamma-E_K$, eV	Γ, eV	α_K^{exp}	$\alpha_K^{\mathrm{theor}}$
Re	72.001	325(2)	38	0.765(8)	0.716
Pa	112.779(7)	175(7)	82	0.212(16)	0.287

Measurements of $\gamma112$ were made by a similar method, with a target made from natural uranium.

The K_β lines of uranium were used as references. Weak components are present in the K_β spectrum.[5] This increases the error, and therefore the measurement accuracy of $\gamma112.8$ is limited to 7 eV. As a result of these measurements, it was found that $E_\gamma(\mathrm{Pa})=112.71(7)$ keV.

3. DISCUSSION OF THE RESULTS

Table I shows the data obtained for α_K^{exp}, $\alpha_K^{\mathrm{theor}}$, and the parameters that characterize the given γ transition. Here Γ is the natural width of the K hole (the calculation was carried out neglecting the L holes), and $\alpha_K^{\mathrm{theor}}$ is the tabulated value of α_K from Refs. 7 and 8. Calculations of $\alpha_K(72)$ were carried out in Ref. 7, and it was found that $\alpha_K=0.702$, neglecting the hole and 0.716 taking it into account. We obtained the α_K value neglecting the hole from the table of Ref. 8.

As can be seen, a systematic error of α_K^{exp} from the theoretical values is observed. In the case with $\gamma72$ in ^{187}Re, this can be associated with an admixture of the $M2$ component mentioned in Ref. 7. An analysis of the $\Delta\alpha=\alpha-\alpha_K=0.195\pm0.017$ value that we obtained and the experimental data concerning the relative internal conversion coefficients at the L subshells[4] shows that the admixture of the $M2$ component is less than 0.02%; i.e., the contribution to α_K is less than 0.014. The difference of the experimental internal conversion coefficient of $\gamma72$ from the tabulated values thus cannot be explained by the $M2$ component. The variation of the α_K value can also be associated with penetration effects. The role of penetration effects was considered in detail in Ref. 9. It follows from this analysis that penetration effects play no substantial role in the internal conversion coefficients corresponding to $\gamma72$, since this is a K-forbidden transition. Such a conclusion also does not contradict the experimental internal conversion coefficients at the L subshells.[4]

The situation is more favorable in the case of ^{234}Pa, since α_K^{exp} is significantly less than $\alpha_K^{\mathrm{theor}}$; i.e., an admixture of the $M2$ component as neglected weak higher-energy γ quanta only increases the discrepancy. It is impossible to directly estimate the role of penetration effects in the given case. However, it follows from an analysis of the diagram of the Pa levels populated when ^{234}Th undergoes β decay that the γ transitions with energies of 92.8 keV and 112.8 keV depopulate states of the same nature to the same low-lying state (see the segment of the diagram in Fig. 1b). The data on the internal conversion coefficients of $\gamma 92$ show that this is an $E1$ transition, the internal conversion of which contains no significant contribution of penetration effects. We conclude that penetration effects cannot change α_K so dramatically in the case of $\gamma 112.8$.

We therefore conclude that the difference of α_K from the tabulated values is caused by correlation effects.

Theoretical estimates of the observed phenomenon can be made, starting from the fact that the interaction of the emerging electron with the electrons of the ion is described similarly in photoionization and internal conversion of γ rays. In Ref. 1, these processes were discussed most completely for photoionization, and it was shown that the probability of the mixing of other electron–hole excitations into the ith excitation under consideration depends in first approximation on the width of the state under consideration and the kinetic energy of the electrons. We have shown that taking correlation effects into account when the same approach to the internal conversion process is used results in the appearance of an additional factor in the internal conversion probability:

$$P=\left(1-\frac{1}{2}\frac{\Gamma}{\Delta E+\Gamma/2}\right)^{-1}. \tag{3}$$

The sign in Eq. (3) is determined from the expression $\delta=(\bar{r})^{-3}-\overline{(\bar{r})^{-3}}$, where $\bar{r}$ is the mean value of the radius that characterizes the distribution of electrons in the atom; i.e., it can be any value.

Using the resulting expression, we determined that $P^{\text{theor}}(\text{Re})=5.8\%$, $P^{\text{exp}}(\text{Re})=6.8(10)\%$, $P^{\text{theor}}(\text{Pa})=23\%$, and $P^{\text{exp}}(\text{Pa})=26(6)\%$.

As can be seen, the agreement between the theoretical and the experimental values is good in Pa. In the case of Re, the experimental value is somewhat overestimated. In our view, this is most probably associated with the necessity of taking into account a small admixture of the $M2$ component. Using a value of 0.02% for the $M2$ component decreases P^{exp} by 2%.

Note also that not only our experimental values but also our theoretical estimates are in good agreement with the calculation carried out in Ref. 2.

Therefore, it can be concluded from everything explained above that the observed deviations of the experimental internal conversion coefficients from the tabulated values can be treated as a manifestation of correlation effects in the internal conversion of γ rays.

[1] M. Ya. Amus'ya, *The Photoelectric Effect* (Nauka, Leningrad, 1987).

[2] M. Ya. Amus'ya, M. A. Listengarten, and S. G. Shapiro, Izv. Akad. Nauk SSSR Ser. Fiz. **32**, 1415 (1968).

[3] E. Browne *et al.*, eds., *Table of Radioactive Isotopes* (Wiley, New York, 1986).

[4] M. Lederer, ed., *Table of Isotopes* (Wiley, New York, 1978).

[5] I. N. Vishnevskiĭ, V. A. Zheltonozhskiĭ, N. V. Stril'chuk *et al.*, Izv. Akad. Nauk SSSR Ser. Fiz. **51**, 863 (1987).

[6] V. A. Zheltonozhskiĭ *et al.*, in *Questions of the Accuracy of Nuclear Spectroscopy* (Izdatel'stvo St. P. Univ., 1994), p. 9.

[7] I. M. Band and T. B. Trzhaskovskaya, Yad. Fiz. **56**, No. 5, 1 (1993) [J. Nucl. Phys. **56**, 573 (1993).

[8] F. Rosel *et al.*, eds., *Atomic Data and Nuclear Data Tables* (Wiley, New York, 1978), p. 21.

[9] M. A. Listengarten, in *Modern Methods of Nuclear Spectroscopy* (Nauka, Leningrad, 1986), p. 142.

Translated by W. J. Manthey