Double K-shell vacancy creation in the decay of ^{109}Cd

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(Received 7 October 1976)

The probability P_{KK}(IC) of double K-shell vacancy creation per K internal conversion of the 88 keV $E3$ transition in the decay of ^{109}Cd has been determined by means of a $K\alpha$-x-ray–K-x-ray coincidence experiment: $P_{KK}(\text{IC}) = (2.8 \pm 0.7) \times 10^{-5}$. The energy shift of the $K\alpha_1$ hypersatellite x-ray line was found to be (532 ± 12) eV.

[RADIOACTIVITY ^{109}Cd; measured $K\alpha$-x-ray–K-x-ray coincidences, deduced double K-shell vacancy probability of 88 keV $E3$ transition and $K\alpha_1$ hypersatellite x-ray shift.]

I. INTRODUCTION

In the decay of ^{109}Cd, K-electron capture as well as K internal conversion of the 88 keV $E3$ transition may give rise to double vacancies in the K shell. Experimental values for the probability of the creation of double K-shell vacancies per K-electron capture, P_{KK}(EC), have not been reported so far. For the probability of the creation of double K-shell vacancies per K internal conversion, P_{KK}(IC), we know of two experimental values. From the results of Knauf and Sommer[1] we find $P_{KK}(\text{IC}) = (7.2^{+0.5}_{-1.5}) \times 10^{-4}$. From a comparison of this value with experimental results for other isotopes (see, e.g., Freedman[2] or Mukoyama and Shimizu[3]) we conclude that it is too high. The more recent result of Nagy, Schupp, and Hurst,[4] $P_{KK}(\text{IC}) = (15.3 \pm 2.4) \times 10^{-5}$, is much lower. Yet, we have an objection against this value as will be explained below. The result has been obtained from a coincidence experiment in which a Si(Li) and a NaI(Tl) detector were employed. The intensity of the Ag $K\alpha^H$- (hypersatellite) x-ray line, which is a measure for the number of double K-shell vacancies, was determined from the Si(Li) coincidence spectrum. The $K\alpha^H$ line was found to be shifted with respect to the $K\alpha$ diagram line by (373 ± 75) eV. Recently, we measured the shift of the $K\alpha^H_1$-x-ray line, emitted in the decay of $^{114}In^m$, by means of a curved-crystal spectrometer.[5] From the result (566 ± 4) eV, we derived a shift of 540 eV for the case of ^{109}Cd, i.e., a value considerably higher than the above mentioned result of 373 eV. This difference might imply a large systematic error in the measured intensity of the $K\alpha^H$ line and consequently in the resulting P_{KK}(IC) value. In view of this we remeasured the intensity of the $K\alpha^H$ line in a coincidence experiment.

II. METHOD OF MEASUREMENT

The experiment was performed by means of a 25 mm^2 Ge x-ray detector of Princeton Gamma-Tech and a 40 mm diam NaI(Tl) x-ray detector. A fast-slow coincidence circuit with a resolving time of 16 nsec was employed. The coincidence spectrum, the accidental coincidence spectrum, and the singles spectrum were recorded simultaneously by means of a routing system. The data were handled by means of a PDP 11/10 computer.

The sources were sandwiched between the two detectors placed at 180°. In order to prevent the detection of Auger electrons and conversion electrons Al foils, 0.25 mm thick, were placed on either side of the source. In order to reduce as much as possible the detection in the Ge detector of iodine K x rays escaped from the NaI(Tl) crystal, a diaphragm of Mo and Cu was placed between the source and the Ge detector. The hole in the diaphragm was slightly larger than the source diameter (≃5 mm).

From the decay scheme[6] we find for the relation between N_c, the number of Ag $K\alpha^H$-x-ray counts in the coincidence spectrum recorded with the Ge detector and N_{NaI}, the number of Ag K x-ray counts recorded in the channel of the NaI(Tl) detector,

$$N_c/(\epsilon N_{\text{NaI}}) = \alpha^H \omega^H_K[(0.34 \pm 0.01)P_{KK}(\text{IC}) + (0.66 \mp 0.01)P_{KK}(\text{EC})]. \quad (1)$$

ϵ is the product of the coincidence efficiency, the detection efficiency, and the solid angle of the Ge detector; α^H is the $K\alpha^H$ fraction of all the K^H x rays, and ω^H_K is the fluorescence yield of the K^H x rays. In the derivation of this formula it was assumed that the fluorescence yield $\omega_K{}^S$ of K^S x rays (satellite K x rays emitted by an atom that has initially one K and one L vacancy) and ω_K of diagram K x rays are identical.

The instrumental constant ϵ has been determined by means of $^{125}Te^m$ sources. The sources were prepared by evaporation of a drop of active solution onto Scotch tape. The source strengths were about 4 and 100 nCi, resulting in ratios of true to accidental coincidences of 15×10^4 and 5×10^3, respectively. With each source ϵ was determined from the intensity of the Te $K\alpha$-x-ray coincidences, N_{cx}, as well as from the intensity of the 35 keV γ-ray coincidences, $N_{c\gamma}$. The applied formulas, derived from the decay scheme,[7] are for the Te $K\alpha$ x rays

$$\epsilon = 2.3 N_{cx}/N_{\mathrm{NaI}} \tag{2}$$

and for the 35 keV γ rays

$$\epsilon = 40 N_{c\gamma}/N_{\mathrm{NaI}} \,. \tag{3}$$

N_{NaI} is the sum of the number of Te K-x-ray counts and the number of 35 keV γ-ray counts recorded in the NaI(Tl) channel. Corrections of, respectively, 6 and 10% were applied in order to take into account the difference in absorption between the Ag $K\alpha^H$ x rays and, respectively, the Te $K\alpha$ x rays and the 35 keV γ rays. The thus obtained four values of ϵ ($\simeq 11\times10^{-3}$) were in agreement with each other within the statistics ($\simeq$5%). The calibrations were repeated after each measurement on ^{109}Cd.

The ^{109}Cd sources were prepared in the same way as the Te sources. Source strengths of 50 and 100 nCi were used. The ratio of true to accidental coincidences was about 5×10^3.

III. RESULTS AND DISCUSSION

Figure 1 shows a coincidence spectrum of the Ag $K\alpha^H$-x-ray region recorded with the Ge detector. The data have been accumulated for three days. Accidental coincidences have been subtracted.

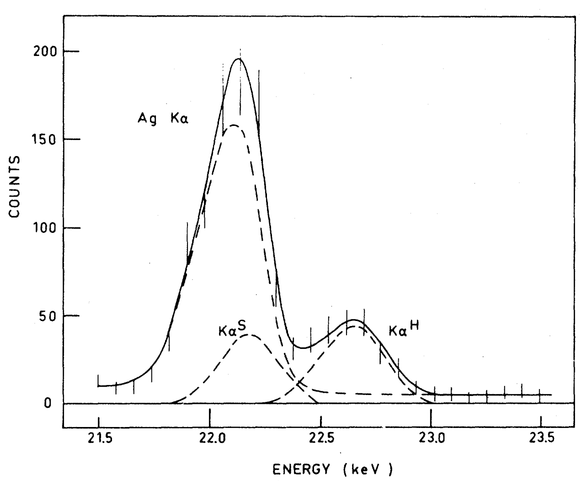

FIG. 1. Ge coincidence spectrum of the Ag $K\alpha$-x-ray region.

The $K\alpha^H$ line is almost completely resolved from the composite line. In consequence of this the inaccuracy in the unfolding procedure does not affect the resulting intensity of the $K\alpha^H$ line significantly.

The composite line has a diagram $K\alpha$-x-ray component arising from coincidences with (1) external bremsstrahlung due to K-conversion electrons and (2) internal Compton photons, and a satellite $K\alpha^S$-x-ray component due to coincidences with $K\alpha^H$ x rays. For simplicity the $K\alpha$-x-ray line due to coincidences with $K\beta^H$ x rays is assumed to coincide with the $K\alpha$ diagram line. One can show that the intensity of the $K\alpha^S$ line is smaller than the intensity of the $K\alpha^H$ line by a factor of α^S (the $K\alpha^S$-x-ray fraction of all the K^S x rays). In the spectrum analysis we used $\alpha^S=\alpha=0.83$, the value for diagram lines.[8] Furthermore, it should be pointed out that the intensity ratio of the $K\alpha_1$ and the $K\alpha_2$ component of the $K\alpha^H$ line is only 1.3,[9] whereas for the $K\alpha$ diagram line this ratio is 1.89.[8] We used this last value also for the $K\alpha^S$ line. In consequence of these different intensity ratios the energy difference of the $K\alpha_1$ diagram line and the $K\alpha_1^H$ line is 36 eV higher than the energy difference of the $K\alpha$ line and the $K\alpha^H$ line as obtained from the Ge spectrum (it is assumed that the energy difference $K\alpha_1-K\alpha_1^H$ is equal to the difference $K\alpha_2-K\alpha_2^H$). As mentioned before, the intensity of the $K\alpha^H$ line does not depend significantly on the spectrum analysis.

From the number of counts in the $K\alpha^H$ line, the number of counts in the NaI(Tl) channel (the correction for noncoincident background was 1.5%), and the instrumental constant ϵ we find the quantity equal to the right side of formula (1). This quantity is presented in Table I. The measurement was performed 7 times. In the first five measurements the 100 nCi source was used, in the last two measurements the 50 nCi source. In Table I we also present the energy difference between the $K\alpha_1$ diagram line and the $K\alpha_1^H$ line. The

TABLE I. Experimental results.

	$\frac{N_c}{\epsilon N_{\mathrm{NaI}}}\times10^5$ [a]	$E(K\alpha_1^H)-E(K\alpha_1)$ (eV)
	2.1	555
	2.2	515
	2.4	515
	2.1	555
	2.1	475
	2.3	555
	1.8	555
Average	2.14 ± 0.07	532 ± 12

[a] For symbols see text [formula (1)].

TABLE II. Comparison of the present experimental result with theory ($P_{KK} \times 10^5$).

Experimental P_{KK}(IC)	P_{KK}(SO) one step	P_{KK}(SO) two step	Theoretical[a] P_{KK}(DC)	P_{KK}(DIC)	P_{KK}(ICIC)
2.8 ± 0.7	0.92	7.4	$\simeq 0.4$	≤ 1.7	47

[a]SO, DC, DIC, and ICIC refer to shakeoff, direct collision, double internal conversion, and internal conversion of internal Compton effect, respectively.

precision in each number is one quarter of the channel width, i.e., 20 eV. The result is in excellent agreement with the expected value of 540 eV mentioned in the Introduction. The energy difference between the $K\alpha$ line and the $K\alpha^S$ line is about 90 eV.

In making the step from the quantity presented in Table I to P_{KK}(IC) we are confronted with two problems: (1) Which value should be used for $\alpha^H \omega_K^H$ and (2) how large is P_{KK}(EC)?

As for the first problem, we consider two cases. (1) We make the assumption that the considered quantities for hypersatellite and diagram lines are identical, i.e., $\alpha^H \omega_K^H = \alpha \omega_K = 0.685$. (2) We make the assumption that only the $K\alpha_1^H$-x-ray transition is suppressed whereas the other hypersatellite transition probabilities are not affected. We then arrive at $\alpha^H \omega_K^H = 0.632$. Although no information is available on whether $K\beta^H$ and hypersatellite Auger transitions are suppressed or not, it is probable that some of them are. We then arrive at an $\alpha^H \omega_K^H$ value probably somewhere in between 0.632 and 0.685. Therefore we used $\alpha^H \omega_K^H = 0.66 \pm 0.03$.

As for the second problem we first make the same assumption Nagy *et al.*[4] made, i.e., $P_{KK}(\text{EC}) = 0$. We then find $P_{KK}(\text{IC}) = (9.5 \pm 0.7) \times 10^{-5}$. This value is considerably lower than the result of Nagy *et al.*, so our statement concerning a large systematic error, made in the Introduction, is proved correct. A possible explanation is that Nagy *et al.* did not take into account the energy shift of the $K\alpha^S$ line in the spectrum analysis.

As mentioned in the Introduction, experimental P_{KK}(EC) values are not available. Theoretical treatments of the shaking process in electron capture decay have been given by several authors.[2] Only Mukoyama and Shimizu give a value for the case of ^{109}Cd (Ref. 3): $P_{KK}(\text{EC}) = 3.47 \times 10^{-5}$. Application of this value yields $P_{KK}(\text{IC}) = (2.8 \pm 0.7) \times 10^{-5}$. The correctness of the different theoretical treatments has not yet been tested sufficiently but all of the predictions are of the order of the experimental data available. Since the predictions of Mukoyama and Shimizu have the lowest values in comparison with the results of other work, it seems probable that the actual P_{KK}(EC) value will not be lower than 3.47×10^{-5} and consequently that P_{KK}(IC) will not be higher than $(2.8 \pm 0.7) \times 10^{-5}$.

The processes which may lead to double K-shell vacancy creation in internal conversion are (1) the shaking process, (2) direct collision, (3) double internal conversion, and (4) internal conversion of internal Compton effect.

Theoretical calculations of the shakeoff probability P_{KK}(SO) have been performed by Mukoyama and Shimizu.[3] They used a one- and a two-step relativistic overlap theory. The results are given in Table II. Comparison with our experimental result shows that the one-step theory seems preferable.

Theoretical values for the shakeup probability are not available. Calculations on electron capture decay[2] show that there the contribution of shakeup is considerable. Consequently, a significant contribution of shakeup in internal conversion should not be excluded. Theoretical work is urgently required.

For internal conversion the direct-collision process has not been treated theoretically. In the case of β^- decay, Feinberg[10] estimated the relative probability of direct collision and shakeoff to be equal to the ratio of the K binding energy and the decay energy. However, from experimental data the conclusion seems justified that the actual contribution from direct collision is much lower.[11] Application of Feinberg's estimate to internal conversion gives only a rough order of magnitude. From $P_{KK}(\text{SO}) = 0.9 \times 10^{-5}$ we find $P_{KK}(\text{DC}) \simeq 0.4 \times 10^{-5}$.

Nagy *et al.*[4] evaluated Eichler's theory of double internal conversion[12] for the case of ^{109}Cd, using the experimental result of Knauf and Sommer[1] for the probability of a double γ process. The result is $P_{KK}(\text{DIC}) \leq 1.7 \times 10^{-5}$.

Internal conversion of internal Compton effect has been treated theoretically by Listengarten.[13] Evaluation by Nagy *et al.*[4] for ^{109}Cd results in a probability of $P_{KK}(\text{ICIC}) = 47 \times 10^{-5}$, i.e., a value which is an order of magnitude higher than the experimental result. Further theoretical treatment of this process is required.

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