Double-K-shell-vacancy creation in the decay of ^{103}Pd

C. W. E. van Eijk, J. Wijnhorst, and M. A. Popelier
Physics Department, Delft University of Technology, Delft, The Netherlands

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The probability P_{KK} of double-K-shell-vacancy creation per K-electron-capture decay of ^{103}Pd has been determined by means of a $K\alpha$-x-ray–K-x-ray coincidence experiment: $P_{KK} = (3.13 \pm 0.31) \times 10^{-5}$. The energy shift of the hypersatellite Rh $K\alpha_1^H$-x-ray line was found to be 512 ± 10 eV.

I. INTRODUCTION

Experimental values of P_{KK}, the probability of double-K-shell-vacancy production per K-electron-capture (EC) decay, are scarce. In 1973 Law and Campbell presented a critical review of experimental work.[1] Only seven experiments, performed on three different isotopes (^{71}Ge, ^{131}Cs, ^{165}Er), were judged to have some degree of reliability.[2-7] Since then, the results of only three new experiments have been published, two of which were on ^{55}Fe and one on ^{109}Cd.[8-10] In addition, the reliability of one of the experiments[3] has become questionable.[11] In general, the accuracy of the experimental P_{KK} values is poor; in some cases the results of different experiments on the same isotope do not overlap.

As for the theoretical side, we refer to the recent reviews by Intemann[12] and by Bambynek *et al.*[13] The most advanced calculations are those due to Intemann[12] and to Mukoyama *et al.*[14] Intemann's calculations result in values for P_{KK}(SO), the "shakeoff" probability, whereas those of Mukoyama *et al.* result in values for P_{KK}(SU), the "shakeup" probability, as well as P_{KK}(SO). The terms shakeoff and shakeup refer to the transition of the noncaptured K-shell electron to a continuum state and an unoccupied bound state, respectively, as the result of the sudden change in the central potential due to the EC process [$P_{KK} = P_{KK}(\text{SO}) + P_{KK}(\text{SU})$]. Owing to the experimental situation sketched above, it is not possible to decide whether one of the theories does predict the correct P_{KK} values.

In an attempt to contribute to the termination of this rather unsatisfactory situation, we report in this paper the result of a measurement of P_{KK} on ^{103}Pd, an isotope not studied yet.

The ground state of ^{103}Pd ($t_{1/2} = 17d$) decays by EC for 99.97% to the 40 keV metastable level in ^{103}Rh.[15] The energy of this level does not allow the production of double K-shell vacancies in the transition to the ground state, and in consequence the K-EC decay to the 40 keV level is the only process in which double K-shell vacancies will be created (the contributions from the weak EC decays to the higher energy levels and from the subsequent transitions are negligibly small).

II. METHOD OF MEASUREMENT

Double K-shell vacancies give rise to the subsequent emission of hypersatellite Rh K^H x rays and satellite Rh K x rays.

These x rays can be observed in a K-x-ray–K-x-ray coincidence experiment, thus supplying a measure for P_{KK}. In the case of ^{103}Pd the weak decay modes to the higher energy levels give a contribution of diagram K x rays to the coincidence spectrum with an intensity which is of the same order of magnitude as the intensity of the lines of interest. Confining ourselves to the most intense hypersatellite line, the Rh $K\alpha^H$ line, we take advantage of the fact that this line is shifted by approximately 500 eV to a higher energy with respect to the diagram Rh $K\alpha$-x-ray line. The application of a high-resolution Ge detector offers the possibility to observe the Rh $K\alpha^H$-x-ray line well separated from the diagram and satellite Rh $K\alpha$ x rays.

The experiments have been performed by means of a 25-mm^2 intrinsic-Ge x-ray detector of Princeton Gamma-Tech and a 40-mm-diam NaI(Tl) x-ray detector. A coincidence circuit was employed with a time window of 50 nsec. The coincidence spectrum, the accidental-coincidence spectrum, and the singles spectrum were recorded simultaneously by means of a routing system. In addition, we recorded the NaI(Tl) spectrum and the time spectrum. The data were handled by means of a PDP 11/10 computer.

The ^{103}Pd activity was obtained from neutron bombardment of ^{102}Pd (enrichment 78%) for 24 hours at a neutron flux of 2.5×10^{14} cm^{-2} sec^{-1}. The sources were prepared by evaporation of a drop of active solution onto Scotch tape. The sources were sandwiched between the two detectors placed at 180°. In order to prevent the detection of electrons arising from double K-shell vacancy production, Auger electrons, and conversion electrons, Al foils were placed on either side of a source. We performed test experiments with Al foils with thicknesses of 0.1, 0.3, and 0.5 mm. We did not observe any difference in $N_c/(\epsilon N_{\rm NaI})$ (see below) at an accuracy level of 10%. Therefore 0.3 mm foils have been used in the experiments.

From the decay scheme we find for the relation between N_c, the number of Rh $K\alpha^H$-x-ray counts in the coincidence spectrum recorded with the Ge detector, and $N_{\rm NaI}$, the number of Rh K-x-ray counts in a gate set on the K x rays recorded with the NaI(Tl) detector,

$$N_c/(\epsilon N_{\rm NaI}) = (0.90 \pm 0.01)\alpha^H \omega_K^H P_{KK}({\rm EC}), \qquad (1)$$

in which 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_K^H is the fluorescence yield for the K^H x rays. In the derivation of this formula it was assumed that the fluorescence yields ω_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 uncertainty in the numerical factor in formula (1) is due to the uncertainty in the intensities of the decay processes.

Before and after each measurement on ^{103}Pd the instrumental constant ϵ has been determined by means of a ^{125}Te^m source ($t_{1/2} = 58d$). The tellurium activity was obtained from neutron bombardment of ^{124}Te (enrichment 96%) for 12 days at a neutron flux of 2.5×10^{14} cm^{-2} sec^{-1}. The sources were prepared by evaporation of a drop of active solution onto Scotch tape. The source strengths varied from 1 to 100 nCi corresponding to counting rates in the Ge and NaI(Tl) detectors varying from, respectively, $\simeq 10$ to $\simeq 10^3$ sec^{-1} and $\simeq 10^2$ to $\simeq 10^4$ sec^{-1}. The tellurium sources were also sandwiched between Al foils and placed between the detectors in a position identical to the one in which the ^{103}Pd source was placed. The instrumental constant ϵ was determined from the intensity of the Te $K\alpha$-x-ray coincidences $N_{c\alpha}$ (energy $E = 27.4$ keV) and the Te $K\beta$-x-ray coincidences $N_{c\beta}$ ($E = 31.0$ keV). The applied formulas derived from the decay scheme[16] are for the $K\alpha$ x rays

$$\epsilon = (2.28 \pm 0.07) N_{c\alpha}/N_{\rm NaI} \qquad (2)$$

and for the $K\beta$ x rays

$$\epsilon = (10.1 \pm 0.3) N_{c\beta}/N_{\rm NaI}, \qquad (3)$$

in which $N_{\rm NaI}$ is the sum of the number of Te K-x-ray counts and the number of 35-keV γ-ray counts in a gate set on the corresponding part of the spectrum recorded with the NaI(Tl) detector. The uncertainty in the coefficients in the formulas is due to the uncertainty in the intensities of the transitions. In formulas (2) and (3) the fluorescence yield, $\omega_K = 0.875 \pm 0.028$ (Ref. 17), does also contribute to the uncertainty. However, this value has been obtained from a fit of a smooth curve to all available ω_K values, and ω_K for the Rh K x rays is also obtained from this fit. As a consequence of this, the latter ω_K value will have approximately the same systematic error as ω_K for Te K x rays. Then, in applying formulas (1)–(3), the errors cancel. For this reason the error in ω_K is not taken into account.

Before we apply ϵ as obtained from formulas (2) and (3) to formula (1), some corrections have to be made. In order to take into account the difference between absorptions in the 0.3-mm-thick Al foil of the Rh $K\alpha^H$ x rays ($E = 20.7$ keV) and respectively the Te $K\alpha$ x rays ($E = 27.4$ keV) and $K\beta$ x rays ($E = 31.0$ keV), we applied corrections of 16% and 19%, respectively. Furthermore corrections have been applied for the difference between the detection efficiencies of the x rays in the Ge detector. These corrections were 3% and 4%, respectively, for the Te $K\alpha$ and $K\beta$ x rays.

The values obtained from formulas (2) and (3) were found to be in good agreement with each other and independent of the source strength within the range from 1 to 100 nCi mentioned above. In consequence, the ^{103}Pd source strengths were chosen well in this range. We obtained $\epsilon = (19.0 \pm 1.6) \times 10^{-3}$. The error is a standard deviation which takes into account the uncertainty in the coefficients in formulas (2) and (3), which amounts to 3%, the uncertainty in the corrections mentioned above (5%), the uncertainty due to the statistics and the positioning of the source between the detectors (3%), and the uncertainty due to a correction of (20 ± 5)%, which has been applied in order to take into account summation effects of Te K x rays in the NaI(Tl) detector. As a check on the independence of the efficiency ϵ of the energy over the energy region of interest, we also did perform a calibration at an energy below the energy of the Rh $K\alpha$ x rays by means of a ^{97}Ru source (Tc $K\alpha$ x rays; $E = 18.3$ keV). A detailed description of this calibration has been presented in Ref. 10. We found excellent agreement with the above result obtained from the ^{125}Te^m source.

Once $N_c/(\epsilon N_{\rm NaI})$ has been determined, we need to know $\alpha^H \omega_K^H$ in order to find P_{KK} [see formula (1)]. It has been pointed out by Åberg *et al.*[18] that in the case of low and intermediate Z, the intensity ratio of the $K\alpha_1^H$- and $K\alpha_2^H$-x-ray transition is considerably smaller than the ratio for the corresponding diagram lines. In the case of Rh the ratio is 1.20 for the hypersatellite lines[18] as against 1.90 for the diagram lines.[17] If we make the assumption that only the $K\alpha_1^H$-x-ray transition is suppressed, we find $\alpha^H\omega_K^H = 0.605$ as against $\alpha\omega_K = 0.670$ for diagram lines.[17] No information is available as to whether other transitions are suppressed. If some of them are, we arrive at a higher value of $\alpha^H\omega_K^H$, probably somewhere between 0.605 and 0.670. Therefore we used $\alpha^H\omega_K^H = 0.64 \pm 0.03$.

III. RESULTS AND DISCUSSION

The measurements on ^{103}Pd have been performed alternately with measurements on other isotopes in five periods of time spread over a year (see Ref. 10).

Figure 1 shows a coincidence spectrum of the Rh $K\alpha$-x-ray complex recorded with the Ge detector in one of the measurements. Accidental coincidences have been subtracted. The height of the subtracted accidental coincidence $K\alpha$-x-ray line was 290 counts.

The diagram $K\alpha$-x-ray line in Fig. 1 is mainly due to coincidences between K x rays from the weak K-electron-capture decay modes to the levels above the 40-keV level and the subsequently emitted K x rays arising from internally converted transitions.

The satellite $K\alpha^S$ line in Fig. 1 is due to coincidences with the $K\alpha^H$ x rays detected in the NaI(Tl) crystal. The intensity of the $K\alpha^S$ line is 0.83 times the intensity of the $K\alpha^H$ line. This is the $K\alpha^S$-x-ray fraction of all the K^S x rays. It has been assumed that this fraction is identical to that of the diagram lines. For the energy shift we used 65 eV.[19] For simplicity we assumed that the satellite line due to coincidences with the $K\beta^H$ x rays has the energy of the diagram line. This satellite line gives only a small contribution to the intensity of the diagram line in Fig. 1.

The $K\alpha^H$ line is due to coincidences with the

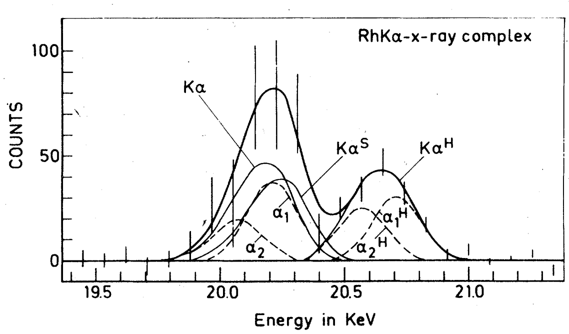

FIG. 1. Ge coincidence spectrum of the Rh $K\alpha$-x-ray region recorded in one of the measurements.

satellite K x rays detected in the NaI(Tl) detector.

In the unfolding procedure, we used the Rh $K\alpha$-x-ray line from the singles spectrum for the diagram and the satellite line. The hypersatellite line has been constructed by means of the $K\alpha_1$- and $K\alpha_2$-x-ray lines as obtained from the unfolding of the diagram line of the singles spectrum (assuming an intensity ratio of 1.90). For the hypersatellite line we used the intensity ratio of 1.20 and we assumed that the energy shift is the same for the two components.

We give the average results of the measurements performed in the five periods of time mentioned above (with time in parentheses at right):

$$N_c/(\epsilon N_{\mathrm{NaI}}) = (1.93 \pm 0.13)\times 10^{-5}\ (20d),$$
$$(1.91 \pm 0.10)\times 10^{-5}\ (26d),$$
$$(1.65 \pm 0.15)\times 10^{-5}\ (9d),$$
$$(1.66 \pm 0.15)\times 10^{-5}\ (19d),$$

and

$$(1.88 \pm 0.18)\times 10^{-5}\ (11d).$$

The errors are standard deviations due to counting statistics only. The results are in reasonable agreement. The unweighted average is given in Table I. Using formula (1) with $\alpha^H\omega_K^H = 0.64 \pm 0.03$ and including an inaccuracy of 8% owing to the inaccuracies in the numerical factors in formulas (1)–(3) and in the corrections applied, we find the value of P_{KK} presented in Table I. The energy differences obtained in the five periods of time are $E(K\alpha_1^H) - E(K\alpha_1) = 529 \pm 15$, 509 ± 10, 517 ± 25, 499 ± 20, and 504 ± 18 eV, respectively. These values are in good agreement. The average value is also presented in Table I. The assignment of an error smaller than 10 eV is not realistic, since a systematic error due to (i) the (unknown) inaccuracy in the ratio $K\alpha_1^H/K\alpha_2^H$ and (ii) the fact that the difference between energy shifts of the $K\alpha_1^H$- and $K\alpha_2^H$-x-ray lines has not been taken into account will start to contribute significantly (see Ref. 10). The present result for the energy shift of the Rh $K\alpha_1^H$-x-ray line is in excellent agreement with the value of 510 eV which is obtained from an interpolation of results for the energy shift of the $K\alpha_2^H$ line for the isotopes ^{26}Fe, ^{28}Ni, and ^{29}Cu,[20] and of the $K\alpha_1^H$ line for ^{114}In^m.[21]

TABLE I. Experimental results.

$N_c/(\epsilon N_{\mathrm{NaI}})\times 10^5$	$P_{KK}\times 10^5$	$E(K\alpha_1^H) - E(K\alpha_1)$ (eV)
1.81 ± 0.06	3.13 ± 0.31	512 ± 10

For a comparison between experiment and theory we consider Fig. 2. The upper solid curve is a smooth fit to the theoretical P_{KK} values for the isotopes ^{55}Fe, ^{71}Ge, ^{109}Cd, ^{131}Cs, ^{165}Er, and ^{169}Yb as obtained by Mukoyama *et al.*[14,22]; the lower solid curve is a smooth fit to the P_{KK}(SO) values for the isotopes ^{55}Fe, ^{71}Ge, ^{131}Cs, and ^{165}Er, calculated by the same authors.[14] The calculations of the P_{KK} for ^{37}A have not been considered since these P_{KK} values are completely off the smooth curves fitting the other results. The dashed curve is a smooth fit to the theoretical P_{KK}(SO) values for the isotopes ^{37}A, ^{71}Ge, ^{131}Cs, and ^{165}Er, as obtained by Intemann.[12] In all cases the difference between a used theoretical P_{KK} or P_{KK}(SO) value and the corresponding point on a curve is less than 10% of the theoretical value.

The experimental points shown are the P_{KK} values reported in Refs. 2, 4–8, and 10; the P_{KK}(SO) value reported in Ref. 9 (^{55}Fe, lower point); and the present result.

For $Z > 26$, P_{KK}(SO) as obtained by Mukoyama *et al.* is smaller than P_{KK}(SO) as obtained by Intemann. If we assume the same tendency for P_{KK}, the present experimental P_{KK} value for the decay of ^{103}Pd seems to favor the approach used by Mukoyama *et al.*

If we consider all experimental data, it is noteworthy that the results obtained from experiments in which semiconductor detectors have been used tend to be relatively lower than the results obtained from other experiments. An explanation may be found in the better energy resolution of a semiconductor detector. This offers the possibility to discriminate against K x rays due to small contaminations, which are difficult to observe in singles spectra but may contribute significantly in coincidence spectra.

On the basis of the results obtained only from semiconductor detector experiments, and considering the suggestion made in Ref. 10 about the dependence of the shaking probability on the ratio of the transition energy and the threshold energy (approximately twice the K-binding energy), the conclusion seems justified that the theoretical P_{KK} values obtained by Mukoyama *et al.* represent an upper limit. It is obvious that still more experimental data are necessary, particularly for EC decays near the threshold energy, in order to establish whether this conclusion is correct.

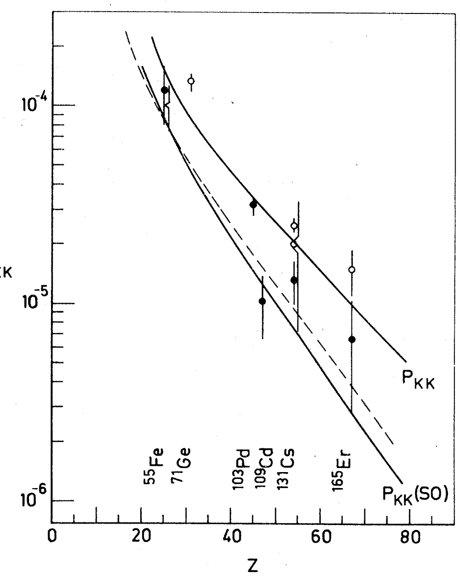

FIG. 2. Comparison of theoretical and experimental P_{KK} and P_{KK}(SO) values. The solid curves represent smooth fits to the theoretical P_{KK} and P_{KK}(SO) values calculated by Mukoyama *et al.*[14,22]; the dashed curve is a smooth fit to the theoretical P_{KK}(SO) values calculated by Intemann.[12] The isotopes for which the experimental values are shown are (references in order of increasing P_{KK} value) ^{55}Fe (Refs. 9 and 8), ^{71}Ge (Ref. 2), ^{103}Pd (present result), ^{109}Cd (Ref. 10), ^{131}Cs (Refs. 6–4), and ^{165}Er (Refs. 6 and 7). Closed circles represent P_{KK} values obtained from semiconductor detector experiments, open circles represent P_{KK} values obtained from other experiments. The lower point for ^{55}Fe is an experimental P_{KK}(SO) value. The points are plotted at Z of the daughter nucleus.

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ray satellite line and the sum of the intensities of the K-x-ray diagram lines. In Ref. 3 this satellite line is identified as the $K\alpha$-x-ray line emitted when the Ga atom has initially one K and one L vacancy ($KL \rightarrow L_{III}L$ and $L_{II}L$). However, the energy difference between the satellite line and the $K\alpha_1$ diagram line is approximately 80 eV, which makes it more probable that this satellite line is due to the transition $KL^2 \rightarrow L_{III}L^2$. If we take into account that the hypersatellite $K\alpha_1^H$-x-ray transition is suppressed ($K\alpha_1^H/K\alpha_2^H \simeq 0.3$; Ref. 18) and assume that the probabilities of other transitions are not affected, we arrive at $P_{KK} = (44 \pm 17) \times 10^{-5}$, an extraordinarily high value.

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