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THE ELECTRON CAPTURE DECAY OF ^{195}Au

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Abstract: The decay of ^{195}Au has been investigated by means of a 4π internal source scintillation spectrometer and a Ge(Li) detector. Electron capture branching ratios and several L/K and M+.../L capture ratios have been measured. The atomic electron capture transition energy and the cross-over decay probability of the 130 keV state have been calculated.

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RADIOACTIVITY ^{195}Au; measured E_γ, I_γ, $I_\gamma+I_{ce}+I_X+I_{Auger}$; deduced L/K, M+.../L capture ratios, Q_{EC}. ^{195}Pt transition deduced cc, cross-over probability. Ge(Li), Si(Li), NaI, CsI detectors. Internal source method.

1. Introduction

The electron capture decay of ^{195}Au and the properties of the low-lying energy levels of ^{195}Pt have been extensively studied by different methods. Nevertheless, there are still differences in the decay schemes. The decay scheme of ^{195}Au is given in fig. 1. Differences between electron capture branching ratios result, among other things, from the difficulties of measuring the electron capture transition to the ground state of ^{195}Pt (see subsect. 3.3.1). Also, a discrepancy exists between the branching ratios to the two excited states near 200 keV as given by several authors (see subsect. 3.3.1). In the decay of the 130 keV level in ^{195}Pt a large discrepancy exists between the cross-over ratio measured from the decay of ^{195}Au and this ratio measured from the decay of ^{195m}Pt (see subsect. 3.3.2).

Concerning the electron capture ratios in the decay of ^{195}Au, only the K-capture fractions in the transitions to the levels at 99 and 130 keV in ^{195}Pt have been measured. The L/K and M+.../L capture ratios have not been measured before. Interest in these ratios is increasing due to the recalculation of the orbital electron wave functions by Zyryanova and Suslov [3, 4]), and by Behrens and Bühring[1]). Recently, Vatai [5]) and Suslov [9]) made new calculations of the exchange and overlap corrections.

In the present work the electron capture decay of ^{195}Au has been re-investigated by two different methods. A 4π internal source scintillation spectrometer has been used to determine the electron capture ratios and the strengths of the transitions to the ground, 99 keV and 130 keV states in ^{195}Pt (see sect. 2). The decay of the 200 and 211 keV levels and the cross-over ratio in the decay of the 130 keV level were determined by means of a Ge(Li) detector (see sect. 3). The carrier-free ^{195}Au activity

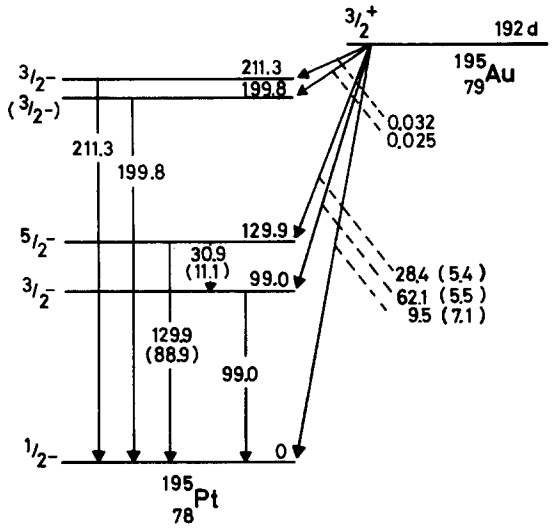

Fig. 1. Decay scheme of ^{195}Au.

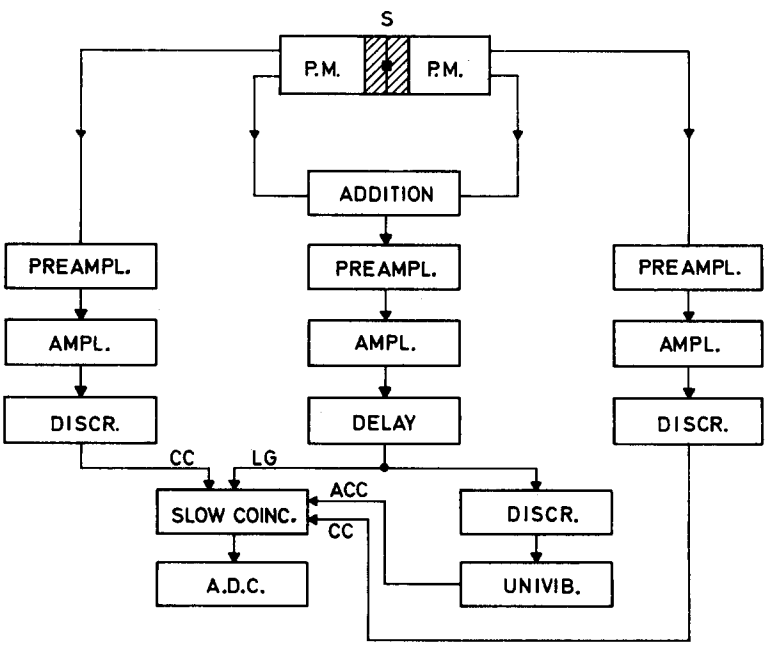

Fig. 2. Block diagram of the experimental arrangement with the 4π internal source scintillation spectrometer.

was bought from N.E.N. Chemicals GmbH and the Radiochemical Centre, Amersham. The principal contamination was ^{196}Au with $T_{\frac{1}{2}} = 6.18$ d. After a few months no effect of this decay was observed.

2. Electron capture ratios

2.1. MEASURING DEVICE

The spectrometer used in this investigation consists of a so-called source crystal, 4π enveloped by two inactive scintillation crystals. A source crystal is a scintillation crystal with a radioactive source dispersed through its volume. Both NaI(Tl) and CsI(Na) crystals were used. The growing and shaping of the source crystals and the assembly of the crystal combinations have been described previously [15]).

A block diagram of the experimental arrangement is given in fig. 2. The photomultipliers (EMI 9635 QA) were selected for their low dark current and high quantum efficiency. The electronic equipment is Ortec designed. The multichannel pulse-height analyser is a Nuclear Data ND 160. An electron capture event detected in any crystal of the assembly causes coincident pulses in both photomultipliers. The anode signals are added in order to get the energy signal. The dynode signals are used as a coincidence criterion in order to discriminate against the photomultiplier and electronic noise. The recording of afterpulses is prevented by an anti-coincidence arrangement. The spectrometer has been surrounded by a Pb shield in order to reduce the background radiation.

2.2. ESCAPE OF X-RAYS AND γ-RAYS

The investigation of the electron capture ratios in the decay of ^{195}Au is rather difficult. Using a proportional counter or an unenveloped source crystal, the K X-radiation has a large probability of escaping because of its high energy (65 keV). Large escape corrections are needed in these cases.

Using a NaI(Tl) crystal in combination with an external ^{195}Au source, it is very difficult to make escape corrections, because the I-escape peaks of the 130 and 99 keV γ-rays and the K X-ray will give a contribution in the photopeaks of the 99 keV γ-ray, the K X-ray and the 31 keV γ-ray respectively [see ref. [12])].

In this work a 4π spectrometer has been used. No escape corrections were necessary because of the dimensions of the crystals.

2.3. ABSORPTION OF CONVERSION AND AUGER ELECTRONS

Although in a source crystal no absorbing materials are present, self-absorption can occur in source atoms clustered on dislocations or other imperfections in the crystal lattice. In order to reduce the probability of clustering the source strength has been chosen to be very low, i.e. < 400 counts/sec $\cdot$ cm^3. The source crystals used meet the physical criterion of Schulz [16]). This has been determined by means of the external calibration sources ^{241}Am and ^{57}Co. Using a 4π geometry it is not relevant whether the dispersion of Au in NaI(Tl) is homogeneous or not [15]).

2.4. MEASUREMENTS

In internal source techniques the orbital electron binding energies are registered instead of the X-ray energies. In the decay of ^{195}Au the de-exciting γ-rays are coincident with the related electron capture events because of the short lifetime of the low-lying energy levels in ^{195}Pt. The light from every kind of radiation involved in this process is summed because of the absence of escape. The energies appearing in the spectrum are not the energies of the first column of table 1 but the energies of the second, third and fourth column.

TABLE 1

Summation of γ-ray energies with the electron binding energies of the K, L_1 and M_1 shell

γ	K	L_1	M_1
0	78.4	13.9	3.3
99.0	177.4	112.9	102.3
129.9	208.3	143.8	133.2

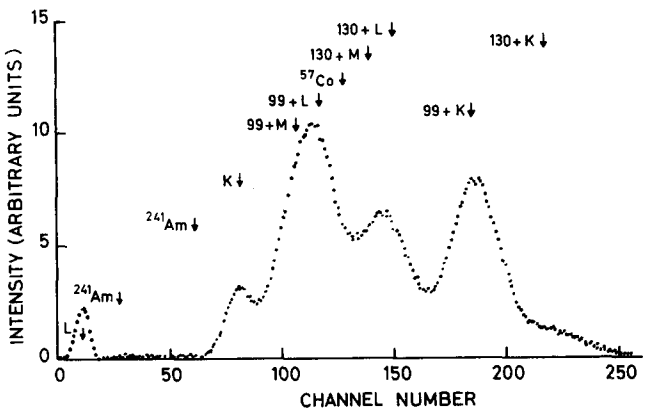

Fig. 3. Typical summation spectrum measured with the 4π internal source scintillation spectrometer. The background has been subtracted.

The measurements have been carried out with NaI(Tl) and CsI(Na) source crystals with different source strengths. The energy spectrum is shown in fig. 3. The background has already been subtracted. This background is determined before and after each measurement. The same gain has been obtained with the help of the calibration sources ^{241}Am and ^{57}Co. This calibration is always necessary because of the breakdown of the optical coupling when removing the source crystal from the spectrometer. The calibration sources are also required to adjust the proper addition of the anode signals from the photomultipliers. Special care has to be taken to ensure an equal light yield from the source crystal and the enveloping crystals since an unequal light yield will result in unreliable summation spectra.

TABLE 2

Experimental electron capture ratios and Q_{EC} values

Author	Ref.	P_K^{130}	L/K; 130	M+. . ./L; 130	P_K^{99}	L/K; 99	M+. . ./L; 99	L/K; g.s.	Q_{EC}
Bisi *et al.*	[8]	0.154±0.021							234±2
Goedbloed *et al.*	[10]	0.153±0.011							226±2 229±1
de Wit and Wapstra	[11]	0.196±0.005							227±1
Harris *et al.*	[12]	0.13 ±0.01			0.40 ±0.09				225±5
Jasinski and Herrlander	[13]	0.166±0.020			0.458±0.012				224±2
Fink and Benczer-Koller	[18]	0.13 ±0.01			0.45 ±0.10				225±5
present work		0.160±0.017	3.055±0.186	0.697±0.078	0.438±0.011	0.871±0.044	0.478±0.020	0.337±0.007	230±1 231±1
Suslov without X_{Suslov} $Q_{EC} = 231 \pm 1$				0.372±0.001		0.845±0.019	0.350±0.001	0.335±0.001	
Suslov with X_{Suslov} $Q_{EC} = 231 \pm 1$				0.393±0.001		0.881±0.019	0.369±0.001	0.349±0.001	
present work			$X_{exp}^{L/K;\ 130}$	$X_{exp}^{M/L;\ 130}$		$X_{exp}^{L/K;\ 99}$	$X_{exp}^{M/L;\ 99}$	$X_{exp}^{L/K;\ g.s.}$	
$Q_{EC} = 230 \pm 1$			1	2.100±0.265		1.012±0.056	1.462±0.072	1.003±0.020	
$Q_{EC} = 231 \pm 1$			1.042	2.107±0.266		1.034±0.056	1.464±0.072	1.006±0.020	

Theoretical electron capture ratios and exchange and overlap corrections, both calculated by means of our Q_{EC} values.

2.5. ANALYSES

The summation spectra have been analysed by means of a computer program. The program makes a least-squares adjustment of the seven peaks to the experimental peak complex with the help of the Marquardt algorithm (see table 1). The input of the program consists of the summation spectrum, the proper background and the starting values of the variables. The variables are the channel number, height and width of each peak. The peak widths have to satisfy the equation of the resolution of scintillation detectors [17]),

$$\left(\frac{\Delta E}{E}\right)^2 = \frac{a^2}{E} + b^2.$$

2.6. RESULTS AND DISCUSSION

The results of the electron capture ratios are given in table 2. The K-capture fraction P_K in the electron capture decay to the 99 and 130 keV levels is compared with other experimental values. They are in agreement with the values given in refs. [12,13]) and [18]) and in refs. [8,10,13]) respectively. The L/K and M+.../L electron capture ratios have not been measured before.

It is interesting to compare these values with the theoretical allowed values. However, the electron capture decay of ^{195}Au is a first-forbidden non-unique decay. Generally the theoretical capture ratios of both kinds of decay are equal to within 3 %, except in the region near the K-capture threshold [21]). In order to make this comparison it is necessary to calculate the atomic electron capture transition energy Q_{EC}. It has been determined from the L/K capture ratio of the 130 keV level. The electron wave functions of Suslov [3,4]) were used in the calculation. The exchange and overlap correction is usually taken to be one in these calculations. The result is $Q_{EC} = 230 \pm 1$ keV. Taking the correction according to Suslov [9]) to be 1.042, Q_{EC} becomes 231 ± 1 keV. Both values are in agreement with Q_{EC} values [10,12,18]) determined from experimental P_K values.

In table 2 the experimental capture ratios are compared with the theoretical ratios calculated with $Q_{EC} = 231 \pm 1$ keV. The error in the Q_{EC} value results in the given error of the theoretical capture ratios. Another comparison with theory is given by X_{exp}, the experimental capture ratio divided by the theoretical one without the exchange and overlap corrections.

The experimental L/K capture ratio of the 99 keV level is, within errors, equal to the theoretical values. The L/K capture ratio of the ground state is in agreement with the theoretical values without the correction according to Suslov. The reason may be:

(i) the correction according to Suslov is too high in the case of ^{195}Au;

(ii) the log ft value of the ground state is higher than the log ft values of the other states.

The experimental M+.../L capture ratios are much larger than the theoretical values. However, at a low transition energy the electron capture from the M-shell becomes more significant in the forbidden decay than in the allowed decay [22]).

3. The decay scheme of ^{195}Au

3.1. MEASUREMENTS

Besides the experiments mentioned in sects. 1 and 2, we performed measurements with a Ge(Li) detector in order to obtain supplementary information about the decay scheme of ^{195}Au. The Ge(Li) detector has been surrounded by a shield as mentioned

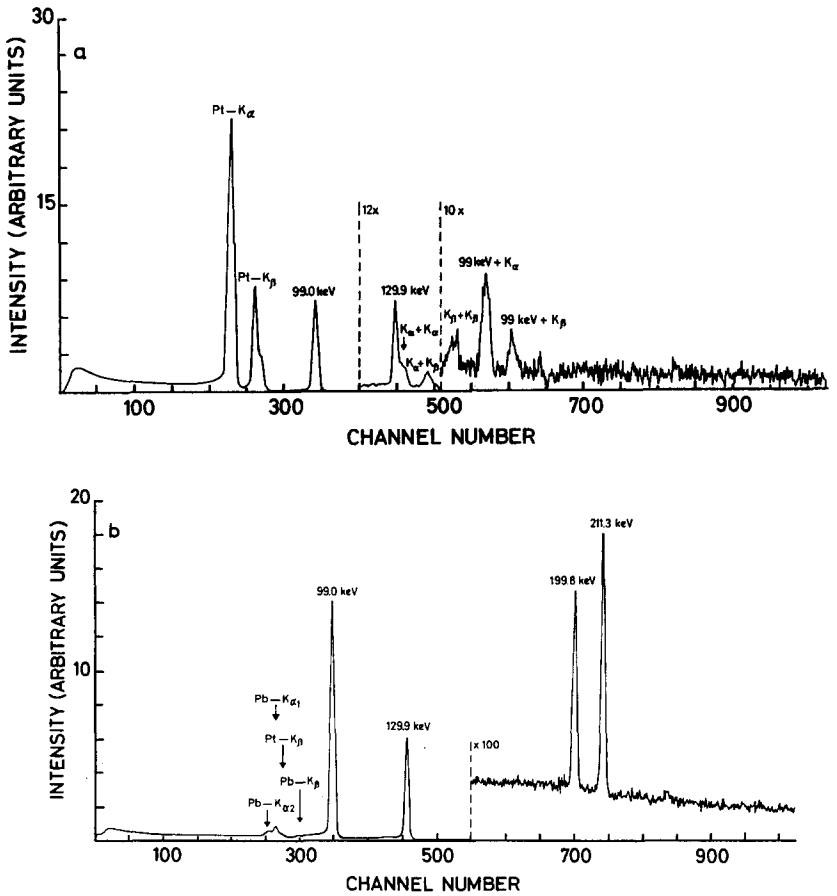

Fig. 4. Gamma-ray spectrum of ^{195}Au measured by means of the Ge(Li) detector. (a) No absorber. (b) With 1.898 g/cm^2 Sn absorber.

before. Single count spectra have been measured with the Ge(Li) detector and an external ^{195}Au source. These measurements were carried out without an absorber as well as with Sn absorbers of 1.898 and 2.665 g/cm^2. Typical runs with and without absorber are shown in fig. 4. The relative efficiency of the Ge(Li) detector has been measured in each experimental set-up by means of NBS calibration sources ^{241}Am, ^{57}Co, ^{113}Sn and ^{203}Hg.

3.2. ANALYSES

3.2.1. The decay to the ground, 99 keV and 130 keV states. The electron capture branching ratios to the ground state and to the first and second excited states of ^{195}Pt have been calculated from the summation spectra obtained with the 4π internal source spectrometer. The decay to the ground state has been determined from the areas of the K- and L-peaks. The contribution from the M- and higher-order electron capture is taken into account by the theoretical M+.../L capture ratio of Suslov [4], *viz.* M+.../L = 0.340. The decay to the 99 and 130 keV levels has been determined in the same way from the areas of the corresponding K-, L- and M+...-peaks.

TABLE 3
Experimental electron capture branching ratios

Author	Ref.	ε_0 (%)	ε_{99} (%)	ε_{130} (%)	ε_{200} (%)	ε_{211} (%)	P_{co}^{130} (%)
De-Shalit *et al.*	[2]		≈ 65	≈ 35			≈ 10
Bisi *et al.*	[7,8]		57, ≈ 50	43, ≈ 50			
Goedbloed *et al.*	[10]	0 $\pm$ 6	59 $\pm$ 4	41 $\pm$4		0.05	4.9$\pm$1.5
Harris *et al.*	[12]	13 $\pm$15	47 $\pm$10	40 $\pm$6		< 0.4	5.2$\pm$1.0
Jasinski and Herrlander	[13]		63	37			6.8
Schöneberg *et al.*	[14]	0 $\pm$ 3	70 $\pm$ 5	30 $\pm$3	0.019	0.025	7.9$\pm$0.5
Fink and Benczer-Koller	[18]	12 $\pm$14	50 $\pm$12	38 $\pm$6		< 0.4	6.0$\pm$0.8
Toburen and Albridge	[19]	< 5	68 $\pm$ 3	32 $\pm$1		0.035$\pm$0.010	5.5
Ahlesten and Bäcklin	[20]	10 $\pm$13	58 $\pm$ 8	32 $\pm$5	0.019	0.025	7.4
Lederer *et al.*	[6]		58	41		0.5	47
present work		9.5$\pm$0.4	62.1$\pm$2.7	28.4$\pm$1.6	0.025$\pm$0.001	0.032$\pm$0.001	11.1$\pm$0.2

3.2.2. The decay to the 200 and 211 keV states. The electron capture branching ratios to the 200 and 211 keV levels in ^{195}Pt have been calculated from the results obtained with the Ge(Li) detector. In order to calculate the ratios between the de-excitation of the 99 and 130 keV levels, and the 200 and 211 keV levels to the ground state, the total internal conversion coefficients of Hager and Seltzer [23] are used. The multipolarities of the transitions are taken from refs. [18,19,20] and refs. [14,24]. The theoretical coefficients are $\alpha^{99} = 6.96$, $\alpha^{130} = 1.72$ and $\alpha^{200} = 0.73$, $\alpha^{211} = 0.71$.

3.2.3. The cross-over decay probability of the 130 keV state. It is possible to calculate the cross-over decay probability of the 130 keV state to the ground state P_{co}^{130}, as a result of the two independent methods used to measure the electron capture branching ratios of the 99 and 130 keV levels. The formulae used are

$$I_{99}(1+\alpha^{99}) = \varepsilon_{99} + (1-P_{co}^{130})\varepsilon_{130}, \qquad I_{130}(1+\alpha^{130}) = \varepsilon_{130}P_{co}^{130},$$

in which I_E is the intensity of the γ-radiation of energy E (keV) measured with the Ge(Li) detector (see subsect. 3.2.2). It is normalized to the total decay. Further, ε_E is the electron capture branching ratio to the level at E keV, measured with the 4π internal source scintillation spectrometer (see subsect. 3.2.1).

Using the conversion coefficients from Hager and Seltzer [23]), the only unknown in these two equations is P_{co}^{130}. The results of the P_{co}^{130} calculations making use of the two different equations are the same. The results from experiments with Sn absorbers also agree with those without absorbers.

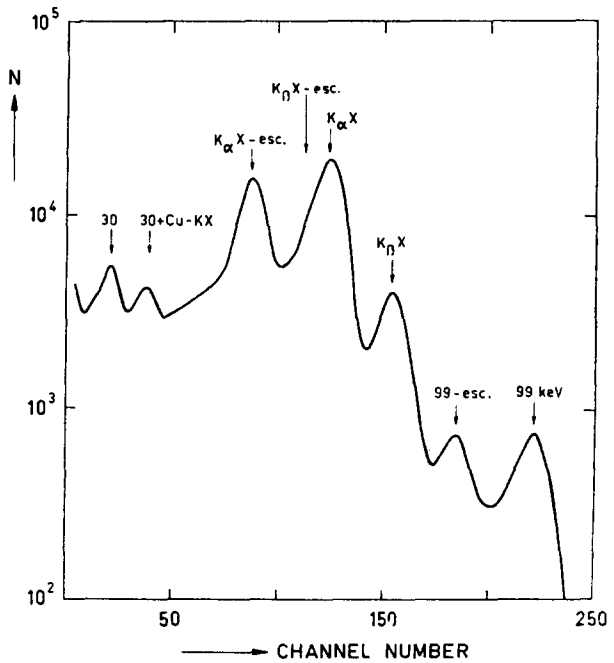

Fig. 5. Spectrum of ^{195}Au measured by means of a krypton-methane counter, taken from Goedbloed *et al.* [10]).

3.3. RESULTS AND DISCUSSION

3.3.1. The electron capture branching ratios. The electron capture branching ratios are given in table 3. Every branching ratio has been measured, including the branching ratio to the ground state of ^{195}Pt. The difference in the ground state branching ratios has to be taken into account in order to make a comparison with other authors. Agreement exists with Schöneberg *et al.* [14]), Toburen *et al.* [19]) and Ahlesten *et al.* [20]).

The 200 keV level is not observed or only weakly observed in Coulomb excitation experiments and is not observed in most decay experiments [10, 12, 18–20]). The level is neither predicted in core excitation theory [25]) nor in the asymmetric rotator model [ref. [26])]. The level has been observed by Schöneberg [14]) and is confirmed in the present work.

3.3.2. The cross-over decay probability of the 130 keV state. The value of the P_{co}^{130} is compared with other experimental values in table 3. A large discrepancy exists between these values [10,12-14,18-20]) and ours. An exception is the value of De-Shalit *et al.* [2]). From all these references only De-Shalit [2]), Goedbloed [10]), Harris [12]) and Schöneberg [14]) have measured P_{co}^{130} themselves. The other authors made use of their work. Coulomb excitation experiments often refer to McGowan *et al.* [24]). Their value has been taken from De-Shalit [2]). The determination of this value was not very accurate. Harris *et al.* [12]) determined the 31 keV γ-intensity to be $(8.2\pm0.7)\,\%$ of the 35 keV I-escape peak of the Pt K X-ray. The product of efficiency and solid angle used in their calculations was taken from calculated tables. Schöneberg *et al.* [14]) determined P_{co}^{130} from the decay of ^{195m}Pt. Goedbloed *et al.* [10]) measured P_{co}^{130} by means of a proportional counter. A spectrum from their work is given in fig. 5. The 31 keV γ-ray intensity was determined by addition of the 31 and 35 keV peaks. The 35 keV peak was explained to be the summation of the 31 keV γ-rays with copper K X-radiation ($\approx$ 8 keV).

In this investigation the experiments of ref. [10]) with copper absorbers were reproduced making use of a Si(Li) X-ray detector (Philips BPX 57, useful volume 3 mm $\times$ 100 mm^2, Be window 250 μm). No evidence for the 35 keV peak was found at all. The cross-over value P_{co}^{130} of Goedbloed has been re-calculated taking only the volume of the 31 keV peak into account. This results in $P_{co}^{130} = (9.3\pm2.8)\,\%$, in agreement with our value. The total internal conversion coefficient of the 99 keV transition was calculated in the work of Goedbloed using their P_{co}^{130} value. They found $\alpha^{99} = 9.9\pm1.0$, in agreement with De-Shalit [2]). Using the new value of P_{co}^{130} this coefficient becomes $\alpha^{99} = 6.6\pm0.7$, in agreement with the theoretical value [23]) and with other experimental values [12,18,24]).

Another piece of evidence for the P_{co}^{130} value calculated in this work can be found in the half-life of the 130 keV level together with Coulomb excitation data. The only independent half-life measurements are from Salling, *viz.* $T_{\frac{1}{2}} = 620\pm30$ ps [private communication to Grodzins *et al.* [27])], and from Bloess *et al.* [28]), *viz.* $T_{\frac{1}{2}} = 620\pm70$ ps. Half-lives deduced from Mössbauer experiments [29-32]) support this value. The B(E2) value of the 130 keV level was measured in several Coulomb excitation experiments [14,24,27,33]). The mean B(E2) value is $2.3\times10^3\ e^2\cdot\mathrm{fm}^4$. This value, together with the half-life and the theoretical α^{130}, results [18]) in a P_{co}^{130} of about 11.5 %, in perfect agreement with the value calculated in this investigation.

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