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Anomalous conversion in the decay of ^{197m}Hg

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Abstract. The total and *K*-shell conversion coefficients of the 165 keV transition in the decay of ^{197m}Hg are determined from intensity balance considerations and a coincidence technique using a Ge(Li) — NaI (Tl) system respectively. The resultant values are $\alpha_T = 274 \cdot 8 \pm 19 \cdot 2$ and $\alpha_K = 47 \pm 12$, while the corresponding theoretical values are 344 and 77 respectively, indicating anomalous conversion. The gamma ray transition probability however, shows a hindrance of only about 6 and cannot be correlated with the present anomalous conversion data. The *K*/*L* ratio of the 130 keV transition, determined using a summing method with a Ge (Li) detector, yielded $0 \cdot 090 \pm 0 \cdot 012$, while the corresponding theoretical value is $0 \cdot 048$, indicating anomalous conversion. The corresponding gamma transition probability shows a hindrance of about 3000, in correlation with anomalous conversion.

Keywords. Internal conversion coefficients; high multipole transitions.

1. Introduction

The agreement between the theoretical and experimental values of the internal conversion coefficients for high multipole transitions was found to be unsatisfactory by Raman *et al* (1972). They found that the experimental values were consistently lower than the theoretical values. Campbell and Martin (1975) reviewed the position more recently and examined the implication of using different types of potentials for the evaluation of the conversion coefficients. They predicted that discrepancies up to about 30% would arise between the theory and experiment for high multipole transitions near threshold in the light element region. It is therefore of interest to undertake systematic studies on high multipole transitions. As a part of this study, the conversion coefficients of the 165 and 130 keV transitions are determined in the decay of ^{197m}Hg. The conversion coefficient of the 165 keV transition is also of interest inasmuch as the 165–134 keV cascade is being used in hyperfine interaction studies. Krien *et al* (1973) recently measured the magnetic hyperfine fields at Hg nuclei in Fe, Ce, and Ni hosts, employing $e - \gamma$ time differential perturbed angular correlation techniques.

Most of the early information on the decay of the radioactive isotope ^{197m}Hg is summarized in the Nuclear Data Sheets by Lewis (1972) and a decay scheme

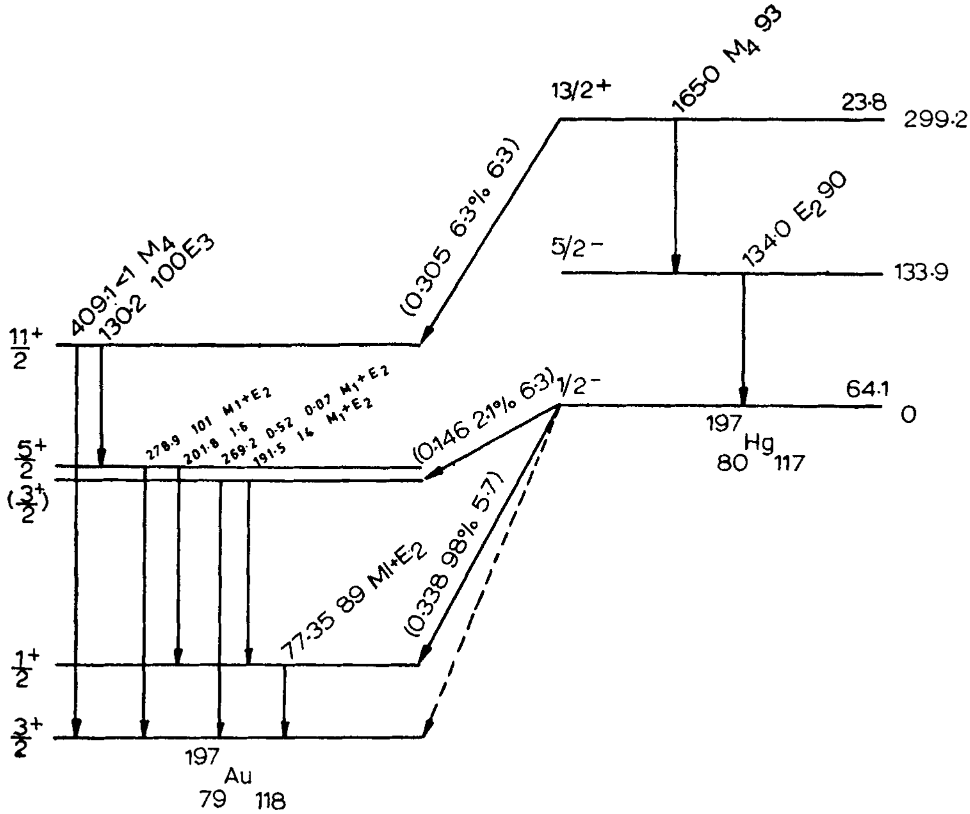

Figure 1. Decay scheme of the radioactive isotope ^{197m}Hg.

is shown in figure 1. Huber *et al* (1951) were the first to determine and *K*-conversion coefficient of the 165 keV transition. But they obtained a value of 4.6 which is very much smaller than the theoretical value expected for an M4 transition (α_k^{theo} (M4) = 77). Coburn *et al* (1957) obtained the total conversion coefficient of the 165 keV transition to be 350 ± 90, consistent with that expected for an M4 transition. The present study is aimed at a direct determination of the *K*-shell conversion coefficient with a better accuracy.

The total as well as the *K*-Shell conversion coefficients of the 130 keV E3 transition were determined by Smend *et al* (1966) by intensity balance considerations. They obtained the values of the *K* and total conversion coefficients to be 0.9 ± 0·3 and 28 ± 4 respectively. The *L*-subshell ratios for this transition were determined by Plajner *et al* (1970) and were found to be consistent with an E3 assignment for this transition. The *K*/*L* ratio, however, is noticed to deviate from the theoretical value by a factor of 2. A redetermination of the *K*/*L* ratio is therefore carried out in the present study.

2. Method

The total conversion coefficient of the 165 keV transition is obtained from intensity balance considerations

$$N_\gamma^{165}\,(1+\alpha_T^{165}) = N_\gamma^{134}\,(1+\alpha_T^{134}) \qquad (1)$$

employing the gamma intensities recorded with a Ge (Li) spectrometer and assuming the total conversion coefficient of the 134 keV transition which is established to be E2 (figure 1). In principle, this method is capable of high accuracy inasmuch as the errors arise in the statistics of counting of gamma rays and the accuracy of the total conversion coefficient of the 134 keV transition can be fairly high. Although a 1% statistical accuracy is usually realisable in counting, the actual errors in the count rates are of the order of 3% due to the uncertainties in photopeak efficiency calibration. The theoretical conversion coefficients for E2 transitions are known to accuracies of the order of nearly 2%. This method is therefore capable of about 4% accuracy. In the present case however, the statistical accuracy in single gamma counting of the 165 keV line is only of the order of 5% for reasons to be explained in section 4.

The K-conversion coefficient of the 165 keV transition is obtained by a coincidence method using a Ge (Li)–NaI (Tl) coincidence system. The spectrum recorded with 134 keV selected in the gate shows peaks corresponding to the 165 keV gamma rays and k-x-rays following the K-conversion of the 165 keV transition. The relative intensities under these two peaks corrected for the relative efficiencies of the detection yield the K-conversion coefficient using the relation

$$\alpha_k\,(165\ \text{keV}) = \frac{N_x}{N_\gamma} \cdot \frac{\epsilon_\gamma}{\epsilon_x} \cdot \frac{1}{\omega_k} \tag{2}$$

where N_x and N_γ are the relative intensities under the x-ray and gamma peaks in the coincidence spectrum, ϵ_x and ϵ_γ are the corresponding relative photopeak efficiencies and ω_k is the K-shell fluorescent yield. The accuracy of this method is determined by statistics of counting in N_x and N_γ and the errors in photopeak efficiency calibration and florescent yield. Usually, 1% statistical accuracy in counting can be achieved, the other errors being of the order of 3%. An overall accuracy of about 5% is realisable by this method. In the present case, however, due to high conversion, gamma intensity of the 165 keV transition is small and the statistical accuracy in coincidence counting better than 20% could not be achieved.

The summing method described by Mukherjee and Das Mahapatra (1974) is employed to determine the K/L ratio of the 130 keV transition. Since K and L x-rays are produced following the conversion of the 130 keV transition, and they occur in coincidence with the following 279 keV transition, the sum peaks are produced in a close geometry. The intensities under these sum lines are employed for the estimation of the K/L ratio using the relation

$$K/L\ (130\ \text{keV}) = \frac{\epsilon_L}{\epsilon_{k\alpha}} \cdot \frac{N_{279+k\alpha}}{K_{279+L}} \cdot \frac{(\omega_L + \omega_{kL})}{f_{k\alpha} \cdot \omega_k} \tag{3}$$

where ϵ's are the relative photopeak efficiencies, N's are the intensities under the sum lines and ω's are the fluorescent yields and $f\,K_\alpha$ is the fraction of the K_α x-rays. The accuracy of the determination of the K/L ratio by this method is mainly decided by the error in the L-fluorescent yield which is of the order of 10%. An overall accuracy of about 15% is realisable in the determination of the K/L ratio.

3. Experimental details

The radioactive source ^{197m}Hg is obtained from the Isotope Division of BARC as $Hg(NO_3)_2$ with a specific activity of 50 mci for gm Hg. The source contained ^{203}Hg as an impurity. But since the decay of ^{203}Hg is well known, its interference could be accounted for. A few drops of the source liquid dried over a mylar foil over a persphex frame are employed for this study.

The isomeric state ^{197m}Hg decays with a half life of 24 hrs. Thus the spectra recorded within 3 or 4 days after producing the source are useful for the investigation of the transitions associated with the isomeric decay. The source radiation after 5 days is that associated with the ground state decay ^{197}Hg (65 hrs). After about 2 weeks, the source radiation is essentially that of ^{203}Hg.

A coaxial 35 cc Ge (Li) detector is employed for the estimation of the single intensities. The output from the detector is passed on for pulse height analysis to a ND 512 channel analyser. The cryostat is of a horizontal type with the Ge (Li) detector situated at 7 mm from the aluminium end casing of the cryostat. The source is directly placed on the casing to record the spectra corresponding to the "close geometry" for the summing method. The efficiency calibration is also carried out in the same geometry using standard radioactive sources ^{152}Eu, ^{133}Ba, ^{57}Co and ^{75}Se sources.

The coincidence system consists of the 35 cc Ge(Li) detector and a $1\frac{3}{4}'' \times 2''$ NaI (Tl) mounted on a DuMont 6292 photomultiplier tube. The NaI (Tl) crystal is placed at a distance of 1 cm from the source mounted on the cryostat of Ge (Li) detector as mentioned above. The detectors are arranged in a conventional slow-fast coincidence system with an effective resolving time of about 30 ns. The coincidence spectrum gated by a selected gamma or x-ray in one of the channels could be recorded on the multichannel analyser.

4. Results and Discussion

4.1. *Total conversion coefficient of the 165 keV transition*

The single spectrum recorded on the Ge (Li) detector with the ^{197m}Hg source is shown in figure 2. The K_α and K_β lines represent the *x*-rays of both Hg and Au. Since the electron capture decay of the ^{197m}Hg is only about 6% and the conversion of the 130 keV transition is high, the intensity under the 134 keV line has a negligible interference from the 130 keV transition. The 165 keV gamma line is of a low intensity because of the high conversion and the line is shown on the expanded scale in the inset of the figure. The 279 keV line includes the contribution from the impurity ^{203}Hg. Since the spectrum is recorded in the close geometry, sum lines as indicated in the figure are recorded.

The intensities under the 134 keV and the 165 keV lines are estimated by a graphical analysis and are employed in relation 1 to obtain the total conversion coefficient of the 165 keV transition as shown in table 1.

An overall error of about 7% is involved in the present result and is contributed mostly by the statistics of the counting associated with the 165 keV line, the error

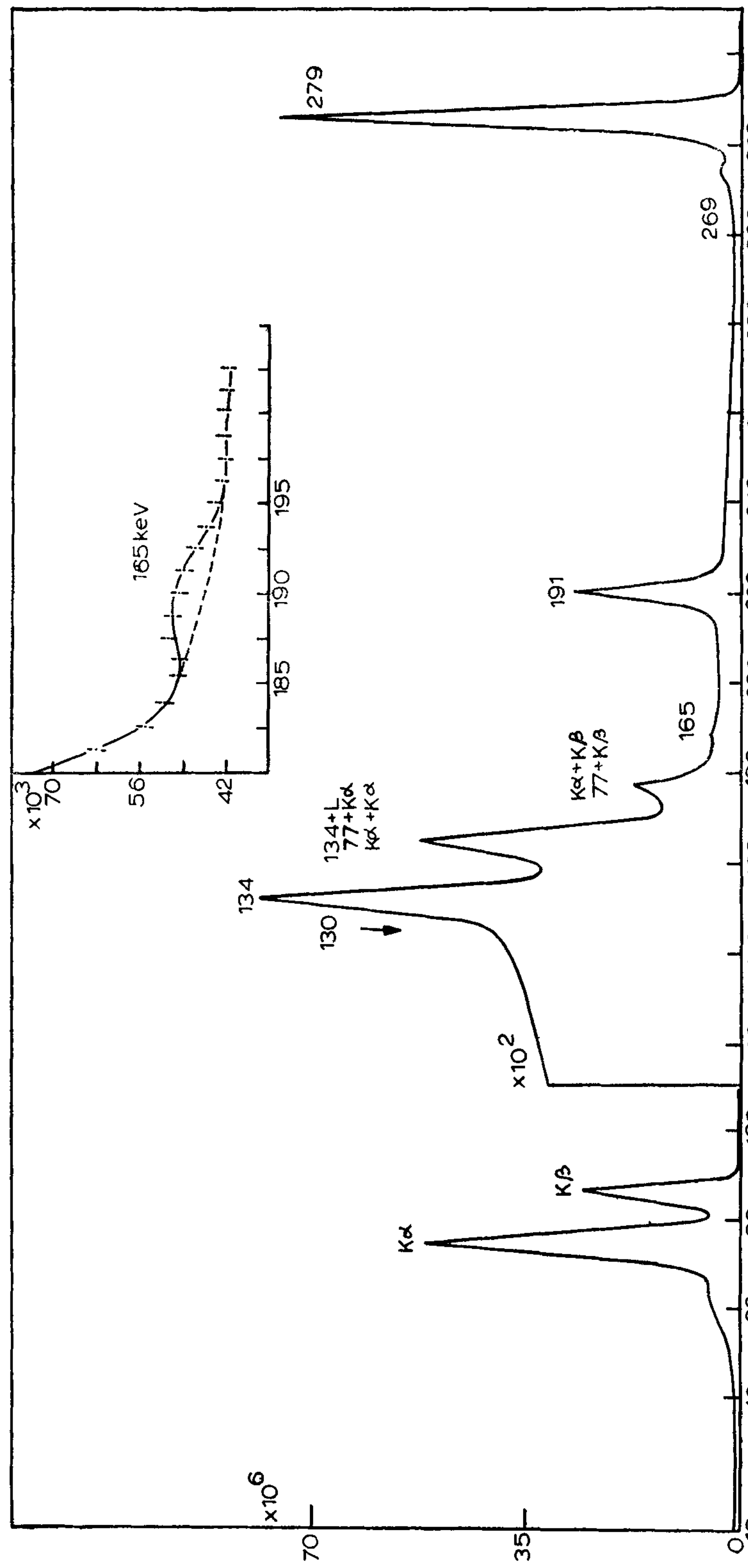

Figure 2. Single spectrum recorded with the Ge(Li) detector in close geometry (The inset shows 165 keV line on an expanded scale).

Table 1. Total conversion coefficient of the 165 keV transition in ^{197}Hg

Description	Value
N_{165}/ϵ_{165}	$0 \cdot 99 \pm 0 \cdot 065$
N_{134}/ϵ_{134}	100 ± 1
α_T (134 keV) *	$1 \cdot 73$
α_T (165 keV)	
Present Value	$274 \cdot 8 \pm 19 \cdot 2$
Coburn *et al* (1957)	350 ± 90
Theory* (M4)	344

*Theoretical value from Hager and Seltzer's data.

in the relative photopeak effiiciencies being only about 3%. The present result agrees with that of Coburn *et al* (1957). The value of Coburn *et al* is associated with a large error, while the present value is much more accurate. The theoretical value of α_T for an M4 transition of energy 165 keV is obtained by the usual computer interpolation programme of Hager and Seltzer (1968), which yielded $\alpha_T = 344$. Thus the present value of the total conversion coefficient is smaller than the theoretical value by $(20 \pm 1 \cdot 5)\%$.

4.2. *K-conversion coefficient of the* 165 *keV transition*

The *K*-conversion coefficient of the 165 keV transition is obtained from the relative gamma and *x*-ray intensities recorded in coincidence with the 134 keV gamma ray detected in the NaI (Tl) counter, which gated the Ge (Li) detector spectrum recorded on the multichannel analyser. The resultant spectrum is shown in figure 3. The intensities under the lines obtained by the graphical analysis are employed in relation 2 to determine the *K*-conversion coefficient, together with other parameters as shown in table 2.

An overall error of about 25% is involved in this measurement and is mostly contributed by the statistics of counting associated with the 165 keV line (figure 3). The source half life is only 24 hrs, and the transit time was about 48 hrs. Hence useful counting time was always restricted to about 48 hrs, although the experiment was repeated with three samples. In addition, the specific activity of the source was rather low. The resolving time was about 30 ns and high source intensities could not be used. It is also undesirable from the absorption effects for the *x*-rays. The present experimental value is lower than the result derived from the relative conversion electron and gamma intensities, as reported in Nuclear Data Sheets ($\alpha_k^{165} = 66$) although assuming a reasonable error in that value gives an overlap with the present result. An underestimation of the *x*-ray intensity in the present work due to lower coincidence efficiency at x-ray energies, is ruled out based on an auxiliary experiment with a ^{133}Ba source where the x-ray energy

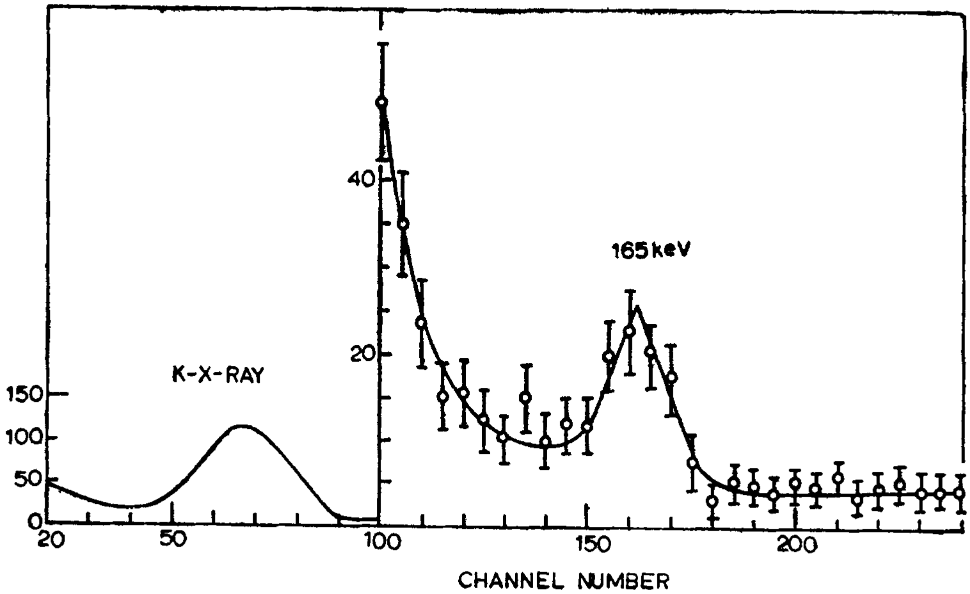

Figure 3. Coincidence spectrum in the Ge(Li) detector with 134 keV selected in NaI(Tl) gate.

Table 2. *K*-conversion coefficient of the 165 keV transition in ^{197}Hg

Description	Value
N_x/ϵ_x	45·8±9
N_{165}/ϵ_{165}	1·0±0·18
ω_k*	0·966±0·02
α_k (165 keV)	
Present Value	47±12
Huber *et al* (1951)	4·6
Nuclear Data Sheets**	66
Theory (M4) Hager and Seltzer	77

*Bambyneck *et al* (1972).
**Derived from relative electron and gamma intensities.

is about 30 keV. The theoretical value of the *K*-conversion coefficient for the 165 keV M4 transition estimated from Hager and Seltzer's data to be 77. The present experimental value is therefore smaller than the theoretical value by (39 ± 10)%.

It therefore appears that the 165 keV transition has an anomalous conversion. The transition is of an established M4 character ($13/2^+ \rightarrow 5/2^-$). The $5/2^-$ state may be viewed by as an $f_{5/2}$ single particle state, while the $13/2^+$ state may be a single particle $i_{13/2}$ configuration. The observed anomaly cannot be explained with this classification. The gamma ray transition probability of the 165 keV transition is estimated after correcting the half life for the electron capture branch and employing the present value of the total conversion coefficient. The resultant value yielded $(2 \cdot 65 \pm 0 \cdot 17) \times 10^{-8}$ transitions per sec, while the corresponding single particle estimate (Moszkowski) is $1 \cdot 69 \times 10^{-7}$ transitions per sec. Thus a hindrance of only about 6 is noticed, suggesting that the assumed classification of the states may represent large parts of the wave functions. Thus while, the internal conversion of the 165 keV transition is anomalous, the gamma transition probability appears to be normal.

4.3. *K/L ratio of the* 130 *keV transition*

The K/L ratio of the 130 keV transition $(11/2^- \rightarrow 5/2^+)$ is estimated from the 279 + K_α and 279 + L peaks as shown in figure 4. The intensities under the peaks $N_{279+k\alpha}$ and N_{279+L} obtained by graphical analysis are employed to estimate the K/L ratio using the relation 4 together with other quantities as shown in table 3.

An overall error of about 13% is involved in the determination of K/L, of which 6% is contributed by statistics of counting and 2% through the K-Shell fluorescent yield, 10% through the L-shell fluorescent yield, 5% through the ω_{kL} and 3% through the relative photopeak efficiencies. The theoretical value of K/L for an E3 transition of energy 130 keV is significantly smaller than the present result as shown in table 3. The two earlier results as furnished in the Nuclear Data Sheets are also

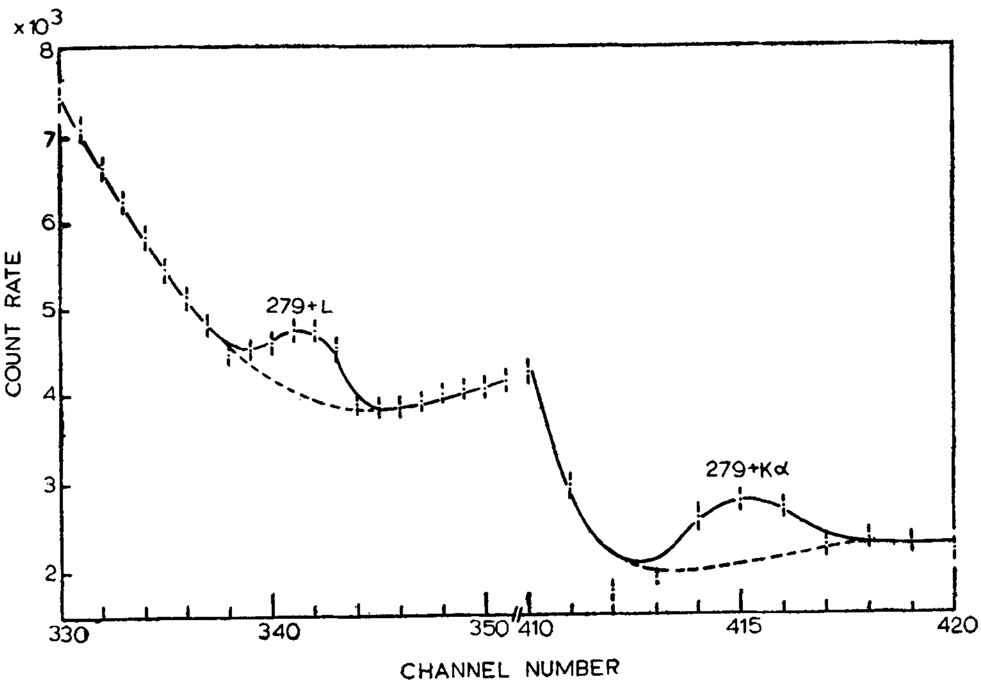

Figure 4. Part of the Ge(Li) spectrum showing 279 + L and 279 + $K\alpha$ lines recorded in close geometry.

Table 3. K/L ratio of the 130 keV transition in ^{197}Au.

Description	Value
$N_{279+K\alpha}/\epsilon_{k\alpha}$	$9\cdot73\pm0\cdot4$
N_{279+L}/ϵ_L	100 ± 4
ω_k (Au)*	$0\cdot964\pm0\cdot017$
ω_L (Au)*	$0\cdot430\pm0\cdot04$
ω_{kL} (Au)*	$0\cdot287\pm0\cdot012$
$f_{K\alpha}$	$0\cdot810\pm0\cdot016$
K/L ratio (130 keV)	
Present value	$0\cdot09\pm0\cdot012$
Earlier results (Nuclear Data Sheets)	0·13 0·11
Theory (E3)**	0·048

*Bambyneck *et al* (1972)
**Interpolated from the data of Hager and Seltzer (1968).

shown in table 3, which are close to the present experimental value. The earlier experimental result of the K-shell conversion coefficient of Smend *et al* (1966) is $0\cdot9 \pm 0\cdot3$ while the theoretical value expected for an E3 transition is 1·02. In view of a large error in the experimental result, it is not possible to say whether there is a real discrepancy between theory and the experiment in the K-shell conversion coefficient. L-subshell ratios of this transition, as determined by Plajner *et al* (1970) however, indicated agreement with the theoretical values. It is therefore likely that the absolute value of the L-shell conversion coefficient and/or the K-shell conversion coefficient show an anomalous behaviour and it is of interest to determine the absolute values accurately.

Assuming the half life of the 409 keV state, the K-conversion coefficient, the present K/L and earlier L/M ratios, the gamma transition probability of the 130 keV transition is estimated to be $2\cdot85 \times 10^{-4}$ transitions per sec, while the corresponding single particle (Moszkowski) estimate is $8\cdot37 \times 10^{-1}$ transitions per sec. Thus a hindrance of about 3000 is observed. Such a hindrance is indicative of the differences in the structures of the connecting states and an anomalous conversion is expected in this case.

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