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SHIFTS IN THE ENERGIES OF GOLD K X-RAYS
AND THE ROLE OF THE EXCITATION MECHANISM

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ABSTRACT

The energies of the gold $K_{\alpha 1}$, $K_{\alpha 2}$ and $K_{\beta 1}$ X-ray lines are larger by 568, 567 and 620 milli-electronvolts, respectively, than those from electron-capture beta decay of $64\text{ h }^{197}\text{Hg}$. It appears likely that these shifts are caused in part by unresolved satellite lines arising from the photo-ionization process.

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The large natural widths of atomic K X-rays conceal a complicated fine structure. In the present work we draw attention to the contribution that must come from unresolved satellite lines associated with outer-shell excitations caused by the primary excitation mechanism. This effect, illustrated in Fig. 1, occurs superimposed on contributions from couplings to electrons in unfilled shells, known to lead to asymmetric lines in light- and medium-weight elements, and on a contribution from the coupling to the nuclear spin (hfs, magnetic hyperfine splitting). Energy shifts from the latter effect have recently been observed [1] in electron-capture beta decay (EC), for which the beta-decay intensity rules lead to a non-statistical population of the hyperfine levels, so that the shifts become directly observable. Here, we exploit another peculiar feature of K-shell excitation in EC beta decay, namely the near-absence of outer-shell excitations during the formation and decay of the $1s_{1/2}$ hole. The reason for this is the almost complete screening of the nuclear charge by the $1s$ electrons, so that the simultaneous disappearance of a proton and of a $1s$ electron leaves the wave functions of the outer electrons essentially unchanged. Thus, in our approach, a comparison of energies of K X-rays from EC beta decay and from photo-ionization (PI) should bring out the contribution made by unresolved^{*)} satellite lines, provided, of course, that the hfs shift is taken into account.

A first attempt to observe effects of the excitation mechanism was made by Hahn et al. [2], who compared the energies of tantalum K X-rays from EC and PI sources consisting of oxides of tungsten and tantalum. The difficulties [1] in interpreting this experiment, to which we shall return at the end of this paper, have stimulated us to investigate another case using metallic sources of isotope-separated radioactivity. The effect of the satellite lines varies slowly with Z , while the hyperfine effect grows rapidly with Z ; consequently, a medium-heavy element would be preferable. However, the high intensities of the radioactive

*) Resolved satellite lines ("non-diagram lines") are well known in X-ray spectroscopy; the existence of components inside the line width is evident, although their contribution to K X-ray energies does not seem to have been discussed before.

beams of mercury at the ISOLDE facility at CERN made an experiment with gold ($Z = 79$) attractive, and have permitted us to reach a statistical precision of 3×10^{-4} of the natural line width for $K_{\alpha 1}$.

The radioactive isotope ^{197}Hg (64 h), produced in the ISOLDE on-line mass separator, was implanted in an aluminium backing in a spot of a few mm^2 . The K X-rays emitted from this radioactivity originate predominantly in the $(\frac{1}{2}^- \rightarrow \frac{1}{2}^+)$ EC transition to the 77.3 keV level, which is below the K threshold so that only 0.7% of the K X-rays result from internal conversion in gold. The photo-ionization source consisted of a 0.05 mm gold foil supported on aluminium placed at distance of 0.5 mm from a 40 Ci ^{169}Yb source. The energies of the $K_{\alpha 1}$, $K_{\alpha 2}$ and $K_{\beta 1}$ X-rays from the two sources were compared by scanning their Bragg reflections in second order with a bent-crystal spectrometer of 4.64 m focal length. The technique for data taking and analysis was outlined previously [1]; new, additional features are a shutter, that makes it possible to adjust the counting time on each source to an optimum, and a micrometer screw for aligning the two sources in the direction of the dispersion. The instrumental asymmetry was lower than in the previous experiments [1] and was found to be $0.93 \times 10^{-6} E$ for a transition of energy E .

The results corrected for asymmetry are given in column 3 of Table 1. The analysis is complicated slightly by the presence of the 24 h $^{197\text{m}}\text{Hg}$ together with the 64 h ^{197}Hg . The Hg $K_{\alpha 2}$ X-ray line from the isomeric transition is so close to the Au $K_{\alpha 1}$ line that its tail gives rise to an additional shift ϵ_{Hg} , which can be calculated (column 5 of Table 1) from the known intensity and shape of the Hg X-ray reflection. A measurement of the Au $K_{\alpha 1}$ line after the 24 h activity had died away (last line in Table 1) confirms that this correction is precise enough. The short-lived isomer furthermore contributes to the hyperfine shift (column 4 of Table 1) through a weak branch to the $\frac{1}{2}^-$ level in ^{197}Au , which was assumed to have $\mu/\mu_N = 6.3$.

The contributions from magnetic hfs can be calculated from the expressions given in Ref. 1. As there is no contribution for X-rays excited in photo-ionization, because of the statistical population of the hyperfine levels, it is

only necessary to consider the electron-capture decays. For the gold K X-rays from the 64 h ^{137}Hg decay, standard data on the decay scheme [3] and magnetic moments [4] yield an increase in the X-ray energy of $-6 - 392(1 + y^2)^{-1}$ meV, where the second term comes from the dominant $\frac{1}{2}^- \rightarrow \frac{1}{2}^+$ (77.4 keV, 94%) EC transition, and where y^2 is its intensity ratio of multipole order zero to multipole order 1. As y is experimentally unknown, it can only be obtained via a nuclear-structure argument. It seems plausible that the dominant beta strength in the $^{197}\text{Hg}(\frac{1}{2}^-) \rightarrow ^{197}\text{Au}(\frac{1}{2}^+)$ transition comes from the $p_{1/2} \rightarrow s_{1/2}$ decay, which, from an analysis of the ^{207}Tl decay [5], is known to have $y^2 \approx 0.05$, the value adopted here. (Note that we are *not* assuming that the states in ^{197}Hg and ^{197}Au are well represented by pure $p_{1/2}$ and $s_{1/2}$ configurations; the low magnetic moment of the $\frac{1}{2}^+$ level [6] as well as the high log ft value clearly point to a more complex structure.) The total magnetic hfs contribution including that of the 24 h activity is given in Table 1. Note that although the magnetic moment of the $\frac{1}{2}^+$ state is known to 1%, the conclusions remain sensitive to the value of y^2 .

The atomic excitation processes are here considered as instantaneous. The K X-rays emitted on a time scale of 10^{-17} s will not be influenced by subsequent atomic rearrangements. The intensities of the satellites, expected to be different for the PI and the EC processes, may be obtained in the sudden approximation. Calculations of vacancy probabilities have been performed by Nestor et al. [7] with single-electron wave functions (Hartree-Fock-Slater) for certain elements and for different excitation processes: β^- -decay, photo-ionization, and electron-capture beta-decay.

The satellite energies, which cannot be obtained in sufficient detail from Nestor's calculation, were calculated separately. This was done in the relativistic Hartree-Fock (Dirac-Fock) scheme, using the programme of Lindgren and Rosén [8]. In the first approximation "frozen orbitals" can be used. Then, for example, the shift in the K_{β_1} line ($3p_{3/2} \rightarrow 1s_{3/2}$) due to a 4s hole is

$$\Delta E(K_{\beta_1}, 4s^{-1}) = \langle 1s \ 4s \ |R| \ 1s \ 4s \rangle - \langle 3p_{3/2} \ 4s \ |R| \ 3p_{3/2} \ 4s \rangle ,$$

where the matrix elements represent the Coulomb interaction with exchange. Somewhat higher accuracy is achieved, if self-consistent wave functions are used for the different hole states, which means that the relaxation effect is taken into account. This was done for the innermost holes, where this effect is significant.

It is clear from Fig. 1 that the satellite lines give rise to an asymmetric line shape; in order to convert this asymmetry into an equivalent energy shift it is necessary to take into account the definition of the location parameter, which we calculate here [1,9] from the centre of gravity of a line truncated from $-a$ to $+a$ around the assumed position, here as in Fig. 1 assumed to be the origin. An unbiased estimate of the true line position is to lowest order $x = N_1/(N_0 \cdot f)$, where N_1 and N_0 represent the first and zeroth moment of the measured intensity and f is the bias correction [9]:

$$f = 1 - 2aI(a) / \int_{-a}^a I(x) dx . \quad (1)$$

With this definition it is easy for a given X-ray transition ν to convert the satellite intensities and energies into an effective energy shift ϵ_{ose}^ν . Calculated values for gold and tantalum are given in Table 2, which also gives a comparison with the measured shifts after reduction by the known contributions from hyperfine shifts, the Hg $K_{\alpha 2}$ impurity line, and the isomer shift calculated from Mössbauer effect data [10].

It is seen from Table 2 that the measured shifts for tantalum are surprisingly large. The authors of [2] explain this as a selective shake-off of 4f electrons in the electron-capture process; as discussed in [1] it is hard to see why such a process is to be expected, especially as electron capture favours s-electron shake-off. Chemical shifts could possibly play a role; although the $K_{\alpha 1}$ shifts Ta (metal) - Ta_2O_5 and W (metal) - WO_3 are both 110 meV [11], so that the data should be equivalent to comparing metal samples, it is possible that the W atom following neutron capture is in a very different chemical state. The gold experiment, on the other hand, compares metallic samples (Au metal with Hg implanted in aluminium), so that chemical shifts are expected to be small.

Our experiments on gold K X-rays show that in addition to the hyperfine shift [1] one finds experimentally a ~ 200 meV shift which we tentatively interpret as the contribution from satellite lines originating in the photo-ionization process. The agreement with a simple calculation is excellent for $K_{\alpha 1}$ and $K_{\alpha 2}$, while less good for $K_{\beta 1}$ (Table 2). We note in this connection that the $K_{\beta 1}$ contribution comes predominantly from the outer electrons, for which the sudden approximation is less precise.

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Table 1

Shifts $\Delta E = E_{PI} - E_{EC}$ in the
energies of gold K X-rays

Line	Time after collection [h] a)	ΔE observed shift (meV)	Calculated contribution		ΔE_{corr} = $\Delta E - \epsilon_{hfs}$ - $\epsilon_{Hg} - \epsilon_{is}$ d)
			ϵ_{hfs} b)	ϵ_{Hg} c)	
$K_{\alpha 1}$	20	286 ± 42	330	-224	168 ± 42 e)
$K_{\alpha 1}$	47	413 ± 25	350	-130	181 ± 25
$K_{\beta 1}$	75	605 ± 52	362	-	231 ± 52
$K_{\beta 1}$	101	560 ± 73	371	-	177 ± 73
$K_{\beta 1}$	126	670 ± 76	373	-	285 ± 76
$K_{\alpha 2}$	159	569 ± 40	378	-	179 ± 40
$K_{\alpha 2}$	189	564 ± 57	378	-	174 ± 57
$K_{\alpha 1}$	238	568 ± 27	378	0	178 ± 27

- a) The time in hours from the end of the 78-hour collection to the mid-time of the measurement.
- b) Contribution from hfs (via E_{EC}). The time dependence is due to Au K X-rays associated with the decay of the 24 h $^{133}_{2}Pb$ isomer.
- c) Contribution to the Au $K_{\alpha 1}$ shift from the tail of the Hg $K_{\alpha 2}$ line from the 24 h isomer.
- d) The isomer shift $\epsilon_{is} = 12$ meV was calculated from Mössbauer effect data [10].
- e) Note that the experimental errors in this column and in column 3 are statistical errors only.

Table 2

Calculated contributions ϵ_{ose} from outer-shell excitations; comparison with experiment

Line	ϵ_{ose} ^{a)} (meV)	ΔE_{corr} (meV)			Ref.
		b)	c)	d)	
Au K _{α_1}	180	178	(± 17)	(± 36)	This work
Au K _{α_2}	180	177	(± 33)	(± 46)	
Au K _{β_1}	440	230	(± 37)	(± 50)	
Ta K _{α_1}	180	1080	e)	(± 130)	[2]
Ta K _{α_2}	180	1370	e)	(± 120)	
Ta K _{β_1}	450	2200	e)	(± 500)	

- a) Contribution calculated for a truncation interval of width $2a = 1.3 \Gamma$. The value given is essentially the increase in energy of the X-ray due to photo-ionization, the contribution from EC being 100 times smaller.
- b) Weighted mean (Table 1).
- c) Error due to counting statistics.
- d) Total error including a contribution from the asymmetry correction (± 15 meV) and from the bias correction ($\pm 0.05 \Delta E$).
- e) Hyperfine shift and isomer shift increase the ¹⁸¹W (EC) tantalum X-ray energy by $(400 + 12)$ meV.

Figure captions

Fig. 1 : Micro-structure of the gold ($Z = 79$) $K_{\alpha 1}$ X-ray ($2p_{3/2} \rightarrow 1s_{1/2}$) showing magnetic hfs and satellite lines due to outer-shell excitations (labelled by the state of the spectator hole). The satellite intensities are those given by Nestor et al. [7] for β^- decay and are except for the $N = 2$ shell, approximately the same as for photoionization. The energies of the satellite lines have been calculated by the method of Rosén and Lindgren [8]. The lower part of the figure shows the low-energy part in an expanded scale. The curves show for both energy scales the natural line shape corresponding to $\Gamma = 62.5$ eV. The diagram leaves out the hfs of the satellite lines. A similar structure exists for the X-rays from EC of mercury, but the outer-shell satellites are expected to be much less intense.

Fig. 1