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SHIFTS IN THE ENERGIES OF HOLMIUM K X-RAYS
AND THE ROLE OF ATOMIC STRUCTURE

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ABSTRACT

Beta decay by electron capture temporarily leaves an atom with atomic number Z in the configuration of the atom $(Z+1)$. Measurements on holmium, including for the first time the KO peak, show that this difference influences the K X-ray energies in a characteristic way; the effect is particularly large when inner shells are filling.

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This paper is the third step in an investigation of new effects influencing the energies of K X-rays. One mechanism peculiar to electron-capture (EC) decay involves a selection rule which leads to a non-statistical population of the atomic hyperfine levels. The resulting energy shifts, which were observed [1] by comparison of X-rays from Xe isotopes following EC decay of Cs, are calculable [1] if the beta-decay pattern and the magnetic moments are known. Another mechanism, which was demonstrated [2] by comparison of X-rays following EC with those following photo-ionization (PI), is the presence of unresolved satellite lines arising from outer-shell excitations, often called shake-off and shake-up. This effect is negligible in the EC decay but contributes typically tenths of an eV in the photo-ionization process [2]. A detailed comparison between the expected and observed [2] shifts of the K_{α_1} , K_{α_2} , and K_{β_1} lines suggests that the measured shifts cannot be explained quantitatively by shake-off and shake-up alone, so that other effects should also be considered.

For assessing such other effects, we consider in the present work the influence of the electronic configuration of the two elements involved as mother and daughter in the EC beta-decay process. The essential point is that the atomic lifetime of the 1s hole is so short (10^{-16} - 10^{-17} s) that the intermediate state to a good approximation should still have the outer-electron configuration of the parent atom. If the two atoms differ only in the presence (in the EC final state) of an additional electron in a state characterized by the quantum numbers $n\ell$, the effect of this electron on the K X-ray energies may be expected to be similar to the chemical shifts [3, 4] observed when an electron initially in the state $n\ell$ is engaged in the formation of an ionic chemical bond. The shift depends mainly on n and is largest for small values of n . The largest effects are therefore expected when inner shells are filling, and especially for the 4f (rare earth) and 5f (actinide) groups.

Hence the pair of elements erbium (Er, $Z = 68$) and holmium (Ho, $Z = 67$) are well suited for an experiment of this kind. These metals both have a *hcp* crystal lattice and differ by less than 1% in the lattice constants. The atoms are believed to differ only in the 4f occupations, $4f^{12}$ and $4f^{11}$, respectively. The radioactive isotope ^{165}Er ($T_{1/2} = 10.3$ h) decays by a single allowed beta branch ($5/2^- \rightarrow 7/2^-$) to the ground state of ^{165}Ho , which is the only constituent of natural holmium.

The shifts in the energies of holmium K X-rays from EC and PI sources were determined by a crystal diffraction technique used in the previous Hg-Au experiment [2] and to be described elsewhere [5]. The photo-ionization source was a foil of holmium metal excited by a strong source of ^{169}Yb (40 Ci). In order to check on possible solid-state effects, two ^{165}Er sources were used. One was a foil of reactor-irradiated erbium metal enriched to 73.6% in ^{164}Er , the other was 30 h ^{165}Tm , which decays into ^{165}Er , implanted at ISOLDE into aluminium as a 60 kV ion beam of 10 min ^{165}Yb [6]. The two sources gave essentially the same results (table 1).

Owing to the high intensities in the present experiment it was possible to obtain the shifts for the X-ray lines K_{α_1} , K_{α_2} , K_{β_1} , $K_{\beta_{24}}$, and $K\text{O}$. The origin and appearance of these lines is illustrated in fig. 1. The measured shifts, summarized in table 1 and displayed in fig. 2, are of a magnitude and show a pattern that are not explicable in terms of the two effects (hyperfine shift and shake-off) discussed in our previous experiments [1, 2]. We now show that an important contribution comes from the additional 4f electron.

For a quantitative understanding of the energies of X-rays in the presence of an additional hole or electron (the "spectator") in the atom, it becomes necessary to go beyond the approximation made in [2]. There it was assumed that the shifts were caused by the direct interaction alone (including exchange) between the spectator and the states involved in the X-ray transition. It is known, however, from studies of chemical shifts [4] that in order to describe

precisely the role played by an individual spectator, it is important to incorporate also the polarization effect that the spectator exerts on the average atomic field. More complete calculations were therefore carried out in a relativistic Hartree-Fock scheme [7]. The change in the K X-ray energy due to an additional 4f electron was calculated from the total energies for the initial and final states involved. The results (energies in millielectronvolts) are 620 (K_{α_1}), 560 (K_{α_2}), 1450 (K_{β_1}), 808 (K_{β_2}), and -3714 (KO).

For the calculation of the shake-off energy shifts, a simpler approach was used: the energy shifts caused by the spectator hole states were calculated from the differences in single-electron removal energies in the (static) Hartree-Fock field. This does not introduce major errors; e.g. for the 4f case quoted above it would give (in the same order) 547, 484, 1296, 751, and -3563 meV. This calculation together with the shake-off probabilities used previously [2] reduces the shake-off effect by a factor of 2-3. Thus for the photo-ionization in holmium the shifts 52, 49, 116, 290, and 790 meV, again in the same order, are obtained.

A comparison of experimental and calculated results for the Ho-Er pair is shown in fig. 2. The dominating contributions are seen to be the hyperfine and "4f" effects. The agreement is good but still not quantitative. In the light of this result we reconsider the Au-Hg pair [2]. The additional electron is here in a 6s state belonging to the conduction band. If its contribution is approximated by that of a 6s state in a free atom, the results given in table 2 and in the inset of fig. 2 are obtained. The agreement is seen to be quantitative. The W-Ta experiments, which have now been repeated with metallic samples [8], on the other hand show residual shifts of 1.2 to 1.4 eV, that is shifts that are much bigger than those expected for a 5d electron. Measurements of $K_{\beta_{24}}$ and KO, which are so important for the whole pattern shown in fig. 2, could probably contribute towards the solution of this puzzle. Solid-state effects may play a role: the difference in crystal structure is larger in the W-Ta case than in the rare earths.

This experiment with intense and short-lived radioactivity produced in Grenoble and measured at CERN has only been possible with help from many sides. We would like especially to thank Messrs. E. Delaguillaumie (CENG, Grenoble) and L.-C. Carraz, R. Deltenre, F. Perroud and J. Tuyn (CERN).

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Table 1

Energy shifts¹⁾ of holmium K X-rays (meV)

Line	¹⁶⁵ Er in Er metal			¹⁶⁵ Er in Al		
	2)	3)		2)	3)	
K _{α1}	505	(14)	(22)	540	(20)	(28)
K _{α2}	604	(28)	(35)			
K _{β1}	-382	(35)	(38)	-399	(45)	(48)
K _{β24}	290	(170)	(170)			
K _O	3840	(180)	(210)			

- 1) Defined as $E_x(^{165}\text{Ho, PI}) - E_x(^{165}\text{Er, EC})$.
- 2) Errors due to counting statistics (standard deviation).
- 3) Total error including a contribution from the asymmetry correction (± 10 meV) and from the bias correction ($\pm 0.03 \Delta E$).

Table 2

Energy shifts¹⁾ for gold K X-rays (meV)

Line	Calculated contributions			Exp. ³⁾
	SO	"6s"	Total ²⁾	
K _{α1}	70	83	543	568 ± 36
K _{α2}	70	94	554	567 ± 46
K _{β1}	124	95	609	620 ± 50

- 1) Defined as $E_x(^{197}\text{Au, PI}) - E_x[^{197}\text{Hg (64 h), EC}]$. The data from [2] have been recalculated to represent the pure 64 h isomer; that is, after corrections for contributions from the 23.8 h isomer and from Hg X-rays.
- 2) The total shifts include a hfs contribution of 378 meV and an isomer shift of 12 meV.
- 3) Total error.

Figure captions

Fig. 1 : Scan of the K X-rays from a photo-ionization source of holmium measured in second order with the 4.64 m bent crystal spectrometer. The line width observed here is essentially the natural line width. The schematic level scheme shows the atomic transitions giving rise to the different peaks.

Fig. 2 : Measured energy shifts (points, full-drawn line) for holmium K X-rays for the transitions from np states to the 1s state as a function of n. Three calculations are shown: the hyperfine shift (hfs) calculated on the basis of a magnetic moment of $4.173 \mu_N$ for the ^{165}Ho ground state, and the sum of hyperfine shift and shake-off effect (so) calculated in the improved approximation discussed in the text; and, finally, a calculation which includes also the effect of the additional 4f electron.

The inset shows on the same scale data [2] and calculations for the ^{197}Au case, where for simplicity we have assumed the additional electron to be in a free-atom 6s state.

Fig. 1

Fig. 2