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PHOTOABSORPTION OF Th AND U BY DIRECT MEASUREMENT AND BY FAST ELECTRON ENERGY LOSS SPECTRA NEAR THE 5d THRESHOLDS

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Résumé. — Le coefficient d'absorption du thorium et de l'uranium a été mesuré dans la région des seuils $O_{IV,V}$ par photoabsorption directe et déduit des spectres de pertes d'énergie d'électrons rapides. Les calculs des sections efficaces 5d effectués dans un modèle à particules indépendantes en introduisant des interactions intra-voies sont utilisés pour proposer une interprétation.

Abstract. — The absorption coefficients of thorium and uranium have been measured by photoabsorption and inferred from electron energy loss spectra, near the O_{IV} O_V thresholds. The results of 5d cross sections calculations with an independent particle model introducing intrachannel interactions are used to give a tentative interpretation.

A common feature in actinides and lanthanides is the occurrence of partially filled 5f and 4f electronic shells, whose localization is not so marked in actinides as in lanthanides. No *ab initio* cross section calculation introducing the multiplet splitting and the coupling between the various channels has yet been performed, concerning the complex structure observed in the 4d absorption spectra of lanthanides [1], but an interpretation was proposed [2]. Some 5d spectra in actinides were therefore investigated for the first time and the absolute total photoabsorption coefficient was determined between 50 and 150 eV in thorium and uranium by two completely independent methods.

The absorption curves obtained by soft X-ray measurements (dotted line) and derived from Fast Electron Energy Loss Spectroscopy « FEELS » (continuous line) are plotted in figures 1 and 2 where we indicate the location of the O_{IV} O_V energy levels as measured in solids by XPS [3].

Absorption measurements in the soft X-ray range were performed by use of synchrotron radiation

FIG. 1.

emitted by the ACO storage ring (536 MeV). The samples (20 to 300 $\mu\text{g}/\text{cm}^2$) were evaporated in clean vacuum (10^{-7} - 10^{-8} torr) on to carbon foil (20 $\mu\text{g}/\text{cm}^2$)

(**) ERA 719 du CNRS.

FIG. 2.

before transfer to the experimental chamber located in front of the spectromonochromator previously described [4]. The apparatus was used with different gratings (ruled and holographic) and several grazing incidence angles (8° to 12°) in order to test the effect of overlapping on absolute values. Special attention was devoted to absorption peak values where overlapping order and diffuse light have important effects. Monochromator mounting with two successive gratings giving negligible false light had been used in this range and leads us to a higher peak value than previously, using one grating mode [5]. Our absolute value, for Th may be compared with other results also obtained with synchrotron light [6]. Agreement is good for the general shape and values except in the main peak range undoubtedly because of the parasitic light effect which we have observed using one grating mode settings. Our result showing a ratio of about two between O_{IV} and O_V peaks in Th and U is confirmed by comparison with recent photoemission and relative photoabsorption cross sections for ThF_4 and UO_2 , UO_3 obtained with X ray tubes [7].

On the other hand, the absorption coefficients μ of self-supporting samples were determined by a Kramers-Kronig analysis of fast electron energy loss spectra (FEELS) after the plural scattering contribution has been calculated and removed as explained in [8]; the experimental methods are described elsewhere [9].

In both metals, a quite satisfactory agreement is observed between the overall shapes of the curves deduced by the two methods. This agreement has been demonstrated over an extended range up to several eV [10] and tests the usefulness of FEELS for absolute measurements. Thorium samples in FEELS and photoabsorption experiments have been

evaporated simultaneously; in this case almost perfect coincidence appears between results obtained by the two methods and may be due to the similar quality and thickness of samples. In uranium, samples were not prepared simultaneously and only previously published spectra [8] were reinvestigated with a new data accumulating system.

Absorption spectra show typical features very similar in the two actinides, mainly two sharp and strong peaks, just a little broader in uranium; the O_{IV} O_V energy levels are located at the onset of the rising absorption curve. In addition, a faint structure appears clearly in the thorium photoabsorption curve just below the O_V energy level, in accordance with [6].

Since no multichannel calculations of the 5d cross sections have been performed yet for elements as heavy as Th and U, only a tentative interpretation of the spectra presented here can be given. However, the non relativistic single channel calculations of σ_{5d} for neutral atoms Th and U (ground state configurations : $(\text{Rn}) 6d^2 7s^2$ and $(\text{Rn}) 5f^3 6d 7s^2$) by F. Combet Farnoux [11] can provide us with some information. They have been performed with an independent particle model using Hartree-Fock wave functions (of the average configurations mentioned above) for both the discrete initial state and the continuum final state. Let us recall that the previous calculations of F. Combet Farnoux [12] relative to σ_{4d} in a series of elements, using the same model, pointed out the existence of a high maximum in the continuum (due to $4d \rightarrow \epsilon f$ transitions) for barium and lanthanum. They also indicated that this maximum was not present in the lanthanides : for these elements, the major part of the oscillator strength is in the discrete spectrum, due to $4d \rightarrow 4f$ discrete transitions, as shown by J. Sugar [13], in the framework of intermediate coupling scheme calculations relative to the multiplet splitting and the oscillator strength for the ionic cores $4d^{10} 4f^N \rightarrow 4d^9 4f^{N+1}$ transitions. Anyway, these preliminary calculations for Th and U allow us to understand that for thorium, the σ_{5d} cross section shows a large maximum due to $5d \rightarrow \epsilon f$ transitions to the continuum, since the 5f states lie in the outer well. On the contrary, the $5d \rightarrow 5f$ discrete transitions are intense in uranium, as some 5f states are already in the inner well. However, the shape of the main peak of the two spectra is very similar : this analogy suggests that an interference effect between the various continua (due to the multiplet splitting of the residual ion) and some discrete final states could broaden widely the discrete lines relative to the $5d \rightarrow 5f$ transitions. Of course, such an interference effect cannot be taken into account in the framework of an independent particle model.

Moreover, it is well known now that even a RPAE (Random Phase Approximation with Exchange) treatment does not take into account some relaxation effects which can be important in the case of an inner

hole; they can not only shift ionization thresholds but also strongly deform the shape of the cross section in the thresholds region, as explained by Wendin [14] in the cases of 4d spectra in Ba and La. Such core relaxation effects were not taken into account in the interpretation proposed previously for the lantha-

nides [2]. For the elements presented in this paper the 5f shell is either empty or filled with only a small number of electrons. Thus for a detailed comparison between actinides and lanthanides, it would be necessary to obtain experimental information for other actinides.

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