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RADIATIVE, AUGER, AND COSTER-KRONIG YIELDS OBTAINED BY THE SYNCHROTON PHOTOIONIZATION TECHNIQUE

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Résumé: Nous avons utilisé une nouvelle technique performante pour mesurer les taux de desexcitation d'atomes lourds ($72 \leq Z \leq 82$) par comblement d'une lacune en couche L. Des valeurs des facteurs de Coster-Kronig f_{23} et $f_{13} + f_{12}f_{23}$ sont présentées et discutées.

Abstract: A novel powerful technique was applied to measure L-shell decay yields of heavy elements ($72 \leq Z \leq 82$). Values of the Coster-Kronig factors f_{23} and $f_{13} + f_{12}f_{23}$ are reported and discussed.

1. Introduction

Reliable accurate values of the inner shell yields are required in numerous fields ranging from fundamental research on atomic collision processes [1] to quantitative surface analysis by spectroscopic techniques [2–4]. Adopted values of L-shell yields [5] partly bear intolerable uncertainties. In particular, for the L_1 -subshell uncertainties amount to about 20% which is due to L_1 -specific problems: experimentally, powerful coincidence techniques are not applicable here [6] and theoretically, the decay is governed by processes with small excess energy [7], where standard calculations [8] fail. Recently, a novel and universal technique for measuring yields of *all* subshells was introduced which makes use of the subshell-selective photoionization with tunable synchrotron radiation [9]. In an exploratory measurement this technique has provided stimulating results for Au ($Z = 79$) [9,10]. Comprehensive measurements for a range of elements $72 \leq Z \leq 82$ have been performed and are now under evaluation. So far, results for Coster-Kronig (CK) factors f_{23} and $f_{13} + f_{12}f_{23}$ are available and are discussed in the present paper. All measurements have been performed at the ROEMO station of the HASYLAB synchrotron facility in Hamburg.

2. Technique

In the applied novel technique, definitive numbers of vacancies in the individual subshells are produced by photoionization and the x-ray fluorescence emitted in the vacancy decay is detected. The creation of vacancies by photoionization offers essential advantages as compared to vacancy creation by other processes: First, the ionization of a particular subshell can be switched on and off by tuning the energy of the primary photons across its threshold. Second, the cross section of an individual subshell is known with comparatively high accuracy since experimentally it can be directly obtained from x-ray absorption studies and theoretically the photoionization process can be accurately predicted. Our experimental attenuation data (Fig. 1) exhibit a smooth dependence on photon energy E_γ by a power law; however, they also reveal minor dispersionlike deviations $\Delta(E_\gamma)$. These are due to electron correlation effects which can be partly quantitatively described in the time-dependent local-density approximation [11]. Corresponding calculations reveal that the deviations are quite similar for the different subshells. Therefore, we decided to express the actual ionisation cross sections $\sigma_{L_i}^{actual}$ of the individual L-subshell i by the formula

$$\sigma_{L_i}^{actual}(E_\gamma) = \sigma_{L_i}^{fit}(E_\gamma)[1 + \Delta(E_\gamma)] \quad (1)$$

where $\sigma_{L_i}^{fit}$ is an analytical fit to tabulated calculated values [12] and Δ is a small empirical correction (at most a few percent) which is independent of the subshell and has been determined by matching the theoretical total absorption cross section (sum over all contributing processes) to the experimental x-ray attenuation data (Fig. 1).

Fig. 1: Photoionization cross section of ${}_{72}\text{Hf}$ versus photon energy E_γ in the regime of the L-edges. Curves in the upper part are a theoretical prediction [12]. Dots are experimental data whose absolute values are normalized to theory. The lower part displays the deviation Δ between experiment and prediction as well as a fitted curve.

In our technique, the intensity of the x-ray fluorescence induced by the photon absorption has to be normalized to the intensity of the primary photons at the various photon energies. This was accomplished by normalization to the K-x-radiation of a lighter element. For this purpose a sample consisting of two elements was used, e.g. Hf and Mn, and the Hf L-x-rays and the Mn K-x-rays were simultaneously detected. The K-x-ray signal is just proportional to the K-shell ionization cross section which in case of Mn behaves for all E_γ measured in the present experiment as $\sigma_K(E_\gamma) \propto E_\gamma^{-2.644}$ as predicted by theory [12] and confirmed by experiment [13].

The L-x-ray spectra show a pronounced energy dependence caused by the onset of the ionization of the individual subshell at their thresholds E_{L_i} , which allows to determine the fluorescence yields $\omega_3, \omega_2, \omega_1$ and the CK factors f_{23}, f_{12}, f_{13} . We now specialize to the evaluation of the L_α -line which bears part of the information: its intensity is proportional to the vacancy creation in the L_3 -subshell, i.e.,

$$\begin{aligned} \sigma_{L_3} & \quad \text{for } E_{L_3} < E_\gamma < E_{L_2} \\ \sigma_{L_3} + f_{23}\sigma_{L_2} & \quad \text{for } E_{L_2} < E_\gamma < E_{L_1} \\ \sigma_{L_3} + f_{23}\sigma_{L_2} + (f_{13} + f_{12}f_{23})\sigma_{L_1} & \quad \text{for } E_{L_1} < E_\gamma < E_K \end{aligned} \quad (2)$$

The first term corresponds to direct L_3 ionization whereas the other two terms describe a CK vacancy transfer from the L_2 and L_1 subshells, respectively. With known ionization cross sections (eq. 1) the measured energy dependence of the L_α -intensity can be fitted (eq. 2). The fits deliver the jumps at the L_2 and L_1 absorption edges and thus the f_{23} and $f_{13} + f_{12}f_{23}$ yields. There are minor deviations between the fits omitting the electron correlations ($\Delta \equiv 0$) and including them. (Fig. 3).

3. Results and discussion

In the following we only consider those yields which have been obtained by fits including electron correlations. Our f_{23} values are compatible with adopted values [5] within the uncertainties, although they are consistently smaller. f_{23} is a parameter which can also be directly measured with a K-x-ray - L-x-ray coincidence technique. By this technique $f_{23}({}_{82}\text{Pb}) = 0.112 \pm 0.002$ has been obtained [14].

Fig. 2: Intensity of the Hf L_α line versus photon energy. Dots are experimental data. Curves are fits according to eq. 2 using subshell ionization cross sections including (full lines) and omitting (dashed lines) electron correlations.

Although the claimed accuracy of 2% is impressive, we mention that by the same technique $f_{23}(s_2 Pb) = 0.130 \pm 0.002$ is reported by another group [15]. One crucial problem of these coincidence measurements is the multidimensionality of the coincidence data with a disturbing low-energy tail in the K-x-ray spectrum. This disturbance requires a correction of the measured f_{23} value which amounts to about 30% of its value. Our method of determining f_{23} is direct; it may suffer from uncertainties in the energydependence of the ionization cross sections caused by electron correlations. Nevertheless we are confident to state the value $f_{23}(s_2 Pb) = 0.091 \pm 0.010$.

In the $f_{13} + f_{12} f_{23}$ CK yield the first term is strongly dominant, and it determines the behaviour of the sum. In the investigated range of elements $72 \leq Z \leq 82$ the yield increases by more than a factor of two (Fig. 3 bottom). This increase originates from the increase of f_{13} caused by the onset of the strong $L_1-L_3 M_{4,5}$ CK channel [8]. Our data lie somewhat above the adopted values [5]. Since (for $Z \geq 76$) the procedure of the compilation gives " f_1 values that are 10–15% lower than the majority of the measured values" [5], we are in perfect agreement with other experimental work.

In a comparison of our data to theory we make the simplifying approximation, that the L_α emission probability is the same if the atom initially has only one L_3 vacancy (direct ionization) or has an additional spectator vacancy (L_3 vacancy production by a Coster-Kronig cascade). Theoretical values of f_{23} [16] are significantly above our experimental ones (Fig. 3). The origin for this discrepancy is not clear; the calculated f_{23} value is dominantly determined by the ratio of the nonradiative decay rates for the processes $L_2-L_3 N$ (CK) and $L_2-M_{4,5} M_{4,5}$ (Auger) [8]. Both processes have excess energies of several hundred eV and should be well predicted by theory. For the $f_{13} + f_{12} f_{23}$ factor the theoretical values lie above the experimental data. This is presumably due to the severe overestimate of the $L_1-L_3 M_{4,5}$ CK transition rate by theory (up to a factor of two) as is known from levelwidth measurements [17] and which is caused by the small excess energy of this transition.

4. Conclusions

The applied technique of measuring inner-shell decay yields provides accurate values. Comprehensive work is now required to obtain data for elements with small atomic number, where available data are scarce. This new material will lead to improved adopted values as required in applications and to an improved theoretical understanding of the intrinsic atomic behaviour in a non-equilibrium vacancy state.

Fig. 3: Coster-Kronig factors f_{23} and $f_{13} + f_{12}f_{23}$. Dots are results of the present work including (full dots) or omitting (open dots) electron correlations. The dashed areas indicate the adopted values with estimated uncertainties [5]; the asterisks denote calculated values [16].

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