Decay mechanisms of 4d core holes in Dysprosium

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The electronic properties of the d and f electrons cause a variety of phenomena such as Kondo resonances, mixed valency and magnetism. The magnetic polarization of the d and f electrons for example gives rise to the magnetism in Lanthanides (as well as in transition metals). Therefore the study of their electronic structure is essential in order to understand the magnetic properties. Our experiments were performed at Beamline 8.0.1 of the Advanced Light Source [1]. In Fig. 1 soft x-ray emission spectra (XES) of Dy_2O_3 are displayed for different selected excitation energies through the 4d-4f resonances. The spectra show strong variations in intensity and structure depending upon which intermediate states are excited.

When exciting below 161.7 eV, the inelastic scattering is highly resonant and the total number of inelastically scattered photons can exceed the number of elastically scattered photons. Arrows in the absorption spectrum (TEY) shown in the insert in the top right of Fig. 1 indicate at which energy the emission spectra are excited. The dipole-forbidden pre-threshold features (below 158 eV) become weakly dipole allowed due to the spin-orbit interaction. The ionization thresholds lie at 152.6 eV and above. We have calculated the spectra excited at different photon energies using the Kramers-Heisenberg description for photon scattering and resonant fluorescence. The calculated emission spectra are shown below each measured spectrum. The energy loss features are due to excitations within the 4f shell $4d^{10}\, 4f^9 \rightarrow 4d^9\, 4f^{10} \rightarrow 4d^{10}\, (4f^9)^*$ in which the 4f shell remains in an excited state. These losses extend over a large energy range from about 0.2 to 7 eV below the elastic peak due to the large number of widely separated terms of the 4f shell filled with 9 electrons. In order to compare the emission processes when exciting at and above the resonance, the emission spectra for excitation energies at threshold (154 and 157.2 eV) and above 4d-4f threshold (208.4 eV) are displayed in Fig. 2 over a wide range of emission energy. When exciting through the resonance, the elastic peak (labeled D) and the energy losses due to 4f inner-shell excitations (labeled C) dominate

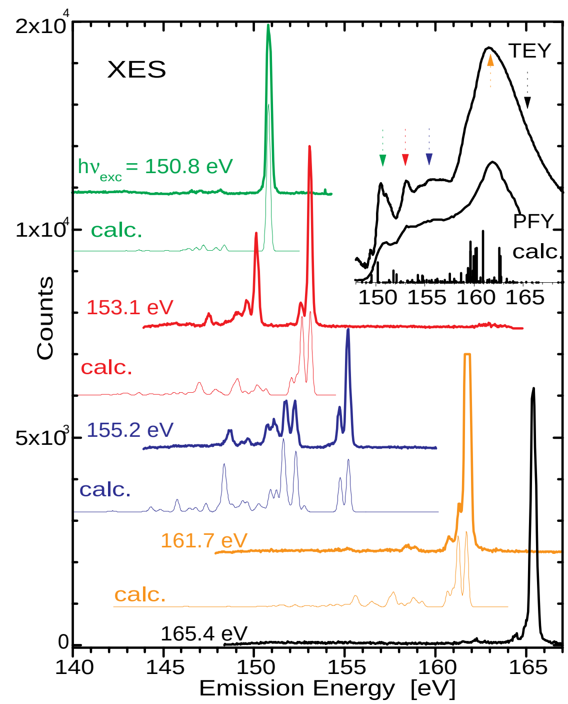

Figure 1. Soft x-ray emission spectra of Dy_2O_3 in the region of Dy the 4d-4f resonances. The excitation energy is given above each experimental spectrum. Absorption (TEY) and partial fluorescence yield (PFY) spectra are shown in the top right insert. The arrows indicate the excitation energies selected for the XES spectra. For clarity the spectra have been offset (in all Figures) and the elastic peak for the spectrum at 161.7 eV is cut off.

the emission. In addition emission from the decay of the 4d hole via the 5p channel is observed (peak A). The inelastic peak labeled as B is due to the transition $4d^{10}\,5p^{6}\,4f^{9} \rightarrow 4d^{9}\,5p^{6}\,4f^{10} \rightarrow 4d^{10}\,5p^{5}\,4f^{10}$ in which a 5p electron is promoted to the 4f shell. The total number of emitted photons is much smaller for above threshold excitation (208.4 eV) than when exciting at or below resonance. The reason is that the core hole decays predominantly by radiationless processes. The dominant radiationless decay processes are 5s, 5p and valence band autoionization for excitation energies below the 4d resonance and for energies at and above resonance the 4f-autoionization becomes dominant. When exciting above resonance the non-resonant fluorescence from the refill of the 4d hole via 5p and 4f is very weak because the excited states decay mainly by radiationless processes. We estimate the total number of emitted photons for above threshold excitation to be at least a factor of 6 smaller than for excitation at threshold [2].

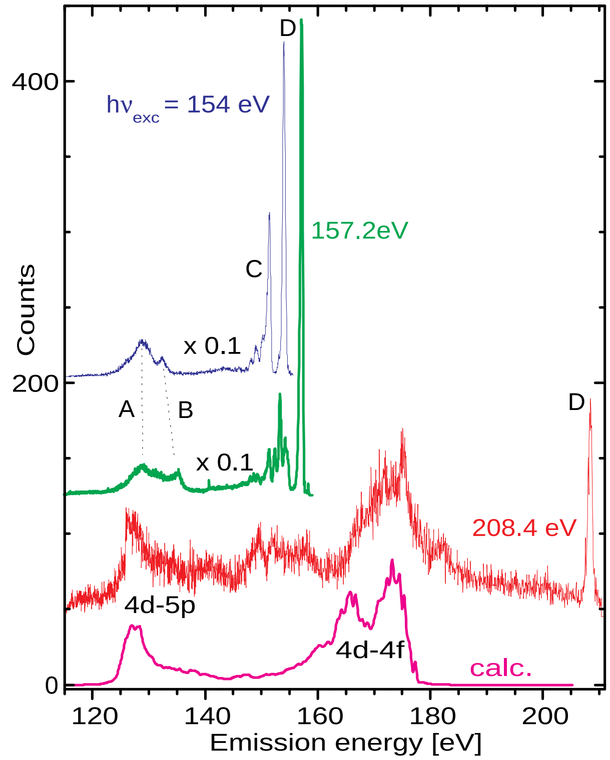

Figure 2. Emission spectra for excitation energies below (154 and 157.2 eV) and above (208.4 eV) 4d-4f resonance. In order to display all data on the same scale, the spectra below threshold are divided by a factor of 10.

REFERENCES

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2. A. Moewes et al., Phys. Rev. B 60, 15728 (1999).

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