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Single photon core ionization with core excitation: a new spectroscopic tool.

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Synopsis . The simultaneous core ionization and core excitation process (or $K^{-2}V$ process) induced by absorption of a single photon provides the basis of a new spectroscopy that offers both advantages of X-ray Photoelectron Spectroscopy (XPS) and near-edge x-ray absorption fine structures (NEXAFS) spectroscopy

In the last few years, double-core-hole states (DCHs) have been extensively studied by single photon double-core ionization at synchrotron centers [1] and, in relation to X-ray two photon double-core spectroscopy [2], at X-ray free electron laser facilities (XFELs). Closely related to double core ionization, the single photon process where a core electron is ionized while another core electron is excited simultaneously, reveals interesting properties on the formation mechanisms of $K^{-2}V$ states and provides a new spectroscopic insight that offers the advantages of both XPS and NEXAFS spectroscopies.

The creation of $K^{-2}V$ states and their subsequent decay are the following:

$h\nu + AB \rightarrow AB^{+}(K^{-2}V) + e^{-}_{ph} \rightarrow AB^{++} + e^{-}_{A1} \rightarrow AB^{3+} + e^{-}_{A2}$.

Since one photoelectron and two Auger electrons are emitted, by detecting in coincidence these three electrons, it is possible to filter out the specific photoelectron spectrum associated with $K^{-2}V$ states. This can be done very efficiently by using a magnetic bottle time-of-flight electron spectrometer under single bunch operation at SOLEIL (France) and Photon Factory (Japan).

The study of the above process for the C_2H_{2n} (n=1-3) series [3] has revealed a NEXAFS signature in the spectra (Fig.1). Two competing channels appear with comparable intensities:

-The first one corresponds to the dipolar ionization of a core electron accompanied by monopolar excitation (direct shake-up) of the remaining core electron and can be interpreted as a "Super" K^{-1} satellite state.

- The second one corresponds to the dipolar excitation of a core electron to a vacant orbital accompanied by monopolar shake-off of the other core electron (conjugate shake-up process) which has a NEXAFS-like character.

Close to the K^{-2} threshold, these two processes appear with comparable intensities. This can be understood as resulting from aborted double-core ionization where one of the outgoing electrons is recaptured in a vacant orbital. At higher photon energy, the conjugate shake-up process is unfavorable since the sudden approximation leads only to the direct process.

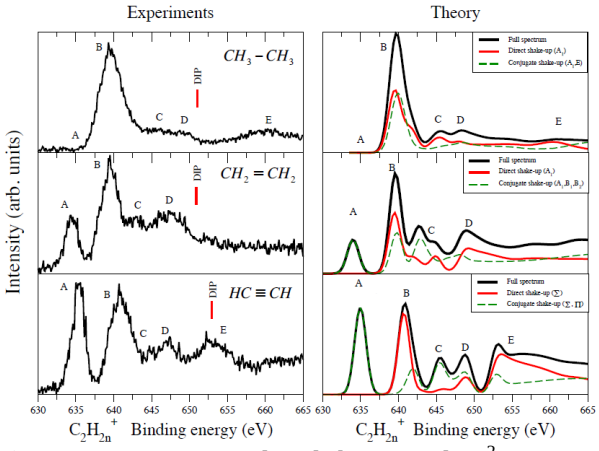

Figure 1: Experimental and theoretical $K^{-2}V$ spectra in C_2H_{2n} series at a photon energy $h\nu$ = 770eV.

Examples on small molecules: H_2O [4], N_2 [5] and many others, have been obtained, which show how powerful this new spectroscopy is and allows also a better interpretation of resonances appearing in NEXAFS spectrum.

References

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