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Double core-hole states in SiX₄ (X = F, Cl, Br, and CH₃) molecules derived by photoelectron and KLL Auger spectroscopy

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Synopsis In recent years double core-hole states are intensively studied since their chemical shifts provide detailed information about initial-state and relaxation effects in a molecule. Most of these measurements are done either by coincidence spectroscopy and single-photon absorption using synchrotron radiation (e.g. [1]) or by two-photon sequential absorption using free electron lasers as light (e.g. [2]) sources. However, some of these double core-hole states can also be obtained by other means. This holds in particular for 2s−2, 2s−12p−1, and 2p−2 core-hole states as a result of KLL Auger decay, which can readily be measured using conventional photoelectron spectroscopy.

Using the new experimental setup for high-resolution HAXPES (HÅrd X-ray PhotoElectron Spectroscopy) in gas phase, which is mounted at the GALAXIES beamline of the synchrotron SOLEIL, we measured the Si KLL Auger spectra of the molecules SiX₄ with X = F, Cl, Br, and CH₃. Complementary photoelectron spectra of these molecules were recorded in order to derive the Si 1s−1, 2s−1, and 2p−1 single core-hole binding energies. Using these binding energies, the chemical shifts of the Si 2s−2, 2s−12p−1, and 2p−2 double core-hole binding energies were derived from the Auger spectra. The obtained binding energies are compared with theoretical results [3] revealing good agreement. Moreover, the results were used to create Wagner plots (i.e. plotting single core-hole energies versus double core-hole energies) for the Si 1s, Si 2s, and Si 2p core hole state, which give a detailed insight in the initial state and relaxation effects [4]. In particular, they show that the various core holes provide similar information. In addition, the Wagner plots reveal that the initial state effects depend only on the electron-accepting (F,Cl,Br) or electron-donating (H,CH₃) character of the ligands. Within these groups differences in the ionization energies are due to varying relaxation energies.

Recently it has been shown for argon that the Ar 1s−12s, p−1 double core-hole shake-up states are accessible with conventional photoelectron spectroscopy [5]. Here, we present similar Si 1s−12s, p−1 spectra for all SiX₄ molecules. In summary, we show that KLL Auger and photoelectron spectroscopy are well suited to derive information about double core-hole states.

References


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