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Double core-hole states in SiX_4 ($X = \text{F}, \text{Cl}, \text{Br}, \text{and CH}_3$) molecules derived by photoelectron and KLL Auger spectroscopy

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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. We derived the Si $1s^{-1}$, $2s^{-1}$, and $2p^{-1}$ binding energies as well as the Si $2s^{-2}$, $2s^{-1}2p^{-1}$, and $2p^{-2}$ double-core hole binding energies of different SiX₄ systems in order to derive the chemical shifts. Based on these results we created Wagner plots, which give insight in the initial state and the relaxation effects in the different molecules.

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 $2s^{-2}$, $2s^{-1}2p^{-1}$, 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 (HARD 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 $2s^{-2}$, $2s^{-1}2p^{-1}$, 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 $1s^{-1}2s, p^{-1}$ double core-hole shake-up states are accessible with conventional photoelectron spectroscopy [5]. Here, we present similar Si $1s^{-1}2s, p^{-1}$ 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

- [1] P. Linusson *et al* 2011 *Phys. Rev. A* **83** 022506 and references therein; P. Lablanquie *et al* 2013 *Phys. Rev. Lett.* **110** 163001 and references therein.
- [2] N. Berrah *et al* 2011 *PNAS* **108** 16912; P. Salen *et al* 2012 *Phys. Rev. Lett.* **108** 153003, and references therein.
- [3] O. Takahashi *et al* 2011 *Chem. Phys.* **384** 28.
- [4] T. D. Thomas 2012 *J. Phys. Chem. A* **116** 3856.
- [5] R. Püttner *et al* 2015 *Phys. Rev. Lett.* **114** 093001.

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