Carbon K-edge X-ray emission of gas phase ethylenic systems

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X-ray emission (XES) is a powerful tool for investigating the electronic structure of molecules, probing not just the energetic separation of occupied electronic states but also determining the symmetries of the occupied orbitals.[1] The combination of element-selectivity and the sensitivity of the X-ray signal to changes in the local environment also mean such techniques are ideally suited to resolving structural dynamics.[2] However, in the soft X-ray regime, where the K-edge transitions for most of the light elements like carbon, nitrogen and oxygen are located, the quantum yield for X-ray fluorescence is significantly lower.[3] The result of this is, while soft X-ray XES has become a routine technique for characterisation of solids, only very limited studies have been carried out on low-density samples[4,5], particularly at the carbon K-edge where the 'carbon hole' results in significantly reduced X-ray flux.

In this work, we present the feasibility of X-ray emission at the carbon K-edge on three ethylenic, gaseous molecules, ethylene, propa-1,2-diene (allene) and *trans*-1,3-butadiene. Non-resonant X-ray emission spectra of ethylene show good agreement with previous high-resolution electronic impact experiments.[6,7] The resonant emission spectra for all three ethylenic systems show significant change from the non-resonant case and allow, in combination with *ab initio* calculations, us to probe the behaviour of different excited, intermediate states. This work was carried out at the GasPhase beamline at the Elettra synchrotron and is a key development for future gas-phase time-resolved experiments with XES.

References

[1] - F. de Groot, *Chem. Rev.*, **101**, 1779-1808, (2011).
[2] - M. Chergui and E. Collet, *Chem. Rev.*, **117**, 11025-11065, (2017).
[3] - J. Nordgren and J.E. Rubensson, *J. Electron Spectros. Relat. Phenomena*, **188**, 3-9, (2013).
[4] - P. Skytt *et al.*, *Phys. Rev. A - At. Mol. Opt. Phys.*, **55**, 146-154, (1997).
[5] - M. Oura *et al.*, *J. Phys. Conf. Ser.*, **235**, 012016, (2010).
[6] - R. Brammer *et al.*, *Chem. Phys. Lett.*, **106**, 425-427, (1984).
[7] - R.A. Mattson and R.C. Ehlert, *J. Chem. Phys.*, **48**, 5465-5470, (1968).