Fragmentation dynamics of CS_2^{4+} after core S1s excitation studied by triple-ion momentum correlation

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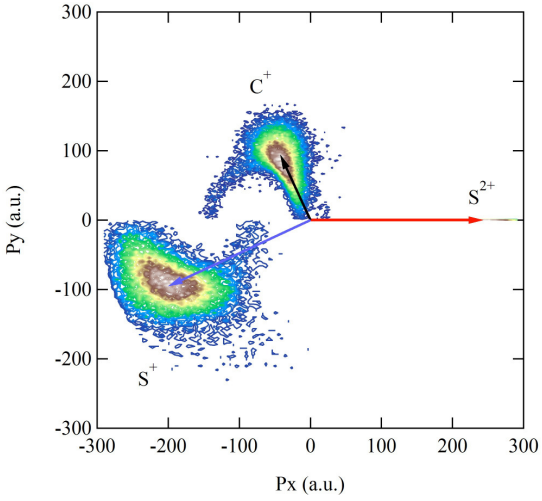
Understanding of the processes resulting from interaction between light and matter is of interest in many diverse fields such as photochemistry, astrophysics, as well as in biology and medicine. The lifetime of electronic states created by the absorption of a photon by an atom or a molecule determines the time scale within which these processes will occur, and the resulting reactions. When X-rays are absorbed, deep electron shells are excited. The significant time scale then becomes of the order of a few femtoseconds. Photoionization of deep core shell is followed by a number of relaxation processes. When a core electron in a free molecule is excited to an unoccupied orbital, molecular deformation can occur within a few femtoseconds during the lifetime of the core-excited state. This ultrafast nuclear motion proceeds in competition with the relaxation of the core hole by radiative or non radiative decay. After relaxation, highly charged ions are produced and the fragmentation takes place by Coulomb explosion. Coincidence techniques and momentum imaging have been shown to be a powerful probe of the dynamics of molecular fragmentation and ultrafast nuclear motion [1,2,3].

We will present experimental results obtained at the French synchrotron source SOLEIL on the LUCIA beam line with a new setup, CELIMENE, designed to provide the full momentum vectors of coincident particles emitted after deep core-level photoexcitation in the "tender" x-ray regime (2 to 10 keV) [4,5].

We have studied the fragmentation dynamics of CS_2 after sulphur 1s core excitation (~2475 eV) and in particular the 3-body dissociation of the CS_2^{4+} ion into S^{2+}+C^++S^+. Our results show that it is possible to follow the molecular deformation and resolve the dynamics of the dissociation pathways. Two channels are disentangled: one corresponding to a concerted fragmentation, where all ionic fragments are emitted simultaneously, and one where the fragmentation occurs sequentially with the emission of the doubly charged ion S^{2+} proceeding first while a long-live CS^{2+} fragment rotates as it recoils from its initial position in the molecular system.



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