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Three-electron collective Auger decay in CH_3F

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Synopsis We report on experimental and theoretical study of collective Auger decay of double inner-valence vacancy in fluoromethane, in which two electrons simultaneously fill the vacancies and a third electron is ejected to continuum. Calculations predict the respective lifetime of the double vacancy state to be in the femtosecond range, typical for two-electron Auger decay. Measured electron spectra indirectly support this rather surprising result.

Inner-shell ionization of atoms and molecules leads to highly-excited ionic states that can relax by electron emission, the Auger effect being the most common example of such a process. The advent of high-intensity free-electron lasers brought into focus study of the non-radiative decay processes under conditions of multiple ionization. Here, we address so-called collective decay, a rather exotic three-electron Auger-like process. In collective decay, two vacancies recombine simultaneously and excess energy is carried away by a third electron that is ejected to continuum.

First such three-electron process reported was probably Auger LL-MMM transition in Ar [1]. More recently, related effects were observed in highly charged carbon atoms [2], three-electron interatomic Coulombic decay in rare-gas systems was studied both theoretically and experimentally [3, 4]. Some evidence that three-electron transitions do not occur exclusively in multiply ionized systems was given by Lee et al on the example of resonantly excited krypton atom [5].

Although the three-electron processes are usually considered to be extremely weak, under favorable conditions they can become dominant decay channels. In the present work, we focus on one such example which is the decay of doubly F($2s$) ionized CH_3F molecule. Excitation energy of this state is well above triple-ionization threshold. However, calculations using the extended second-order algebraic diagrammatic construction (ADC) [6] show that all triply-ionized decay channels with at least one F($2s$) vacancy are closed which is the condition we seek for the study of collective decay process. The figure shows triple-ionization spectrum of the $CH_3F(2s^0)^{2+}$ metastable state obtained by coincidence measurement of electrons emitted in a two step cascade initiated by C(1s) core ionization. The spectrum is compared to the theoretical spectra calculated using Fano-ADC method with and without the nuclear dynamics along C-F bond taken into account. Excellent agreement between the theoretical and experimental spectra supports the calculated lifetime is 2.5-3.0 fs, which is in the range of typical two-electron Auger lifetimes.

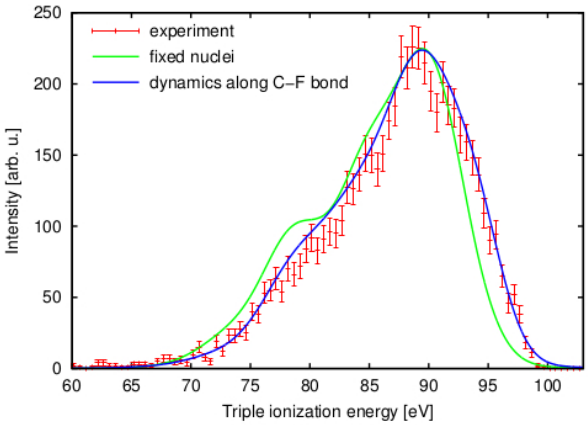

Figure 1. Measured and calculated spectrum of the $CH_3F(2s^0)^{2+}$ metastable state.

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