ULTRAFAST MOLECULAR THREE-ELECTRON COLLECTIVE AUGER DECAY

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A new class of many-electron Auger transitions in atoms was initially proposed over 40 years ago[b], but the first tentative evidence for its real existence was only adduced by Lee et al.[c] in 1993, on the basis of the resonant Auger spectrum of Kr. Using a multi-electron coincidence technique with synchrotron radiation, we unambiguously showed very recently that the transition suggested by Lee et al. in Kr really does take place, but with a rather small branching ratio[d]. Related inter-atomic three-electron transitions in rare gas clusters were recently predicted by Averbukh and Kolorenč[e] and demonstrated by Ouchi et al.[f]. From consideration of the energy levels involved it seems that the basic three-electron process could occur in molecules too, wherever a double inner-valence shell vacancy lies at a higher energy than the molecular triple ionisation onset. Experiments on CH_3F reveal for the first time the existence of this new decay pathway there[g], and calculations show that despite its three-electron nature, its effective oscillator strength is orders of magnitudes higher than in atoms, allowing an efficient competition with both molecular dissociation and two-electron decay channels on the ultrafast time scale. The dramatic enhancement of the molecular three-electron Auger transition can be explained in terms of a partial breakdown of the molecular orbital picture of ionisation. We predict that the collective decay pathway will be significant in a wide variety of heteroatomic molecules ionised by extreme UV and soft X-rays, particularly at Free-Electron-Lasers where double inner-shell vacancies can be created efficiently by two-photon transitions.

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