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Time-Dependent Quantum Model for Attosecond X-ray Non-Linear Spectroscopy

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The recent capacity of X-ray free-electron lasers (XFELs) to produce ultrashort and intense X-ray pulses gives access to the observation of electron motion in molecules with high temporal (attosecond) and spatial (angström) resolutions. In the present study, we use a promising nonlinear technique called impulsive stimulated X-ray Raman scattering (ISXRS) to produce a coherent superposition of neutral excited electronic states in nitric oxide (NO) [1]. An attosecond X-ray pulse first core-excites the O atom, promoting a core electron to a $2\pi^*$ orbital. Before this short-lived core-excited state decays, a second photon is absorbed from the same X-ray pulse and creates a coherent superposition of neutral valence-excited states. A time-delayed femtosecond UV pulse probes the induced dynamics by ionizing the molecule, and the NO+ yield allows us to quantify the population transfer induced by the ISXRS process.
We developed a sophisticated quantum model to interpret this first experimental demonstration of ISXRS in a molecule. It is based on the time-dependent Schrödinger equation for the electrons, and takes into account the interaction of the X-ray pulse with the molecule, the Auger decay, as well as the strong electron correlation effects in the presence of a core vacancy. With this model, we are able to provide a quantitative and qualitative interpretation of the ISXRS process, as well as to characterize the X-ray pulse parameters. This work
demonstrates the possibility to induce electronic population transfer via ISXRS using a single attosecond X-ray pulse, and sheds light on the role of electronic coherences at the earliest stage of chemical processes. Moreover, this study is relevant for future two color attosecond X-ray pump / X-ray probe set-ups that will also permit to probe site-selectively the induced dynamics at a remote atom in the molecule. This joint experimental and theoretical investigation is thus a stepping stone towards studying electronic dynamics in more complex systems and opens a path for investigation of transient electronic phenomena in matter at XFEL facilities.

[1] J. T. O'Neal et al., Phys. Rev. Lett., 125 073203 (2020)

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