Theoretical approaches for time-resolved inner-shell physics and coherent control

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New capabilities at accelerator-based and laser-based sources are continuously being developed, being possible to nowadays generate two-color XUV/x-ray pulses with controlled time delay. In the optical regime, in which lasers have achieved a high-degree of spatio-temporal coherence several decades ago, we find many control techniques for atoms and molecules based on coherence. The novel two-color XUV/x-ray sources will allow exploring novel quantum control schemes by using the intrinsic site-specificity excitation of high-energy photons.

After x-ray excitation a core-hole state is produced. Core-hole states decays, between hundreds of attoseconds and few femtoseconds, mainly by fluorescence and Auger processes. A theoretical approach that accounts those processes concurrently with the light-matter interaction is needed to describe the coherence of the system and the measured observables in a time-resolved experiment.

The FERMI FEL has unique laser-seeding capabilities to produce high-degree spatio-temporal coherent pulses [1], and most recent developments at FERMI allow the possibility to reach photon energies in the soft x-ray regime (carbon K-edge) [2]. A novel developed theoretical approach was used to demonstrate Stimulated Raman Adiabatic Passage (STIRAP) [3] at the FERMI FEL. Using the 4d core-hole state in Xe, we can use a three-level system, in which the initial state *(i)* is the ground state of Xe atom, the intermediate state *(e)* is the core-hole state $4d^{-1}6p$, and *(f)* is the excited final state $5p^5 6p$. Preliminary calculations, see Fig. 1, show that a maximum transfer of around 30% is possible to reach with the experimental conditions at FERMI, which is a significant and experimentally measureable effect.

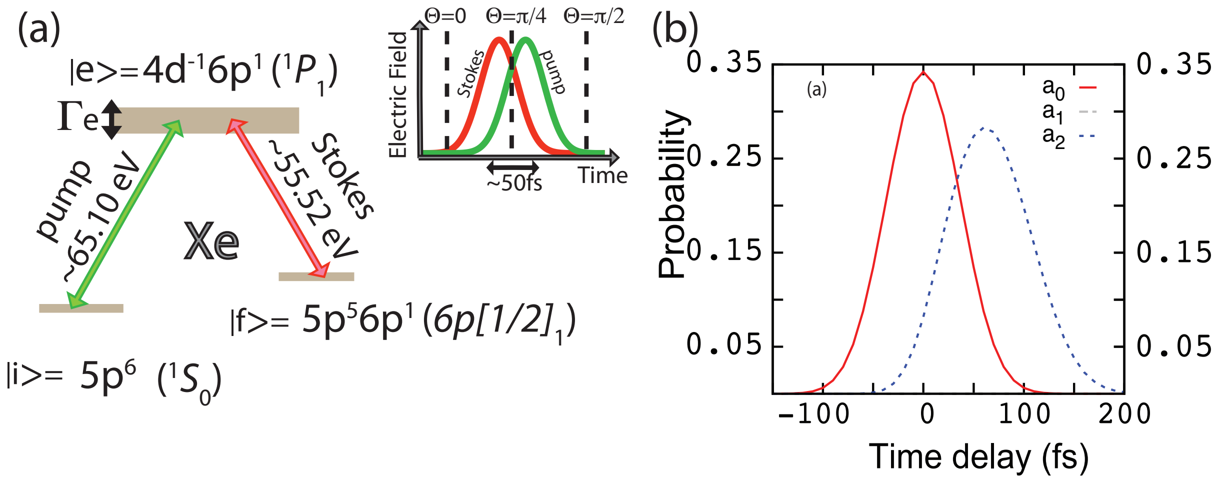

Fig. 1 (a) STIRAP scheme for Xe, the three main states are shown. (b) Numerical simulations for the populations of the Xe ground state (a_0), the 4d→6p intermediate state (a_1), and the $5p^5 6p$ excited final state (a_2).

Also a novel theoretical approach was developed to describe the dynamics of an x-ray photoelectron experiment with two-color x-ray pulses at the Linac Coherent Light Source (LCLS). Preliminary theoretical results show the potential of x-ray photoelectron spectroscopy to follow both electronic and nuclear motion in the femtosecond time scale for x-ray-induced molecular phenomena.

References

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[3] Nikolay V. Vitanov, Andon A. Rangelov, Bruce W. Shore, Klaas Bergmann, "Stimulated Raman adiabatic passage in physics, chemistry and beyond", arXiv:1605.00224 (2016).