Watching and steering few- to many-electron bound-state dynamics

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Synopsis Time-domain XUV absorption spectroscopy and x-ray diffractive imaging are employed to study and control how electrons in atoms and molecules respond to (multiple) excitations and strong laser fields.

Much attention in current molecular physics and chemistry research focuses on the measurement and understanding of the structure and (electronic) dynamics within increasingly complex, large, (even bio-)molecules. In these experiments, questions arise as to how the fundamental interactions used for probing, such as the absorption of a single (or multiple) photons, influence the structure and dynamics to be observed. Recent experiments remind us that the interaction of a few-electron atom or molecule, even in their bound (neutral) states, with only moderately strong laser fields is still far from understood. This calls for a new class of experiments on simple (few-body) atomic and molecular systems, which can be modeled sufficiently well to develop and test observational methods and extract fundamental dynamical mechanisms that can then be scaled up to more complex systems.

This talk will cover measurements and conceptual models for understanding and laser controlling fundamental quantum dynamics from few- to many-electron excited states. The focus will be mainly on atoms and small molecules. Both freely evolving [1] and laser-driven internal dynamics [1,2] can be reconstructed within certain limitations, and the mechanisms [3] initially observed for two excited electrons in helium atoms can be shown to apply even to larger molecular systems, such as organic dye molecules in solution [4].

Another general model of impulsive (short pulsed) strong-field interaction [5] was recently applied to temporally resolve the emergence of a Fano resonance in time [6]. In a time-resolved absorption spectroscopy experiment, attosecond extreme ultraviolet (XUV) pulsed light was used for excitation and a strong near infrared laser pulse at controllable delays as a temporal gate for cutting off the natural autoionization decay by laser-induced ionization.

In our effort to understand and control time-dependent structural changes of many-body molecular systems we performed an x-ray diffractive imaging experiment on fullerene molecules at the x-ray free-electron laser LCLS. In the time-dependent x-ray diffraction images we identified different dynamical regimes of C_{60} interacting with intense near-infrared laser pulses and were able to temporally follow the initial coherent expansion of the molecular cage into the statistical (incoherent) breakup of smaller fragments.

References

[1] C. Ott, A. Kaldun, L. Argenti, P. Raith, K. Meyer, M. Laux, Y. Zhang, A. Blättermann, S. Hagstotz, T. Ding, R. Heck, J. Madronero, F. Martín, and T. Pfeifer.
Nature **516**, 374 (2014)

[2] A. Kaldun, C. Ott, A. Blättermann, M. Laux, K. Meyer, T. Ding, A. Fischer, and T. Pfeifer.
Phys. Rev. Lett. **112**, 103001 (2014)

[3] C. Ott, A. Kaldun, P. Raith, K. Meyer, M. Laux, J. Evers, C. H. Keitel, C. H. Greene, and T. Pfeifer.
Science **340**, 716 (2013)

[4] K. Meyer, Z. Liu, N. Müller, J.-M. Mewes, A. Dreuw, T. Buckup, M. Motzkus, and T. Pfeifer.
Proc. Natl. Acad. Sci. USA (PNAS) **112**, 15613 (2015)

[5] A. Blättermann, C. Ott, A. Kaldun, T. Ding, and T. Pfeifer, *Two-dimensional spectral interpretation of time-dependent absorption near laser-coupled resonances*, *J. Phys. B: At., Mol. Opt. Phys.* **47**, 124008 (2014).

[6] A. Kaldun, A. Blättermann, V. Stooß, S. Donsa, H. Wei, R. Pazourek, S. Nagele, C. Ott, C. D. Lin, J. Burgdörfer, and T. Pfeifer.
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