

Understanding Spectral Shapes from Atoms to Molecules: Laser Control of Two and Many Electrons

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When intense laser pulses interact with atoms and molecules, they can strongly modify their internal electronic structure. Acting on very short time scales of few femtoseconds down to attoseconds, they outrun other processes such as electronic or internuclear relaxation that pull the system back to thermal equilibrium. This ultrashort window of coherence can thus be used to induce and characterize novel states of matter which may otherwise hardly exist in nature.

Here, we discuss some of our past and recent experiments and their results on the modification of matter on short time scales. Experimentally, in these activities we employ the combination of high-frequency light with intense infrared (IR) laser fields. This allows observing electronic dynamics in atoms and molecules after extreme-ultraviolet (XUV) excitation in a state-selective manner.

For two interacting electrons in helium atom, we uncovered that ultrashort laser induced quantum-level shifts right after pulsed excitation lead to phase shifts of coherently excited quantum-state amplitudes. The latter leave their imprint in the spectral line shape of an absorption spectrum, recorded after passage of light through an absorbing medium. The spectral shape can thus be used as a metrology tool to quantify the coupling of intense fields with these few-electron states. This phase shift of quantum states also characterizes the Fano resonance, caused by configuration interaction of electronic bound and continuum state. The interaction with a laser field can thus be used to undo the natural phase shift originating from configuration interaction, turning a Fano resonance back into a symmetric Lorentzian resonance, and vice versa.

Moving on to multiple electrons in recent work, we focus on another fundamental quantum-mechanical effect, namely exchange interaction. It arises due to the fact that only one electron (due to its Fermionic nature) can occupy a certain quantum state, thus modifying the energy of multi-electron states depending on their spin configuration. Among many different realizations and applications in various fields of physics and chemistry, electronic exchange interaction plays a major role in molecules and their bonding dynamics. In our experiments, employing a few-cycle infrared(IR)-tunable laser system, we found that the relative line strength of a core-level resonance absorption doublet in the sulfur hexafluoride (SF_6) molecule is modified by its interaction with the intense fields from a 1.5 micron few-cycle laser pulse.

It turns out that this effect of line-strength variation can be understood within a model Hamiltonian by the mixing of two effective states exhibiting different magnitudes of exchange interaction. The laser intensity can thus be used to tune the effective exchange interaction, despite the substantial complexity of multi-electron IR laser interaction with many valence electrons in the molecule. A state-of-the-art simulation of the group of Maurits Haverkort (Heidelberg University) confirms the results of our measurement and the model description of the main physical mechanism.

These results of laser steering the fundamental quantum dynamics of two, or even multiple, electrons even for the complex case of poly-atomic molecules, open exciting future perspectives. They lay the foundation for controlling molecular dynamics and chemical reactions directly on the electronic level.