Attosecond-resolved Quantum Chemistry. Predictions from the early years and comparison with recent experiments

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A variety of processes originating from the interaction of atomic or molecular N-electron states with strong and/or hypershort radiation pulses can be understood quantitatively only by first determining with good accuracy the solutions of the many-electron time-dependent Schrödinger equation (METDSE) that describe the corresponding physics. The METDSE is solvable nonperturbatively via the *state-specific expansion approach* (SSEA), introduced in the early 1990s [1]. SSEA solutions have been used, or can be used, for quantitative explanation and numerically reliable predictions of quantities that have been measured or are measurable in modern experiments that can track, with hypershort time resolution, the effects of electron dynamics in atoms and molecules. The calculations take into account in a transparent way the interplay between the phenomena and the electronic structures of the physically significant states in discrete and multichannel continuous spectra, including multiply- and inner-hole–excited resonance states.

Soon after the announcement in 2001 of the first generation of attosecond pulses in the laboratory, in a series of papers starting in 2002 [2,3], we proposed and demonstrated from first principles, via the implementation of the SSEA, that coherent excitation and decay of strongly correlated resonance (autoionizing, Auger) states constitute a fertile domain for the then nascent attosecond spectroscopy, where effects of state-superpositions, of electron correlations and of electron rearrangements can be resolved on attosecond scales.

The lecture will discuss briefly key elements of the SSEA, and will include a comparison of our predictions from 2007 [4] and 2010 [5] with quantitative attosecond-resolved measurements of novel phenomena, such as, 1) the time-dependent build-up of the profile of the He $2s2p\ ^1P^o$ resonance state during photoionization [4,6], and, 2) the recognition that there is a *relative time delay* of the order of about 10 attoseconds in the photoejection of the ($2s, 2p$) electrons of Neon [3,5,7].

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