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ATOMIC AND MOLECULAR SYSTEMS IN INTENSE ULTRASHORT LASER PULSES

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Abstract. The full quantum mechanical treatment of atomic and molecular systems exposed to intense laser pulses is a so far unsolved challenge, even for systems as small as molecular hydrogen. Therefore, a number of simplified qualitative and quantitative models have been introduced in order to provide at least some interpretational tools for experimental data. The assessment of these models describing the molecular response is complicated, since a comparison to experiment requires often a number of averages to be performed. This includes in many cases averaging of different orientations of the molecule with respect to the laser field, focal volume effects, etc. Furthermore, the pulse shape and even the peak intensity is experimentally not known with very high precision; considering, e. g., the exponential intensity dependence of the ionization signal. Finally, experiments usually provide only relative yields. As a consequence of all these averagings and uncertainties, it is possible that different models may successfully explain some experimental results or features, although these models disagree substantially, if their predictions are compared before averaging. Therefore, fully quantum-mechanical approaches at least for small atomic and molecular systems are highly desirable and have been developed in our group. This includes efficient codes for solving the time-dependent Schrödinger equation of atomic hydrogen, helium or other effective one- or two-electron atoms as well as for the electronic motion in linear (effective) one-and two-electron diatomic molecules like H_2.Very recently, a code for larger molecular systems that adopts the so-called single-active electron approximation was also successfully implemented and applied.

In the first part of this talk popular models describing intense laser-field ionization of atoms and their extensions to molecules are described. Then their validity is discussed on the basis of quantum-mechanical calculations. Finally, some peculiar molecular strong-field effects and the possibility of strong-field control mechanisms will be demonstrated. This includes phenomena like enhanced ionization and bond softening as well as the creation of vibrational wavepacket in the non-ionized electronic ground state of H_2 by creating a Schrödinger-cat state between the ionized and the non-ionized molecules. The latter, theoretically predicted phenomenon was very recently experimentally observed and lead to the real-time observation of the so far fastest molecular motion.