Multi-color attosecond control of single ionization

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Synopsis We present the results of *ab initio* 6-D calculations of atomic He in intense laser fields. In particular we investigate the effect of a delay between an infrared field and an attosecond pulse train on single ionization. By casting the combined electric field of the infrared and attosecond pulse train in terms of multiple colors, an analytic framework is developed to interpret the numerical results.

At the heart of atomic and molecular physics is the motion of bound electrons. Since the relevant time scale for movement of an electron wavepacket between energy levels in atoms and molecules is on the order of a few atomic units (1 a.u. $\approx$ 24 as), detailed experimental studies on electron motion require forces that act on such time scales. With such ultra-fast forces, the avenue for control of the electron dynamics is opened up. When synchronized to an infrared field, attosecond-scale UV pulses which occur every IR period can provide such quick forces, and therefore the possibility of macroscopic control of electronic dynamics.

Because an attosecond pulse is often produced via high harmonic generation, the resultant attosecond pulse train (APT) will contain a large of harmonics of the fundamental frequency of an intense IR laser. With selective filtering, a specific range of harmonics can be isolated and used to illuminate the target. Control is exercised by varying the delay between the APT and IR field on a tens-of-attoseconds scale. The experimental demonstration of control of ionization probability using this method has been realized [1], as well as control of the angular distribution of the ionized electrons [2, 3].

In this work we account for the observed modulation in the ion yield of helium in an IR+APT field with respect to delay with an analytic framework verified by fully time-dependent 6-D calculations. Toward this end, we recast APT+IR experiments in terms of a many-color control problem. By employing a Floquet-like approach [4, 5], the control parameter (the delay between colors) is analytically separated from the wavefunction. This picture also gives us the ability to remove all of the time-dependence from the Hamiltonian other than the relatively slowly-varying envelopes of the pulses.

Our nonperturbative, *ab initio*, 6-D calculations utilize the adiabatic hyperspherical representation. This approach is appealing because it requires no modeling for helium — it is a full two-electron calculation and can be made exact by increasing the number of included channels. Another benefit of this approach is the natural incorporation of singly-excited states and their effect on ionization.

Ultimately, the solution to the time-dependent Schrödinger equation and the analysis of observed phenomena boils down to using dipole selection rules, energy conservation, and counting photons. Using these natural ideas, the attosecond control of single-ionization can be explained by the interference between various photon pathways. While we examined the APT+IR on helium experiments, our picture is general enough to analyze any system that uses the delay between different harmonics as a control mechanism.

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