Unified Time and Frequency Picture of Ultrafast Atomic Excitation in Strong Laser

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Synopsis We report on experiments on strong-field excitation in both the tunneling and multiphoton regimes and their rigorous interpretation by time dependent Schrödinger equation calculations. The study readily consolidates the seemingly opposing strong-field regimes with their complementary pictures. Most strikingly, we observe an unprecedented enhancement of excitation yields, which opens new possibilities in ultrafast strong-field control of Rydberg wave packet excitation and laser intensity characterization.

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In a combined experimental and theoretical study on ultrafast excitation of atoms in intense short pulse laser fields we show that the prevailing and seemingly disparate intuitive pictures that are usually used to describe the interaction of atoms with intense laser fields, the multiphoton picture and the tunneling picture, can be finally united and ascribed to a single nonlinear process [1].

In the experiments we measure directly the excitation yield of Ar and Ne atoms, as a function of the laser intensity covering both the multiphoton and tunneling regimes by choosing 400 nm and 800 nm wavelengths. In the multiphoton regime, characterized by the Keldysh parameter $\gamma>1$, we observe pronounced resonant enhancements in the excitation yield. These are particularly striking in the case of Ar, where the sharp step in yield with an increase by a factor of 100 occurs in the vicinity of the 6 photon channel closing. We note that this enhancement provides a nearly model-independent intensity calibration. In the tunneling regime, $\gamma<1$, no structure due to channel closings is apparent.

By solving the time dependent Schrödinger equation (TDSE) numerically, we obtain the probability for excitation, which, taking into account focal-volume averaging is in excellent agreement with the data. An important question that arises is whether channel-closing signatures persist in the tunneling- and over-the-barrier-ionization regime and, if so, how do they evolve from the multiphoton to the tunneling regime.

In the tunneling regime the theoretical results show regularly spaced spikes due to channel closings. Obviously, focal-volume averaging hinders their experimental observation. At higher intensities the spacing becomes irregular, but is clearly present in the spectrum. As detailed in [1], the transition from regular to irregular spacing can be understood within the time picture. At each laser cycle maximum an electron wave packet is launched with the respective tunneling probability. It can be shown that the electron wave packets pick up a phase between two consecutive cycles. In the multiphoton regime we find that the electron wavepackets are predominantly produced at the maximum of the laser pulse with a constant phase difference $2\pi\frac{U_p}{\omega}$, where U_p is the ponderomotive potential and ω is the laser frequency. This results in constructive interference in the time domain. It gives rise to peaks at the channel closings in the frequency domain. In the tunneling regime electron wave packets are produced in the rising part of the laser pulse, so that the phase is modified by large contributions sufficient to change the sense of the interference, as observed.

Finally, we have also measured and analyzed the n-distribution of Rydberg state. We are able to observe hints of intensity-dependent control of the excited Rydberg state distributions, which are strongly supported by our TDSE results.

References

[1] H. Zimmermann *et al.* 2017 *Phys.Rev.Lett.* **118**, *013003*

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