Atomic wavefunctions probed through strong field light-matter interactions

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The spatial properties of atomic wavefunctions were probed through the strong field process of high harmonic generation. Using elliptically polarized laser fields and two color fields we were able to manipulate the 2D motion of free electrons and probe the atomic wavefunctions from different angles. The atomic orbitals in turn were used as spatial probes to investigate the dynamics of the ionization process.

Strong field light-matter interactions can encode the spatial properties of the electronic wave functions which contribute to the process. Recently it has been established that the broadband harmonic spectra, measured for a series of molecular alignments, can be used to create a tomographic reconstruction of molecular orbitals [1]. We take tomography one step forward, extending the approach to systems that cannot be naturally aligned. We demonstrate this ability by probing the two dimensional properties of atomic wavefunctions. By manipulating an electron ion recollision process, we are able to resolve the symmetry of the atomic wavefunction with notably high contrast. Finally we demonstrate how such approach can be extended to resolve the ionization mechanism.

Manipulating the 2D dynamics of the free electron can be achieved using either an ellipticaly polarized single color field or an orthogonally polarized two color field. Tunnel ionization which is a selective process that determines the wave function to be probed [2] occurs along the instantaneous electric field direction. The free electron accelerates in the electric field and is then driven to recollide with the parent ion at an angle. By manipulating the parameters of the field such as the two fields delay, in a two color scheme, or the laser ellipticity, in a single color scheme, we can tune the recollision angle and perform a 2D scan of the atomic wavefunction.

We have recently demonstrated this approach experimentally, using a two color scheme, by probing the wavefunction of He (1s state) and Ne (2p state) [3]. The experimental results show a striking difference between the two atomic wavefunctions. We conclude that the symmetry of the ground state is imprinted in the high harmonic spectrum.

Finally, we analyzed the high harmonic spectrum to resolve the orbital. Specifically, by analyzing the measurements using a simple classical model we resolve the degree of selectivity achieved during tunnel ionization. We find that tunnel ionization of Ne atoms generates almost a pure aligned state, with a relative population of 90% in the main quantization axis.

Our measurement provides a direct insight into the outcome of tunnel ionization. Therefore, we can extend our approach and apply the probing mechanism to resolve the ionization process. In this approach the atomic wavefunction serves as a spatial probe utilizing its simple structure and static nature.

Ionization in strong laser fields is commonly described by the ADK theory relying on the fundamental work of Keldysh [4]. The Keldysh theory separates two distinct ionization mechanisms: tunnel ionization and multiphoton absorption, characterized by the Keldysh parameter. We perform a systematic study of the ionization process by manipulating the fundamental parameters of the interaction – the laser intensity and wavelength. By using the atomic wavefunction as a probe while tuning the Keldysh parameter we study the two regimes of the ionization process with extremely high contrast.

References:

[1] J. Itatani *et al.*, Nature **432**, 867 (2004).
[2] L. Young *et al*., Phys. Rev. Lett. **97,** 083601 (2006).
[3] D. Shafir *et al.*, accepted to Nature physics (2009).
[4] L. V. Keldysh, Sov. Phys. JEPT **20** (1964).

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