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Watching the emergence of a Fano resonance in doubly excited helium

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Synopsis We report on the experimental observation of the buildup of the 2s2p Fano resonance in helium in the time domain, which has been under theoretical investigation for more than a decade. The emergence of the absorption line is temporally resolved by interrupting the natural decay of the excited state via saturated strong-field ionization at a variable time delay. We compare the experimental data with full *ab-initio* simulations to validate the time-gating by strong-field ionization and thereby confirm the recently developed theory for the formation of Fano line-profiles.

Asymmetric Fano line shapes occur in a variety of research fields ranging from nuclear and atomic physics to condensed-matter physics and photonics. Among the most well-known examples are the resonances in the extreme ultraviolet (XUV) absorption spectrum of doubly excited helium. From the early works on attosecond dynamics in helium [1,2] until today, there has been substantial theoretical interest on how these spectral lines emerge and evolve after the transition is triggered and the subsequent process of autoionization takes place [3-9], also in the presence of strong near-infrared (NIR) laser fields.

Here, we report the experimental observation of the ultrafast formation of a Fano resonance, namely the helium 2s2p spectral line via high-resolution XUV absorption spectroscopy [10]. In order to monitor the buildup of the absorption line, we apply an intense few-cycle near-infrared laser pulse which rapidly depletes the excited state via strong-field ionization. This, in turn, terminates the optical response of the atom, and therefore limits the resonant contribution to the measured absorption spectrum providing a temporal gate between XUV excitation and NIR depletion (see Fig. 1). By realizing rise/fall times of the gate much shorter than the lifetime of the state, and by controlling the time delay between the XUV and NIR pulses with sub-femtosecond precision, we are able to sample the transient buildup of the 2s2p line. In the experiment, we use 7 fs NIR pulses created by a hollow-core fiber and chirped mirror compressor from the 30 fs output of a commercial Ti:sapphire laser system. The NIR pulses are used to generate XUV light in the spectral region around 60 eV, i.e., where the doubly excited states of helium are located with respect to the 1s² ground state, via high-order harmonic generation (HHG) in a neon-filled gas cell. While the XUV pulse duration has not been measured in the current transient absorption setup, due to the high nonlinearity of HHG, it is expected to be considerably shorter than the NIR pulse duration. Therefore, its duration will affect the temporal resolution of our method very little. After HHG, the NIR pulse intensity is controlled by a motorized aperture, which defines the NIR beam diameter but leaves the low-divergence XUV beam unaffected. The phase-locked and co-propagating NIR and XUV pulses are then temporally separated with sub-fs precision by a piezo-driven concentric split-mirror where the inner mirror is predominantly hit by the low-divergence XUV beam and the outer mirror by the NIR beam. Afterwards, a spatial filter unit removes the residual NIR light reflected by the inner mirror and the XUV light hitting the outer mirror.

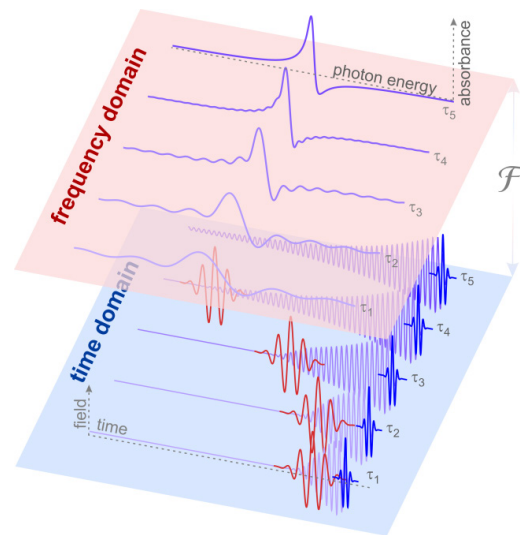


Figure 1. Schematic of the experimental approach. The two planes depict the frequency domain (top) and time domain (bottom) representation of the emerging Fano resonance. The time delay τ between XUV excitation and NIR termination defines the width of the gate within which the resonance is allowed to build up.

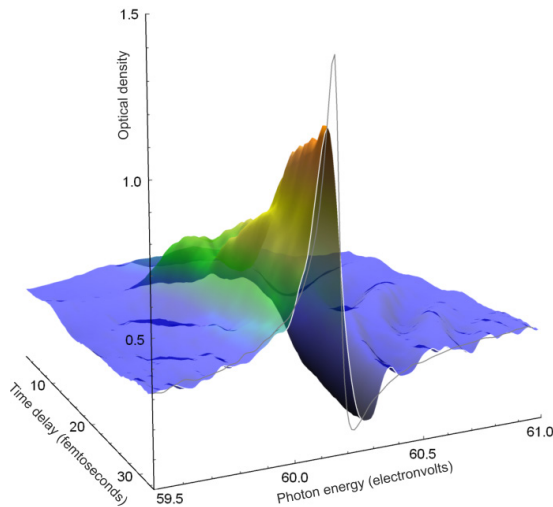


Figure 2. Experimental buildup of the 2s2p absorption line in terms of the time-resolved absorption spectrum given by the optical density of the helium target (colored surface). The reference (gray line) was recorded with minimum NIR laser intensity, which corresponds to the asymptotic line shape for $\tau \rightarrow \infty$. We averaged the experimental data over one NIR cycle in order to suppress the fast beating of the 2s2p state absorbance, which is caused by resonant coupling to the sp_{23+} state [12].

A toroidal mirror is used to refocus the two beams into the target gas cell filled with helium. Finally, the transmitted XUV light is separated from the NIR beam by a metal (here aluminum) filter and spectrally resolved by a flat-field spectrometer. Our spectrometer consists of a variable-line-space grating (producing a flat image plane) and a CCD camera.

Upon excitation of the helium atom by the XUV pulse, the dynamic buildup of the Fano resonance is initiated. The oscillating dipole moment that is driven by the XUV light and associated with the evolution of the two-electron system gives rise to the optical response of the atom. It is the interference of the atomic response with the XUV pulse that results in the spectral signatures in the transmitted light [13]. The absorption cross-section and the optical density of the target are proportional to the imaginary part of the frequency-dependent dipole moment, i.e., the frequency components oscillating out-of-phase with the driving field by $\pi/2$. Quantum-mechanically, the oscillating dipole is caused by a superposition of the ground state and the excited state. An intense and time-delayed NIR pulse is now used to deplete the doubly excited state by means of saturated strong-field ionization, thereby also terminating the dipole response. This confines the contribution of the decaying dipole to the measured spectrum to a temporal gate between the XUV and NIR pulses. By scanning the width of the gate, we can thus monitor the buildup of the Fano absorption profile in time.

Figure 2 depicts the formation of the 2s2p spectral line. For time delays small compared with the lifetime of the 2s2p state (17 fs), i.e., in the temporal region below 10 fs, the measured spectrum is very broad since the XUV-optical response of the doubly excited helium contributes only for a very short time to the formation of the spectral line. This is in accordance with the time-energy uncertainty principle. The line formation does not start at zero delay because of the finite duration of the NIR pulse. Only when the strong-field pulse is delayed by a few femtoseconds the atom is allowed to radiate. Extending the duration of the gate by increasing the NIR delay, the excited atom is allowed to contribute to the absorption spectrum for longer and longer times, which enables the interference between the direct ionization and the autoionizing pathway to build up. This causes the resonance line to become more and more pronounced as the energy is redistributed towards the resonance energy of 60.15 eV. At the maximum experimentally accessible delay of 32 fs, the observed line shape already closely resembles the natural Fano resonance line, which is indicated by the gray line profile in Fig. 2. However, the natural Fano line width is affected by the spectrometer resolution of approximately 50 meV (FWHM) in the photon energy region around 60 eV.

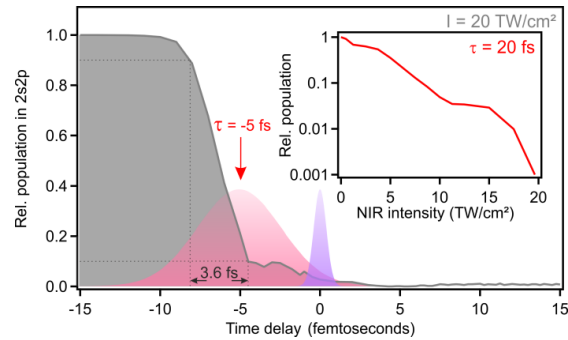


Figure 3. Calculated relative population of the 2s2p state surviving the depletion by a delayed 20 TW/cm^2 NIR pulse. The population for each time delay τ is obtained by projection of the wave function onto the field-free 2s2p state 40.5 fs after excitation. Relative population refers to the actual population relative to the case without NIR interaction. As an example, the NIR preceding the XUV pulse by 5 fs is illustrated. The inset shows the exponential intensity dependence of the survival for a fixed delay of 20 fs .

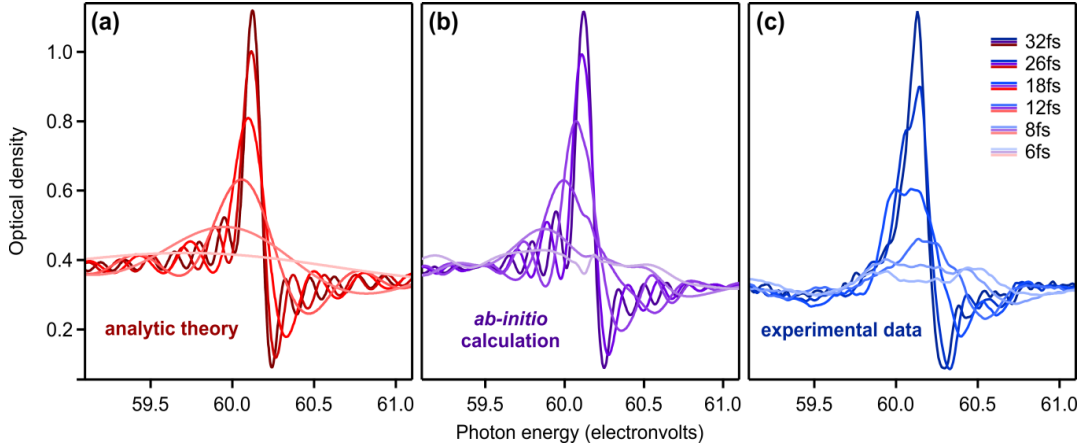


Figure 4. Comparison of the 2s2p Fano line formation in theory and experiment as snapshots for a series of six time delays. (a) Analytically calculated absorption spectra according to Eq. 2. (b) Numerical simulation of absorption spectra where a **7 fs** and **20 TW/cm²** NIR pulse was used to establish the gate closure. (c) Experimental absorption spectra. The spectral amplitude of each set of theory data is scaled to match the experiment at **32 fs**. The analytic spectra are shifted in time delay by **4.5 fs** to compensate for the delayed beginning of the buildup in the case of a finite pulse duration.

From our analytic theory, we predict a 95% match of the peak amplitude of the emerging line and the asymptotic limit to be reached after about 100 fs. Nevertheless, our experiments capture the essential buildup of the Fano resonance despite the currently limited delay range.

Our measurements are compared with numerical calculations and an analytic theory of Fano resonances in order to analyze and confirm the experimental observations [1]. Therefore, we solved the full 3D two-electron Schrödinger equation from first principles using the time-dependent close-coupling method [14,15]. The absorption spectrum is evaluated by means of the dipole spectrum $\tilde{d}(\Omega)$, which is in turn obtained from the resulting time-dependent wave function:

$$\sigma(\Omega) = 4\pi \alpha \Omega \operatorname{Im} \left[\frac{\tilde{d}(\Omega)}{\tilde{F}(\Omega)} \right]. \quad (1)$$

Here, Ω is the frequency and $\tilde{F}(\Omega)$ is the Fourier transform of the combined light fields. In the *ab-initio* simulation we used a 7 fs Gaussian NIR pulse and a 160 as Gaussian XUV pulse.

First, the method of creating a temporal gate by XUV excitation and NIR depletion of the 2s2p state is verified. For this, we study the residual population in the 2s2p state surviving the strong-field ionization as a function of the XUV-NIR delay τ as depicted in Fig. 3. For large negative delays, i.e., $\tau < 10$ fs, the system is not affected by the NIR pulse since the intensity of 2×10^{13} W/cm² is insufficient to modify helium in its ground state. However, already around 5 fs before XUV-NIR overlap, the trailing edge of the NIR pulse leaves only 10% of the 2s2p population bound to the atom. The fall time associated with this depletion, i.e., time to go from the 10% to the 90% level of depletion, is less than 4 fs, which is short compared with the state lifetime of 17 fs. Thus, the gate closure can be expected to be sufficiently fast in order to sample the buildup of the resonance. From around $\tau = 4$ to 5 fs, the surviving population breaches the 1% level and near-complete depletion is achieved, which demonstrates the efficiency of strong-field ionization. In addition, the inset of Fig. 3 shows that the intensity dependence of the depletion process is approximately exponential as would be expected from strong-field ionization. To reach survival levels below 1%, intensities on the order of 2×10^{13} W/cm² are necessary.

With the efficiency of the gating technique demonstrated, we compare the experimental absorption spectra to results calculated from first principles and to an analytic theory developed during the past decade [4,6,9] based on the description by Chu and Lin in Ref. [7]. In order to be directly comparable to the experiment, we employ the following expression which describes the time-dependent formation of the Fano absorption profile:

$$\sigma(\epsilon, \tau) \propto \operatorname{Re} \left[1 + \frac{(q-i)^2}{1-i\epsilon} \left(1 - e^{-\frac{\Gamma}{2}\tau + i\epsilon\tau} \right) \right]. \quad (2)$$

Here, τ is the time delay, q the Fano parameter, and Γ the resonance width. ϵ denotes the scaled photon energy with $\epsilon = 2(E - E_R)/\Gamma$, where E is the photon energy and E_R is the resonance energy. In this theory, the temporal gate is closed suddenly, which corresponds to a Dirac- δ shaped pulse.

Figure 4 presents a comparison between the absorption spectra, employing the analytic theory in (a), the *ab-initio* calculation in (b), and the measured spectra in (c), for a series of time delays. The main conceptual difference among the data is the way the depletion is described. As pointed out already, an infinitesimally short event is used in the analytic calculation. In contrast, a Gaussian laser pulse of 7 fs duration is used in the numerical calculation mimicking the experimental pulse shape which is, however, only approximately known. The three spectra agree well with respect to the line shape and the peak-to-baseline ratios especially at the later delay values where the influence of the NIR pulse shape on the recovered Fano line becomes small. For small time delays, the XUV pulse and NIR pulse partially overlap. Here, we cannot expect perfect agreement with the analytic theory because pulse-overlap effects are absent in this model. In this region, the sequential picture of XUV excitation and NIR depletion does not hold, since the presence of the NIR pulse influences the excitation step already, as the 2s2p state is modified in the strong field. Fig. 4(b) and (c) exhibit good agreement as they display a broad-band winged structure with comparable amplitude whereas the analytic spectrum becomes virtually flat. The good agreement between experiment and *ab-initio* theory across the whole delay range shows that the physics of the time-resolved emergence of the 2s2p Fano resonance is captured by the experiment, and in turn, verifies the theoretical understanding of the buildup.

A complementary study by an independent team of researchers allowed to observe the formation of the Fano resonance in the photoelectron spectrum of the very same transition, which has been reported simultaneously [11].

To summarize, we demonstrated the experimental observation of the femtosecond-buildup of the Fano resonance line shape of the 2s2p autoionizing state in atomic helium. In order to validate the experimental approach, the data is compared with analytical and *ab-initio* calculations, which show very good agreement. Thereby, we confirm the theory on the buildup of Fano resonances, which has been developed over the last decade. The presented method of establishing a temporal gate by excitation and laser-driven saturated ionization is also a promising technique for future studies of ultrafast phenomena, e.g., the creation of quasi particles in solids and the emergence of electron–electron or electron–internuclear correlations in molecules.

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