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SUPPLEMENTARY MATERIALS

www.sciencemag.org/content/354/6313/734/suppl/DC1
Supplementary Text
Figs. S1 to S7
Movie S1
References (*31*–*43*)

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CHEMICAL PHYSICS

Observing the ultrafast buildup of a Fano resonance in the time domain

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Although the time-dependent buildup of asymmetric Fano line shapes in absorption spectra has been of great theoretical interest in the past decade, experimental verification of the predictions has been elusive. Here, we report the experimental observation of the emergence of a Fano resonance in the prototype system of helium by interrupting the autoionization process of a correlated two-electron excited state with a strong laser field. The tunable temporal gate between excitation and termination of the resonance allows us to follow the formation of a Fano line shape in time. The agreement with ab initio calculations validates our experimental time-gating technique for addressing an even broader range of topics, such as the emergence of electron correlation, the onset of electron-internuclear coupling, and quasi-particle formation.

Fano resonances generally occur in the course of excitation of discrete quantum states embedded in and coupled to a continuum (*1*, *2*). As such, they play a fundamental role in nuclear, atomic, molecular, and condensed-matter physics as well as photonics (*3*–*12*). In the prominent example of helium, the discrete doubly excited states are located within different sets of continua, where the prominent 2s2p state is coupled only to the continuum of singly ionized ground-state Helium He^+ (1s). Coulomb interaction among the two electrons leads to autoionization, thus coupling the discrete state and the 1s continuum and giving rise to the famous asymmetric Fano line profiles. Following the early scientific work on attosecond dynamics in laser-driven helium (*13*, *14*), several recent theoretical calculations have predicted the time-dependent formation of Fano resonances (*15*–*20*). However, up to now no such experiment has been performed.

Here, we report a measurement of the time-dependent formation of a Fano resonance in the prototype system of helium. We observed the transient buildup of the 2s2p doubly excited state via extreme ultraviolet (XUV) absorption spectroscopy using high-harmonic radiation. Monitoring the formation of the Fano line was achieved by rapidly terminating the coherent dipole response of the atom via saturated strong-field ionization (SFI) by use of an intense near-infrared (NIR) laser pulse. The key idea is that the NIR pulse acts as a temporal gate of the Fano resonance decay. By varying the time delay between the XUV and the NIR pulse with subfemtosecond precision, we tracked the evolution of the Fano line shape in real time (Fig. 1). To that end, we used laser intensities beyond the Fano-phase control regime discussed in previous work (*7*) to fully deplete the doubly excited state by means of SFI at variable time delays instead of just shifting its phase at a constant (near zero) time delay.

Although recent theoretical work has concentrated on the photoelectron spectrum for accessing the autoionization process, we made use of the fact that state-of-the-art optical spectrometers (*7*, *21*) attain at least an order of magnitude better energy resolution as compared with that of electron spectrometers (*22*), thus allowing for the observation of subtle changes in the spectral profile.

Upon excitation, the XUV pulse triggers the dynamic buildup of the Fano resonance by inducing an oscillating dipole moment, which in turn gives rise to the optical dipole response of the transition. Signatures in the transmitted XUV spectrum are related to the imaginary part of the frequency-domain dipole response (*7*). The time-delayed strong-field NIR pulse is then used to ionize the system, depleting the autoionizing level and ending the buildup process of the spectral line. The experimental results in Fig. 2 show the time-dependent formation of the 2s2p Fano absorption line. For the unperturbed case—in the absence of the NIR pulse as depicted in Fig. 2 (gray line)—we measured the original Fano line shape. The intensity of the 7-fs full width at half maximum (FWHM) NIR pulse was set high enough (~10^{13} W/cm^2) so that the doubly excited states were completely ionized. The dipole oscillation, and with that the resonant optical response of the atom, was thus terminated within the NIR pulse duration. Because this interruption due to SFI is considerably shorter than the lifetime of the state, we can sample the time-dependent formation of the line shape (*20*). For positive time delays τ, the terminating NIR pulse arrives after the XUV excitation pulse. When τ is small as compared with the state's lifetime of ~17 fs, the short duration in which radiation is emitted by the XUV-triggered dipole oscillation is insufficient to form a well-defined Fano line, as can be seen in Fig. 2 for τ less than 10 fs. At $\tau \approx 6$ fs, the effect of the NIR is strongest, and the spectral line is smeared out completely. When the autoionizing state is immediately depopulated after its excitation, the spectral response is mainly determined by the excitation process driven by the attosecond XUV pulse and, because of the fast termination by the NIR pulse, spans several electron volts. This result agrees with several theoretical studies (*15*, *16*) that show that the energy distribution of the electrons ejected within one third of the state lifetime (corresponding to 6 fs in the case of the 2s2p state in helium) after the initial excitation is governed by the frequency range of the excitation pulse. Now, by increasing the time delay τ between excitation and ionization, the doubly excited state has time to decay, and the interference with the direct contributions builds up; the oscillating dipole is granted more and more time to emit the optical response. This gives rise to a narrower

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