



Controlling the XUV Transparency of Helium Using Two-Pathway Quantum Interference

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Atoms irradiated with combined femtosecond laser and extreme ultraviolet (XUV) fields ionize through multiphoton processes, even when the energy of the XUV photon is below the ionization potential. However, in the presence of two different XUV photons and an intense laser field, it is possible to induce full electromagnetic transparency. Taking helium as an example, the laser field modifies its electronic structure, while the presence of two different XUV photons and the laser field leads to two distinct ionization pathways that can interfere destructively. This work demonstrates a new approach for coherent control in a regime of highly excited states and strong optical fields.

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High-order harmonic generation (HHG) is a unique source of ultrashort-pulse light spanning the ultraviolet and soft x-ray regions, that is ideal for initiating and probing fast dynamics in atoms, molecules, and materials [1–5]. Since the high harmonic field is perfectly synchronized to the generating laser field, complex reaction pathways relevant to radiation physics and chemistry can be probed [6–8]. In such highly excited processes, the laser field plays a crucial role in initiating, interrupting, or probing a reaction. However the full influence of the laser field on highly excited reaction pathways is still being uncovered [9,10], despite a wealth of theory and experiments exploring these processes. For example, calculations that model how dynamics are probed by HHG generally ignore the dynamic modification of the electronic structure of the atom or molecule by the laser field itself. Moreover, although He is fully accessible to state-of-the-art theoretical calculations, even a simple two-color ionization process has to date proven to be far from simple to understand [11–16].

In this work we study one of the simplest multiphoton processes: two-color multiphoton ionization of helium in combined infrared (IR) laser and extreme ultraviolet (XUV) high harmonic fields in a regime where two different ionization pathways can interfere coherently: $13\text{th} + 3\omega$ and $11\text{th} + 5\omega$. We show both theoretically and experimentally that this two-color multiphoton ionization process can be viewed intuitively as a double-slit interference experiment in the frequency domain, and that ionization can be turned on or off by manipulating the two interfering pathways (see Fig. 1). In addition to participating in the multiphoton absorption process, the strong IR field also alters the He excited state electronic structure by Floquet state (or dressing state), resulting in energy sidebands [11]. This allows us to adjust the position, amplitude and phase of the two multiphoton

ionization channels, by tuning the XUV fields to the laser-dressed He $2p$ absorption resonance. Most importantly, by analyzing the multiphoton ionization process in the frequency domain, we develop an intuitive understanding of strong field two-color ionization to show, for the first time, that full laser-induced XUV electromagnetic transparency in He is possible.

In the two-photon absorption schemes widely used for coherent control using visible laser fields [17–23], the phase of a weak, nonperturbative IR pulse can control absorption through interfering two-photon transitions. A multicolor, multiphoton XUV + IR ionization process is similar in some respects, except that in this case, the final state is in the continuum. Moreover, we show that a strong IR laser field can play multiple roles in a multicolor, multiphoton, XUV-IR ionization process, as is illustrated schematically in Fig. 1. Specifically, by coherently populating laser-dressed excited states of He with the two tunable XUV harmonics (11th and 13th), and simultaneously using the same IR laser to couple the excited states to the continuum with equal amplitudes but with opposite phases, it is possible to make a He atom fully transparent to multiphoton XUV + IR ionization. The phases of the two interfering channels are controlled by introducing suboptical-cycle delays between the XUV and IR pulses, while the amplitudes are controlled by using the IR field to modulate the electronic structure of the He atom, thus changing the absorption cross sections of the two XUV harmonics through ac and dc Stark shifts that move and broaden the atomic resonances [13].

We note that the modulation of the He⁺ ionization yield, when irradiated by a combined laser and comb of XUV harmonic fields, can also be explained as a time-domain process resulting from interferences between transitions to many different states [12]. This time-domain picture is useful for attosecond (asec) phenomena, whereas the

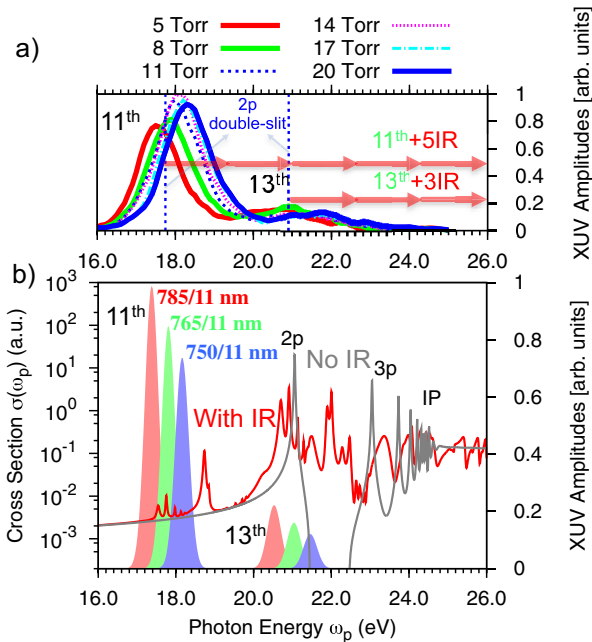


FIG. 1 (color online). (a) Tuning of the harmonics in the frequency domain into the IR-created $2p$ double slit by varying the phase-matching conditions in a xenon-filled waveguide. The dashed vertical lines represent the calculated sidebands of the dressed Floquet $2p$ state of He. The two interfering quantum pathways are $11\text{th} + 5\omega$ and $13\text{th} + 3\omega$. (b) Calculated He excitation cross section, with and without the presence of an IR field of intensity $4 \times 10^{12} \text{ W/cm}^2$. The relative amplitudes of the harmonics are shown on the right axis. By changing the absorption cross section of He using the IR laser field, the amplitudes of the two interfering quantum pathways are controlled.

frequency domain approach presented here is best suited for understanding how to coherently control highly excited processes, including for example, how to induce XUV transparency in atoms.

Our experimental setup consists of a high-power (30 W), high repetition rate (10 kHz) 30 fs Ti:sapphire laser system (785 nm), a gas-filled waveguide for generating harmonics, and a COLTRIMS apparatus which allows for simultaneous detection of ion and electron 3D momenta [13,24–26]. Using part of the laser output, high harmonics are generated in Xe gas and then refocused into a separate He gas target using a pair of multilayer XUV mirrors which reflect photon energies up to $\approx 22 \text{ eV}$. A 100 nm thick Al filter is inserted in the XUV beam to attenuate the driving IR pulses reflected from the XUV mirrors, and to eliminate harmonics lower than the 11th order. The amplitudes of the harmonics can be controlled using different combinations of XUV mirror coatings, while the exact energies of the harmonics can be continuously tuned by the phase-matching conditions via pressure tuning of Xe. Figure 1(a) shows that we can adjust the relative amplitudes of the XUV harmonics to be 4:1 in favor of the 11th harmonic, and change the phase-matching conditions in the waveguide to continuously drive the energy of the 13th

harmonic across the $2p$ resonance in He. Another part of the laser beam is temporally and spatially recombined with the XUV beam in a collinear fashion, and focused onto the He target at a laser intensity of $\approx 4 \times 10^{12} \text{ W/cm}^2$. A linear stage, with 260 assec resolution, was used to delay the IR pulse relative to the XUV pulse. The XUV pulse duration was approximated to be 10 fs.

To predict the amplitude of a multicolor or multiphoton absorption or ionization channel and to compare with our experimental data, we calculate how a weak IR field modifies the absorption cross section of He (at the same IR intensity used in the present experiment), using an approach outlined in [27]. This calculation, shown in Fig. 1(b), provides a photoabsorption map that helps us to compare the relative amplitudes of the two ionization channels. In general the intuitive picture of how an IR field broadens the resonances is confirmed by this calculation: the presence of the IR field shifts and broadens the He resonances, and modifies the electronic structure, leading to a drop in the absorption cross section for the resonant XUV energies, and an increase in the absorption cross section between the resonances. We note that the width of the resonance represents the lifetime or the ionization rate of the dressed state, and the ionization probability is proportional to the absorption cross section. Therefore, in the following discussion, we will not distinguish absorption and ionization. We also see that the IR field modifies the electronic structure of He by inducing fine-structure features in the absorption cross section. For our coherent control of multiphoton XUV + IR ionization, the IR wavelength defines the width of the $2p$ double slit (two IR photons), while the IR intensity defines the position of the double slit in the frequency domain.

To experimentally observe how the IR field modifies the amplitude of the $13\text{th} + 3 \text{ IR}$ photons ionization pathway as a function of the exact XUV photon energy, we monitor the time dependent He^+ yields on the femtosecond time scale, as the 13th harmonic is scanned across the $2p$ resonance. Figure 2(a) plots the He^+ yield for three different XUV 13th harmonic energies below (red [medium gray] curve), at (green [light gray] curve), and above (blue [dark gray] curve) the He $2p$ resonance, as a function of IR laser time delay. The easiest way to see how the IR pulse modifies the absorption cross section of He is to tune the 13th harmonic into the He $2p$ resonance (green [light gray] curve corresponding to a $765/13 = 58.8 \text{ nm}$ XUV wavelength), and compare the ion yields for different time delays of the IR laser pulse. At zero time delay between the XUV and IR fields, absorption of the 13th harmonic occurs in the presence of the IR-modified resonance [Fig. 1(b)]. This significantly decreases the He^+ yield compared with when the IR laser arrives well after the XUV field (positive IR delays). The inset of Fig. 2(a) shows the experimentally observed and theoretically calculated photoelectron momenta at IR laser time delays of 200 fs. A g -wave angular distribution with 4 nodes in the positive p_x direction confirms that the XUV pulse excites

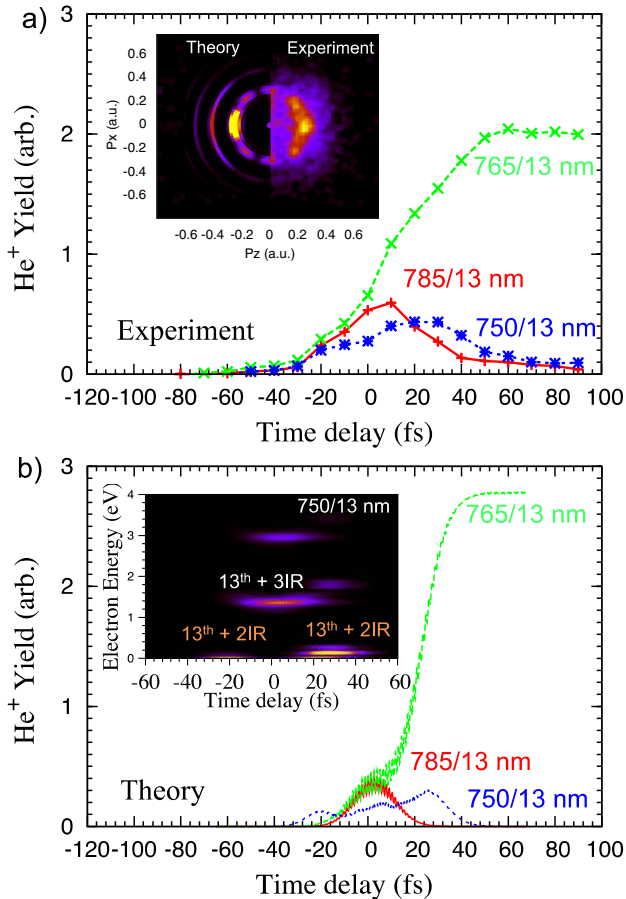


FIG. 2 (color online). (a) He^+ yields for XUV and IR illumination (laser intensity of $4 \times 10^{12} \text{ W/cm}^2$), in the case of the three different two-color XUV spectra of Fig. 1(a). The inset shows the theoretical and experimental photoelectron momenta at 200 fs IR delay, along the negative and positive p_z axis, respectively. (b) Theoretical calculation of the He^+ yield under the same conditions as (a). The inset shows calculated photoelectron energies corresponding to the 750/11 nm and 750/13 nm XUV beams. The 13th + 2ω (2 IR photon) channel appears at ± 25 fs IR delays.

only the $2p$ eigenstate of He, which is then sequentially promoted into the continuum by absorption of at least 3 IR laser photons.

When the XUV photon energy is tuned above the $2p$ resonance (blue [dark gray] curve of Fig. 2(a) corresponding to a $750/13 = 57.7 \text{ nm}$ XUV wavelength), the He^+ yield reveals a slight asymmetry around zero time delay. The enhancement of ionization for the positive IR delays is rather counterintuitive, and has not been observed previously. However, it has very recently been predicted theoretically [13], and can be explained as follows. In combined XUV and IR laser fields of duration 10 fs and 30 fs, respectively, small negative IR time delays correspond to the case of ionization of He on the leading edge of the IR laser pulse (IR pulse arrives later in time than the XUV pulse). For small positive IR delays, He is ionized on the trailing edge of the IR laser pulse (IR arrives before the XUV

pulse). When He is ionized as the laser intensity is increasing, the IR field ponderomotively drives the electron to periodically reencounter its parent ion, making it possible for the continuum photoelectron to recombine. This reduces the overall level of ionization by a surprisingly substantial amount. When He is ionized on the trailing edge of the laser pulse (i.e., 20 fs delay), electrons are released after the peak intensity is gone. This shuts off the recombination channel that effectively reduces the ionization rate.

Figure 2(b) plots the theoretical He^+ yield as a function of time delay between the XUV and IR fields. Good qualitative agreement with the experimental data can be seen. For the three XUV photon energies spanning the He $2p$ resonance, the calculated ion yield exhibits the observed yield asymmetry around zero delay. To explain different asymmetric yields for the two nonresonant XUV photon energies [red (medium gray) and blue (dark gray), Fig. 1(a)], we calculated the photoelectron energies corresponding to the 750/13 nm XUV wavelength. The inset of Fig. 2(b) shows that the additional enhancement at ± 25 fs delay comes from absorption of 2 IR photons. The 13th + 2 IR photons ionization channel only turns on for long relative time delays between the XUV and laser fields. This channel shuts off at higher intensity (around zero delay) because the He ionization potential is ponderomotively increased [13,14].

Armed with the knowledge of the roles the XUV and IR fields play in 2-color multiphoton ionization, we now demonstrate experimentally and theoretically that we can very sensitively control multicolor multiphoton ionization of He by interfering two ionization pathways, with the potential to completely shut off the ionization process. Varying the time delay between the XUV and IR pulses with subcycle resolution introduces ultrafast modulations of the He^+ yield. Experimental and theoretical results for different XUV photon energies and amplitude ratios are plotted in Fig. 3. Figures 3(a)–3(c) show how the amplitude of the oscillation in the ion yield changes as the 11th–13th harmonic pair is scanned across the He $2p$ resonance that forms the Floquet double slit in the laser field, for different amplitude ratios of the 11th and 13th harmonic. Figure 3(d) plots the experimentally observed oscillation in the He^+ yield when the 13th harmonic is below the $2p$ resonance. This measurement is in very good agreement with the superimposed calculated ion yield for the same IR laser intensity of $4 \times 10^{12} \text{ W/cm}^2$, and for a 4:1 amplitude ratio of the 11th and 13th harmonics. Thus, we have confidence in our ability to predict the ionization yield in the two-color, laser-dressed, multiphoton XUV + IR ionization process.

At the IR-dressed $2p$ resonance and for an 11th:13th harmonic amplitude ratio of 256:1, we observe a near perfect modulation of the He^+ yield, with the maximum-to-minimum ratio of ≈ 40 [red curve of Fig. 3(a)], thus demonstrating the potential to completely shut off the multiphoton ionization process. For 785/13 nm wavelength XUV beams [red curve Fig. 3(b)], the ratio

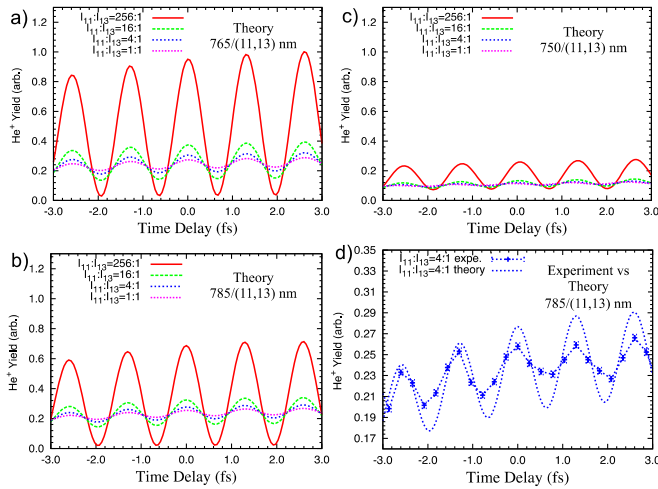


FIG. 3 (color online). (a)–(c) Calculated modulation of the He ionization probability for different ratios of the 11th:13th harmonic fields, and for two-color 765/(11, 13) nm, 785/(11, 13) nm, and 750/(11, 13) nm XUV beams, respectively. (d) Experimental and theoretical modulation of the He⁺ yield for a laser intensity of 4×10^{12} W/cm², and a 785/(11, 13) nm XUV field. The red curve of (a) demonstrates that a ratio of 256:1 for the 11th vs the 13th harmonic can result in nearly complete cancellation of XUV absorption.

is ≈ 30 , while for 750/13 nm XUV beams and 11th:13th ratio of 256:1 [red curve Fig. 3(c)], the modulation of the ion yield drops to only 5. We note that in the latter case, the maximum yield is slightly shifted from zero delay, unlike the other two cases that both maximize at zero. The highest amplitude of the oscillation, leading to complete electromagnetic transparency at the correct relative time delays, occurs for the case when the two XUV photons excite the He 2*p* Floquet states exactly through two sidebands (two Fourier components), as shown in Figs. 1 and 3(a).

The intuitive Young double-slit picture is that for harmonic photon energies of 765/11 nm and 765/13 nm, the two XUV photons go through a double-slit interference in frequency space that is created by the IR laser field [Fig. 1(a)]. Below the resonance, the photon pair does not go fully through the double slit, but still partly overlaps with the 2*p* sideband at 17.7 eV and 20.9 eV, as seen in Fig. 1(b) [and more robustly in Fig. 1(a)]. Above the 2*p* resonance, the two XUV photons almost completely miss the double slit [Figs. 1(a) and 1(b)], which results in a small modulation in the ionization yield in Fig. 3(c). Moreover, in this case the 3*p* Floquet sidebands start playing a role, making the two XUV photons to go through two double slits in series, which are displaced with respect to each other in the frequency domain. This can be seen in Fig. 1(b), where the 13th harmonic overlaps with the 3*p* (and higher) IR-dressed states at 21.5 eV. This also results in shifting of the maximum yield from zero delay [Fig. 3(c)].

In conclusion, we demonstrate that full electromagnetic transparency to XUV light in He can be achieved by destructively interfering two distinct multiphoton ionization

pathways. Through combined theory and experiment, we show how a laser field can modify the electronic structure of an atom, optimizing the destructive interference between two ionization pathways under the correct conditions. This approach opens up new possibilities for coherent control of highly excited states, and emphasizes the important and complex role the IR laser field plays in strong field ionization. We believe that this concept can be applied to induce and control the outcome of chemical reactions, as well as in condensed matter physics where the concept of the resonance dressing, shifting, and broadening can be applied to the electronic band structure.

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