Attosecond Electron Wave-Packet Interference Observed by Transient Absorption

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We perform attosecond time-resolved transient absorption spectroscopy around the first ionization threshold of helium and observe rapid oscillations of the absorption of the individual harmonics as a function of time delay with respect to a superimposed, moderately strong infrared laser field. The phase relation between the absorption modulation of individual harmonics gives direct evidence for the interference of transiently bound electronic wave packets as the mechanism behind the absorption modulation.

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The availability of light pulses in the extreme ultraviolet (XUV) spectral region and with attosecond duration [1,2] enables the direct observation of the dynamics of electrons on their natural time scale. Isolated attosecond pulses [1] are usually preferred for traditional pump-probe measurement schemes. For example, they have been used for studying the dynamics of various ionization processes in atoms [3–5] and on the surface of solids [6]. Attosecond pulse trains (APTs, [2]) on the other hand allow for probing the interference of electron wave packets initiated by the individual pulses in the train [7–9]. The interference of subsequent wave packets enhances the sensitivity of the method and enables the extraction of phase information.

Recently, the ionization probability of helium atoms with APT photon energies below the ionization threshold in the presence of a time-delayed infrared (IR) field was studied [8]. It was found that the ion yield is modulated with twice the IR driving laser frequency. The result was explained by the interference of transiently bound electron wave packets (EWPs). We investigated this physical system with the all-optical approach of attosecond time-resolved transient absorption spectroscopy. Instead of detecting ions or electrons, we spectrally resolve the transmitted harmonic radiation in the presence of a time-delayed IR field. Our experiment probes a different physical observable than electron or ion detection and reveals more insight into the time-dependent perturbation of the atom by the IR field including contributions from bound-bound transitions. While transient absorption is a well-known measurement technique in the femtosecond domain (see, e.g., [10] for transient absorption with high-order harmonic sources), it was only recently extended into the attosecond regime using isolated attosecond pulses [11]. The different aspects probed by our optical technique compared to the experiment by Johnsson *et al.* [8] allows us to verify the plausibility of the previously introduced intuitive theoretical model and to demonstrate attosecond control of the relative absorption of the individual spectral constituents of an APT. This rapid absorption modulation represents the fastest dynamics resolved by all-optical methods so far.

Figure 1 shows the experimental configuration (a) and the spectral location (b) of the APT with respect to the energetic structure of helium. The APT is generated by high-order harmonic generation (HHG) in a xenon target with 30-fs pulses from a Ti:sapphire laser centered at 800 nm wavelength. The plasma induced spectral blueshift in the xenon target results in an apparent driving wavelength of 785 nm. The remaining IR radiation is removed after the HHG source by passing the beam through a thin aluminum filter. A small part of the original Ti:sapphire laser beam is sent over a second independent optical path of the setup that can be time delayed with respect to the

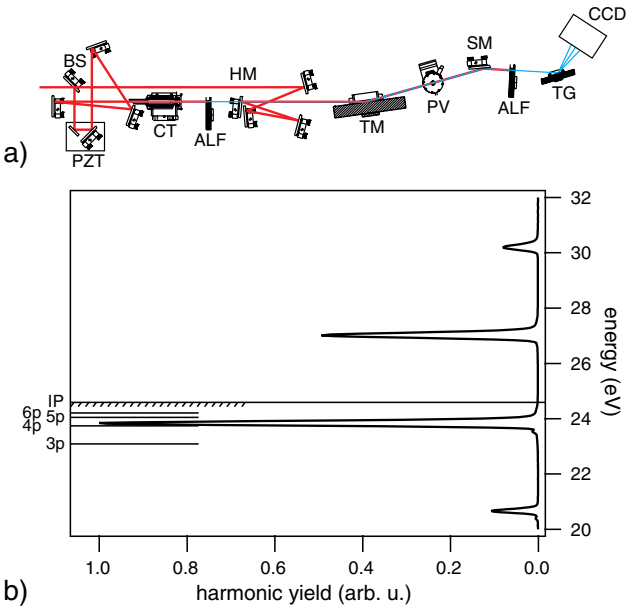

FIG. 1 (color online). Experimental configuration. (a) Setup for attosecond transient absorption. BS, beam splitter; PZT, piezo translation stage; CT, capillary target; ALF, Al-filter; HM, mirror with center hole; TM, toroidal mirror; PV, pulsed valve; SM, spherical mirror; TG, toroidal grating; CCD: XUV-CCD. (b) Optical spectrum of the APT in relation to the energetic structure of helium. Only the field-free p levels and first ionization potential are shown.

APT. This relatively weak IR beam is recombined collinearly with the APT on a mirror with a center hole and then focused into a dense helium target (particle density $\approx 5 \times 10^{17}$ cm^{-3}). The harmonic photons transmitted through the target are collected and sent into a XUV spectrometer equipped with a CCD camera. It is important to note that the infrared intensity chosen in the helium target (in the range from 0.7×10^{13} W/cm^2 to 1.7×10^{13} W/cm^2) alone is not sufficient to induce strong-field processes in ground-state helium. Furthermore, our APT consists mainly of harmonic orders 13 and 15, which are energetically located below the field-free ionization potential (IP) of helium and harmonic 17, which is above the IP [see Fig. 1(b)]. As a result, harmonic 13 and 15 are not absorbed in the absence of the infrared field unless they overlap with internal resonances of helium. The latter is the case only for the weak spectral wings of harmonic 15, which can populate the *1s4p* and the *1s5p* from the helium ground state by single-photon absorption. The helium pressure is set such that about 50% of the photons at harmonic 17 are absorbed in the field-free case. The pulses in the APT were characterized with the RABITT technique (reconstruction of attosecond beating by interference of two-photon transitions, [2,12]) to assure that the individual harmonics indeed form an APT. The average pulse duration in the train was found to be ≈ 380 as. The full width at half maximum of the pulse train envelope can be estimated to be on the order of 10–12 fs, which would contain 8–10 attosecond pulses.

With an experimental setup and parameters similar to ours, Johnsson *et al.* observed periodic modulations of the He^+ ion yield as they changed the relative delay between the APT and the IR field [8]. The modulation period corresponds to twice the fundamental frequency of the IR pulse. If they replace the helium target gas by argon, the modulations are absent. Johnsson *et al.* explain their observation by two effects. First, the oscillating IR field distorts the atomic potential and thereby modifies the absorption probability for the APT photons depending on the relative delay between the two fields. Second, transiently bound EWPs initiated by the bursts forming the APT are accelerated by the IR field and are returned with a certain probability back into the vicinity of the ion when a subsequent attosecond pulse excites a next wave packet. These processes are repeated with a half-cycle periodicity due to the symmetry of the system. The EWPs are only transiently bound because the potential barrier may be sufficiently lowered for proper delay between the infrared and the APT such that the wave packets can escape into the continuum. The interference between wave packets excited by subsequent attosecond pulses significantly alters the ionization probabilities with delay. It is found that the second effect dominates the first one by an order of magnitude. This theoretical model is schematically pictured in Fig. 2. In this study, Johnsson *et al.* also found theoretically

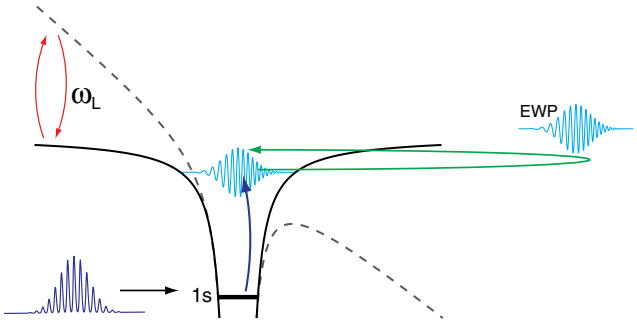

FIG. 2 (color online). Schematic of the electron wave-packet dynamics generated by an APT in the presence of an additional IR field. A pulse in the APT creates a transiently bound EWP in the potential modified by the IR field. This EWP interferes with the EWP created by a subsequent pulse in the APT, rendering the process sensitive to the phase accumulated in the IR field.

that only 60%–70% of the population transferred out of the ground state by the XUV radiation is ionized in the presence of an IR field, whereas roughly 30%–40% of the population promoted by the XUV light remains in an excited state. The probabilities of absorbing a photon, generating a continuum electron, or promoting the electron into a bound excited state furthermore exhibit a phase shift with respect to the pump-probe delay. Since the detection of He^+ ions is only sensitive to processes leading to the ionization of an atom, no complete information on the relative phases and amplitudes of the absorption of photons can be derived from such a measurement. Rivière *et al.* introduced a minimal analytical model for the total photon absorption probabilities based on the strong-field approximation [13]. They found that the replicated EWPs can be factorized into two terms: A term that depends on the number of IR cycles the APT spans (which is a comb function and acts as a momentum filter) and a term that depends on the number of attosecond pulses per laser cycle. In their model, the modulation of the absorption probability is assigned exclusively to the interference of electron wave packets. An alternative theoretical description based on Floquet theory arrives at similar conclusions [14]. It directly addresses the differences in the behavior with argon and helium target gas and compares the single attosecond pulse to APT excitation.

Two main observations in the ion yield experiment were considered indicative for the transiently bound wave-packet interference model. First, the modulation contrast was considerably higher than that expected for a single attosecond excitation pulse. Calculations showed that the interferences enhance this contrast by an order of magnitude for an APT with a 10 fs FWHM envelope and bring it to the level observed experimentally. Second, the strongly directional emission seen in the two-dimensional photoelectron momentum distribution along the direction of the IR field polarization was found to be consistent with the fact that the electrons are ejected preferentially along

the directions where the potential barrier is lowered by the IR field. For electrons ionized well above the ionization potential, such directionality is not observed as can be verified by replacing the helium target gas with argon.

With transient absorption we can now examine the impact of the IR field on the absorption probability of the XUV photons and thus the influence of a time dependence in the transition operator much more completely. Transient absorption allows us to see which photon energies get absorbed and to what extent and at which APT-IR delays. Figure 3 shows the transmitted HHG photons as a function of delay. It can be clearly seen that the transmission (or absorption) of the harmonics is modulated at twice the infrared fundamental frequency. This modulation survives spectral integration [see Fig. 3(a)] even though the individual modulations exhibit a phase shift with respect to each other. This must be the case because the total absorption is directly related to the ion yield detected in the experiment by Johnsson *et al.*. However, the relative phases seen in the spectrally resolved case yield additional information allowing the verification of the proposed theoretical models. Figures 3(b) and 3(c) show the modulations on the individual harmonics 13 and 15, respectively. The plotted values were each detected by a single CCD column located on the respective harmonic spectral peak. The pixel columns cover approximately 20 meV photon energy in width. From the simple picture shown in Fig. 2 (as well as the analytical model and the Floquet-theory based approach), we can derive the following predictions for the interference of subsequent wave packets: The temporal phase of each pulse in the APT directly maps onto the EWP upon its birth. The so-called ''atto-chirp'' [15] describes phase contributions that are identical for all pulses in the train. The interference of wave packets generated by subsequent attosecond pulses should thus not depend on the atto-chirp. On the other hand, the dominant influence on the interference of a previous wave packet with a newly created one should come from the phase accumulated on its excursion into the continuum. The phase picked up by the EWP during the acceleration in the electric field of the infrared pulse depends sensitively on the laser intensity [16]. Any change in the interference conditions between EWPs from subsequent attosecond pulses should manifest itself in a change in the observed absorption modulation pattern.

In order to test the first prediction, we varied the thickness of our aluminum filters used to separate the APT from its generating infrared field. Varying the filter thickness by hundreds of nanometers is known to change in good approximation only the spectral phase difference between neighboring harmonics [17]. In the time domain, this harmonic-to-harmonic phase variation corresponds to the atto-chirp. Larger amounts of dispersion that would lead to non-negligible pulse-to-pulse phase variation were not investigated in our experiments. Figures 3(b) and 3(c) compare the transmitted photon yield for harmonics 13 and 15 as a function of APT-to-IR delay for aluminum filter thicknesses of 500 nm and 100 nm. Both data sets were recorded at identical infrared intensity of 1.3×10^{13} W/cm^2 with a step size of 107 as over 1400 steps and an exposure time of 50 ms and 10 ms per spectrum, respectively. For the 500 nm case, harmonics 13 and 17 are oscillating in phase (phase difference 0.1 rad), whereas harmonics 13 and 15 are shifted by 2.5 rad with respect

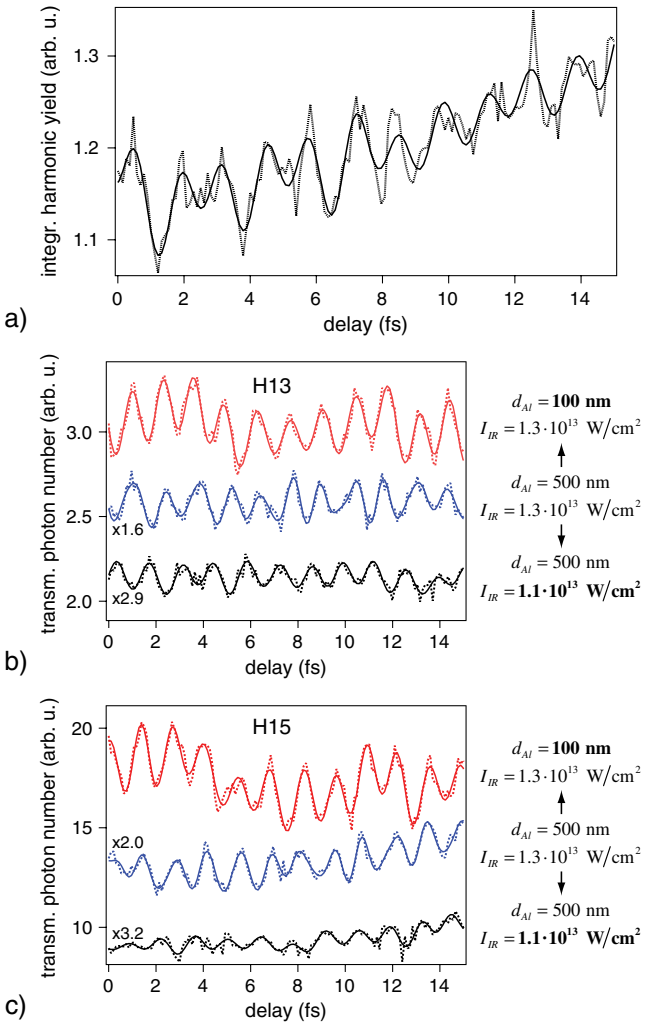

FIG. 3 (color online). Transmitted harmonic photon yields with respect to APT-IR-pump-probe delay. All harmonics are modulated at twice the fundamental laser frequency. These modulations are robust against spectral integration (a), which is expected from the He^+ yield measurements by Johnsson *et al.* [8]. In the spectrally resolved data, it can be seen that the phase of the modulations on individual harmonics remains unchanged for different thicknesses of the Al filter used [(b), harmonic 13; (c), harmonic 15]. The phase, however, sensitively reacts to a change in the IR intensity. The dotted lines correspond to the measured data, the solid lines to a signal smoothed by Fourier low-pass filtering. The black and blue traces in (b) and (c) have been rescaled for easier visual comparison as indicated next to the corresponding trace.

to each other. If the filter is replaced by the 100 nm version, then the relative phase between harmonics 13 and 17 amounts to 0.2 rad, while a phase shift of 2.2 rad is observed between harmonics 13 and 15. The differences between these two cases are within experimental uncertainty as was verified by repeating the measurement with the 500 nm filter after the 100 nm filter run. The phase values have been extracted from the measured data by Fourier analysis and were found to be reproducible within 0.5 rad. For comparison, the linear dispersion of Al changes the spectral phase difference between harmonics 13 and 15 of the incoming APT by 1.3 rad and between harmonics 13 and 17 by 1.74 rad. The observation of negligible phase change in the absorption modulations for significant filter thickness change is consistent with the theoretical pictures outlined above.

In order to verify the predicted sensitivity of the modulations towards IR intensity changes, we performed one measurement run with the 500 nm aluminum filter but at a lower intensity of 1.1×10^{13} W/cm^2 [black lines in Figs. 3(b) and 3(c)]. This time, even with the intensity being lowered by merely 15%, the relative phase between the transmitted photon yield modulations changes considerably by an amount well beyond experimental uncertainty. The relative phase between harmonics 13 and 17 is now determined to be 1.6 rad, while the modulations on harmonic 13 and 15 are shifted by 3.0 rad with respect to each other. As expected qualitatively from the theoretical models and the schematic picture in Fig. 2, the infrared laser intensity has a strong effect on the electron wave-packet interference conditions. This is due to the fact that the interference occurs between an EWP having accumulated phase during acceleration in the IR field and an EWP having just been excited from the ground state by the following attosecond pulse. The relative phase is thus always being governed by the contribution from the IR driven EWP excursion. The atto-chirp phase contribution, on the other hand, is identical for each EWP and thus cancels upon interference.

Our experiment thus provides strong evidence for the transiently bound wave-packet interference picture and is in agreement with all three theoretical descriptions developed so far. This makes this experiment the first all-optical observation of attosecond wave-packet interferences. Transient absorption offers a valuable alternative view of the physical system as compared to photo-ion or photoelectron detection. In addition, we find that the sensitive optical detectors allow for rapid data acquisition with good signal-to-noise ratio. The insensitivity towards space charge effects enables us to work with higher target gas densities for an additional boost in signal levels. It needs to be noted, however, that the theoretical frameworks needed for the calculation of transmitted photon spectra in strong-field laser-matter interactions do not yet exist, particularly not in a regime close to the ionization threshold where many traditional approximations break down.

In conclusion, we have used attosecond transient absorption of an APT in the presence of a moderately strong infrared field in a dense helium target to investigate the interference of transiently bound electron wave packets. Our measurements provide more complete insight into the influence of the ultrafast perturbation of XUV transitions in an atom irradiated by an IR field. We find that the relative phases between the rapid modulations of the transmitted photon yield at harmonics 13, 15, and 17 sensitively depend on the infrared intensity but remain unchanged within experimental uncertainty for changing atto-chirp of the APT. By changing the IR-APT delay and the IR intensity, the relative absorption of the individual harmonics can be controlled with high precision. This can be considered a form of absorptive pulse shaping of the original APT. Furthermore, our experiment demonstrates that transient absorption can provide more physical insight into the attosecond dynamics of suitable systems compared to the more traditional electron or ion detection. With improved theoretical approaches for the modeling of the optical response of physical systems on this time scale, it will become possible to extract even more information from such data and to obtain a better quantitative understanding of the processes under investigation.

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