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Subcycle laser control and quantum interferences in attosecond photoabsorption of neon

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The dynamics of an atom in a strong infrared laser field (1013 W/cm2) result in substantial changes to the field-free electronic energy levels, which can be probed on time scales shorter than the laser cycle using isolated attosecond pulses. Here, we measure the transient absorption of an isolated attosecond pulse by laser-dressed bound states of neon near the first ionization threshold. The observed subcycle changes in the absorption spectrum result from both laser-induced ac Stark shifts and from quantum interferences between different multiphoton excitation pathways. We further demonstrate the ability to experimentally turn off the quantum interference mechanism by eliminating one of the interfering pathways.

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I. INTRODUCTION

Photoabsorption has long been used for spectroscopic study of atomic and molecular structure and for remote identification of chemical species. With the advent of ultrafast laser pulses, time-resolved transient absorption spectroscopy made it possible to study molecular processes on the ∼10-fs time scale [1]. In transient absorption spectroscopy, a relatively weak probe laser pulse is used to monitor the changes in the absorption spectrum induced by a strong pump pulse with a variable time delay. Recently, the development and application of isolated attosecond pulses [2] has allowed probing of the atom with unprecedented time resolution, and attosecond transient absorption spectroscopy (ATAS) has been demonstrated to time-resolve electron dynamics on the femto- and even suboptical-cycle [7–9] time scales.

Full numerical simulation and intuitive physical understanding of ATAS experiments has so far proved difficult. For this reason, most experiments were interpreted more generally, using a three-level atom [4,10–13] or other model systems [7] to explain the dominant features of the experiments. For recent ATAS experiments in bound states of helium [14–16], however, ab initio solutions of the time-dependent Schrödinger equation (TDSE) have also been obtained [17], which have helped to unify the existing theory and to demystify several previously unexplained features. However, helium is still a relatively simple (three-body) system, and it is unclear whether the theoretical models can be applied to more complex targets which are not exactly solvable. Therefore, before ATAS can be applied to molecular systems or condensed matter, further experiments on many-electron atomic systems are needed in order to test the current theoretical models. In this work, we extended the attosecond transient absorption technique to the neon atom, which contains ten electrons, and which can be solved numerically only by using a model potential in the single-active-electron (SAE) approximation. With neon as the absorption gas, we observed large laser-induced ac Stark shifts, laser-induced absorption structures corresponding to multiphoton excitation of dipole-forbidden transitions, and subcycle oscillations of the absorption strength, which in most cases can be explained using the same theoretical description as in helium ATAS experiments. These experimental features can also be reproduced quite well by numerical calculation of the TDSE under the SAE approximation. Additionally, we confirmed by both experiment and calculation that the subcycle oscillations of the absorption strength result from quantum interference between two distinct pathways of excitation to a final state.

II. EXPERIMENT

The experiments were carried out on the same ATAS setup base that was used previously by our group [15]. The single isolated attosecond pulses (SIAPs) with pulse duration of ∼200 as were generated by the interaction of 4–6 fs near-infrared (NIR) laser pulses centered at 730 nm (photon energy ωL ≈ 1.7 eV) with a xenon-filled gas cell by using the generalized double-optical-gating (GDOG) technique [18]. After being separated from the residual NIR laser by a 200-nm aluminum filter, the SIAPs were focused by a toroidal mirror into a second neon-filled gas cell with inner diameter of 1.5 mm. A hole-drilled mirror was placed between the toroidal mirror and the second gas cell to combine a time-delayed replica of the driving NIR pulse with the SIAP. After the absorption cell, the transmitted portion of the SIAP was sent to a home-built XUV spectrometer [19], in which the SIAPs were dispersed by a flat-field grating and detected by a microchannel plate (MCP)–phosphor detector which was imaged onto a thermoelectrically cooled CCD. The XUV spectrometer resolution was better than 50 meV in the energy range of interest, as determined by measuring the bound-state absorption lines near the ionization thresholds of neon, argon, and helium.

The absorption cell was backed with 40 Torr of neon gas to absorb ∼75% of the spectrum above the neon ionization threshold in the absence of the NIR laser. The intensity of the NIR laser was controlled by an iris and determined from the measured laser pulse energy, duration, and focal spot size. The delay between the NIR and SIAP was introduced by

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a piezoelectric transducer (PZT), and a cw green laser was copropagated with both the SIAP and the delayed NIR laser to stabilize and control the delay \[20\] with a delay step size of 140 as and a stability of 50 as rms. At each delay step 10 000 shots were accumulated to increase the signal-to-noise level. The carrier-envelope phase of the driving laser was of no importance in our measurement, since the SIAP generated using the GDOG technique is naturally synchronized to the field oscillation of the driving NIR pulse \[21\].

The ground state of neon can be described by the electron configuration \(1s^22s^22p^6\), and higher bound energy levels are formed by excitation of one valence electron resulting in a configuration of \(1s^22s^22p^n l\). The energy levels of interest along with the measured absorption cross section are shown in Fig. 1. Due to dipole selection rules, absorption of an XUV photon can lead to excitation of a \(2p\) electron to the discrete \(ns\) or \(nd\) energy levels or to the \(s\) or \(d\) continua above the ionization threshold \(I_p\), as indicated in Fig. 1. The introduction of a moderately intense NIR laser field, as in ATAS experiments, results in energy-level splitting and shifting, which can be observed through changes in the positions and shapes of the absorption features \[17\]. Therefore ATAS measurements enable us to resolve the evolution of laser-perturbed atomic energy levels on time scales comparable to or even shorter than the laser optical cycle.

Figure 2(a) shows the measured absorbance (proportional to the absorption cross section) of neon atoms perturbed by a \(\sim 10\) TW/cm\(^2\) laser field as a function of the time delay between the SIAP and the NIR field. For negative delays, where the SIAP arrives on the target before the NIR pulse, we can observe absorption lines corresponding to electron transitions from the ground state to excited states, as indicated in the figure. The broad continuous spectrum of the SIAP allows us to observe the absorption features corresponding to each bound state, as well as the above-threshold absorption, simultaneously, while the high resolution of the XUV spectrometer allows us to resolve the contribution of each state individually. As the delay between the two pulses approaches zero, the energy levels exhibit several interesting features. Firstly, the absorption cross section in the vicinity of the \(1s^22s^22p^3s\) state oscillates with a period of half of the NIR cycle \(\sim 1.3\) fs. Additionally, new laser-induced absorption structures are observed above the ionization threshold (near 22 eV) near zero delay. Finally, strong subcycle ac Stark shifts are observed near zero delay for higher excited states, similar to those previously observed in helium atoms \[7\].

For negative delays, there is no clear observation of the ionization threshold, with a steplike increase in the absorption. Instead, we observe that the near-threshold absorption lines exhibit energy- and delay-dependent oscillations, appearing to shift above the ionization threshold as the delay approaches zero. The absorption edge associated with the ionization threshold appears only at positive delays. This feature was also observed in previous experiments in helium \[15\], and is likely because the near-threshold excited states of neon can be easily ionized by the delayed NIR field by single-photon absorption. This process is remarkably similar to the perturbed free polarization decay previously observed in femtosecond transient absorption experiments \[22\] and in the vicinity of the \(1s2p\) state of helium \[14\], which results in the formation of absorption sidebands with energy spacing inversely proportional to the time delay. In order to investigate this process in more detail, we increased the NIR laser intensity to \(\sim 75\) TW/cm\(^2\), so that the perturbation to the highly excited states is much stronger. The delay-dependent absorption spectrogram is shown in Fig. 2(b), wherein multiple absorption sidebands are observed between \(\sim 21\) and \(\sim 22.5\) eV in negative delay.

III. THEORETICAL CALCULATIONS AND DISCUSSIONS

The theoretical calculations were performed by solving the TDSE in the integral form \[23\] under the SAE approximation, with the model potential obtained from the density functional theory with self-interaction correction \[24\]. The Fourier transform of the autocorrelation function (overlap of the time-dependent wave function with the group state weighted by transition dipole) is proportional to the photoabsorption cross section as detailed in \[25\] for a given time delay and...
external field. With the obtained time-dependent wave function $\Psi(t)$, we calculated the photoabsorption cross section as

$$
\sigma(\omega) = \frac{4\pi}{cS(\omega)} \left| \int_{-\infty}^{\infty} \langle \Psi(t) = -\infty | \Psi(t) \rangle e^{i(\omega t + \epsilon_{gs}) t} dt \right|,
$$

with $\omega$ the photon energy, $S(\omega)$ the Fourier component of SIAP, and $\epsilon_{gs}$ the ground-state energy. Different from Ref. [25], here we do not need to average the autocorrelation function over one IR cycle, since short pulses were used in the experiment. In the simulation, 1000 radial grid points and 32 partial waves were used. The numerical convergence has been checked by varying simulation parameters. The calculated photoabsorption cross sections of neon as a function of the time delay are shown in Figs. 2(c) and 2(d) for NIR intensities of 10 TW/cm$^2$ and 75 TW/cm$^2$, respectively. All the prevalent features observed experimentally in Figs. 2(a) and 2(b) also appear in the calculated results. We note that the absorption lines corresponding to the excited states of neon are slightly shifted in energy from those in the experiment, which is due to the model potential used in the calculations. However, the behavior of each energy level is remarkably similar to that observed in the experiment.

Although many interacting states are present in both the experiment and calculation, the dominant features can be explained in terms of the interactions of relatively few states. For example, the origin of the half-cycle oscillation in the 1s$^2$2s$^2$2p$^3$3s state (16.8 eV), which extends to negative delays, is illustrated schematically in Fig. 3(a). Here, there are two distinct pathways for electronic transitions between the ground state and 3s state. When the SIAP arrives on the target, the absorption of XUV photons within the spectrum of the SIAP can result in excitation of several ns and nd states, one of which is 3s. Additionally, the 3d state (20.1 eV), which is energetically separated from the 3s state by 2$\omega_{L}$, can be populated. When the delayed NIR laser arrives, the 3d state population can be transferred to the 3s state by emission of two NIR photons, assisted by the intermediate 3p state. These two pathways finally lead to interference in the delay-dependent absorption cross section. Because both of these pathways are allowed as long as the SIAP arrives on the target before the NIR laser, the interference oscillations in the measured absorption strength extend to large negative delays, as indicated by the black curve in Fig. 3(c). On the other hand, the laser-induced 4p$^+$ absorption structure shown in Figs. 2(a) and 2(b) near 22 eV is localized near zero delay. This is because this structure arises from a dipole-forbidden transition (2p to 4p), which cannot be populated by the attosecond pulse alone, as illustrated in Fig. 3(b). In the moderately intense NIR field, the 4p state forms two Floquet-like sidebands separated from the 4p$^-$ by $\omega_{L}$, which are marked as 4p$^+$ and 4p$^-$ in Figs. 2 and 3. Therefore the 4p state can be populated by simultaneous absorption of an XUV photon with energy $E_{4p} \pm \omega_{L}$ and absorption or emission of an NIR photon, resulting in laser-induced absorption structures near zero delay. In both experimental and theoretical spectrograms, only the 4p$^+$ structure could be observed, since 4p$^-$ (18.6 eV) is not close to any ns or nd bound states while 4p$^+$ overlaps with the continuum states. The half-cycle oscillations in the absorption strength near the 4p$^+$ structure, shown in the gray curve in Fig. 3(c), also result from interference between multiple quantum paths, since the 4p$^+$ state can be reached either directly by absorption of one XUV photon or indirectly by initial excitation of the 4p$^-$ state, followed by absorption of two NIR photons. Note that the Floquet-like sidebands exist only if the laser is present, so the light-induced structures as well as the corresponding interferences appear only near zero delay.

Previously, the role that a particular state plays in ATAS experiments was identified through the use of test calculations which dynamically eliminated the state during the time propagation of the TDSE [14,15]. The same method was adopted here to confirm the origin of a 4p$^+$ structure by individually removing 3p, 4p, and 4f states in the test calculations. These calculations confirm that 4p$^+$ absorption results from the laser-induced sideband of the 4p state. We additionally adopt an experimental technique to confirm the origin of the interference in the vicinity of the 3s state by suppressing the indirect pathway. In order to achieve this,
the aluminum filter used to filter out the SIAPs was replaced with a 200-nm-thick indium filter, which allows transmission of XUV pulses only within the spectral range from 11 to 17 eV. In this case, no delay-dependent quantum path interferences are observed.

In conclusion, we measured the transient absorption of an isolated attosecond pulse by laser-dressed bound states of neon atoms. The observed subcycle changes in the neon absorption spectrum result from both subcycle ac Stark shifts and from quantum interferences between different multiphoton excitation pathways. New laser-induced absorption structures corresponding to multiphoton excitation of dipole-forbidden transitions were also observed. Furthermore, we experimentally demonstrated the ability to turn off the quantum interference observed in the 3s state by removing the indirect excitation pathway via the 3d state. Although direct numerical simulation of the ten-electron neon atom is still impossible, we demonstrate that the dynamics can still be interpreted through TDSE simulations by relying on the SAE approximation and through relatively simple models involving few interacting states. The applicability of these existing theoretical models to the present neon ATAS experiments paves the way to extend the ATAS technique to more complex systems.

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