Local enhancement in transient absorption spectroscopy by gating the resonance in the time domain

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The concept of resonant perfect absorption, enabled by the combined action of pulse propagation and an auxiliary gate pulse, was recently proposed and demonstrated in a group of two-level systems [Y. He *et al.*, Phys. Rev. Lett. **129**, 273201 (2022)]. Here we exploit this method in a more realistic scenario by solving the coupled time-dependent Schrödinger equation and the Maxwell wave equation in helium. Through emptying the population of the $1s2p$ excited state after its excitation, we explore the evolution of the spectral profile with time delay and propagation distance and link the observations to the controlled interference between the original field and the gated new field. We find that resonant absorption for higher-lying states can also be strongly enhanced, in spite of the congestion of multiple resonances and the presence of complex laser-induced couplings. Our results show that interferometric control of absorption using intense laser fields can be applied selectively in both the temporal and spatial domains.

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I. INTRODUCTION

Transient absorption spectroscopy (TAS) has become an established technique for tracking and steering ultrafast quantum dynamics. Facilitated by the development and widespread availability of extreme-ultraviolet (XUV) and even higher-frequency sources from high-harmonic generation and free-electron lasers, steady progress has been made in the observation and understanding of many fundamental phenomena, including spectral lineshape modifications [1–13], formation of light-induced states [14–19], reconstruction of the dipole response [20–23], and molecular dissociation [24–27] and vibrational [28–30] dynamics. In contrast to photoelectron measurements, TAS often deals with a large ensemble of systems and light propagation through the target medium generally needs to be considered.

Resonant pulse propagation describes a universal phenomenon when a short pulse passes through a medium with a long-lived resonance [31,32]. Modern laser technology has promoted the related investigation from near-infrared (NIR) [33–36] to the XUV [37–46] and even x-ray region [47]. While propagation effects have been observed and utilized in a wide range of applications, they have been rarely exploited to manipulate the absorption property of the target system. For the reported experimental [35–39] and theoretical [42–46] TAS studies, it is generally understood that resonant pulse propagation would complicate the light-matter interaction and distort the results predicted by the single-atom response. Effectively harnessing the induced temporal reshaping to control the absorption process remains to be explored, and would constitute a novel tool for future applications.

One approach to absorption control is the so-called coherent perfect absorption, which describes the time-reversed process of lasing at the first threshold and has been theoretically proposed [48] and experimentally realized in several setups [49–52]. In such devices, perfect absorption occurs when full destructive interference between the light transmitted and reflected from the cavity is realized. In this work we discuss a complementary method to achieve efficient absorption, based on a time-dependent interferometric effect, as was proposed recently in TAS framework [53,54]. This method relies on shaping the newly generated electric field by the medium to counteract the incoming field. It describes a cavity-free phenomenon and works for ultrashort laser pulses, which allows rapidly switching the absorption property of the target medium on ultrafast timescales.

The demonstration of resonant perfect absorption in Ref. [54] yielded matching numerical and analytical results, and the underlying concept was revealed using simple two- and three-level systems. In this article, we go beyond the previous few-level study and perform a more comprehensive description of the light-matter interaction. The absorption process is modulated interferometrically, by controlling the new electric field developed and temporally tailored during pulse propagation. In spite of the presence of nearby states and complex laser-induced couplings [55], we find the local enhancement of resonant absorption with time delay and propagation distance is clearly exhibited. Our results demonstrate the applicability and generality of switching the resonant absorption property by gating the resonance in the time domain,

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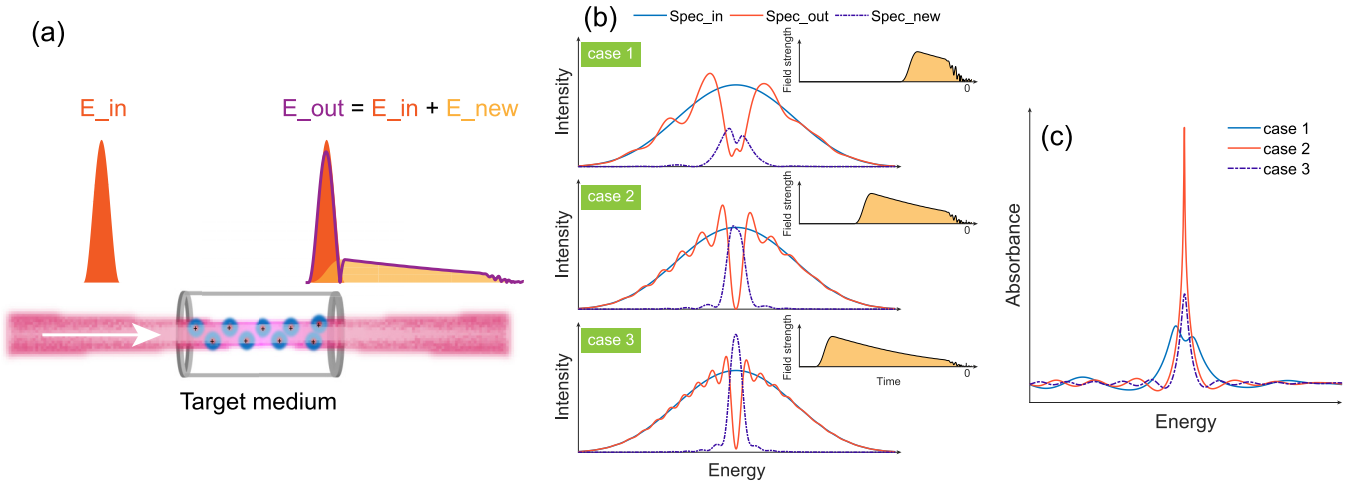

FIG. 1. Schematic illustration of the underlying interferometric mechanism. (a) Sketch of the evolution of the XUV pulse. The incident pulse (orange shaded area) transmits through the medium and interacts with the target system, which stimulates new coherent emission (yellow shaded area). The output field (purple line), composed of the incoming field and the newly generated one, gets temporally reshaped with a tail as compared to the input one. (b) Spectra of the input, output, and newly generated pulse for different time delays. The insets show the corresponding temporal profiles of the new field, which share the same initial shape, but the trailing ends are truncated differently by the time-delayed gate pulse at $t = 0$. (c) Absorption profiles of these three cases.

and open up new prospects for temporal engineering of high-frequency responses.

II. PRINCIPLE AND NUMERICAL METHOD

From a wave perspective, photoabsorption stems from the interference between the incident pulse and the new electric field stimulated by the polarization response during transmission through the target medium. Figure 1 presents the underlying interferometric mechanism for controlling the absorption process. After passing through the medium, the output field gets prolonged with a tail as shown in Fig. 1(a); the tail is temporally truncated by a time-delayed gate pulse. When this tail is dominated by a single subpulse, the spectrum of the new field gradually builds up with the increase of time delay. As this new field is out of phase with the incident one, they will interfere destructively and make a hole around the transition energy in the output spectrum as shown in Fig. 1(b). The absorption of the medium peaks when the spectral component of the new field counteracts that of the incident pulse, while weaker and stronger contributions of the new field yield less absorption [cf. the three cases displayed in Fig. 1(c)]. Accordingly, the increase of time delay between the input XUV and the gate pulse will lead to a local enhancement in resonant absorption. The presence of a second subpulse, which is out of phase with the first one, would enable the appearance of a second local enhancement region as shown in Ref. [54].

The numerical calculations in this work are performed by solving the three-dimensional coupled time-dependent Schrödinger equation (TDSE) in the single active electron approximation and the Maxwell wave equation (MWE) in helium [42]. This approach allows us to describe the absorption and dispersion of both the XUV and NIR pulses in a self-consistent manner. In the calculation, we choose the laser and medium parameters to be close to those used before [54]. The NIR pulse is centered at 764 nm, has a full width at half maximum (FWHM) duration of 7 fs, and the peak intensity is 2×10^{13} W/cm^2. The XUV pulse has a peak intensity of 1×10^{11} W/cm^2, and is centered at 21.1 eV, in accordance with the $1s2p$ state energy predicted with the pseudopotential used in the TDSE calculations (for a description of how the pseudopotential is constructed, see Ref. [56]). Unless otherwise specified, the confocal parameters for the XUV and NIR pulses are 3.0 and 1.6 cm, respectively, which yields a NIR beam waist that is 2.6 times as large as that of the XUV beam; a gas medium of 0.3-mm length and an atomic number density of 2×10^{17} cm^{-3} is considered. The delay- and space-dependent absorption spectrum is characterized by the optical density (OD) $\mathrm{OD}(\omega, \tau, z) = -\log_{10}[I(\omega, \tau, z)/I_{\mathrm{in}}(\omega)]$, where $I(\omega, \tau, z)$ and $I_{\mathrm{in}}(\omega)$ are the transmitted and incident laser spectra, respectively. ω, τ, and z denote the frequency, the time delay between the two pulses, and the spatial position along the pulse propagation direction. A window function of $\sim$230 fs width is imposed on the time-dependent polarization response, which ensures its decay after the excitation and mimics the experimental dephasing effect.

III. RESULTS AND DISCUSSIONS

For a demonstration of the concept illustrated in Fig. 1, we start by investigating the evolution of OD with respect to the interpulse time delay, and Fig. 2 shows the calculated results in the time and frequency domain. The computations are performed with a 2.55-fs FWHM duration of the XUV pulse whose spectrum is relatively narrow and only the $1s2p$ bright state is predominantly excited. As shown in Fig. 2(a), the tail of the XUV pulse developed during propagation is effectively truncated by the NIR pulse, which is centered at $t = 0$ fs. Since the tail in this case is mostly contributed by the dipole emission of the $1s^2$-$1s2p$ transition, it suggests that the $1s2p$ state population excited by the preceding XUV pulse

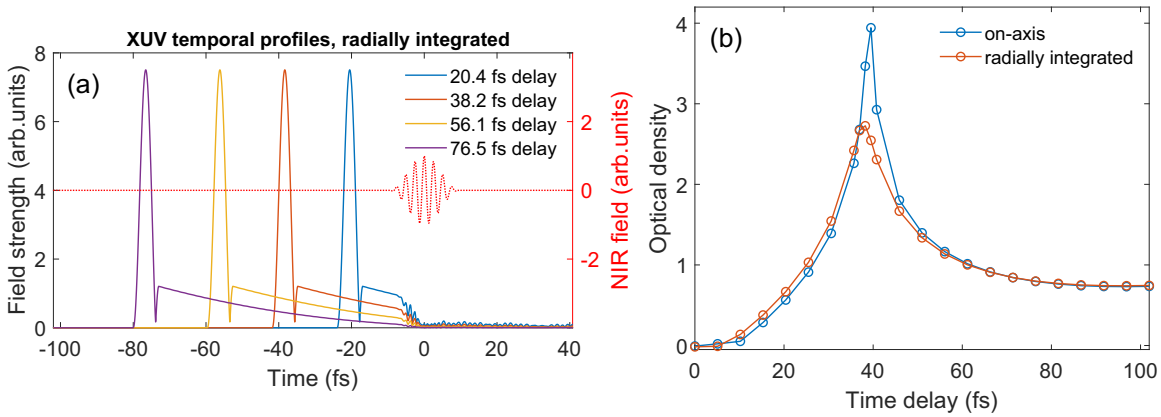

FIG. 2. (a) XUV temporal profiles at the end of the medium for a set of time-delay values computed by the TDSE-MWE simulation. The XUV tail basically terminates around time zero, where the NIR pulse (red dotted line) arrives. (b) Optical density at the $1s^2$-$1s2p$ transition frequency at the end of the medium as a function of time delay, computed by the on-axis and radially integrated laser spectra. The slight difference between them should be attributed to the radial inhomogeneity of the NIR field. The local enhancement of resonant absorption with increasing time delay is clearly exhibited.

is basically emptied out via ionization during the interaction with the NIR pulse, which is further verfied in Appendix A. The XUV tail is truncated in sub-NIR-cycle steps, near the oscillation peaks of the ionizing laser field. This truncation is completed before the peak of the NIR field at $t = 0$ fs due to the nonlinearity of the ionization process, which enables a gate width that is shorter than the NIR pulse duration. As the time delay increases from 0 fs, in contrast to the gradual buildup of a Lorentzian profile as one would expect within the single-atom response, the OD at the resonant frequency in Fig. 2(b) increases initially to a maximum, followed by a decrease with even larger time delay, in agreement with the illustration in Fig. 1. We wish to note that to detect high OD results in future experiments, a low noise level and a high spectrometer resolution are required. The somewhat limited frequency resolution in this large-scale calculation restricts higher peak OD values to be obtained. Nevertheless, this local enhancement in photoabsorption, either calculated by the on-axis or radially integrated laser spectra, closely resembles our earlier findings in a reduced-dimensionality model [54]. This implies that the mechanism of rapid switching of the absorption by time-gating the resonance is preserved also in the presence of the transverse degree of freedom and when more complex quantum-level structures are at work.

In the present case, the new XUV field is mainly influenced by two key factors: the time delay limits its temporal width and the propagation distance (when the target density is fixed) determines its strength. Thus control over the resonant absorption can also be achieved by varying the propagation distance. To illustrate how the absorption profile evolves as the laser pulses propagate through the medium, we plot the OD spectra as a function of propagation distance for three different time delays as shown in Fig. 3. For a small time delay at which the temporal width of the new field is very limited, its spectral contribution is too weak compared with that of the incident field, thus the absorption increases steadily with further propagation inside the medium as shown in Fig. 3(a). A larger time delay permits a longer XUV field to be generated, which leads to stronger absorption profiles as shown in Fig. 3(b). While in Fig. 3(c), the even larger time delay enables a new field whose spectral part could counteract that of incident pulse at the resonant frequency. The OD spectrum reaches a local maximum after transmitting through a part of the medium, and further propagation results in decreased photoabsorption.

The results presented above are calculated with a NIR beam larger than the XUV beam; the radial dimension perpendicular to the propagation direction thus predominantly serves to average the response and to describe the focusing and defocusing of both beams. An interesting situation arises when the NIR pulse intensity varies substantially across the XUV beam, so that the absorption dynamics varies across the radial dimension [57–59]. We explore this in Fig. 4, where we show the radial variation of the propagating XUV field for two different relative sizes of the XUV and NIR beams. In contrast to Fig. 4(a), the results in Fig. 4(b) exhibit complex radial variations of the XUV intensity at the $1s^2$-$1s2p$ transition frequency, reflecting the different dynamical processes occurring at each radial position. In particular, the XUV intensity at the resonant frequency gets hollowed out on axis at 38.2 fs time delay in Fig. 4(b). The corresponding radial-spectral beam profiles are shown in Figs. 4(c) and 4(d), in which the XUV beam is considerably larger than the NIR beam. By contrast, its counterpart in Fig. 4(a) basically stays zero along the radial direction. These results suggest that a larger gate beam is favorable for achieving significant attenuation of specific frequencies, while a smaller gate beam enables to create a dark spectral core, which resembles the dark-hollow beams [60–63] at the transition frequency. Future explorations by using spatially offset beams as shown in Refs. [57–59] would further enrich the toolkit for controlling the photoabsorption process with more knobs.

Finally, to further test the generality of the above mechanism, the examination of its application to other resonances is important. By employing a much shorter XUV pulse of 0.51-fs FWHM duration, one can excite a range of higher-lying $1snp$ ($n \geqslant 3$) states of helium, allowing us to follow the ultrafast buildup of these resonances simultaneously. Figures 5(a) and 5(b) show the transient absorption spectra calculated for

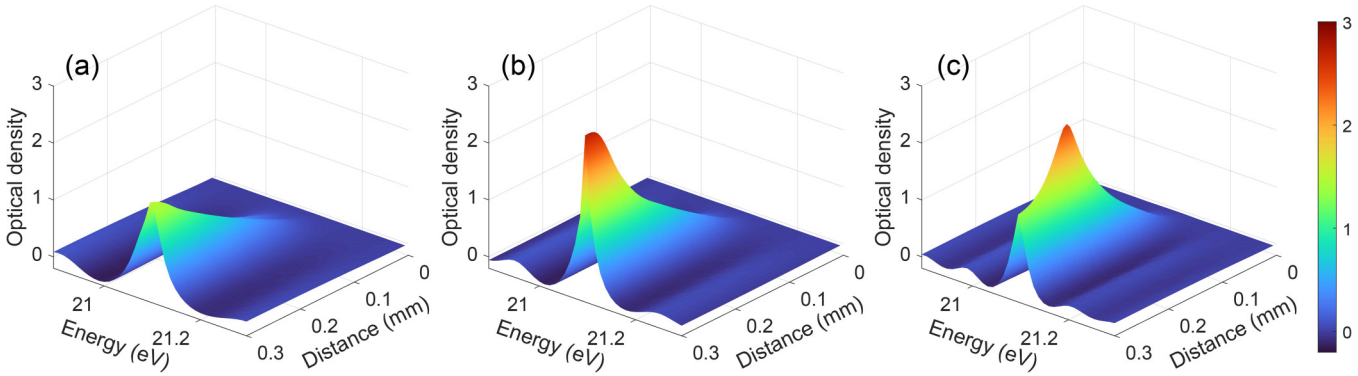

FIG. 3. Evolution of the absorption spectrum with propagation distance for three different time delays of (a) 30.6, (b) 38.2, and (c) 51.0 fs, corresponding to the positions before, around, and after the peak position in Fig. 2(b).

the $1snp$ ($n \geqslant 3$) series for two different densities, 2×10^{17} (as used above) and 6×10^{17} cm^{-3}, respectively. The results around the $1s2p$ resonance are shown in Appendix B, in which the further propagation leads to the appearance of a second local enhancement region, in accordance with the results in Ref. [54]. In the lower-density case, the OD increases smoothly with time delay for these resonances, similar to the results measured for the Rydberg series of the doubly excited states in helium [21]. This can be understood in terms of the smaller dipole matrix elements for XUV excitation of these states compared to that of the $1s2p$ state, and the correspondingly weaker single-atom response. Similar observations of the difference between these resonances were experimentally and theoretically reported before [38,39]. The oscillating structures at small time delays are due to the interference from the wide hyperbolic sidebands of neighboring resonances [55], and are thus the result of the congestion of multiple excited states. When we increase the density as in Fig. 5(b), we can compensate for the smaller dipole matrix elements. This is clearly exhibited in the characteristic rise and fall of the sharp OD peak for $1s3p$ state around 50-fs time delay. The observation of a similar enhancement for the $1snp$ ($n \geqslant 4$) states would require even higher atomic densities, as the dipole matrix elements between them and the ground state are even smaller. Similarly, the difference in the dipole matrix elements also explains why no second local enhancement appears for the $1snp$ ($n \geqslant 3$) series for the employed parameter range in Fig. 5, in contrast to the results for the $1s2p$ resonance shown in Appendix B. With the exemplary results presented here, it is reasonable to anticipate that the underlying mechanism of achieving local enhancement in resonant photoabsorption is generally applicable to other resonances and atomic species.

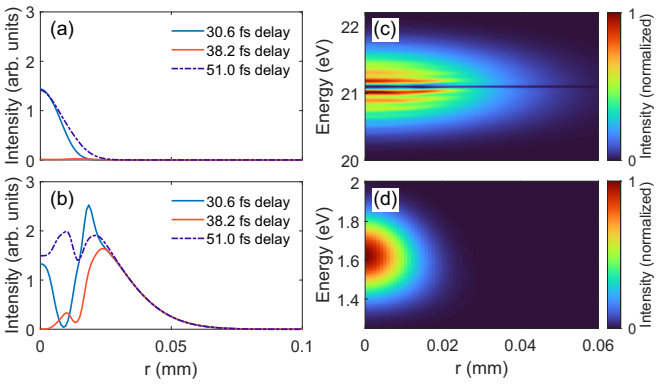

FIG. 4. The output XUV intensity at the $1s^2$-$1s2p$ transition frequency as a function of radial coordinate for three time delays, (a) computed with the previous beam parameters and (b) for the new case in which the XUV and NIR beam waists exchanged. Radial-spectral profiles of the output (c) XUV and (d) NIR beams for the case in (b) at 38.2-fs time delay.

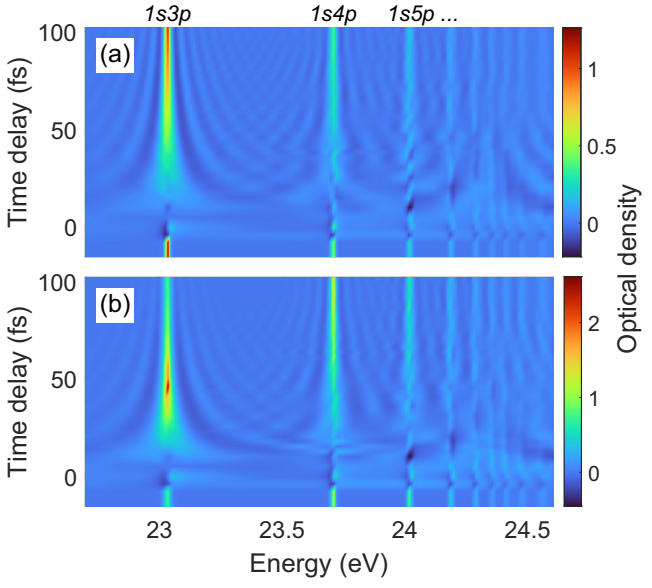

FIG. 5. Transient absorption spectra of helium excited by the broadband XUV pulse, showing the ultrafast buildup of the $1snp$ series for atomic densities of (a) 2×10^{17} and (b) 6×10^{17} cm^{-3}. For large negative time delays when the XUV pulse arrives after the NIR pulse, unperturbed absorption lineshapes manifest. For the higher-density case, the local enhancement at the $1s3p$ resonance near 23 eV becomes visible.

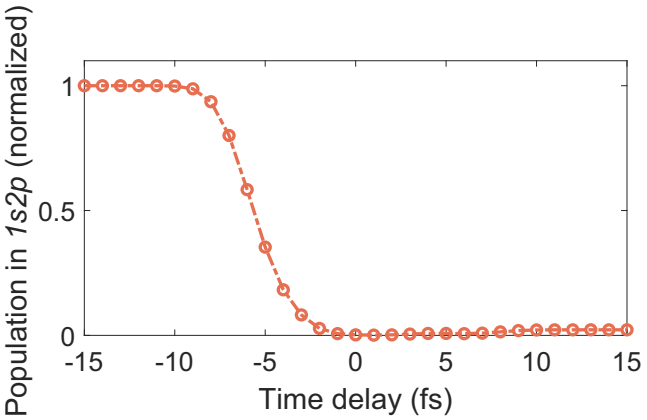

FIG. 6. Calculated relative population of $1s2p$ state as a function of time delay at a fixed time of 20 fs. Note that the NIR pulse is centered at $t = 0$.

IV. CONCLUSION

In conclusion, we validated the concept of resonant perfect absorption by performing the large-scale TDSE-MWE calculations in helium. The new field developed in the target medium is governed by the concerted action of pulse propagation and an auxiliary gate pulse, which interferes controllably with the incident pulse. It is remarkable that the local enhancement of resonant absorption with time delay persists even in a more realistic description of the light-matter interaction, proving the feasibility of the proposed method. With similar phenomena observed for the $1s3p$ state, the mechanism behind the absorption modulation is promising to be generally applicable to other resonances and atomic systems in any region of the spectrum. An interesting case arises when the NIR pulse is overintense in this scheme, so that the high-order harmonics generated during the interaction with the medium will interfere with the incoming field jointly, which stimulates future explorations. The presented results yield interferometric insights into understanding and controlling the macroscopic absorption property through the newly generated electric field by the medium, and are expected to trigger new prospects for engineering high-frequency radiation.

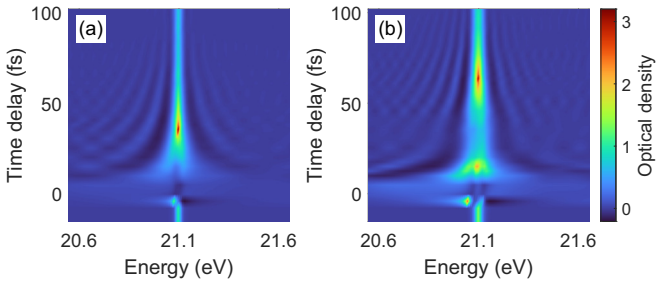

FIG. 7. (a), (b) Transient absorption spectra in the vicinity of the $1s2p$ resonance calculated with the same parameters as in Figs. 5(a) and 5(b) in the main text, respectively.

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APPENDIX A: EVOLUTION OF THE $1s2p$ STATE POPULATION WITH TIME DELAY

The gating mechanism is verified in Fig. 6, in which the evolution of the $1s2p$ state population as a function of time delay is presented. The population remains unaffected for large negative time delays and gets significantly depleted during the interaction with the NIR pulse centered at $t = 0$. For positive time delays, it gets nearly emptied out.

APPENDIX B: TRANSIENT ABSORPTION RESULTS AROUND THE $1s2p$ RESONANCE

Figure 7 shows the transient absorption spectra for the calculations performed in Fig. 5 in the main text, here in the vicinity of the $1s2p$ resonance. Compared to the lower-density case in Fig. 7(a), the enhanced absorption peak in Fig. 7(b) shifts to a smaller time delay and a second local enhancement region appears, which signals that a second subpulse in the time domain comes into play [54].

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