Resonant Perfect Absorption Yielded by Zero-Area Pulses

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We propose and study the manipulation of the macroscopic transient absorption of an ensemble of open two-level systems via temporal engineering. The key idea is to impose an ultrashort temporal gate on the polarization decay of the system by transient absorption spectroscopy, thus confining its free evolution and the natural reshaping of the excitation pulse. The numerical and analytical results demonstrate that even at moderate optical depths, the resonant absorption of light can be reduced or significantly enhanced by more than 5 orders of magnitude relative to that without laser manipulation. The achievement of the quasicomplete extinction of light at the resonant frequency, here referred to as resonant perfect absorption, arises from the full destructive interference between the excitation pulse and its subpulses developed and tailored during propagation, and is revealed to be connected with the formation of zero-area pulses in the time domain.

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The ability to control light underlies the possibility of steering quantum dynamics. Femtosecond pulse-shaping techniques using programmable spatial light modulators have made great success in coherent quantum control [1–3]. A direct extension of these methods developed in the optical range to shorter wavelengths is hampered by the small diffraction efficiencies for spectral dispersion and the limited transparency range of the modulator materials [4]. Alternatively, control over the radiation can be realized in the time domain by modulating the induced response of the system. By attosecond transient absorption spectroscopy using femtosecond near-infrared (NIR) laser fields to exert the modulation, the compression [5] and redirection [6,7] of extreme-ultraviolet (XUV) radiation have been demonstrated. Very recently, by virtue of the frequency-dependent refractive indices in the vicinity of a single and closely spaced resonances, the deflection [8] and spectral compression [9] of XUV pulses have been reported. Coupled with recent advances and the resulting widespread availability of XUV and x-ray sources from high-harmonic generation and free-electron lasers, the capability of shaping short-wavelength pulses promises to create new opportunities for controlling atomic and nuclear quantum dynamics [10,11].

The coherent interaction between ultrashort laser pulses and resonant media is a fundamental issue in quantum optics. Among many pulse forms, of particular interest are zero-area (0π) pulses that produce no net excitation in a two-level system on exact resonance, which have attracted considerable theoretical [12–14] and experimental [15–21] interest. According to the pulse-area theorem [22–24], a zero-area pulse can be produced by natural reshaping via passing a weak pulse through a resonant medium with a narrow bandwidth. During transmission, the temporal shape of the laser pulse experiences strong modification, creating negative and positive lobes in the tail that make the pulse area approach zero after a sufficient distance. While propagation effects often complicate the consideration of light-matter interaction, the generated zero-area pulses have been harnessed to enhance the transient excitation of atoms [18,19], and behave as a clear sign of strong interaction between single photons and atoms [20,21]. In addition to the reshaping on the waveform of the laser pulse itself, the frequency-domain modifications induced by propagation effects have come in for considerable investigations recently. By altering the polarization of the system, the broadening of the resonance profiles and the emergence of spectral substructures in attosecond transient absorption spectra have been observed [25–27].

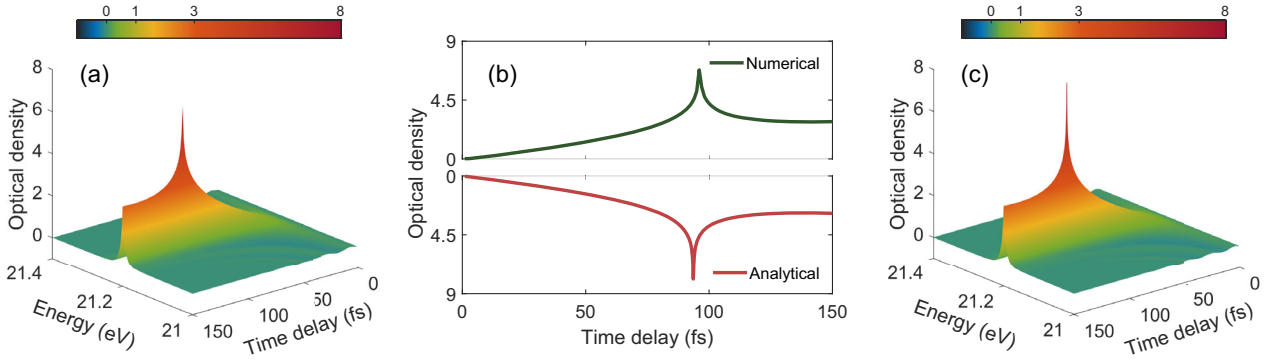

FIG. 1. (a) Numerical and (c) analytical spectra for the transient absorption in the vicinity of the resonance energy, showing the buildup of the Lorentzian profile in a macroscopic medium. The corresponding delay dependence of optical density at the resonant frequency is shown in (b).

In this Letter, a general scheme for manipulating the absorption properties of a resonant medium is revealed by attosecond transient absorption spectroscopy. By applying the time-gating approach [28–30], we isolate and benchmark a novel and counterintuitive phenomenon, namely the resonant perfect absorption in a macroscopic medium by utilizing only part of the electric field. This observation is connected with the controlled formation of zero-area pulses, a hallmark of quantum optics. It is achieved by tailoring the XUV pulse in a way that its subpulses developed during propagation interfere destructively with the main pulse, fully canceling the resonant spectrum out. Note that this phenomenon is reminiscent of the investigations of coherent perfect absorption (time-reversed lasing) [31–33], in which the incident coherent monochromatic illumination can be perfectly absorbed under specific conditions. Both methods rely on the interplay of interference and absorption, but ours presented here involves a resonant medium while an optical cavity is not required. The demonstrated control of the excitation pulse via spectral phase manipulation and temporal engineering, would facilitate the extension of coherent-control schemes to the high-frequency regime.

The numerical procedure essentially corresponds to addressing the formation and reshaping of a Lorentzian resonance in a macroscopic medium. Briefly, the wave function of the model system is expanded as $|\Psi(t,\tau)\rangle = c_1(t,\tau)|1\rangle + c_2(t,\tau)e^{-i\omega_{12}t-\Gamma t/2}|2\rangle$, where $|1\rangle$ and $|2\rangle$ represent the ground and excited states, respectively. ω_{12} denotes the transition frequency and Γ the decay rate of the excited state. Atomic units are used unless otherwise noted. The system is driven by a weak attosecond XUV pulse expressed as $E(t) = F(t)e^{i\omega_X t} + c.c.$, whose central frequency ω_X is chosen to match the transition. We employ the rotating wave approximation for the XUV coupling for its low intensity and the zero detuning [34], which facilitates subsequent extraction of the field envelope $F(t)$ during pulse propagation. The dynamical evolution of the system in the presence of the XUV pulse only can be computed by directly solving the time-dependent Schrödinger equation, whereas the rapid depletion of the upper state's coefficient by the femtosecond NIR field delayed by τ is incorporated through a phenomenological decay rate $\Gamma_n(t,\tau) = \alpha_n I_{\mathrm{NIR}}^n(t,\tau)$, i.e., $\langle 2|H(t,\tau)|2\rangle = \omega_{12} - i\Gamma/2 - i\Gamma_n$. The NIR intensity I_{NIR} is high enough in our model to fully ionize the excited electrons (with n denoting the required number of photons), and thereby α_n is taken to achieve a complete extinction of the polarization after the passage of the NIR pulse. As ionization depends on the instantaneous electric field, we employ $I_{\mathrm{NIR}}(t,\tau) = E_{\mathrm{NIR}}^2(t,\tau)$ rather than a pure intensity envelope to account for the half-cycle ionization bursts. The nonlinearity of the ionization enables an effective truncation window shorter than the NIR pulse duration. By introducing a local frame moving with the pulses, the spatial evolution of the XUV pulse in one dimension can be approximated by [35]

$$\frac{\partial}{\partial x}\tilde{E}(\omega,\tau,x) = -2\pi i\frac{\omega}{c}\tilde{P}(\omega,\tau,x), \tag{1}$$

with the delay- and space-dependent polarization response in the frequency domain denoted by $\tilde{P}(\omega,\tau,x)$, and the spectral representation of the XUV pulse $\tilde{E}(\omega,\tau,x)$. We characterize the absorption spectrum by the optical density (OD) defined as the decadic logarithm of the intensity ratio, $\mathrm{OD}(\omega,\tau) = -\log_{10}[I(\omega,\tau)/I_0(\omega)]$, where $I(\omega,\tau)$ and $I_0(\omega)$ are XUV spectra at the exit and the entrance of the medium, respectively.

Figure 1(a) depicts the transient absorption spectrum calculated with a medium length of 0.3 mm and an atomic number density of $N = 1.2\times10^{17}$ cm^{-3} (about 5 mbar pressure at room temperature of an ideal gas). The spatial propagation is performed with a step size of 0.15 μm. The atomic parameters are $\omega_{12} = 21.2$ eV, $\mu_{12} = 0.42$, and $n = 3$, which resemble a helium atom with its ground and $1s2p$ excited states for the levels $|1\rangle$ and $|2\rangle$. We choose a decay rate of $\Gamma = 1/60$ fs^{-1} (i.e., natural linewidth of ∼11 meV) and $\alpha_3 = 8\times10^8$ below. A 500 as full

width at half maximum (FWHM) duration XUV pulse with a peak intensity of 10^{11} W/cm^2 is applied, which is centered at $t = 0$. The delayed NIR pulse has an FWHM duration of 7 fs, a central wavelength of 760 nm, and a peak intensity of 20 TW/cm^2. Temporal Gaussian envelopes are input for both pulses. Of particular interest are positive time delays, at which the XUV excitation and NIR truncation occur sequentially. For small time delays, the resonant profiles are severely suppressed, exhibiting a broad and weak absorption spectrum. With increasing time delays, the polarization is allowed to evolve longer and more information of the resonance survives. The absorption line gradually builds up and becomes narrower. The most intriguing feature is the emergence of a narrow, sharp peak at around 100 fs before seeing a decline for even larger time delays. In comparison with the spectrum in the dilute-gas limit shown in Supplemental Material [36], the nonlinear spectral evolution clearly implies that we go beyond the single-atom response and enter a new regime where collective light-matter interactions play an important role. To highlight this peculiar spectral evolution with time delay, in Fig. 1(b) (upper line) we plot the optical density at the resonance frequency, which represents the strongest absorption obtained along the energy axis for each time delay. Recalling our definition of optical density, the reached OD values exceeding 6 means most of the injected intensity has been absorbed by the resonant medium, attenuated by more than 6 orders of magnitude. The present observation of the quasicomplete extinction of incident light at the resonant frequency with a narrow linewidth, may be regarded as cavity-free resonant perfect absorption.

To understand the calculated spectral behavior, it is instructive to give a conceptual explanation of the process and the associated XUV pulse evolution along its propagation direction. At the beginning of the medium, the short XUV pulse arrives and excites the resonance. A long-lived coherence ensues, giving rise to an electric field at the resonance frequency after the original pulse (free induction decay). The electric field in the tail is π out of phase with the original field, thus the two fields interfere destructively and result in a spectral hole at the transition energy. As the pulse propagates, a secondary polarization could further be excited by the tail, resulting in a newly generated electric field with components both in and out of phase with the original one. The process repeats continuously along the propagation path until the pulse exits the medium. By confining the temporal dynamics with the subsequent NIR pulse, the phase of the preceding dipole response and hence that of the subpulses are effectively unaffected compared to the free-evolution case. Accordingly, the consequence of the interference between the main pulse and the subpulses remains a simple dip in the transmitted spectrum, which translates into the symmetric absorption profiles persisted in Fig. 1(a). No complex spectral substructures manifest on

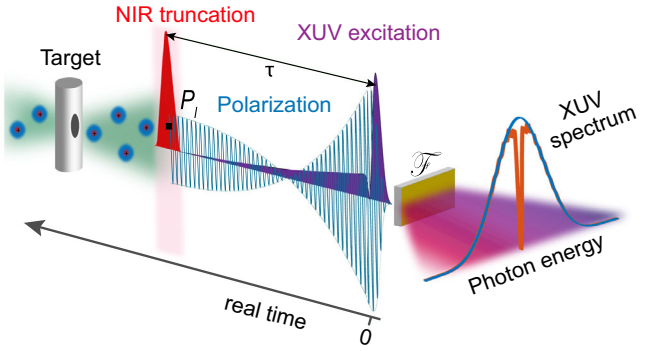

FIG. 2. Conceptual illustration of a laser-gated resonance in a macroscopic medium. A weak resonant XUV pulse comes first and triggers a long-lived polarization response decaying exponentially, giving rise to an electric field emitted for a long time after the original pulse. After a certain distance of propagation through the medium, the XUV pulse develops a long tail (purple) and results in a complex-shaped polarization (blue). The following intense NIR pulse (red) truncates the evolving polarization via complete ionization of the excited state. Tuning the time delay allows us to confine the polarization and hence the tail of the XUV pulse in a desired manner. P_l represents the polarization response at $t = \tau$ at each spatial position and is explained further in the main text.

the shoulder of the absorption lines as a result of the sustained phase relation of the subpulses, in contrast to the previously reported transient absorption studies in macroscopic media involving a phase shift to the dipole moment either induced by external disturbance [25–27] or by internal configuration interaction [30]. The emergence of the resonant perfect absorption peak is discussed later.

The above consideration can be appreciably simplified with some appropriate assumptions. The schematic illustration of the analytical model is shown in Fig. 2. The interaction of the XUV pulse with the system is described in first-order perturbation theory, well applicable for the light-matter interaction of high-frequency pulses produced from lab-based high-order harmonic generation. By taking the NIR truncation as infinitesimally short, the polarization response in the frequency domain is given by (for the derivation of the analytical model, see Supplemental Material [36])

$$\tilde{P}(\omega,\tau,x) = \chi(\omega)\tilde{E}(\omega,\tau,x) - \frac{i\chi(\omega)}{\sqrt{2\pi}N\mu_{12}^2} P_l e^{-i\omega\tau}, \quad (2)$$

with the linear susceptibility defined as $\chi(\omega) = N\mu_{12}^2/(\omega_{12} - \omega + i\Gamma/2)$. P_l denotes the polarization response at the time when the truncation occurs [i.e., $P(t = \tau, \tau, x)$] and can be expressed as

$$P_l(\tau,x) = \frac{1}{\sqrt{2\pi}} \int_0^\infty \chi(\omega)\tilde{E}_0(\omega,x)e^{i\omega\tau}d\omega. \quad (3)$$

$\tilde{E}_0(\omega, x)$ represents the freely propagating field and its evolution can be described by the solution of the optical Helmholtz equation in a local frame

$$\tilde{E}_0(\omega, x) = \tilde{E}(\omega, 0) e^{-2\pi i \frac{\omega}{c} \chi(\omega) x}. \tag{4}$$

Control over the amount of spectral phase accumulated during pulse propagation could hence be afforded by tuning the atomic density and medium length. Nevertheless, the pressure-length product is kept fixed here, and we are more concerned with the temporal control of the natural reshaping and the resulting spectral properties.

Figures 1(c) and 1(b) (lower line) show the analytical transient absorption spectrum and the optical density at the resonant frequency as a function of time delay, respectively. Compared with the numerical counterparts, essential features have been captured. The general agreement proves the validity of the analytical model and the two approximations, which will be further employed in our analysis. Remaining discrepancies are mostly ascribed to the fact that the NIR truncation is treated instantaneously here, whereas a finite-duration laser gate is applied in the numerical model. It is noteworthy that the analytical approach described in the frequency domain has circumvented the need for a time integration, relaxing the computational cost also for future applications in more complex targets. A time delay step of 1 fs is used in Figs. 1(a) and 1(b) (upper line) for computation reasons, while a much finer step of 0.2 fs is implemented in Figs. 1(c) and 1(b) (lower line).

The field envelope of the XUV pulse at each spatial position, denoted by $F(t, \tau, x) = E(t, \tau, x)/e^{i\omega_X t}$, can be obtained from $\tilde{E}(\omega, \tau, x)$ by inverse Fourier transformation. Particularly, in the limit of δ-like truncation, it can be approximated by directly cutting off the freely evolving envelope $F_0(t, x)$ in time. For the on-resonance case where the temporal fields of the main pulse and the subpulses oscillate at the same frequency ($\omega_X = \omega_{12}$), we can arrive at a simple formula for the electric field at the resonance frequency

$$\begin{aligned}\tilde{E}(\omega_{12}, \tau, x) &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} F(t, \tau, x) dt \\ &= \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\tau} F_0(t, x) dt.\end{aligned} \tag{5}$$

This expression reveals a subtle connection between the pulse spectrum at the resonant frequency and the temporal envelope of the XUV pulse, which is composed by the main pulse and propagation-induced subpulses. The described resonant perfect absorption manifests when the modulus of $\tilde{E}(\omega_{12}, \tau, x)$ drops to zero during pulse propagation. This corresponds to the situation that the envelope of the developed and truncated subpulses counteracts that of the main pulse, making a net zero area enclosed by the pulse envelope. It is a direct consequence of the full destructive interference between the main pulse and the generated subpulses, perfectly canceling the resonant frequency out. Additionally, it is interesting to note that apart from a numerical factor of $2\sqrt{2\pi}\mu_{12}$, Eq. (5) is essentially the expression for the pulse area of the truncated XUV pulse. Accordingly, the observation of resonant perfect absorption in the frequency domain suggests the formation of a zero-area pulse in the time domain. The degree of perfect absorption is intimately connected with the accuracy of the zero-area pulse formed in the process. The presented ionization-induced gate effectively acts as an ultrafast shutter on the XUV pulse envelope, confining it in a finite duration. The shutter speed is connected with the depletion time of the excited state, while its position can be controlled by the time delay. Unlike traditional methods that integrate the whole temporal envelope, this time-gating approach allows the excitation pulse area down to zero even at moderate optical depths.

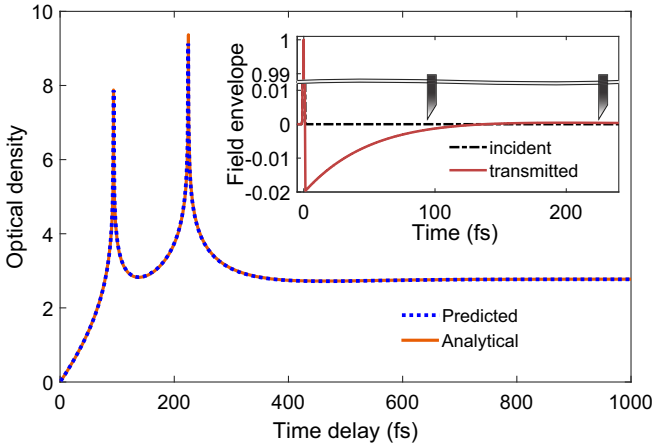

FIG. 3. The evolution of optical density at the resonant frequency with time delay. Regarding the stable OD value over a long delay range as the result for the free-evolution case, the macroscopic absorption of the medium to the resonant frequency can be reduced or enhanced by more than 5 orders of magnitude with the adoption of temporal gating. This is further validated by the orange solid line representing the analytical results. The inset shows the incident XUV field envelope (real part, normalized) together with the transmitted one in the absence of NIR truncation. A break is made in the vertical axis to skip the featureless range. Shutter closure positions at which resonant perfect absorption can be achieved are marked with knife symbols.

The calculated optical density at the resonance frequency by integrating the truncated field envelope for different time delays is shown in Fig. 3 (blue dotted line). The curve accords closely with the preceding results shown in Fig. 1(b) but for a longer delay range. However, we notice the prediction of the appearance of another perfect absorption peak at a larger time delay, indicating the zero-area condition is satisfied once more for the given optical depth,

which is confirmed by our subsequent analytical calculation shown together in Fig. 3 (orange solid line). The procedure based on Eqs. (4) and (5) turns out to be an effective avenue for predicting the emergence of resonant perfect absorption with given parameters of the driving laser and target medium. The weak tail of the envelope far from the pulse center contributes little to the pulse area, and as expected, the optical density changes barely for even longer integration time (time delays). Hereby, the near-constant OD value with increasing time delay suggests it can be essentially regarded as the free-evolution case. Consequently, by effectively gating the two-level resonance, the amount of resonant radiation absorbed by the medium can be reduced, or significantly enhanced by more than 5 orders of magnitude compared with the free-evolution counterpart. This cavity-free phenomenon would be particularly relevant when aiming at efficient resonant absorption of light in only a moderately dense medium.

Although only a set of parameters are discussed in the main text, the influence of different pulse and atomic parameters has been further explored, including the cases of non-Gaussian temporal pulse shapes and the presence of a nearby excited state [36]. We find that the described method for achieving resonant perfect absorption by cutting off the laser field is generally applicable to various settings. The feasibility of this method has been further tested in a more realistic scenario by solving the coupled time-dependent Schrödinger equation in the single active electron approximation and the Maxwell wave equation [38]. From the preliminary calculation results, we find the described phenomenon persists in a macroscopic helium gas. Refining this approach would stimulate many interesting prospects, but goes beyond the scope of this work. The controlled temporal and spectral reshaping, the consequence of collective light-matter interactions, is enabled by the resonant ensemble and external fields. In view of its resonant character, the underlying mechanism is expected to be generally applicable at different frequencies wherever the resonance lies. The ability to rapidly switch the resonant spectrum in the time domain by another pulse would provide a promising toolbox for the development of future opto-optical modulators [6,7].

In conclusion, we have demonstrated a general scheme for manipulating the macroscopic transient absorption of an ensemble of two-level systems and shaping XUV radiation. It is remarkable that a temporal gate enables a rapid switching of the medium's absorption properties, and the resonant perfect absorption can be achieved at certain conditions. Moreover, the latter is proved to be connected with the formation of zero-area pulses in the time domain when driven by resonant pulses. The presented concept is robust against different settings and persists even in a realistic scenario, whose further exploration opens promising directions for future ultrafast switching devices. We envision an organized act of more complex light-matter interactions and pulse propagation in macroscopic media to open new routes for the temporal engineering of high-frequency responses and pulse shapes.

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