Transmission of an isolated attosecond pulse in a strong-field dressed atom

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The optical properties of matter can be manipulated by coupling a strong field to an optical transition. The concepts of Autler-Townes splitting and electromagnetically induced transparency, where a control field manipulates the transmission of a probe field, have recently been extended to control the transmission of x rays. The use of a strong femtosecond pulse to control the transmission of an isolated attosecond extreme ultraviolet or soft x-ray pulse opens a new field of investigation, since the duration of the probe pulse is only a fraction of one optical cycle of the control pulse. In this paper a three-state model beyond the rotating wave approximation is formulated that treats the attosecond pulse as an initial value problem in the time domain. Two effects for spectroscopic measurements are elucidated: first the absorption profile of the attosecond pulse is split into multiple lines rather than a doublet in the high-field regime, and second the transmission of the attosecond pulse depends on the relative timing within the optical cycle of the control pulse. An analytical treatment is carried out in the time domain in contrast to the more common approach in the frequency domain. This is appropriate for the field of attosecond physics where the main interest lies in the time evolution of the fields, rather than spectroscopic studies.

The idea of using strong electromagnetic radiation for the coherent manipulation of atoms and molecules has led to the development of media with radically new optical properties [1]. Strong electromagnetic radiation tuned to the transition energy of a two-level system induces oscillatory population transfer, so-called Rabi flopping. If the system is probed to a third level by a relatively weak field, then the optical properties at the probe wavelength are controlled by the strong field, and effects like the Autler-Townes splitting [2] and electromagnetically induced transparency (EIT) [3] are observed.

Recently, those concepts were extended to the extreme-ultraviolet (XUV) [4–9] and x-ray regime [10,11]. Core-hole states that are accessed by x-ray transitions have a lifetime of only a few femtoseconds, which requires the control field to be extraordinarily strong for the maintenance of coherence. The control field needs to be sufficiently intense to induce Rabi flopping between the core-hole states on a timescale faster than their decay [11]. Presently, only laser pulses in the femtosecond regime fulfill this requirement.

The use of isolated attosecond pulses [12] as the probe field raises further questions and can lead to new opportunities. In this case, the duration of the probe pulse is only a fraction of one optical cycle of the control field, which gives importance to the relative timing of the attosecond pulse with respect to the phase of the control pulse. The picture of instantaneous Stark shifts is currently discussed [13,14]. A treatment in the time domain seems more appropriate than the usual approach in the frequency domain based on the Floquet theorem.

In this paper a simplified three-level model is analyzed for the transmission of an isolated attosecond pulse in an atom in the presence of a strong control field. We regard the high-field regime, characterized by a Rabi frequency that is on the order of the laser frequency or higher. An equation is derived for the time evolution of the atomic polarization response, which is the

source for the optical effects observed in a macroscopic gas-phase medium. The traditional rotating wave approximation (RWA) is unsuitable when the field strength of the control field approaches the high-field regime. We show two effects that are not reproduced within the RWA: First, the absorption profile splits up into a variety of lines in the high-field regime instead of the doublet characteristic for weaker field strengths. Second, the absorption depends on the relative timing of the two pulses. This paper focuses on the analysis of these two effects in the simple environment of a three-level system in order to elucidate the physical origin and to give an analytical treatment. However, the same effects are found if more than three levels are considered in the simulation, as well as limited lifetimes of the states and depletion by strong-field ionization [7].

An analysis of the problem in the time domain rather than the approach in the frequency domain based on the Floquet theorem is necessary for two reasons. First, the decay of the core-hole states accessed by x rays will damp the atomic polarization response within a few femtoseconds. Second, the electric field amplitude in a strong-field pulse, which is necessary to reach the required field strength, also changes within a few femtoseconds. A treatment in the frequency domain without accounting for these two effects is not meaningful, and a frequency-domain analysis that includes the effects without the traditional RWA is complicated. In contrast, the solution in the time-domain presented here yields a valid and useful prediction for the first few femtoseconds after the action of the attosecond pulse. In light of possible pulse-shaping techniques for x rays, as proposed in Ref. [10], the analytic solution in the time domain is a valuable tool.

Analytical derivations for three-level systems typically ignore the phase between the two electromagnetic fields. That is a reasonable approximation for pulses with similar pulse duration and without a phase lock. This approximation is not
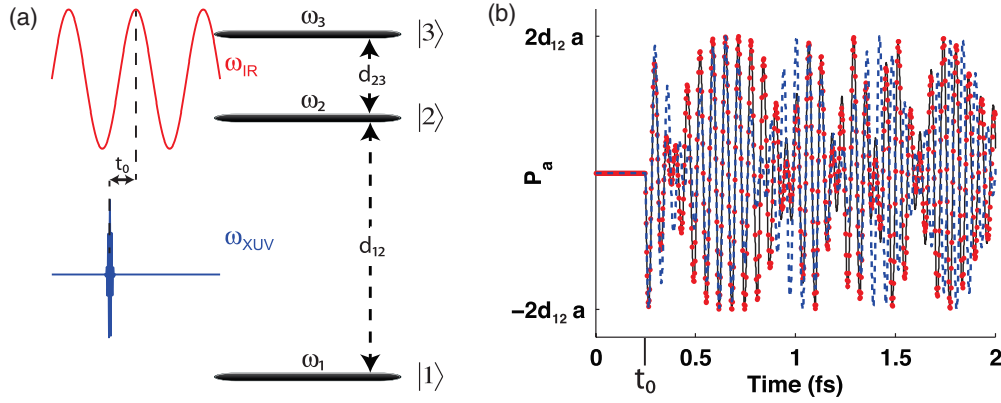
FIG. 1. (Color online) Three-level model for the time-dependent transmission of an attosecond pulse in the presence of strong coupling pulse. (a) An attosecond pulse (angular frequency ω_{XUV}) induces transitions from the ground level $|1\rangle$ to an excited level $|2\rangle$ via the transition matrix element d_{12}. A strong control pulse (angular frequency ω_{IR}) couples the two upper levels $|2\rangle$ and $|3\rangle$ via the transition matrix element d_{23}. The relative timing of the two pulses is given by t_0. (b) The time evolution of the atomic polarization response P_a as given by Eq. (6) for $t_0 = 0.25$ fs. Three different ways for the evaluation of ρ_{12} are considered: the numerical integration of Eq. (2) with RWA (blue dashed line), the approximation given by Eq. (4) (red dots), and the numerical integration of Eq. (2) without RWA (black solid line). The coupling pulse is assumed to have an infinite pulse duration, a wavelength of 800 nm at an intensity of 7×10^{14} W/cm². The probe pulse is assumed to be infinitely short. The atomic parameters are $\omega_2 = 65.11$ eV, $\omega_3 = 63.73$ eV, $d_{12} = 0.02$, $d_{23} = 2.7$ (this resembles a xenon atom in its ground state $^1S_0$ and its core-hole states $4d^{-1}(^2D_{5/2})6p(^2P_{3/2})$ and $4d^{-1}(^2D_{5/2})6s(^2S_{1/2})$ for the levels $|1\rangle$, $|2\rangle$, and $|3\rangle$).

valid for probing with isolated attosecond pulses, where the duration of the probe pulse is only a fraction of one optical cycle of the control pulse (Fig. 1). In the following we seek to elaborate a model that incorporates the relative timing of the attosecond pulse with respect to the phase of the control pulse.

The Hamiltonian of the three-level system shown in Fig. 1 is given by (atomic units are used throughout the paper)

$$\mathbf{H} = \begin{pmatrix} 0 & d_{12}F(t) & 0 \\ d_{12}F(t) & \omega_2 & d_{23}F(t) \\ 0 & d_{23}F(t) & \omega_3 \end{pmatrix}, \quad (1)$$

where $F(t)$ is the instantaneous electric field strength and where the ground-state energy ω_1 is set to zero. The time evolution of the density matrix ρ is given by the von Neumann equation

$$i\dot{\rho} = [\mathbf{H}, \rho]. \quad (2)$$

In the following analytical derivation it is assumed that the attosecond pulse acts only on the coupling between levels

$|1\rangle$ and $|2\rangle$ and that the femtosecond pulse acts only on the coupling between levels $|2\rangle$ and $|3\rangle$. The femtosecond pulse is assumed to be infinitely long such that its electric field is given by $F_{\mathrm{IR}}(t) = f_{\mathrm{IR}}\cos(\omega_{\mathrm{IR}}t)$, with f_{IR} and ω_{IR} being the amplitude and angular frequency. It is assumed that the system is initially in its ground state and that the attosecond pulse is infinitely short such that it populates the off-diagonal element ρ_{12} of the density matrix via the transition matrix element d_{12} at a time t_0 with a population a. In this approximation, the action of the attosecond pulse is an initial value problem for the solution of Eq. (2):

$$\begin{aligned} \rho_{11}(t_0) &= 1, \\ \rho_{12}(t_0) &= -\rho_{21}(t_0) = ia, \quad (3) \\ \rho_{mn}(t_0) &= 0 \,\forall\, (m,n) \notin \{(1,1),(1,2),(2,1)\}. \end{aligned}$$

In the high-field limit, here defined by $\Omega_{\mathrm{Rabi}} \gg \omega_{\mathrm{IR}}$ and $\Omega_{\mathrm{Rabi}} \gg |\omega_3 - \omega_2|$ where $\Omega_{\mathrm{Rabi}} = d_{23}f_{\mathrm{IR}}$ is the Rabi frequency, the time evolution of the off-diagonal matrix element between the ground state and the first-excited state can be approximated by

$$\rho_{12}(t) = ia\exp\left(i\frac{\omega_2 + \omega_3}{2}(t - t_0)\right)\left(\frac{\omega_2 - \omega_3 + W(t_0)}{2W(t_0)}\exp(i\vartheta) + \frac{\omega_3 - \omega_2 + W(t_0)}{2W(t_0)}\exp(-i\vartheta)\right),$$

$$\vartheta = \frac{\sqrt{(2\Omega_{\mathrm{Rabi}})^2 + (\omega_2 - \omega_3)^2}}{2\omega_{\mathrm{IR}}}[E(\omega_{\mathrm{IR}}t, m) - E(\omega_{\mathrm{IR}}t_0, m)], \quad (4)$$

$$W(t) = \sqrt{[2\Omega_{\mathrm{Rabi}}\cos(\omega_{\mathrm{IR}}t)]^2 + (\omega_3 - \omega_2)^2},$$

where $E(\varphi, m)$ is the elliptic integral of the second kind, defined by

$$E(\varphi, m) = \int_0^\varphi \sqrt{1 - m\sin^2(\varphi')}\, d\varphi'$$
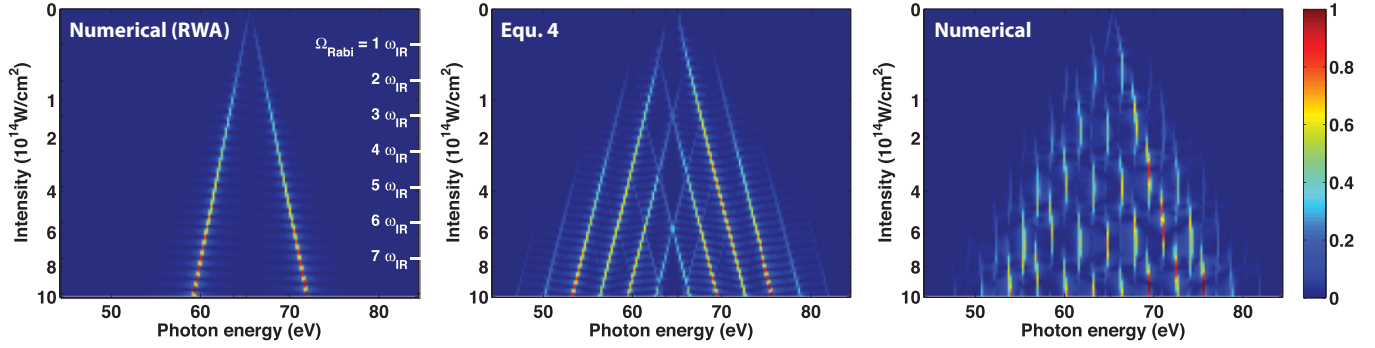
FIG. 2. (Color online) Frequency spectrum of atomic polarization response P_a as a function of intensity for a delay time $t_0 = 0$. The numerical solution with RWA, the analytical approximation given by Eq. (4), and the numerical solution without RWA are shown in the respective panels. See Fig. 1 for other parameter values. The intensity values where the Rabi frequency equals multiples of the IR frequency are indicated in the left panel.

and

$$m = (2\Omega_{\text{Rabi}})^2/[(2\Omega_{\text{Rabi}})^2 + (\omega_2 - \omega_3)^2].$$

Here, the effect of the pulse duration of the control pulse and lifetimes is neglected, and the equation is expected to be valid for the first few femtoseconds after the action of the attosecond pulse.

In the following sections it is shown that the RWA is unsuitable in the high-field regime. For this purpose, the numerical solution of the von Neumann equation will be compared to the numerical solution within the RWA, where the terms that oscillate rapidly are neglected and the Hamiltonian is given by

$$\mathbf{H}^{\text{RWA}} = \begin{pmatrix} 0 & \frac{1}{2}d_{12}F_c(t) & 0 \\ \frac{1}{2}d_{12}F_c^*(t) & \omega_2 & \frac{1}{2}d_{23}F_c(t) \\ 0 & \frac{1}{2}d_{23}F_c^*(t) & \omega_3 \end{pmatrix}. \quad (5)$$

where $F_c(t)$ is the complex instantaneous electric field strength such that $F(t) = \text{Re}\{F_c(t)\}$. The atomic polarization response P_a can be calculated from the density matrix and the dipole transition matrix $\mathbf{d}$:

$$P_a = \text{Tr}(\rho \cdot \mathbf{d}) = 2d_{12}\text{Re}\{\rho_{12}\} \quad (6)$$

(the contribution from ρ_{23} is neglected here because $\rho_{23} \ll \rho_{12}$ in our regime). As shown in Fig. 1, the atomic polarization response exhibits numerous modulations on top of the carrier frequency. The approximation given by Eq. (4) (red dots) reproduces those modulations with high fidelity over the first few femtoseconds after t_0. The modulations correspond to the wide frequency spectrum that is generated for high field strengths (see next paragraph and Fig. 2). In contrast to that, the solution with the RWA (blue dashed line) deviates from the numerical solution (black solid line) already after a short time.

Here it is determined whether the approximation for the atomic polarization response given by Eq. (4) is also useful for spectroscopic studies. Figure 2 depicts the frequency spectrum of P_a as a function of intensity. Within the RWA, the spectrum shows two branches that diverge with growing intensity. This corresponds to the usual Autler-Townes doublet in absorption measurements. Without the RWA, the spectrum splits up into

multiple lines. This effect is partially captured by Eq. (4). The range of covered frequencies is well reproduced, but the exact way in which the frequency lines evolve is different for the numerical solution and the approximation. An interpretation of the multiple absorption lines can be given in the Floquet picture. The multiple absorption lines correspond to transitions from the ground state to the Floquet states, which are separated by multiples of the photon energy of the coupling field.

The appearance of multiple absorption lines for certain conditions was already found in microwave spectroscopy, where a strong radiofrequency field is coupled to a transition in a molecule [15]. The observation of a multiple line splitting in the close-to-optical–wavelength region, as depicted in Fig. 2, is hindered by the extreme field strengths that are required for the observation of the effect. While the necessary field strengths are now readily obtained with femtosecond pulses, the target gas will usually be ionized before the effect can be observed. However, the onset of the effect shown in Fig. 2, which is the observation of one peak in addition to the normal doublet in the absorption spectrum, was already observed in simulations (see, for example, Ref. [4]), and is also visible in recent experimental data [7].

A unique feature for the transmission control of attosecond pulses is the expected dependence of optical properties on the subcycle timing with respect to the femtosecond pulse. The prerequisite for the observation of this effect is that the probe pulse needs to be short compared to one optical cycle of the control field and can repeatedly be generated at a fixed time delay with respect to the control field, a condition that is met for attosecond pulses. Figure 3 depicts the dependence of the atomic polarization response to the subcycle timing of the control field. Within the RWA, the spectrum shows no subcycle dependence. In contrast to that, Eq. (4) reflects the dependence on t_0 with a half-cycle periodicity with respect to the control field, in accordance with the numerical solution.

The experimental observation of the subcycle dependent transmission of attosecond pulses will be complicated by the steep envelope of the femtosecond pulse because the electric field amplitude will always vary together with the time delay and also influence the polarization response. Another complication will arise from the contribution of other mechanisms, such as electron wave-packet interference [16]
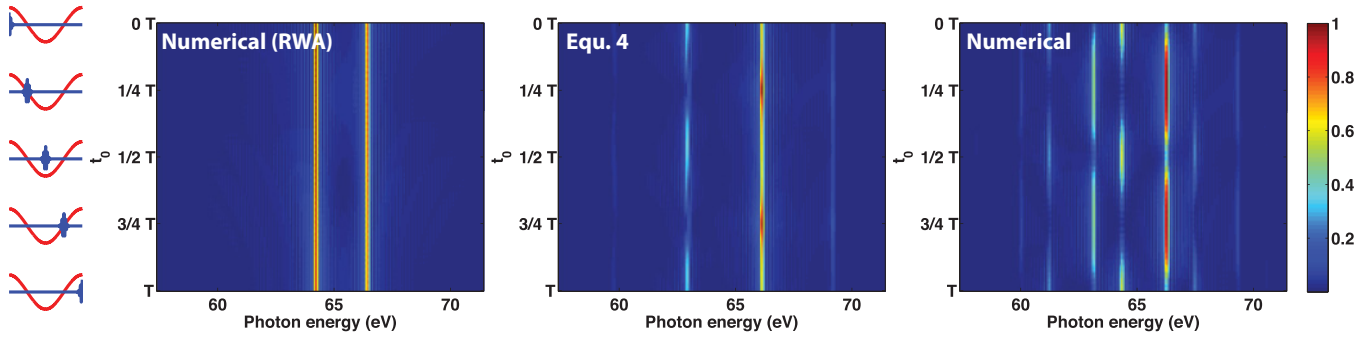
FIG. 3. (Color online) Frequency spectrum of polarization response P_a as a function of the delay time t_0 at an intensity of 0.3×10^{14} W/cm^2. The numerical solution with RWA, the analytical approximation given by Eq. (4), and the numerical solution without RWA are shown in the respective panels. One optical cycle of the control field T is 2.67 fs. See Fig. 1 for other parameter values.

in attosecond pulse trains (see Ref. [17] for an interpretation of the effect in the Floquet picture). These have already been shown to modulate the transient absorption spectrum with a half-cycle periodicity [18]. Also, Fano interference of auto-ionizing states can lead to half-cycle periodicity in photoelectron and absorption measurements [8,19].

While a future application of the effects described here might be pulse shaping of x rays, the easiest way to observe

these effects would be transient absorption experiments. The measurement of the optical spectrum before ($|\widetilde{E}_{\text{in}}(\omega)|^2$) and after ($|\widetilde{E}_{\text{out}}(\omega)|^2$) an interaction gas cell, with a setup like that in Refs. [5] or [18], determines the absorption signal A:

$$A = \ln(|\widetilde{E}_{\text{in}}(\omega)|^2/|\widetilde{E}_{\text{out}}(\omega)|^2). \quad (7)$$

The absorption signal is closely related to the atomic polarization response. Assuming that the polarization response
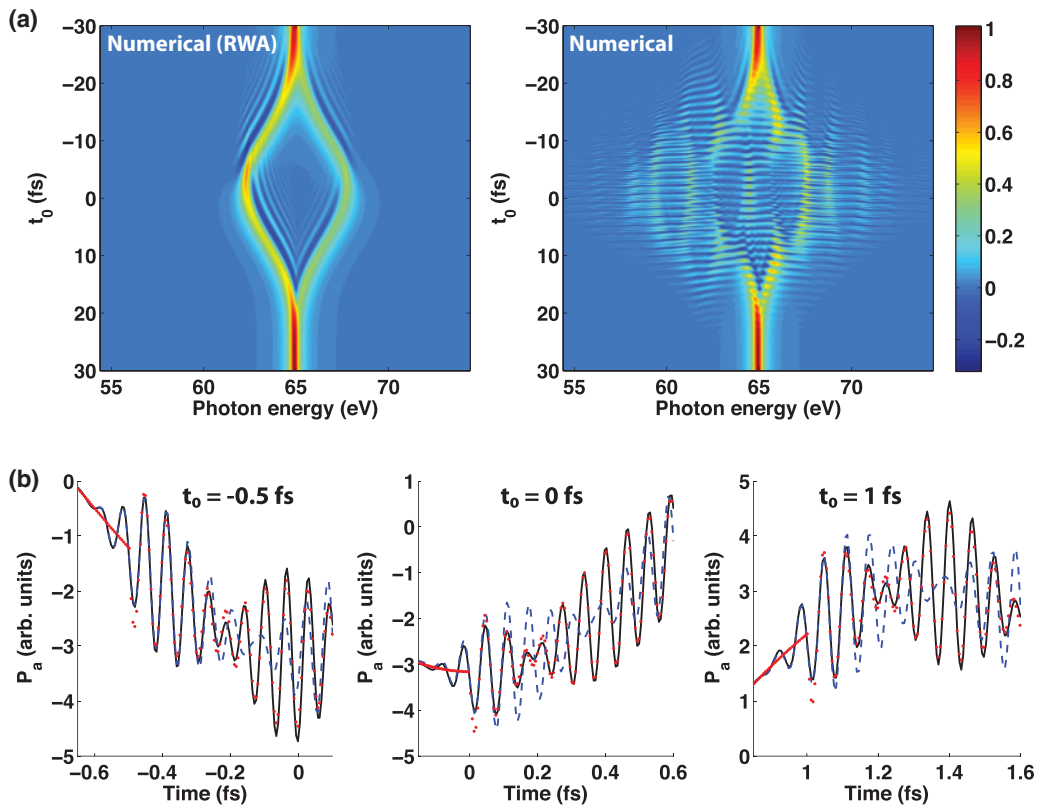

FIG. 4. (Color online) (a) Transient absorption signal A [Eq. (7)] dependent on the delay time t_0. The panels display the numerical solution with RWA and without RWA, respectively. The IR pulse has an intensity of 2×10^{14} W/cm^2 and a pulse length of 15 fs, the XUV pulse has an intensity of 1×10^{11} W/cm^2 and a pulse length of 100 as (the exact parameters of the XUV pulse do not change the result significantly). See Fig. 1 for other parameter values. (b) The atomic polarization response P_a [Eq. (6)] for selected values of the delay time t_0. The numerical solution with RWA, extracted from the calculations displayed in panel (a) and shifted by $0.5 P_a^{\text{static}}$ for better comparison with the other curves, is shown by the blue dashed line. The analytical approximation ($P_a + P_a^{\text{static}}$) given by Eqs. (9) and (4) is also shown (red dots). The numerical solution without RWA, extracted from the calculations displayed in (a), is shown by the black solid line.

is proportional to the electric field and neglecting reshaping effects as discussed in Ref. [4] (that is, assuming that Beer's law is valid), the spectrum after a thin interaction cell can be calculated as

$$\widetilde{E}_{\text{out}}(\omega) = \widetilde{E}_{\text{in}}(\omega) \exp\left[i\frac{2\pi\omega}{c}\frac{N\widetilde{P}_a(\omega)}{\widetilde{E}_{\text{in}}(\omega)}L\right], \qquad (8)$$

where N is the atomic number density in the gas cell and L is the length of the interaction region. Figure 4 displays the transient absorption of a 100-as pulse in the three-level system specified in Fig. 1. The spectroscopic effects described above, which are the splitting into multiple absorption lines and the subcycle modulations, are found in this simulation with realistic pulse parameters instead of the assumption of an infinitely short probe pulse and an infinitely long control

pulse that was used for the analytical derivation. Both effects are missing when the RWA is made. The asymmetry of the absorption spectrum depends on whether $\omega_2 > \omega_3$ (this is the case in the simulation shown in Fig. 4) or $\omega_2 < \omega_3$ (this assumption would cause the images shown in Fig. 4 to be mirrored around the y axis).

The atomic polarization response in the simulation with realistic pulse parameters (shown in Fig. 4) shows oscillations on two different time scales: there are slow oscillations that follow the electric field strength, and there are fast oscillations that arise after the action of the attosecond pulse. The fast oscillations have been discussed above; the slow oscillations are due to the dipole induced by the strong electric field in the three-level system. The slow oscillations can be calculated in the static field limit ($\omega_2 \gg \omega_{\text{IR}}$ and $\omega_3 \gg \omega_{\text{IR}}$):

$$P_a^{\text{static}} = -2\frac{d_{12}^2 F_{\text{IR}}(t)}{\omega_2}\left(1 - \frac{d_{23}^2 F_{\text{IR}}(t)^2(\omega_3 - \omega_2)}{\left[d_{23}^2 F_{\text{IR}}(t)^2 - \omega_2\omega_3\right](\omega_3 - \omega_2) + d_{12}^2 F_{\text{IR}}(t)^2\omega_2}\right). \qquad (9)$$

Figure 4(b) shows that the atomic polarization response directly after the attosecond pulse is reliably reproduced by $(P_a + P_a^{\text{static}})$ given by Eqs. (4) and (9), whereas the RWA solution deviates significantly. The results show that the analytical equations derived in this paper are also valid for realistic pulse parameters. They predict the burst of radiation emitted by the atomic dipole in response to an attosecond pulse, and therefore they might become useful for x-ray pulse-shaping applications.

In this paper a simplified model is discussed for the interaction of an isolated attosecond pulse with a three-level system that is dressed by a strong femtosecond pulse. An analytical equation for the time evolution of the polarization response is obtained, valid in the high-field limit. Two spectroscopic effects are predicted: the splitting of the absorption spectrum into a variety of lines rather than into a doublet for high intensities and the dependence of the absorption spectrum on the relative timing of the two pulses.

The effect of multiple lines in the absorption spectrum is visible in recent experimental data [7]. A time-dependent change of the absorption profile of isolated attosecond pulses has not been reported yet, but subcycle modulated absorption

features in transient absorption measurements using attosecond pulse trains have been reported [18].

The prospect of laser-mediated x-ray pulse shaping puts the focus on the time evolution of the fields, rather than spectroscopic studies. The treatment in the time domain pursued in this paper is particularly useful for that purpose, because the rapid decay of the core-hole states that are populated by x rays limits the atomic polarization response to the first few femtoseconds after excitation.

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