Carrier-envelope phase-sensitive inversion in two-level systems

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We theoretically study the carrier-envelope phase-dependent inversion generated in a two-level system by excitation with a few-cycle pulse. On the basis of the invariance of the inversion under time reversal of the exciting field, parameters are introduced to characterize the phase sensitivity of the induced inversion. Linear and nonlinear phase effects are numerically studied for rectangular and sinc-shaped pulses. Furthermore, analytical results are obtained in the limits of weak fields, as well as strong dephasing, and by nearly degenerate perturbation theory for sinusoidal excitation. The results show that the phase-sensitive inversion in the ideal two-level system is a promising route for constructing carrier-envelope phase detectors. © 2005 Optical Society of America

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1. INTRODUCTION

The two-level system is a fundamental and widely used model to describe the interaction of electromagnetic waves with matter. The temporal evolution of the system is usually treated within the framework of the rotating-wave approximation (RWA), in which the driving electric field enters the equations of motion only through its complex envelope and center frequency.[1] For a complete description of the electric field, the carrier-envelope (CE) phase, specifying the position of the envelope with respect to the rapidly oscillating carrier wave, also has to be taken into account. The RWA breaks down for strongly driven systems,[2] giving rise to new effects that not only depend on the pulse envelope and carrier frequency but also on the CE phase.

The phase-sensitive dynamics of the driven two-level system beyond the RWA has been the topic of several papers. The phase dependence of the inversion was carefully examined for a sinusoidal excitation, serving as a model for the interaction of atoms and molecules with continuous-wave laser radiation.[3–8] The discussion was extended to rectangular pulses, which can be obtained by abruptly switching on and off the sinusoidal excitation. The phase sensitivity of the inversion was investigated and experimentally demonstrated in the radio-frequency regime by researchers' exciting the anticrossing of the potassium 21*s*–19*f* states.[8]

Following the arrival of laser pulses consisting of only a few optical cycles, there has been considerable interest in phase-sensitive effects in the pulsed optical regime,[9] and various approaches have been used for detecting the CE phase and frequency.[10–16] In this context, the CE phase-dependent emission of two-level systems[17] and semiconductors[18] interacting with few-cycle pulses has been theoretically investigated, and the effect has experimentally been observed in GaAs.[19] So far, less attention has been given to the CE phase dependence of the inversion, with a few exceptions studying the interaction with Gaussian pulses.[20,21]

Whereas for sinusoidal excitation the generated inversion shows a CE phase dependence even for weak fields, the phase-sensitive dynamics relies completely on nonlinear effects for pulsed optical excitation. In this paper, both linear and higher-order phase-dependent inversion effects in two-level systems are theoretically investigated, using analytical approximations and numerical simulations with properly chosen test pulses. In addition, the influence of dephasing on the phase sensitivity is studied. General properties of the steady-state inversion are discussed, and approximate expressions are derived in the linear and the nonlinear regimes. The paper is organized as follows. In Section 2, the equations of motion for an excited two-level system are given in a fixed and a rotating reference frames. In Section 3, general properties of the steady-state inversion are discussed. In Section 4, the phase-dependent inversion in the weak-field limit is analytically examined, and, in Section 5, the discussion is extended beyond the linear regime for rectangular and sinc-shaped pulses, using numerical simulations and nearly degenerate perturbation theory. In Section 6, the influ-

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ence of the phase relaxation is studied on the basis of the strong dephasing approximation and numerical results. We conclude in Section 7.

2. EQUATIONS OF MOTION

A two-level system is characterized by its dipole matrix element d and resonance frequency $\omega_{ba}=2\pi f_{ba}=(E_b-E_a)/\hbar$, where E_a and E_b are the eigenenergies associated with the low- and high-energy states $|a\rangle$ and $|b\rangle$, respectively. Dissipative effects, which arise owing to the interaction of the ideal two-level system with its environment, can be taken into account in a statistical approach. The density matrix is represented by the components of the Bloch vector $\mathbf{s}$. The components s_1 and s_2 are related to the real and imaginary parts of the off-diagonal density-matrix elements by $\rho_{ab}=(s_1+\mathrm{i}s_2)/2$, and $\rho_{bb}-\rho_{aa}=s_3=w$ is the population inversion. In this paper, the relaxation processes are described by phenomenological parameters, the energy relaxation rate $\gamma_1=1/T_1$, and the dephasing rate $\gamma_2=1/T_2$. Frequently, relaxation is dominated by processes that lead to a destruction of the phase coherence in the quantum system without affecting the inversion, resulting in a dephasing time T_2, which is much shorter than the energy relaxation time T_1. For example, this is typically the case in a gas due to collision broadening.[22] Thus, in the following, we set $\gamma_1=0$, assuming that the energy relaxation processes are slow compared with the interaction time with the field, whereas we do allow for dephasing processes occurring on a time scale comparable with the duration of the exciting pulse.

Assuming linear polarization of the exciting field and a vanishing static dipole moment, the dynamics of the system is described by the Bloch equations[1]

$$\dot{s}_1=-\omega_{ba}s_2-\gamma_2 s_1,$$

$$\dot{s}_2=\omega_{ba}s_1+2\Omega s_3-\gamma_2 s_2,$$

$$\dot{s}_3=-2\Omega s_2. \tag{1}$$

The overdot denotes a time derivative. The electric field $E(t)$ is parametrized in terms of the instantaneous Rabi frequency $\Omega(t)=dE(t)/\hbar$, which can be written as

$$\Omega(t)=\Omega_R[\epsilon(t)\exp(-\mathrm{i}\omega_c t+\mathrm{i}\phi_{\mathrm{CE}})+\epsilon^*(t)\exp(\mathrm{i}\omega_c t-\mathrm{i}\phi_{\mathrm{CE}})]/2, \tag{2}$$

where the asterisk denotes the complex conjugate. Here, $\epsilon(t)$ is the normalized, in general, complex envelope function; $\omega_c=2\pi f_c$ is the carrier frequency; and ϕ_{CE} is the CE phase. In this paper, we refer to $\Omega_R=dE_0/\hbar$ as the (peak) Rabi frequency, with the maximum value of the electric field envelope E_0.

For some applications, it is convenient to transform the Bloch equations, Eqs. (1), into a rotating reference frame. The transformation between the Bloch vector $\mathbf{s}$ and the vector in the rotating frame $\mathbf{u}=(u,v,w)^{\mathrm{T}}$, where T denotes the transposed vector, is given by $\mathbf{u}(t)=A(t)\mathbf{s}(t)$:

$$A(t)=\begin{bmatrix}\cos(\omega_{ba}t) & \sin(\omega_{ba}t) & 0\\ -\sin(\omega_{ba}t) & \cos(\omega_{ba}t) & 0\\ 0 & 0 & 1\end{bmatrix}. \tag{3}$$

For $\gamma_1=0$, we obtain the equations of motion in the rotating frame:

$$\dot{u}=2\Omega\sin(\omega_{ba}t)w-\gamma_2 u, \tag{4a}$$

$$\dot{v}=2\Omega\cos(\omega_{ba}t)w-\gamma_2 v, \tag{4b}$$

$$\dot{w}=-2\Omega\sin(\omega_{ba}t)u-2\Omega\cos(\omega_{ba}t)v. \tag{4c}$$

3. STEADY-STATE INVERSION

We consider a two-level system excited by an electric field pulse, parametrized in terms of $\Omega(t)$, and neglect energy relaxation, i.e., $\gamma_1=0$. During interaction with the field, the inversion in the system evolves and reaches a steady-state value w_s, staying constant after interaction with the pulse. In this paper, we investigate the CE phase sensitivity of the steady-state inversion, which is readily accessible to experiments. We assume that the dipole moment of the system is initially vanishing, corresponding to the initial condition $s_1=s_2=0$ and $s_3=w_0$. This is always fulfilled if the system starts from equilibrium, where w_0 corresponds to the equilibrium inversion.

A. Inversion Invariance under Time Reversal of the Field

Strong excitation of the two-level system results in the breakdown of the RWA and of related features such as the area theorem.[2] By way of contrast, the property discussed in the following is valid even in the strong-field limit.

We take the Bloch equations, Eqs. (4), assuming $\gamma_1=0$ and an initially vanishing dipole moment. Under these conditions, the steady-state inversion w_s, after interaction with an arbitrary pulse $\Omega(t)$, is the same as for excitation with the time-reversed pulse $\Omega(-t)$. The proof is given in Appendix A. This feature is remarkable, in that the temporal evolution of the system from its initial state is completely different in both cases and should not be confused with the time-reversed dynamics of the system. In Fig. 1,

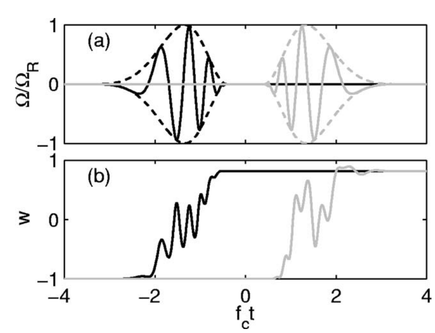

Fig. 1. Dynamics of the two-level system for excitation with a field $\Omega(t)$ (black curve) and the corresponding time-reversed field $\Omega(-t)$ (gray curve). (a) Time-dependent electric field, parametrized in terms of the normalized Rabi frequency. (b) Evolution of the inversion for a Rabi frequency $\Omega_R=1.3\ \omega_c$, a transition frequency $\omega_{ba}=1.7\ \omega_c$, and a dephasing rate $\gamma_2=f_c/10$.

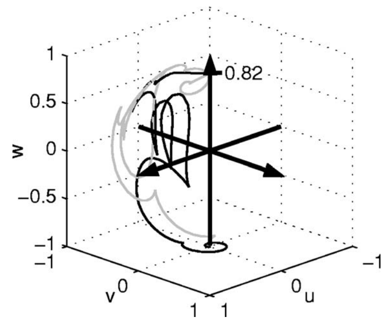

Fig. 2. Bloch vector trajectories for excitation of a two-level system with a driving field (black curve) and the time-reversed field (gray curve). The fields and system parameters are the same as for Fig. 1.

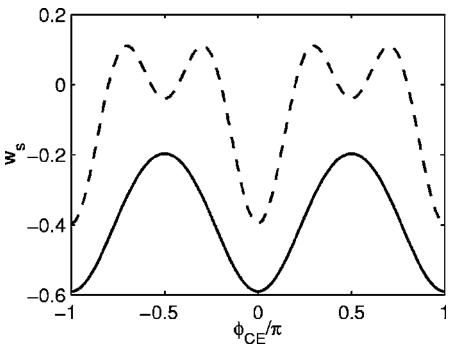

Fig. 3. Phase-dependent steady-state inversion w_s after interaction with a two-cycle sinc pulse, for Rabi frequencies $\Omega_R=1.5\ \omega_c$ (solid curve) and $\Omega_R=2.3\ \omega_c$ (dashed curve). The transition frequency is $\omega_{ba}=1.5\ \omega_c$, and the dephasing time is $T_2=10/f_c$.

the invariance of w_s with respect to time reversal of the field is illustrated by means of an example. The time-dependent field of an exciting pulse is shown in Fig. 1(a), as well as the corresponding time-reversed field. The pulse shape is, in principle, arbitrary; the only condition is that the pulse has a finite energy, and thus the field approaches zero for $t\to\pm\infty$. For a propagating electric field, we furthermore require a vanishing dc component. This condition, however, is not crucial for the inversion invariance discussed here. In Fig. 1(b), the time-dependent inversion is displayed, starting at the initial value $w_0=-1$. The evolution of the inversion is completely different for excitation with the field and the time-reversed field, but, still, the same steady-state value is reached in both cases. This can also be seen in the Bloch sphere representation. In Fig. 2, the trajectories of the Bloch components u,v, and w in the rotating frame are shown, with the south pole of the sphere corresponding to the initial state. The Bloch vectors evolve in completely different ways, but the same steady-state inversion is reached. In the presence of dephasing, the case that is shown here, the trajectories approach the same point for $t\to\infty$ because the dipole moment decays; i.e., u and v tend to zero for $t\to\infty$. By way of contrast, for $\gamma_2=0$, u and v approach different steady-state values for excitation with the field and the time-reversed field, respectively. Thus, for $t\to\infty$, the two trajectories still reach the same value for w but not for u and v.

B. Steady-State Inversion for Symmetric Pulses

In Sections 5 and 6, we discuss the two-level dynamics for excitation by sinc-shaped and rectangular pulses, which are chirp free and have symmetric intensity envelopes. For such pulses [or, more generally, for pulses with $\epsilon(t)=\epsilon^*(-t)$], the time reversal of the field corresponds to a mere sign change of the CE phase, as can be seen from Eq. (2). As a consequence of the time-reversal invariance discussed above, we have $w_s(\phi_{\mathrm{CE}})=w_s(-\phi_{\mathrm{CE}})$ for these pulses. Owing to the centrosymmetry of the two-level system, the inversion is furthermore invariant with respect to a sign change of the electric field, corresponding to a π shift in the CE phase, i.e., $w_s(\phi_{\mathrm{CE}})=w_s(\phi_{\mathrm{CE}}+\pi)$. Thus, the CE phase-dependent occupation probability can be represented by a π periodic Fourier series. If $w_s(\phi_{\mathrm{CE}})=w_s(-\phi_{\mathrm{CE}})$ holds, only the even (i.e., cosine) terms remain. For nonresonant excitation and nonexcessive field strengths, the series can be truncated after the first-order term, and we obtain

$$w_s(\phi_{\mathrm{CE}})=\bar{w}_s+\Delta\cos(2\phi_{\mathrm{CE}}). \tag{5}$$

The parameters used here are the CE phase-averaged inversion,

$$\bar{w}_s=[w_s(0)+w_s(\pi/2)]/2, \tag{6}$$

and the modulation amplitude

$$\Delta=[w_s(0)-w_s(\pi/2)]/2, \tag{7}$$

with $-1\leqslant\bar{w}_s,\Delta\leqslant 1$.

Figure 3 shows the phase dependence of w_s after interaction of the two-level system with a sinc pulse. For $\Omega_R=1.5\omega_c$, the phase dependence of w_s can be well approximated by Eq. (5). For $\Omega_R=2.3\omega_c$, higher-order terms in the Fourier expansion result in a bump around the symmetry point. For excitation near the two-level resonances $\omega_{ba}=(2n+1)\omega_c$, $n=0, 1, 2, \ldots$, the approximation Eq. (5) already breaks down for smaller Rabi frequencies; i.e., more terms in the Fourier series are necessary for a full description.

4. WEAK-FIELD LIMIT

As mentioned in Section 1, linear phase-sensitive effects have been observed for custom-tailored microwave and radio-frequency pulses, whereas, for laser-generated optical pulses, a CE phase dependence can be obtained only in the nonlinear regime. The phase-dependent dynamics of an excited two-level system can be treated analytically by a time-dependent perturbation-series expansion, with the first-order approximation describing the evolution in the linear regime. We use the Bloch equations in the rotating reference frame; see Eqs. (4). As the initial condition at $t\to-\infty$, we choose $\mathbf{u}_0=(0,0,w_0)^{\mathrm{T}}$. By formally integrating Eqs. (4a) and (4b) and inserting the results into Eq. (4c), we arrive at an integral equation for $w(t)$:

$$w(t)=w_0-4\int_{-\infty}^{t}\mathrm{d}t'\int_0^{\infty}\mathrm{d}\tau\cos(\omega_{ba}\tau)\times\exp(-\gamma_2\tau)\Omega(t')\Omega(t'-\tau)w(t'-\tau). \tag{8}$$

In the weak-field limit, the time-dependent inversion changes only slightly with respect to its initial value. The first-order approximation (which is second order in Ω_R) is obtained by one's inserting the zeroth- order solution $w^{(0)}(t)=w_0$ into the right-hand side of Eq. (8):

$$w^{(1)}(t) = w_0\left[1 - 4\int_0^{\infty} \mathrm{d}\tau \cos(\omega_{ba}\tau)\exp(-\gamma_2\tau) \times \int_{-\infty}^{t} \mathrm{d}t'\,\Omega(t')\Omega(t'-\tau)\right]. \tag{9}$$

The result for the steady-state inversion $w_s=w(t\to\infty)$ is favorably expressed in terms of the Fourier transform

$$\Omega(\omega) = \int_{-\infty}^{\infty} \mathrm{d}t\,\Omega(t)\exp(\mathrm{i}\omega t), \tag{10}$$

yielding

$$w_s^{(1)} = w_0\left[1 - \int_0^{\infty} \mathrm{d}\omega |\Omega(\omega)|^2 H(\omega)\right]. \tag{11}$$

Thus, in the linear regime, the relationship between w_s and the power spectrum of the pulse $|\Omega(\omega)|^2$ can be described in terms of a spectral filter function $H(\omega)$, which is given by

$$H(\omega) = \frac{4}{\pi}\frac{\gamma_2\left(\omega^2 + \gamma_2^2 + \omega_{ba}^2\right)}{\left(\omega^2 + \gamma_2^2 + \omega_{ba}^2\right)^2 - 4\omega_{ba}^2\omega^2}. \tag{12}$$

For $\gamma_2 \ll \omega_{ba}$, we can make the approximation

$$H(\omega) = \frac{4}{\pi}\frac{\gamma_2/2}{(\omega - \omega_{ba})^2 + \gamma_2^2}, \tag{13}$$

and, in the limit of no dephasing, $\gamma_2 \to 0$, we obtain

$$H(\omega) = 2\delta(\omega - \omega_{ba}). \tag{14}$$

The Fourier transform of $\Omega(t)$, Eq. (10), can, with Eq. (2), be written as

$$\Omega(\omega) = \Omega_R[\epsilon(\omega - \omega_c)\exp(\mathrm{i}\phi_{\mathrm{CE}}) + \epsilon^*(-\omega - \omega_c)\exp(-\mathrm{i}\phi_{\mathrm{CE}})]/2. \tag{15}$$

The power spectrum $|\Omega(\omega)|^2$ is only phase independent if $\epsilon(\omega)=0$ for $\omega \leqslant -\omega_c$, which, for chirp-free pulses, corresponds to the condition $\epsilon(\omega)=0$ for $|\omega| \geqslant \omega_c$.[12]

Formally, Eq. (8) is a Volterra integral equation of the second kind, as can be shown by exchanging the order of integration. The weak-field approximation, Eq. (9), is thus equivalent to a first-order Neumann-series expansion,[23] yielding $(w_s - w_0) \propto \Omega_R^2$ and also $\Delta \propto \Omega_R^2$ for pulses with phase-dependent power spectra. Thus, the amount of generated inversion and its phase dependence scale linearly with the peak intensity of the pulse. This is different for pulses with phase-independent power spectra, where, in the weak-field limit, the phase dependence of w_s is provided by the second-order term. In this case, we obtain $\Delta \propto \Omega_R^4$ (or $\Delta \propto \Omega_R^3$ for two-level systems with a static dipole moment[21]); i.e., the generated inversion becomes phase insensitive for weak fields.

In the microwave regime, pulses with the desired envelopes and CE phases can be custom tailored. For example, pulses with durations far below one carrier cycle are available, giving rise to linear CE phase-dependent processes.[24] For the experimental demonstration of the phase-sensitive inversion, pulses in the radio-frequency regime with approximately rectangular envelopes, which have a phase-dependent power spectrum, were applied.[8] For such pulses, linear filter effects can be used to determine the CE phase, and a phase-dependent inversion w_s is obtained even in the weak-field limit. Strictly speaking, this is also true for Gaussian or hyperbolic secant pulse shapes, often used as a model for laser pulses.[20,21,25] In contrast, for optical pulses emitted by current laser systems, the power spectrum is phase independent. As a consequence, a CE phase-dependent inversion w_s can be observed only in the nonlinear regime.

5. SYSTEMS WITHOUT DISSIPATION

In the following, we investigate the phase dependence of the steady-state inversion for interaction with rectangular and sinc pulses, serving as typical model pulses for excitation in the radio-frequency and optical regimes, respectively. In this section, we neglect dissipation; i.e., we set $\gamma_1=\gamma_2=0$. The rectangular pulse corresponds to a sinusoidal field of finite duration and is thus closely related to continuous-wave excitation, which plays an important role in atomic and molecular physics and quantum chemistry.[3–8] This particularly basic pulse shape features a piecewise-constant envelope. The periodicity of the field can be exploited for approximately solving the equations of motion, and basic features of the solutions can be derived. In the radio-frequency regime, rectangular pulse shapes can be approximately generated by one's switching on and off a sinusoidal field, and they were used for the experimental demonstration of the phase-sensitive inversion.[8] As already pointed out, rectangular pulses have a phase-dependent power spectrum, yielding phase-sensitive effects even in the linear regime. Such pulses are currently not available in the optical regime from laser sources. Sinc pulses, on the other hand, feature a rectangular, phase-independent power spectrum and have proven to be a fairly good description of few-cycle laser pulses.[25]

A. Excitation with Rectangular Pulses

For rectangular pulses, the envelope in Eq. (2) is given by $\epsilon(t)=1$ for $-T/2 \leqslant t \leqslant T/2$ and $\epsilon(t)=0$ otherwise, where T is the pulse duration. For one to avoid a nonpropagating dc component for arbitrary CE phases ϕ_{CE}, the pulses must contain an integer number of optical cycles; i.e., T is an integer multiple of the carrier period $T_0=1/f_c$. For $\phi_{\mathrm{CE}} \neq (n+1/2)\pi$, $n \in \mathbb{Z}$, the field strength exhibits discontinuities, giving rise to a CE phase-dependent power spectrum. In Fig. 4, the power spectrum $|\Omega(\omega)|^2$ of a rectangular two-cycle pulse is displayed. The CE phase dependence of the power spectrum also becomes clear from the spectral properties of the pulse. Because the pulse spectrum does not vanish identically for $|\omega| \geqslant \omega_c$, the two components in Eq. (15), centered around ω_c and $-\omega_c$, have a region of spectral overlap, resulting in a CE phase-dependent interference.

In the following, we investigate the phase sensitivity of the inversion by numerically solving the Bloch equations, Eqs. (1). Figure 5 shows the average inversion $\bar{w}_s$ and the modulation amplitude Δ as a function of Ω_R and ω_{ba}. In Figs. 5(a) and 5(b), $\bar{w}_s$ is displayed for excitation with rectangular single-cycle and two-cycle pulses, respectively. For weak fields, the inversion is largest for near-resonant excitation. This resonance is more pronounced for the two-cycle pulses, owing to their narrower spectrum as compared with single-cycle pulses. For Rabi frequencies approaching or even exceeding ω_c, regions with strong inversion can also be found for off-resonant excitation due to higher-order transitions. Δ is shown in Figs. 5(c) and 5(d), again for excitation with rectangular single-cycle and two-cycle pulses. During interaction with the pulse, the inversion performs oscillations due to carrier-wave Rabi flopping,[2] and the steady-state value after interaction with the pulse w_s strongly depends on the pulse and system parameters. As a consequence, for $\bar{w}_s$, as well as Δ, regions with positive and negative values alternate. For rectangular pulses, the generated inversion exhibits a phase dependence even in the linear regime, as discussed in Section 4. We introduce the relative modulation amplitude or modulation depth

$$\delta = \Delta/(\bar{w}_s - w_0), \tag{16}$$

which is helpful to discuss the phase sensitivity in the weak-field limit. A value of $\delta = \pm 1$ indicates maximum possible CE phase modulation of the generated inversion, and $\delta = 0$ indicates no modulation. Although the amount of generated inversion vanishes for low field strengths, the relative phase sensitivity of w_s does not disappear in the case of phase-dependent power spectra, yielding a finite value for δ in the weak-field limit.

For interaction with a rectangular pulse shape, δ does not depend on the pulse duration T. This is a consequence of the periodic excitation, as shown in Appendix B, and is true only in two-level systems without dissipation. An approximate solution beyond the weak-field limit can be obtained by using almost degenerate perturbation theory,[26–28] where the perturbation parameter is the normalized Rabi frequency Ω_R/ω_c. The zeroth-order perturbation theory, corresponding to the RWA,[28] does not contain any CE phase dependence and thus fails in predicting phase-sensitive effects.[21] Following the procedure in Ref. 28, we obtain the first-order perturbative result

$$\delta_p = \frac{(\omega_{ba}^2 - \omega_c^2)(\omega_{ba} + \omega_c) + \omega_c \Omega_R^2}{(\omega_{ba}^2 + \omega_c^2)(\omega_{ba} + \omega_c) - \omega_c \Omega_R^2}; \tag{17}$$

see Appendix C. Figure 6 shows the perturbative approximation and the numerical solution for the modulation depth. The perturbative result is in good agreement with the exact solution, especially for moderate field strengths and moderate detuning, where the almost degenerate per-

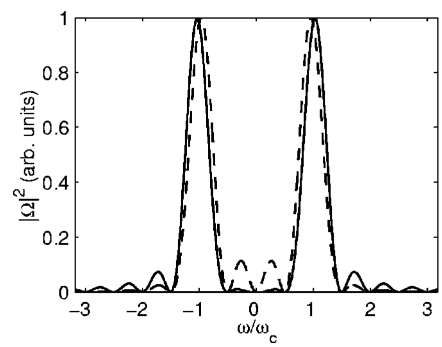

Fig. 4. Rectangular two-cycle pulse: power spectrum for CE phases 0 (solid curve) and $\pi/2$ (dashed curve).

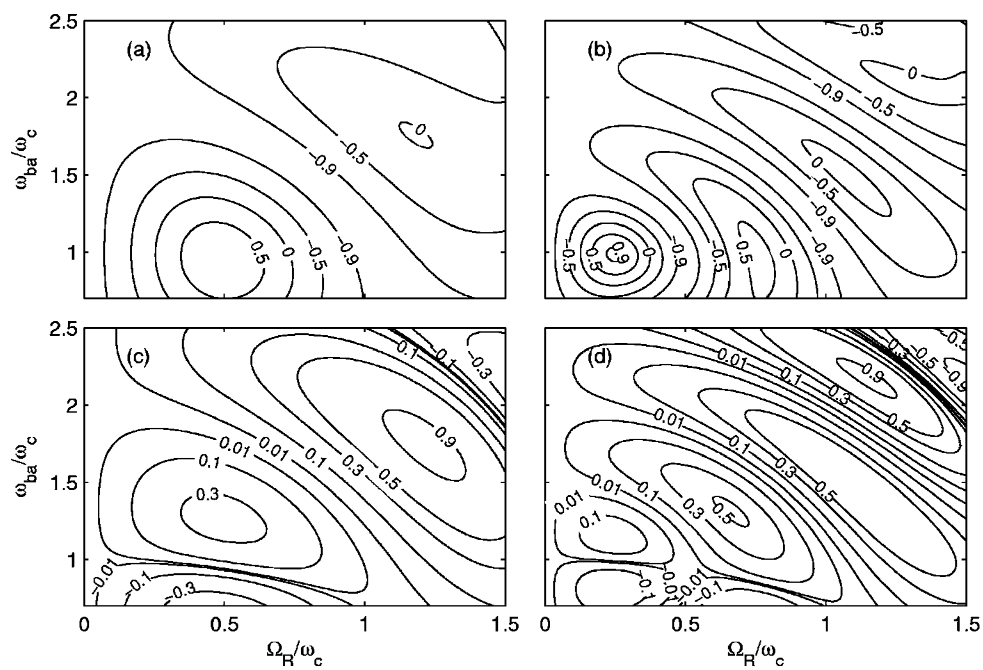

Fig. 5. Average inversion $\bar{w}_s$ and modulation amplitude Δ as a function of the Rabi frequency Ω_R and the transition frequency ω_{ba} in units of ω_c. Displayed are $\bar{w}_s$ after interaction with rectangular (a) single-cycle and (b) two-cycle pulses and Δ after interaction with rectangular (c) single-cycle and (d) two-cycle pulses.

turbative treatment converges best.[28] For small Rabi frequencies Ω_R, w_s approaches zero, but the relative phase sensitivity of the inversion does not disappear; i.e., the modulation depth stays finite. In the limit $\Omega_R \to 0$, where the perturbation theory becomes exact and coincides with the weak-field approximation [see Eqs. (11) and (14)], we obtain

$$\delta_p = (\omega_{ba}^2 - \omega_c^2)/(\omega_{ba}^2 + \omega_c^2) \tag{18}$$

(except for odd multiphoton resonances, where δ becomes degenerate for $\Omega_R \to 0$ and the approximation breaks down). Thus, in the weak-field limit, $\delta > 0$ for excitation below resonance, and $\delta < 0$ for excitation above resonance. For resonant excitation ($\omega_c = \omega_{ba}$), where the population transfer reaches its maximum, we obtain $\delta = 0$; i.e., the inversion does not show a phase sensitivity.

As already mentioned, the RWA does not predict any phase dependence in the inversion. This might be counterintuitive, especially for pulses with CE phase-dependent power spectra, where the generated inversion depends on the CE phase even in the weak-field limit. A straightforward explanation is that the validity of the RWA is restricted to near-resonant excitation,[22] whereas, on the other hand, the modulation depth in Eq. (18) indicates a phase sensitivity of the inversion just for excitation off resonance.

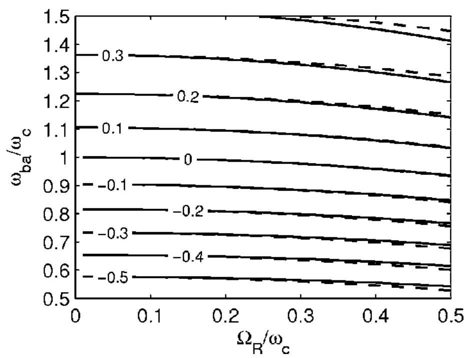

Fig. 6. Modulation depth δ as a function of the Rabi frequency Ω_R and the transition frequency ω_{ba} in units of ω_c: perturbative (dashed curves) and exact numerical result (solid curves).

B. Excitation with Sinc-Shaped Pulses

Sinc pulses are characterized by the full width at half-maximum (FWHM) value T of the intensity envelope and the Rabi frequency Ω_R. The normalized envelope function $\epsilon(t)$, introduced in Eq. (2), is given by $\epsilon(t) = \mathrm{sinc}(t/\tau) = \sin(t/\tau)/(t/\tau)$. The FWHM pulse duration is $T = 2.783\tau$. These pulses have a rectangular spectrum, extending from $f_c - 1/(2\pi\tau)$ to $f_c + 1/(2\pi\tau)$. We require $\tau > 1/(2\pi f_c)$ in order to avoid unphysical dc components. Under this condition, the power spectrum is CE phase independent; see discussion after Eq. (15). Thus the inversion does not exhibit a CE phase sensitivity in the linear regime. We note that Gaussian or hyperbolic secant pulse descriptions feature unphysical dc components and a phase-sensitive power spectrum. This can lead to erroneous results, especially in the weak-field limit or for strong dephasing (see Section 6), in which the remaining phase sensitivity is due to the phase-dependent power spectrum, as opposed to nonlinear electric field effects.

The Bloch equations, Eqs. (1), are numerically solved. In Fig. 7, the average inversion $\bar{w}_s$ and the modulation amplitude Δ are displayed as a function of Ω_R and ω_{ba}. In Figs. 7(a) and 7(b), $\bar{w}_s$ is shown for excitation with sinc-shaped single-cycle and two-cycle pulses, and the corresponding Δ is displayed in Figs. 7(c) and 7(d). As in the case of excitation with rectangular pulses (see Fig. 5), regions with positive and negative values alternate for both

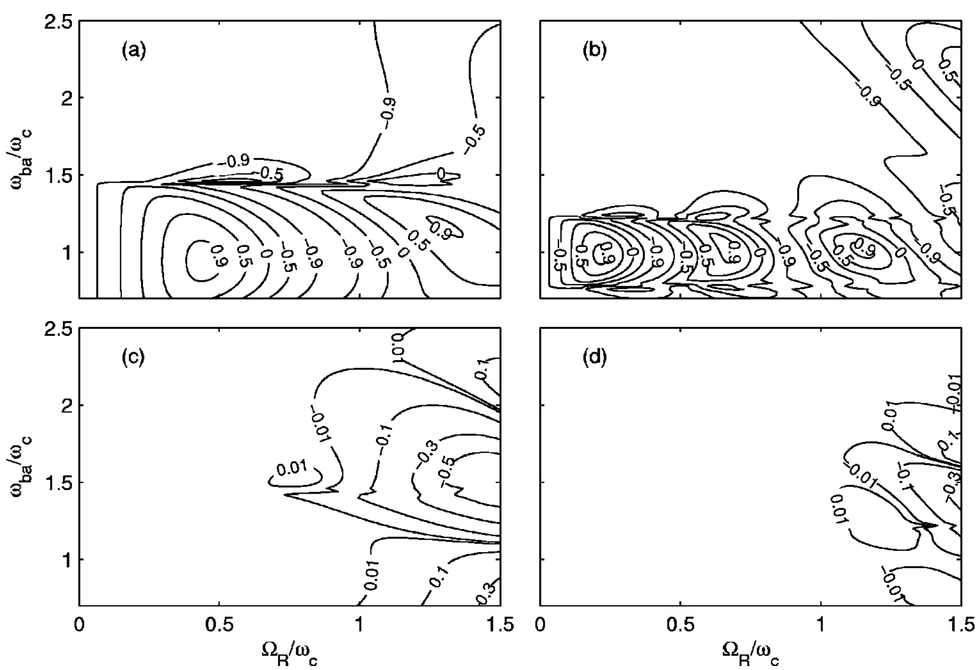

Fig. 7. Average inversion $\bar{w}_s$ and modulation amplitude Δ as a function of the Rabi frequency Ω_R and the transition frequency ω_{ba} in units of ω_c. Displayed are $\bar{w}_s$ after interaction with sinc-shaped (a) single-cycle and (b) two-cycle pulses and Δ after interaction with sinc-shaped (c) single-cycle and (d) two-cycle pulses.

$\bar{w}_s$ and Δ. The inversion $\bar{w}_s$ is largest for near-resonant excitation, and the resonance is more pronounced for two-cycle pulses, which have a narrower spectrum. Higher-order transitions become relevant if Ω_R approaches or even exceeds ω_c, leading to regions with strong inversion also for off-resonant excitation. As discussed in Section 4, the steady-state inversion shows no phase dependence in the linear regime; i.e., the modulation depth δ [see Eq. (16)] approaches zero for sinc pulses. A significant phase sensitivity can only be found for field strengths in the regime of carrier-wave Rabi flopping, in which nonlinear effects play a significant role. For sinc-shaped pulses, which have smooth envelopes unlike rectangular pulses, the phase sensitivity of w_s depends considerably on the pulse duration in that the phase dependence of the field is more pronounced for shorter pulses. Thus, higher field strengths are necessary to obtain a significant modulation amplitude Δ for excitation by two-cycle pulses as compared with single-cycle pulses.

6. INFLUENCE OF DEPHASING

In the following, we investigate the influence of phase-relaxation processes on the CE phase-dependent inversion. In the limit of strong dephasing, an approximate expression can be derived. We start from the integral equation Eq. (8). Because the Rabi oscillations of the inversion are significantly dampened and the kernel decays rapidly owing to strong dephasing, we can approximate $w(t'-\tau)\approx w(t')$. By differentiation with respect to t, we obtain the first-order differential equation

$$\dot{w}(t) = -4w(t)\int_0^\infty \mathrm{d}\tau \cos(\omega_{ba}\tau)\exp(-\gamma_2\tau)\Omega(t')\Omega(t'-\tau), \tag{19}$$

with the solution

$$w(t) = w_0 \exp\left[-4\int_0^\infty \mathrm{d}\tau \cos(\omega_{ba}\tau)\exp(-\gamma_2\tau) \times \int_{-\infty}^{t} \mathrm{d}t'\,\Omega(t')\Omega(t'-\tau)\right]. \tag{20}$$

For weak fields, where the exponential can be approximated by a first-order Taylor series, this expression coincides with the weak-field approximation; see Eq. (9). In analogy to Eq. (11), we can write the steady-state inversion as

$$w_s = w_0 \exp\left[-\int_0^\infty \mathrm{d}\omega |\Omega(\omega)|^2 H(\omega)\right], \tag{21}$$

with the spectral filter function $H(\omega)$ defined in Eq. (12) and the Fourier transform $\Omega(\omega)$ given in Eq. (10). As discussed in Section 4, this expression contains a CE phase sensitivity only for pulses with a phase-dependent power spectrum. If γ_2 is large compared with ω_{ba}, ω_c, and the spectral width of the electric field, the steady-state inversion is given by

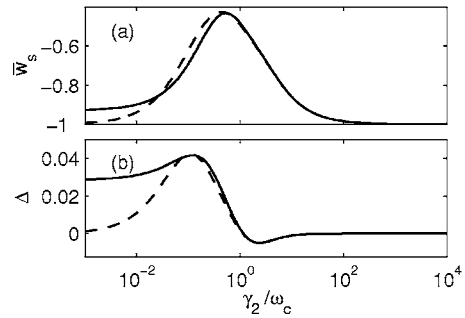

Fig. 8. Excitation with a rectangular two-cycle pulse: exact numerical result (solid curves) and strong dephasing approximation (dashed curves) for the (a) average inversion $\bar{w}_s$ and (b) modulation amplitude Δ as a function of the normalized dephasing rate γ_2/ω_c. The Rabi frequency is $\Omega_R = 0.25\ \omega_c$, and the transition frequency is $\omega_{ba} = 1.5\ \omega_c$.

$$w_s = w_0 \exp\left[-\frac{4}{\pi\gamma_2}\int_0^\infty \mathrm{d}\omega |\Omega(\omega)|^2\right] = w_0 \exp\left[-\frac{4}{\gamma_2}\int_{-\infty}^\infty \mathrm{d}t |\Omega(t)|^2\right], \tag{22}$$

which does not depend on the CE phase or the pulse shape but only on the total pulse energy. This result can also be obtained by adiabatic elimination of the polarization in Eq. (4).

In Figs. 8–11, the results of the numerical simulation (solid curves) and the strong dephasing approximation (dashed curves) for the CE phase-dependent steady-state inversion are shown as a function of γ_2/ω_c. In Fig. 8, the phase-averaged inversion $\bar{w}_s$ and the modulation amplitude Δ are displayed for interaction with a rectangular pulse, assuming a moderate Rabi frequency $\Omega_R = 0.25\ \omega_c$. The results of the approximation and the numerical simulation agree well, especially for increased dephasing, $\gamma_2 \gtrsim 0.1\omega_c$. Figure 9 shows the phase-averaged inversion $\bar{w}_s$ for excitation by sinc-shaped pulses with the same parameters as before. Here, even better agreement between numerical and analytical results is found. The CE phase dependence of the inversion is negligible in this case, $|\Delta| < 10^{-7}$, because nonlinear phase-sensitive effects do not play a significant role at those field strengths. Figures 10 and 11 show again the results for excitation by rectangular and sinc-shaped pulses, respectively, but now for a higher Rabi frequency $\Omega_R = \omega_c$. In this case, the validity range of the strong dephasing approximation is reduced as compared with $\Omega_R = 0.25\ \omega_c$, and good agreement with the exact result is obtained only for $\gamma_2 \gtrsim \omega_c$.

The inversion dynamics under the influence of phase relaxation can be classified into different regimes. In the Rabi flopping regime, associated with moderate dephasing, the inversion dynamics is governed by carrier-wave Rabi oscillations. As discussed in Section 5, the steady-state inversion exhibits a significant phase sensitivity for sufficiently short and strong pulses or for pulses with phase-dependent power spectra. For higher dephasing rates, the near-transparency regime is reached, characterized by an almost CE phase-independent steady-state inversion $w_s \approx 0$. This regime can be associated with dephasing values of roughly $\gamma_2/\omega_c \approx 0.5 \ldots 5$ in Fig. 10

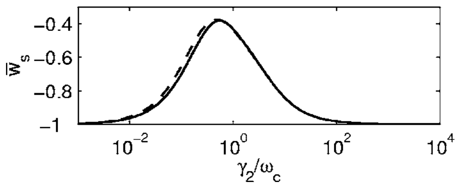

Fig. 9. Excitation with a sinc-shaped two-cycle pulse: exact numerical result (solid curve) and strong dephasing approximation (dashed curve) for the average inversion $\bar{w}_s$ as a function of the normalized dephasing rate γ_2/ω_c. The Rabi frequency is $\Omega_R = 0.25\ \omega_c$, and the transition frequency is $\omega_{ba} = 1.5\ \omega_c$.

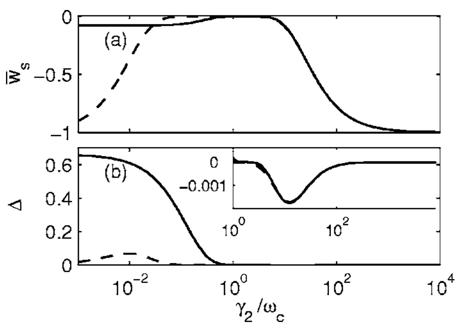

Fig. 10. Excitation with a rectangular two-cycle pulse: exact numerical result (solid curves) and strong dephasing approximation (dashed curves) for the (a) average inversion $\bar{w}_s$ and (b) modulation amplitude Δ as a function of the normalized dephasing rate γ_2/ω_c. The Rabi frequency is $\Omega_R = \omega_c$, and the transition frequency is $\omega_{ba} = 1.5\ \omega_c$. The inset shows Δ at an increased scale.

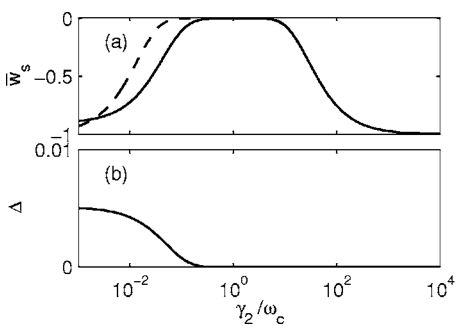

Fig. 11. Excitation with a sinc-shaped two-cycle pulse: exact numerical result (solid curves) and strong dephasing approximation (dashed curve) for the (a) average inversion $\bar{w}_s$ and (b) modulation amplitude Δ as a function of the normalized dephasing rate γ_2/ω_c. The Rabi frequency is $\Omega_R = \omega_c$, and the transition frequency is $\omega_{ba} = 1.5\ \omega_c$. For Δ, only the numerical result is shown because the approximation is not CE phase sensitive for sinc pulses.

and $\gamma_2/\omega_c \approx 0.2 \ldots 5$ in Fig. 11. It does not exist if the excitation is too weak; see Figs. 8 and 9. In the limit of strong dephasing, w_s approaches its initial value w_0 because the evolution of the inversion is suppressed. The freezing of the quantum states can be interpreted as the quantum Zeno effect,[29] in which the decoherence of the states is induced by frequent measurement or, more generally, by rapid system-environment interactions. In the transition regime between transparency and freezing of the states, the phase sensitivity slightly recovers for the rectangular pulse, reaching an extremal value of $\Delta = -1.8 \times 10^{-3}$ in the inset of Fig. 10. In the quantum Zeno regime, corresponding to $\gamma_2/\omega_c \gtrsim 5 \times 10^3$ in Figs. 10 and 11, the modulation amplitude Δ and also the modulation depth $\delta = \Delta/(\bar{w}_s - w_0)$ asymptotically approach zero.

7. CONCLUSION

In conclusion, we have studied the carrier-envelope phase sensitivity of the steady-state inversion w_s, remaining in a two-level system after interaction with a pulse. We examined the invariance of w_s under time reversal of the field. On the basis of this property, we introduced as parameters the phase-averaged inversion $\bar{w}_s$ and the modulation amplitude Δ. We discussed the two-level dynamics analytically in the weak-field limit, in which a phase sensitivity of the inversion could be found only for pulses with phase-dependent power spectrum. Beyond the linear regime, the phase sensitivity of w_s was studied numerically for excitation with rectangular and sinc-shaped pulses. For the rectangular pulse, it was furthermore proven that the modulation depth $\delta = \Delta/(\bar{w}_s - w_0)$ does not depend on the pulse length, and an approximate analytical expression for δ was derived by application of almost degenerate perturbation theory. The influence of phase relaxation on the inversion was investigated on the basis of the strong dephasing approximation and numerical simulations.

For interaction with few-cycle laser pulses, the two-level model predicts a considerable phase dependence of w_s if the Rabi frequency approaches or even exceeds the carrier frequency; see Fig. 7. The necessary field strengths can be reached even for unamplified femtosecond pulses directly out of the laser oscillator when the light is tightly focused onto the sample.[18] These model calculations show that the phase-dependent inversion is a promising route for the CE phase detection of few-cycle laser pulses. One has to be aware of the fact that the applicability of the two-level model to an atomic, molecular, or solid-state system in strong fields has its limitations. Nevertheless, recent theoretical and experimental research shows that basic features of the two-level model are preserved for the strong-field excitation with few-cycle laser pulses.[18,19]

The generated inversion can be detected optically, either by an additional probe beam or by the short-wavelength spectral component of the pulse itself, which is delayed against the excitation part of the pulse and can, for example, be chosen at resonance with the two-level system. It is also possible to read out the excited electrons electronically with a field-ionization pulse.[8] Experimentally, such a detector could be realized by a gas or a metal vapor. For example, in the two-level approximation, we obtain with $\Omega_R = 1.5\omega_c$ a modulation amplitude of $\Delta = -0.34$ for interaction of two-cycle Ti:sapphire laser pulses with a transition around 589 nm, corresponding to the famous D lines in sodium. Another option might be the use of artificial quantum systems such as quantum dots.

APPENDIX A: INVERSION INVARIANCE UNDER TIME REVERSAL OF THE FIELD

We now want to show that, for $\gamma_1 = 0$ and vanishing initial dipole moment, $s_1 = s_2 = 0$ for $t \to -\infty$, the remaining inversion after passage of the pulse is invariant under time reversal of the field $\Omega(t)$. This should not be confused with the time-reversed dynamics of the system. The Bloch equations in the rotating frame, Eqs. (4), can in matrix form be written as

$$\dot{\mathbf{u}} = 2\Omega M\mathbf{u} - \gamma_2(u_1,u_2,0)^{\mathrm{T}}, \tag{A1}$$

with

$$M(t) = \begin{bmatrix} 0 & 0 & \sin(\omega_{ba}t) \\ 0 & 0 & \cos(\omega_{ba}t) \\ -\sin(\omega_{ba}t) & -\cos(\omega_{ba}t) & 0 \end{bmatrix} \tag{A2}$$

and $\mathbf{u}=(u_1,u_2,u_3)^{\mathrm{T}}$. We assume that the pulse extends from $-t_0$ to t_0. As initial condition at $t=-t_0$, we choose $\mathbf{u}_0=(0,0,w_0)^{\mathrm{T}}$. First we assume $\gamma_2=0$. The formal solution of Eq. (A1) is then given by

$$\begin{aligned}\mathbf{u}(t_0) &= \hat{T}\exp\left[\int_{-t_0}^{t_0} 2\Omega(t)M(t)\mathrm{d}t\right]\mathbf{u}_0 \\ &= \lim_{N\to\infty}\prod_{n=-N}^{N}\exp[2\Omega(t_{-n})M_{-n}t_0/N]\mathbf{u}_0 \\ &= \lim_{N\to\infty}\prod_{n=-N}^{N}[I+2\Omega(t_{-n})M_{-n}t_0/N]\mathbf{u}_0,\end{aligned} \tag{A3}$$

with the time-ordering operator $\hat{T}$, the unity matrix I, $t_n=t_0n/N$, and $M_n=M(t_n)$. By formally multiplying out the last line of Eq. (A3), we see that the solution is only invariant under time reversal of an arbitrary field $\Omega(t)$ if

$$M_{-n_1}M_{-n_2}\ldots M_{-n_\ell}\mathbf{u}_0 = M_{n_\ell}\ldots M_{n_2}M_{n_1}\mathbf{u}_0, \tag{A4}$$

for any subset of indices n with $-N\leq n_1<n_2<\ldots<n_\ell\leq N$. For an even number ℓ, we obtain

$$\begin{aligned}M_{n_\ell}\ldots M_{n_2}M_{n_1}\mathbf{u}_0 = w_0(-1)^{\ell/2} &\times\prod_{m=1}^{\ell/2}\cos[\omega_{ba}(t_{n_{2m-1}}-t_{n_{2m}})] \\ &\times(0,0,1)^{\mathrm{T}}.\end{aligned} \tag{A5}$$

If ℓ is odd, we obtain

$$\begin{aligned}M_{n_\ell}\ldots M_{n_2}M_{n_1}\mathbf{u}_0 = w_0(-1)^{(\ell-1)/2} &\times\prod_{m=1}^{(\ell-1)/2}\cos[\omega_{ba}(t_{n_{2m-1}}-t_{n_{2m}})] \\ &\times[\sin(\omega_{ba}t_\ell),\cos(\omega_{ba}t_\ell),0]^{\mathrm{T}}.\end{aligned} \tag{A6}$$

Thus for odd ℓ Eq. (A4) is fulfilled only for the third vector component, and thus only $s_3(t_0)$ is invariant under time reversal of the field.

The case with $\gamma_2\neq 0$ can be handled analogously by using the transformation $u_1=\tilde{u}_1\exp(-\gamma_2 t), u_2=\tilde{u}_2\exp(-\gamma_2 t)$. Equation (A4) must now be fulfilled for the matrix

$$\tilde{M}(t) = \begin{bmatrix} 0 & 0 & \sin(\omega_{ba}t)\exp(\gamma_2 t) \\ 0 & 0 & \cos(\omega_{ba}t)\exp(\gamma_2 t) \\ -\sin(\omega_{ba}t)\exp(-\gamma_2 t) & -\cos(\omega_{ba}t)\exp(-\gamma_2 t) & 0 \end{bmatrix}. \tag{A7}$$

For an even number ℓ, we obtain

$$\begin{aligned}\tilde{M}_{n_\ell}\ldots\tilde{M}_{n_2}\tilde{M}_{n_1}\mathbf{u}_0 = w_0(-1)^{\ell/2} &\times\prod_{m=1}^{\ell/2}\{\exp[\gamma_2(t_{n_{2m-1}}-t_{n_{2m}})] \\ &\times\cos[\omega_{ba}(t_{n_{2m-1}}-t_{n_{2m}})]\} \\ &\times(0,0,1)^{\mathrm{T}}.\end{aligned} \tag{A8}$$

If ℓ is odd, we obtain

$$\begin{aligned}\tilde{M}_{n_\ell}\ldots\tilde{M}_{n_2}\tilde{M}_{n_1}\mathbf{u}_0 = w_0(-1)^{(\ell-1)/2} &\times\prod_{m=1}^{(\ell-1)/2}\{\exp[\gamma_2(t_{n_{2m-1}}-t_{n_{2m}})] \\ &\times\cos[\omega_{ba}(t_{n_{2m-1}}-t_{n_{2m}})]\} \\ &\times\exp(\gamma_2 t_\ell)[\sin(\omega_{ba}t_\ell),\cos(\omega_{ba}t_\ell),0]^{\mathrm{T}}.\end{aligned} \tag{A9}$$

Again, Eq. (A4) is fulfilled only for the third vector component, and thus only $s_3(t_0)$ is invariant under time reversal of the field.

APPENDIX B: MODULATION DEPTH FOR THE RECTANGULAR PULSE

In the following, we want to show that, for interaction with a rectangular pulse shape, the modulation depth δ, defined in Eq. (16), does not depend on the pulse duration T. Assuming no dissipation, we can describe the state of the two-level system by the ket $|\psi(t)\rangle=c_a(t)|a\rangle+c_b(t)|b\rangle$, with the state vector $\mathbf{c}(t)=[c_a(t),c_b(t)]^{\mathrm{T}}$. The initial condition $\rho_{ab}=(s_1+is_2)/2=c_a^*c_b=0$ (see Section 3) is here implemented by one's choosing the initial state vector $\mathbf{c}_0=(1,0)^{\mathrm{T}}$, corresponding to $w_0=-1$. The evolution of the system can be described by the Schrödinger equation. For an exciting field with period T_0, we can bring the solution into the form

$$\mathbf{c}_n = U^n\mathbf{c}_0,$$

where U is the unitary transformation matrix and $\mathbf{c}_n$ is the state vector after n periods.[8] We diagonalize U by means of a unitary matrix $T, \mathbf{c}_n'=T\mathbf{c}_n, U'=TUT^{-1}$ and obtain for the jth component of $\mathbf{c}'_n$

$$c'_{nj} = \exp(-\mathrm{i}nW_jT_0/\hbar)c'_{0j}.$$

Here, W_j are the Floquet energies, which do not depend on the CE phase ϕ_{CE} of the exciting field. By back transformation, we obtain the occupation probability of the upper level:

$$|c_b(nT_0)|^2 = \left| \frac{[\exp(-\mathrm{i}nW_bT_0/\hbar) - \exp(-\mathrm{i}nW_aT_0/\hbar)]T_{aa}T_{ba}}{T_{aa}T_{bb} - T_{ab}T_{ba}} \right|^2, \tag{B1}$$

and the inversion is given by $w(nT_0)=2|c_b(nT_0)|^2-1$. The matrix elements T_{ij} depend on ϕ_{CE} but not on the time nT_0. Thus, the time dependence cancels out in the expression for the modulation depth; see Eq. (16). We note that for the initial condition $s_1=s_2=0$ the modulation depth is invariant with respect to w_0 in that the choice of w_0 just leads to a scaling of the Bloch vector solution $\mathbf{s}(t)$ in Eqs. (1). Thus, the proof is also valid for $w_0 \neq -1$.

APPENDIX C: ALMOST-DEGENERATE PERTURBATION THEORY

Following the formalism in Ref. 28, we can derive a perturbative result for the phase-dependent inversion. In this appendix, the references to equations refer to Ref. 28 unless otherwise stated, and we adopt the nomenclature used there, with $\omega_0=\omega_{ba}, \omega=\omega_c$, and $\lambda=-\Omega_R/2$. As initial condition, we assume $w_0=-1$ as in Appendix B (also see remark at the end of Appendix B), corresponding to $\rho_{\alpha\alpha}(t_0)=1$, $\rho_{\beta\beta}(t_0)=\rho_{\alpha\beta}(t_0)=\rho_{\beta\alpha}(t_0)=0$ in Ref. 28. From Eq. (1.33), we get after a few straightforward manipulations the upper-state population $\rho_{\beta\beta}(t)=[w(t)+1]/2$ in a two-level system:

$$\begin{aligned}\rho_{\beta\beta}(t) &= \left|\sum_m \langle\beta,m|\chi_+\rangle\exp(\mathrm{i}\omega mt)\right|^2 \left|\sum_m \langle\alpha,m|\chi_+\rangle\exp(\mathrm{i}\omega mt_0)\right|^2 \\ &+ \left|\sum_m \langle\beta,m|\chi_-\rangle\exp(\mathrm{i}\omega mt)\right|^2 \left|\sum_m \langle\alpha,m|\chi_-\rangle\exp(\mathrm{i}\omega mt_0)\right|^2 \\ &+ 2\Re\left\{\exp[2\mathrm{i}q(t-t_0)]\left[\sum_m \langle\alpha,m|\chi_+\rangle\exp(\mathrm{i}\omega mt_0)\right]\right. \\ &\times\left[\sum_m \langle\chi_-|\alpha,m\rangle\exp(-\mathrm{i}\omega mt_0)\right] \\ &\times\left[\sum_m \langle\chi_+|\beta,m\rangle\exp(-\mathrm{i}\omega mt)\right] \\ &\left.\times\left[\sum_m \langle\beta,m|\chi_-\rangle\exp(\mathrm{i}\omega mt)\right]\right\}.\end{aligned} \tag{C1}$$

Here, $|\alpha,m\rangle, |\beta,m\rangle$ are the pseudostates, and $|\chi_+\rangle, |\chi_-\rangle$ are the Floquet modes of the two-level system. The frequency q is given in Eq. (1.71). With Eqs. (1.66), (1.73), (1.74), and (1.76), we obtain in first-order approximation

$$\begin{aligned}|\chi_+\rangle &= N[\cos(\theta)(|\alpha,0\rangle - \eta|\beta,1\rangle) + \sin(\theta)(|\beta,-1\rangle + \eta|\alpha,-2\rangle)], \\ |\chi_-\rangle &= N[-\sin(\theta)(|\alpha,0\rangle - \eta|\beta,1\rangle) + \cos(\theta)(|\beta,-1\rangle + \eta|\alpha,-2\rangle)],\end{aligned} \tag{C2}$$

with the normalized field $\eta=\lambda/(\omega+\omega_0)$. The normalization constant N is given in Eq. (1.80b), and $\sin(\theta),\cos(\theta)$ are defined in Eq. (1.72). Furthermore, by inserting Eq. (1.77) in Eq. (1.73), we obtain within the first-order approximation

$$\begin{aligned}\epsilon_{jj} &= -\hbar\omega_0/2 - \eta^2\hbar(\omega_0+\omega), \\ \epsilon_{kk} &= \hbar\omega_0/2 - \hbar\omega + \eta^2\hbar(\omega_0+\omega), \\ \epsilon_{jk} &= \epsilon_{kj} = \eta\hbar(\omega_0+\omega) - \eta^3\hbar(\omega_0+\omega).\end{aligned} \tag{C3}$$

The case $\phi_{\mathrm{CE}}=0$ corresponds to a cosine pulse extending from $t_0=-T/2$ to $t=T/2$, where T is equal to or an integer multiple of the carrier period $T_0=2\pi/\omega$. We obtain for the remaining population after passage of the pulse

$$\begin{aligned}\rho_{\beta\beta}(T/2) &= 2N^4[\sin(\theta) - \eta\cos(\theta)]^2 \\ &\times[\cos(\theta) + \eta\sin(\theta)]^2[1-\cos(2qT)].\end{aligned} \tag{C4}$$

The case $\phi_{\mathrm{CE}}=-\pi/2$ corresponds to a cosine pulse extending from $t_0=-T/2-T_0/4$ to $t=T/2-T_0/4$. We obtain

$$\begin{aligned}\rho_{\beta\beta}(T/2-T_0/4) &= 2N^4[\sin(\theta) + \eta\cos(\theta)]^2 \\ &\times[\cos(\theta) - \eta\sin(\theta)]^2[1-\cos(2qT)].\end{aligned} \tag{C5}$$

With Eq. (16) in our paper, we get the modulation depth

$$\delta = \frac{2\eta\sin(\theta)\cos(\theta)[\sin(\theta)^2 - \cos(\theta)^2](1-\eta^2)}{\sin(\theta)^2\cos(\theta)^2(1-6\eta^2+\eta^4)+\eta^2}. \tag{C6}$$

We now insert Eq. (1.72), making use of the relations $\sin^2(\theta)-\cos^2(\theta)=-\Delta/q, \sin(\theta)\cos(\theta)=\epsilon_{jk}/(2q\hbar)$. The frequencies q and Δ are given in Eq. (1.71). We multiply the numerator and denominator of our expression for δ by q^2/η^2 and subsequently keep only the terms up to second order in η. Making the transition to the nomenclature used in our paper, with $\omega_0 \to \omega_{ba}$, $\omega \to \omega_c$, and $\eta \to -\Omega_R/[2(\omega_{ba}+\omega_c)]$, we finally obtain the perturbative result given in Eq. (17) in our paper.

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