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Interference-induced transparency and coherent control of quantum systems by frequency-chirped pulses

A. Nazarkin, R. Netz, and R. Sauerbrey
Institut für Optik und Quantenelektronik, Friedrich-Schiller-Universität, Max-Wien-Platz 1, D-07743 Jena, Germany
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A selective excitation technique based on light interference is proposed to control quantum systems by frequency-chirped laser fields. Interference of two identical, delayed and phase-shifted pulses is used to modulate the laser spectrum and project it onto the time domain. By adjusting the delay and phase shift, selected transitions can be brought into the ''holes'' of the spectrum and thus remain nonexcited. The possibility to selectively manipulate or even ''shut down'' resonant transitions, making the medium transparent to the field, is shown for the Rb atom.

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The manipulation of quantum systems using phase-amplitude shaped optical pulses has been a field of intense studies in recent years. An important goal is to bring an ensemble of atoms or molecules into a specific quantum state in order to facilitate selective breaking of chemical bonds [1–3]. Femtosecond-laser pulses represent an effective solution to this problem, since the bandwidth of the pulses may be large enough to contain all the frequency components of the multistep quantum transition of interest [2,4]. By stretching a femtosecond pulse in dispersive elements one produces a frequency-swept field which is applied to excite atoms or molecules in the regime of adiabatic rapid passage (ARP) [5–7]. The approach was demonstrated for quantum systems of different (electronic, vibrational, rotational) nature and different (ladder-, V-, and Λ-type) level configurations [5–9]. The problem of selectivity of population transfer by the ARP technique has been the focus of these investigations. In the ideal situation, the instantaneous laser frequency follows the spacing between the successive transitions of the quantum path, resulting in an enhanced population transfer. Actually, many quantum systems show a strong anharmonicity, so that the optimum quantum pathway cannot be ideally fitted by a smooth monotonous frequency sweep. It has been found that in some simple systems (such as a three-level ladder [6]) the selectivity can be achieved even though the frequency sweep does not exactly follow the frequencies of the successive transitions. Selective population transfer in atomic systems with closely lying upper sublevels was demonstrated [7]. These studies revealed that the mechanism responsible for the selectivity in the ARP regime is essentially nonlinear, basically arising from the redistribution of adiabatic quantum paths in the strong field. The results suggest that to achieve the selectivity in systems with a strong anharmonicity, one needs, correspondingly, a stronger driving field, roughly, in proportion to the anharmonicity of the transitions involved in the interaction [6,7,9]. Experimentally, the strong-field regime might bring into existence additional quantum paths, cause self-focusing and medium ionization, leading to a reduction of the selectivity. Therefore, alternative mechanisms which can provide selectivity at much lower intensity level (say, in the linear interaction regime) are of great physical interest, especially in the spectral regions [e.g., in UV and XUV (extreme ultraviolet)] where available coherent sources are not sufficiently energetic, and, in addition, the conventional pulse shaping technique [3] can hardly be applied. The recent studies [10,11] on high-order harmonic generation (HHG) revealed that harmonics are produced in the form of attosecond pulses with frequency modulation close to the linear one, and thus development of appropriate selective excitation mechanisms with chirped pulses can give direct access to coherent quantum control experiments with attosecond time resolution.

In this paper, we propose an alternative approach to the problem of selectivity with broadband frequency-chirped radiation, which principally does not need increase of intensity. The idea consists in the using a superposition of two identical time-delayed and phase-shifted pulses. As a result of optical interference, the laser spectrum becomes sinusoidally modulated, and the frequency-chirping projects this modulation onto the time domain. By monitoring the delay and phase shift between the pulses, certain transitions can be brought in the ''holes'' of the pulse spectrum and remain nonexcited. This enables one to suppress parallel channels and, thus, to reach the target state via the optimum quantum path. The potential of this approach is demonstrated by modeling the population dynamics in atomic Rb. The feasibility to selectively excite or ''shut down'' resonant transitions, thereby making the atoms transparent to the laser field, is shown.

To get insight into the physics of the discussed approach, we consider a coherent superposition of two identical laser fields characterized by the relative temporal delay $\Delta\tau$ and phase shift $\Delta\psi$. Assuming that the laser beams have identical transverse profile, we have $E_\Sigma(t)=E_1(t)+E_2(t)=\frac{1}{2}A_0(t)\exp[i\omega_0 t]+\frac{1}{2}A_0(t-\Delta\tau)\exp[i\omega_0(t-\Delta\tau)+i\Delta\psi]+\text{c.c.}$, where $A_0(t)$ is the complex amplitude of each pulse. By performing Fourier transformation of $E_\Sigma(t)$, we get the spectral intensity of the total field:

$$|F_\Sigma(\omega)|^2=4|F_0(\omega)|^2\cos^2[(\omega\Delta\tau+\Delta\psi)/2], \qquad (1)$$

where $F_0(\omega)$ is the spectrum of a single pulse. As follows from Eq. (1), the spectrum of the total field has the same envelope as the single-pulse spectrum but is modulated due to optical interference. The modulation picture depends on the delay between the pulses and the phase shift. In particu-

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lar, when the phase in Eq. (1) is such that $(\omega\Delta\tau+\Delta\psi)=\pi(2k+1), k=0,\pm1,\ldots$, then the intensity of the spectral components at frequency $\omega_k=[\pi(2k+1)-\Delta\psi]/\Delta\tau$ equals zero. By increasing or decreasing the delay $\Delta\tau$ between the pulses the spacing between the zeros of the modulated spectrum can be made smaller or larger, and variation of the phase shift translates the whole interference structure along the frequency axis. The technique of spectral manipulation, first proposed by Ramsey [12], was later applied to study ultranarrow resonances in atoms [4,13] and normally uses two spectrally limited (free of phase modulation) laser pulses. This method provides information about the structure of transitions in terms of the absorption linewidth, but does not allow one to localize interaction with a selected transition in the time domain, which is of prime importance for the control of the evolution of the system.

The new point is that the temporal dynamics can be introduced in this technique by using a superposition of temporally overlapped frequency-chirped pulses. We consider that the combined field is passed through a dispersive element, where the phase of the field acquires a frequency dependent shift: $\varphi(\omega)=\varphi(\omega_0)+\varphi'(\omega_0)(\omega-\omega_0)+\frac{1}{2}\varphi''(\omega_0)(\omega-\omega_0)^2$. Assuming a Gaussian shape for both input pulses, $A_0(t)\propto\exp[-(t/\tau_0)^2]$, the complex amplitude of each pulse after the element is proportional to $A_0^{str}(t)\propto\exp[-(t/\tau_S)^2+i\alpha t^2]$ and describes a phase-modulated pulse with duration $\tau_S=\tau_0\sqrt{(1+4\varphi''^2/\tau_0^4)}$ and a chirping parameter $\alpha=2\varphi''/(\tau_0\tau_S)^2$. We assume that the dispersive element provides a sufficient stretching, $\tau_S\gg\tau_0$, so that the relative contribution of the phase modulation in the spectrum is much greater than that of the amplitude one. If the delay is small, $\Delta\tau\ll\tau_S$, the total field is approximated by

$$E_\Sigma^{str}(t)\approx 2|A_0^{str}(t)|\cos\left[(\omega_0+2\alpha t)\frac{\Delta\tau}{2}+\frac{\Delta\psi}{2}\right]\times\cos[\omega_0 t+\alpha t^2+\psi_\Sigma], \tag{2}$$

where $\psi_\Sigma=\alpha\Delta\tau^2/4+\Delta\psi/2$. As can be seen, the combined field is a frequency-chirped pulse whose amplitude is temporally modulated at frequency $\Omega_{mod}=\alpha\Delta\tau$. By introducing the instantaneous frequency of the field, $\omega(t)=\partial\Phi/\partial t=(\omega_0+2\alpha t)$, where $\Phi(t)$ is the global phase, we find that the zeros of the amplitude modulation are determined by the same condition as the zeros of the spectral amplitude (1). This reflects the fact that the chirping projects the spectrum (1) onto the time domain, and the excitation dynamics can now be controlled in time. By placing selected quantum transitions at the zeros of the spectral modulation, one can switch-off the parallel quantum channels, thereby allowing the system to evolve in the given quantum-mechanical direction [Figs. 1(a) and 1(b)].

From the context of the problem it follows that to provide the selectivity, the combined field has to be not too strong, so that the target state population can be controlled by the parameters of the modulation. The relevant region of laser field intensities is determined by the condition

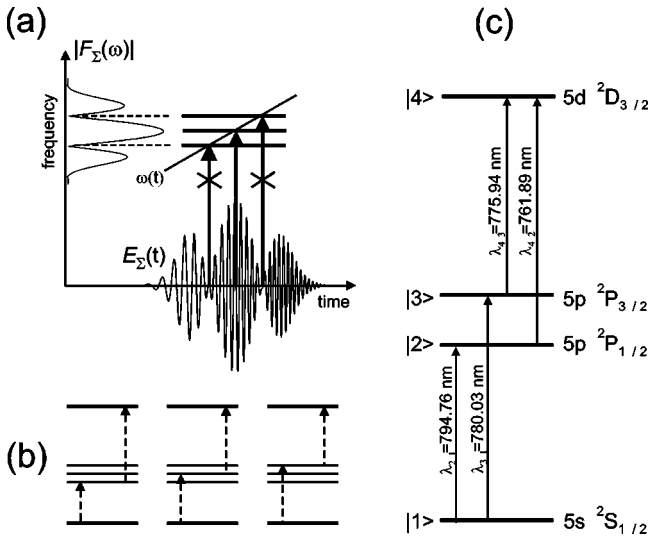

FIG. 1. (a) Interference of two identical delayed chirped pulses result in a sinusoidal modulation of the spectrum which is projected onto the time domain. (b) By placing certain transitions in the ''holes'' of the spectrum one can reach the target state via a selected path. (c) Two parallel quantum paths in Rb.

$$\gamma=\frac{\Omega_{Rabi}^2}{|d\omega(t)/dt|}\equiv\frac{|\mu_{mn}E_\Sigma|^2}{2|\alpha|\hbar^2}\leqslant 1, \tag{3}$$

where γ is the parameter of adiabaticity of the process [16] and μ_{mn} is the dipole moment of the transition. Since the regime (3) is far from the ARP regime ($\gamma\gg1$), the dynamics of population transfer is governed both by resonant and nonresonant interaction, leading to fast oscillation of the populations near the asymptotic value after the level crossing [17,18]. This imposes limitations on the rate of frequency sweeping and the modulation frequency. The time interval between subsequent level crossings [$T_{cr}\simeq\Delta\omega/(2\alpha)$, where $\Delta\omega$ is the anharmonicity of the system] and the modulation period ($T_{mod}=2\pi/\Omega_{mod}$) have to be long compared to the transition time τ_{tr}:

$$\tau_{tr}=(\tau_{jump}+\tau_{relax})\ll T_{mod}\leqslant T_{cr}. \tag{4}$$

Here the jump time (τ_{jump}) is the time the system takes to be first populated to the level of the asymptotic value $N_{exc}(\infty)$, and the relaxation time is the time it takes for the oscillations to damp to a small value $\varepsilon N_{exc}(\infty)$, where $\varepsilon\ll1$ [18]. Analysis of condition (4) based on the Landau-Zenner (LZ) model [16,18] shows that in the region defined by Eq. (3), the jump time is close to the low-intensity value: $\tau_{jump}=\sqrt{2\pi/\alpha}$, while the relaxation time, being independent of the field at low intensities ($\tau_{relax}=(1/\varepsilon)\sqrt{1/\pi\alpha}$), rapidly decreases with increasing the field. Assuming 5% criterion ($\varepsilon=0.05$) for the relaxation time, we find that the rate of frequency sweeping is limited by $\alpha<0.01\Delta\omega^2$, and the optimum delay lies in the region $\Delta\tau\simeq2\pi/\Delta\omega$.

If conditions (3) and (4) are fulfilled, interaction of the modulated field with the system can be treated as a sequence of LZ transitions. For example, by applying LZ formula

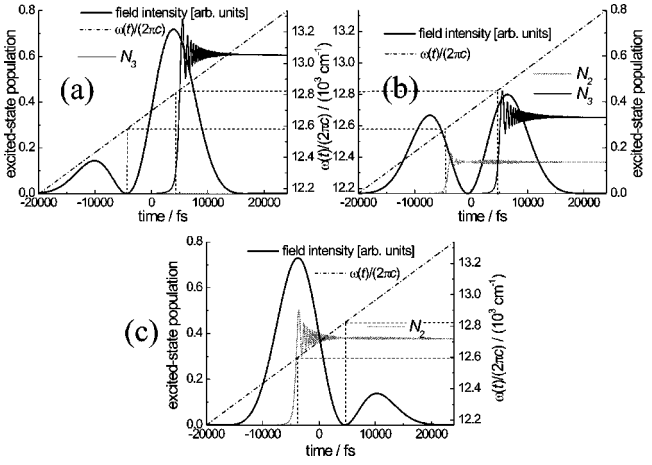

FIG. 2. Temporal control of the D-line transitions in Rb ($A_0 = 5.0\times10^6$ V/m, $\Delta\lambda = 24$ nm, $\Delta\tau = 59.67$ fs, $\varphi'' = 2.0\times10^5$ fs^2). (a) $\Delta\psi = 0$: $|1\rangle\rightarrow|3\rangle$ is switched on, while $|1\rangle\rightarrow|2\rangle$ is suppressed. (b) $\Delta\psi = \pi/2$: both transitions are switched on. (c) $\Delta\psi = \pi$: $|1\rangle\rightarrow|2\rangle$ is switched on, $|1\rangle\rightarrow|3\rangle$ is suppressed.

[16,18] to a V-type system with multiple upper sublevels, the population on the mth sublevel after the interaction is found to be

$$N_m = \exp\left(-\frac{\pi}{|\alpha|}\sum_{k=1}^{m-1}\Omega^2(t_k)\right)\left(1-\exp\left[-\frac{\pi}{|\alpha|}\Omega^2(t_m)\right]\right), \tag{5}$$

where $\Omega(t_k) = (2|\mu_{0k}|/\hbar)|A_0^{str}(t_k)|\cos[\omega(t_k)\Delta\tau/2 + \Delta\psi/2]$ is the Rabi frequency associated with transition from the ground (0) to the upper (k) state, which is calculated at the moment of the level crossing: $\omega(t_k) = (\omega_0 + 2\alpha t_k) = \omega_{0k}$ and $t_1 < \cdots < t_{k-1} < t_k$. By properly adjusting the free parameters of the modulation ($\Delta\tau$ and $\Delta\psi$), so that at the time of crossing the Rabi frequency is zero, the interaction with selected transitions can be suppressed. The variable phase shift can be produced in different ways: for example, using the carrier-envelope phase control technique [14], or by pulse scattering on a variable phase volume diffraction grating [15].

To demonstrate the concept of selective excitation, we consider interaction of laser field with Rb atom. The schematic of levels involved in the interaction is shown in Fig. 1(c). The initial population begins in the ground state $5s\,^2S_{1/2}$. The excited states $5p\,^2P_{1/2}$ and $5p\,^2P_{3/2}$ are coupled by the laser field and form a V-type system. If the laser spectrum is sufficiently broad coupling of the two excited $5p\,^2P_{1/2,3/2}$ states to the upper lying $5d\,^2D_{3/2}$ state occurs. Although there exists also coupling to the closely lying $5d\,^2D_{5/2}$ state, this channel is not shown in Fig. 1(c), because practically it does not affect the selective population of $5d\,^2D_{3/2}$ state. To model a coherent control experiment in Rb, the standard approach based on the solution of the time-dependent density-matrix equations [16] for the multilevel system was used. Figure 2 shows the dynamics of the populations in the V system and the behavior of the field intensity and the instantaneous frequency for two interfering Gaussian

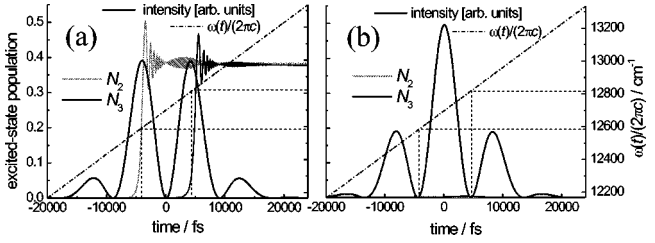

FIG. 3. Interference-induced transparency in Rb [$\Delta\tau = 139.21$ fs (see Fig. 2)]. (a) $\Delta\psi = \pi$. (b) $\Delta\psi = 0$: the medium is transparent to the laser field.

pulses with spectral width $\Delta\lambda = 24$ nm. For the delay $\Delta\tau = 59.67$ fs and phase shift $\Delta\psi = 0$ [see Fig. 2(a)] between the pulses, transition $5s\,^2S_{1/2}\rightarrow 5p\,^2P_{1/2}$ falls on the zero of the spectral intensity, and level $5p\,^2P_{1/2}$ remains nonexcited. As the phase shift is increased, the interference structure is shifted along the spectral and the temporal axis, resulting in a gradual change of the populations on the levels [Fig. 2(b)]. For $\Delta\psi = \pi$, another transition $5s\,^2S_{1/2}\rightarrow 5p\,^2P_{3/2}$ of the V system is brought in the zero of the interference. As can be seen, the excitation dynamics is characterized by a rapid raise of the upper-level population to the maximum value followed by fast oscillations relaxing to the asymptotic value. Although some modulation of the oscillation amplitude due to the interaction with the nonresonant part of the

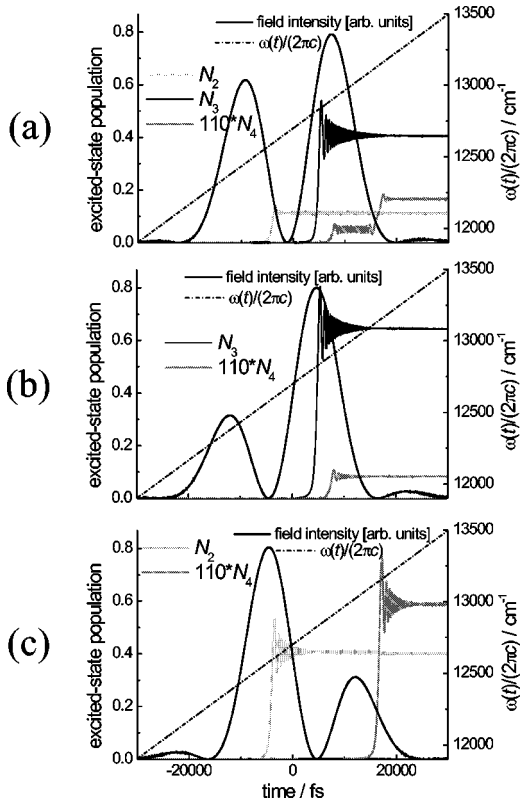

FIG. 4. Control of the quantum paths in Rb [see Fig. 1(c)] ($\Delta\lambda = 35$ nm, $\Delta\tau = 59.67$ fs). (a) $\Delta\psi = \pi/2$: path I ($|1\rangle\rightarrow|3\rangle\rightarrow|4\rangle$) and path II ($|1\rangle\rightarrow|2\rangle\rightarrow|4\rangle$) are opened. (b) $\Delta\psi = \pi$: path I is opened, path II is suppressed. (c) $\Delta\psi = 0$: path II is opened, path I is suppressed.

field is observed, the asymptotic populations are seen to be independent of the modulation and not affected by crossing with another transition. In Fig. 3, the temporal delay between the pulses was chosen such that the spacing between the zeros in the spectrum of the combined field be equal to the energy difference between the upper $5p\,^2P_{1/2}$ and $5p\,^2P_{3/2}$ levels of the system. By adjusting the value of the phase shift, the both transitions can be placed in the holes of the interference spectrum. This shuts down interaction with the atoms and thereby makes the medium transparent to the laser field. When the laser spectrum is sufficiently broad ($\Delta\lambda > 30$ nm) the approach can be applied to select the optimum pathway in Rb [Fig. 1(c)]. Figure 4(a) shows the population dynamics when the two channels are open and thus both contribute to the population of the target state $5d\ ^2D_{3/2}$ as a result of the first ($t_1 = 8$ ps) and second ($t_2 = 17$ ps) level crossing. Due to the small value of dipole moment on the transition $5p\,^2P_{3/2} \rightarrow 5d\,^2D_{3/2}$, its contribution to the total population is correspondingly smaller. Selection of a quantum pathway is achieved through adjusting the phase shift [Fig. 4(b,c)] or by varying the delay, so that one or another upper level of the V system is shut down by the spectral modulation. As can be seen, the target state is now populated only once (at $t = t_1$ or $t = t_2$), suggesting that only one (selected) channel contributes to the excitation. We also note that the modeling revealed that the simple analytical expression like Eq. (5) based on the LZ formula gives very good (within a few percent accuracy) estimation for the level populations.

The results presented above suggest that the interferometric mechanism can be used to control multilevel quantum systems with broadband-chirped radiation. The method is very general and, in principle, applicable to quantum systems of different nature and level configuration. In contrast to the traditional ARP technique, neither increase of the driving field intensity nor change of the chirp direction is required to achieve the selectivity. The important feature of the method is that the modulation law (1) and the position of zeros in the spectrum are independent of the spectral characteristics of the two optical fields used, and depend only on the relative delay and phase shift. Therefore, this dispersionless selectivity mechanism can as appropriately be used with the low-fluence attosecond XUV radiation produced by HHG. Since harmonics of moderately high orders are phase locked [19], by splitting the pump beam into two beams and generating two properly time-delayed and phase-shifted beams at the harmonic frequency, a wide range of optical processes in the XUV can be selectively controlled. Note that a time-resolved spectroscopy of inner-shell excitation and relaxation using attosecond XUV pulses has been recently demonstrated [20].

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