Observation of Selectivity of Coherent Population Transfer Induced by Optical Interference

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A fs time-resolved selective control of multilevel systems using superposition of two identical, frequency-chirped fields is proposed and demonstrated. By adjusting the delay between the pulses, a selected transition of the Rb doublet was brought into the "holes" of the interference pattern and remained nonexcited, thus allowing to manipulate another transition by the laser field as if it were an isolated two-level system. Based on light interference, this technique needs neither strong driving field intensities nor controlling the chirp direction to achieve the selectivity.

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Preparation of an ensemble of atoms or molecules in a specific state using laser light is of fundamental importance not only for spectroscopy, but also for the control of chemical and biological processes [1,2]. In recent years it has become apparent that the optical field shaping is an effective solution to this problem. By tailoring the time behavior of the optical phase and amplitude, a great variety of systems can be manipulated [3]. Coherent control of spin evolution in atomic and molecular systems [4], ionization and dissociation [5], atomic and molecular fluorescence [6,7], and ultrafast dynamics in semiconductors [8] have been demonstrated. Furthermore, it has been found that the pulse-shaping technique allows not only to enhance the excitation selectivity, but also maximize the excitation cross section [7].

The idea of using frequency-chirped optical pulses, e.g., pulses whose instantaneous frequency drifts in time, has proven to be of particular value for coherent control [9–12]. By stretching a fs pulse in a dispersive medium one produces a frequency-swept field which is used to excite a quantum system in the regime of adiabatic rapid passage (ARP) [9–11]. Since the required preparation time may be sufficiently short (in the fs range), a central problem arising here is how to achieve selectivity of target state population when multiple transitions are coupled by the broadband laser field. Taking the D lines of sodium as a model V-type system, it has been shown [11] that frequency-chirped pulses provide much more selectivity of population transfer than transform-limited pulses with the same bandwidth. The mechanism of the observed selectivity was found to be essentially nonlinear, originating basically from the specific redistribution of the adiabatic quantum pathways in the strong field. By applying even higher laser fields [10], several quantum systems were selectively controlled even though the frequency sweep did not exactly follow the frequencies of successive transitions. The results suggest that to achieve selectivity in multilevel systems with essentially stronger anharmonicity (for instance, an atom in the field of an attosecond pulse [13]), one needs, correspondingly, much stronger driving fields. This could bring into existence additional quantum paths, cause self-focusing and medium ionization [10], eventually leading to a lost of selectivity. On the other hand, the possibility to operate in the regime of moderate and low intensities would be of considerable importance for controlling transitions in the spectral regions (such as extreme ultraviolet and x ray) where intense coherent sources are presently not available.

In this Letter we demonstrate experimentally an alternative approach to the problem of selective excitation. Based on interference of light, this approach needs neither increasing the driving field intensity nor controlling direction of the frequency chirp to achieve enhanced excitation on a selected transition. Using a superposition of two identical, time-delayed and frequency-chirped optical pulses we could selectively control, on the fs-time scale, the doublet lines in rubidium. By monitoring the delay between the two interfering pulses, either of the two upper states of the Rb doublet could be brought in the holes of the laser spectrum, thus allowing to independently manipulate another one by the laser field. Unlike the common ARP technique, the approach enables one to completely decouple a selected transition from the neighboring one, and, as a result, achieve any prescribed absolute population on the level.

To give an idea of the proposed approach, consider a superposition of two identical time-delayed and phase-shifted pulses: $E_\Sigma(t) = E_1(t) + E_2(t) = \frac{1}{2}A_0(t)\exp[i\omega_0 t] + \frac{1}{2}A_0(t-\Delta\tau)\exp[i\omega_0(t-\Delta\tau) + i\Delta\psi] + \text{c.c.}$ where $A_0(t)$ is the complex amplitude of each pulse. As a result of optical interference, the spectrum of the combined field becomes sinusoidally modulated: $|F_\Sigma(\omega)|^2 = 4|F_0(\omega)|^2 \times \cos^2[(\omega\Delta\tau + \Delta\psi)/2]$, where $F_0(\omega)$ is the Fourier spectrum of $A_0(t)$. When the phase of the modulation is such that $(\omega\Delta\tau + \Delta\psi) = \pi(2k+1)$, $k = 0, \pm 1, \ldots$, 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 $\Delta\psi$ translates the whole interference structure along the

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frequency axis. The technique of modulation of optical spectrum, first proposed by Ramsey [14], was later applied to study ultranarrow resonances in atoms [15], 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 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 demonstrated in the present paper is that this can be done by using a superposition of temporally overlapped frequency-chirped pulses, $A_0(t) \propto \exp[-(t/\tau_S)^2 + i\alpha t^2]$ (Gaussian temporal shape is assumed for simplicity). If the pulses were phase modulated to second order $[\varphi''(\omega_0)]$ in a dispersive element, the chirping parameter α and the pulse duration τ_S are given by $\alpha = 2\varphi''/(\tau_0\tau_S)^2$ and $\tau_S = \tau_0\sqrt{(1 + 4\varphi''^2/\tau_0^4)}$ where τ_0 is the width of the transform-limited input pulse. Assuming that the dispersive element provides a sufficient stretching, $\tau_S \gg \tau_0$, so that the relative contribution of the phase modulation in the pulse spectrum is much greater than that of the amplitude one, and considering small temporal delays, $\Delta\tau \ll \tau_S$, the total field can be approximated by

$$E_\Sigma(t) \approx 2|A_0(t)|\cos\left[(\omega_0 + 2\alpha t)\frac{\Delta\tau}{2} + \frac{\Delta\psi}{2}\right] \times \cos[\omega_0 t + \alpha t^2 + \psi_\Sigma], \quad (1)$$

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 $\Omega_{\mathrm{mod}} = \alpha\Delta\tau$. 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 modulation of the field amplitude has the same form $(\cos[(\omega(t)\Delta\tau + \Delta\psi)/2])$ as the modulation of the field spectrum. This reflects the fact that the chirping projects the sinusoidally modulated spectrum onto the time domain, and therefore allows to control the evolution of the system in time.

Below we focus on selective excitation in terms of the V-type system shown in Fig. 1. It is assumed that the pulse spectrum is sufficiently broad to cover both the upper levels. As was shown previously [11], the selectivity in such a system is achieved by operating in the ARP regime. The level excited first by the frequency-chirped field becomes selectively populated, and thus the selectivity is controlled by changing the direction of chirp. At lower intensities, however, no selectivity is achieved and the levels are populated in proportion to the oscillator strengths of the transitions. In the proposed scheme (Fig. 1), by adjusting the parameters of the modulation [Eq. (1)] the selected transition is placed at the zero of the interference and thus remains nonexcited during all the interaction time. This offers the way to manipulate another transition by the field as if it is an isolated two-level system. As a result, any predetermined population on the selected level can be achieved, without operating in the strong field regime and changing the direction of the chirp.

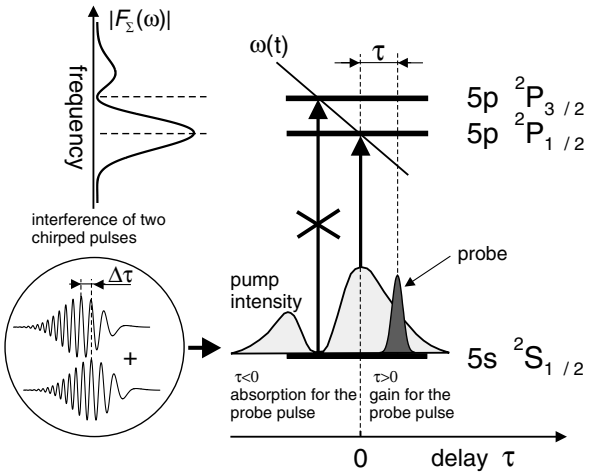

FIG. 1. Interference-induced selectivity: Superposition of two identical, time-delayed and chirped pulses (circular inset) results in a sinusoidal modulation of the spectrum $[|F_\Sigma(\omega)|]$ which is projected onto the time domain $[|E_\Sigma(t)|]$. When a selected transition is placed at the "holes" of the spectrum, the upper state remains nonexcited during all the interaction time.

To demonstrate experimentally the proposed technique, we performed pump-probe experiments in atomic rubidium vapor (Fig. 1). Both the $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ and $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transitions of the Rb doublet were controlled by a superposition of two identical, time-delayed and frequency-chirped laser pulses. The excited-state dynamics was traced by measuring the stimulated emission on the transitions with a weak replica of the reference fs-laser pulse. The reference laser pulses were delivered by a conventional chirped pulse amplification Ti:sapphire laser system. The oscillator pulses were stretched, amplified, and finally recompressed to generate 60 fs pulses with 0.85 mJ energy and a spectral bandwidth of 24 nm at a center wavelength of 790 nm and a repetition rate of 1 kHz. The main part of the laser output was phase modulated to second order, $\varphi''(\omega_0) \approx -2.1 \times 10^5$ fs^2, using a grating compressor, while a small part of the output was directed into the probe channel. The desired temporal and spectral modulation was generated in a Michelson interferometer equipped with a piezo-driven delay stage (pump-pump delay). Because of the relative simplicity of the system under study, the tunable phase shift was not implemented [16]. One more delay stage was used to take the probe pulse spectra as a function of the delay τ between the pump and probe (Fig. 1). With the same delay stage, the frequency-resolved cross correlation of the pump and probe pulse could be simultaneously measured allowing to relate the dynamics of the probe pulse spectra to the time behavior of the pump pulse. The intensity range relevant to our experiments can be characterized by the parameter

of adiabacity of the interaction [18] $\gamma = [\Omega_{\mathrm{Rabi}}^2/|d\omega(t)/dt|] \equiv [|\mu_{\mathrm{mn}}E_\Sigma|^2/(2|\alpha|\hbar^2)]$, where μ_{mn} is the dipole moment of the transition and Ω_{Rabi} is the corresponding Rabi frequency in the field $E_\Sigma(t)$. To observe the interference-induced selectivity, e.g., the feasibility to activate, say, the $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ transition and suppress the $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transition, it is necessary that the ground state population is not completely depleted by the first level crossing before the second level crossing occurs [by level crossing is implied the moment where the instantaneous frequency $\omega(t)$ matches the frequency of the transition]. Therefore the working laser intensities were chosen in the region $\gamma \leq 1$ where the excited-state population is still sensitive to the driving field amplitude. The temperature of the Rb cell (100 °C) and the corresponding particle density (about 6×10^{12} cm^{-3}) was set so that the propagation effects had only insignificant effect on the measurements. The polarization dephasing time T_2 of the transitions was measured to be approximately 50 ps. The use of the noncollinear propagation scheme for the pump-probe measurements enabled to avoid phase locking of the pump and probe fields [19]. Thus, by tracing the temporal behavior of the probe pulse gain we could get information about the dynamics of the population transfer to the upper levels. By varying the temporal delay between the long pump and short probe pulse (60 fs), positive or negative gain for the probe field could be observed depending on the current value of population inversion on the levels (Fig. 1). Figures 2(a) and 2(c) present the results of the pump-probe measurements at the resonant wavelengths (780.03 and 794.8 nm) of the Rb doublet lines as a function of the delay τ. Figures 2(b) and 2(d) show the corresponding cross correlations of the pump and probe pulses. In the measurements shown in Fig. 2(a), the delay between the two channels (54.9 fs) of the interferometer was chosen so that the driving field intensity had a "hole" near $\tau = 0$ fs, where the instantaneous frequency $\omega(t)$ is resonant to the frequency of the $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transition [Fig. 2(b)]. As a result, the population transfer on this transition was completely suppressed, while another transition ($5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$) could be manipulated by the laser field. The suppression effect can be clearly seen in Fig. 2(a) where no gain is observed until the moment $\tau \approx$ 9000 fs at which resonant crossing with the level $5p\,^2P_{1/2}$ occurs. Since the excitation of the state $5p\,^2P_{1/2}$ in Fig. 2(a) [or state $5p\,^2P_{3/2}$ in Fig. 2(c)] results in the depopulation of the common ground state $5s\,^2S_{1/2}$, the gain appears simultaneously at both the resonance frequencies. As can be seen, it exhibits a similar temporal behavior for both the (excited and nonexcited) transitions, which is a strong evidence that the observed behavior is governed by the population dynamics. By setting the delay between the two pulses in the interferometer equal to 53.7 fs, the $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ transition could be placed at the zero of the driving field intensity near

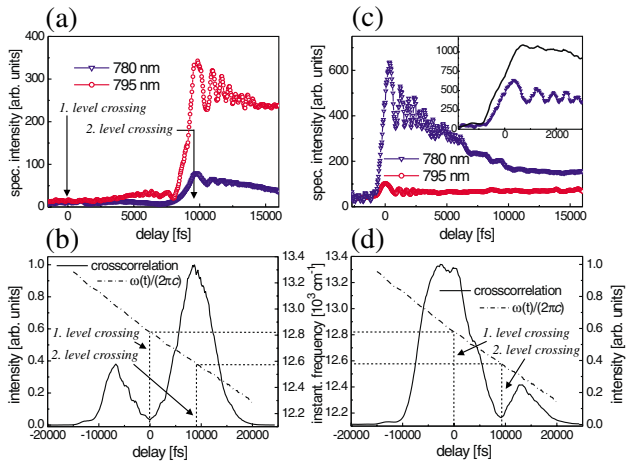

FIG. 2 (color online). (a),(c) The probe pulse gain at the resonance wavelengths 780.03 and 794.8 nm as a function of the pump-probe delay τ. (b),(d) The corresponding cross correlation and instantaneous frequency of the combined driving field. (a) Pump-pump delay $\Delta\tau = 54.9$ fs: $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ is suppressed, $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ is switched on. (c) $\Delta\tau =$ 53.7 fs: $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ is switched on, $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ is suppressed. The solid line in the inset to (c) shows dynamics of the $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transition for laser intensity close to the ARP regime.

$\tau \approx 9000$ fs [Figs. 2(c) and 2(d)]. Now interaction with the first transition is "switched on" and results in the gain appearing at both the resonant frequencies at the moment of level crossing ($\tau = 0$ fs), while the second transition is completely suppressed (no change in the probe pulse spectrum after the level crossing at $\tau \approx 9000$ fs is observed).

As can be seen, the excitation dynamics is characterized by a steep growth of the population to the maximum value at the moment of level crossing followed by oscillations relaxing to the mean value which, in turn, slowly decreases. Specifically, the frequency of the damped oscillations gets higher with increasing time after the level crossing. Such dynamics of population transfer is typical for the regime of intermediate laser intensities, where $\gamma < 1$ (which is the case in our experiments) and arises from the interference of resonant and nonresonant contributions of the frequency-chirped field to the level excitation [6,20].

The results clearly demonstrate the main features of the proposed method. Unlike the traditional approach employing single frequency-chirped pulse, the interference of two pulses provides a complete decoupling of a selected transition from another one within the laser spectrum. As a result, the multilevel system is controlled by the frequency-chirped field as if it is an isolated two-level system. The inset in Fig. 2(c) shows the dynamics of selective excitation of the $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transition for laser intensity close to the ARP regime (solid line). One can clearly see the smooth (without oscillations)

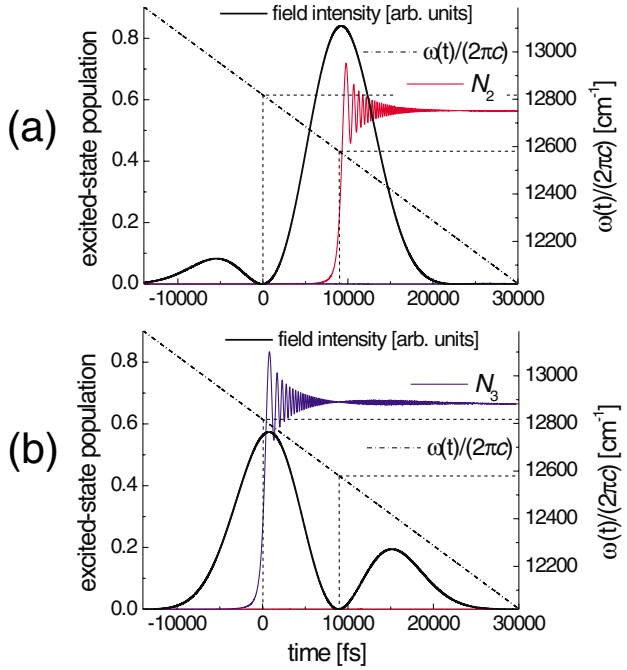

FIG. 3 (color online). Calculated dynamics of the upper level populations (N_2, N_3) of the Rb doublet for the pump amplitude $A_0 = 6.5 \times 10^6$ V/m (fluence 100 μJ/cm^2), $\Delta\lambda = 24$ nm, $\varphi'' = -2.1 \times 10^5$ fs^2. (a) Pump-pump delay $\Delta\tau = 54.9$ fs: only the $5p\,^2P_{1/2} \leftarrow 5s\,^2S_{1/2}$ transition (N_2) is on. (b) $\Delta\tau = 53.7$ fs: only the $5p\,^2P_{3/2} \leftarrow 5s\,^2S_{1/2}$ transition (N_3) is on.

temporal dynamics of population transfer which is typical for the ARP excitation of a two-level system [20].

To analyze the observed behavior quantitatively, we performed numerical modeling of the coherent control experiments in Rb. The standard approach based on the solution of the density matrix equations for the three-level system [18] in a laser field was used. Figure 3 presents the dynamics of population on the upper levels $5p\,^2P_{1/2}$ (N_2) and $5p\,^2P_{3/2}$ (N_3) in the regime where either of the transitions is suppressed [Figs. 3(a) and 3(b)]. Comparison of the results with the data in Fig. 2 reveals that the main features of the interaction with the temporally modulated frequency-chirped laser field are clearly observable in the measurements. The analysis showed that there exists a minimum width of the spectral hole (or a maximum modulation frequency) which still provides a "switch off" of the transition. Far from saturation ($\gamma < 1$), the frequency resolution $\Delta\omega_{\rm res}$ of the switch off is inversely proportional to the time $\tau_{\rm exc} = (2\pi/\alpha)^{1/2}$ it takes for the upper level to be populated to the asymptotic value [20] and is given by $\Delta\omega_{\rm res} \approx 2(2\pi\alpha)^{1/2}$. In the measurements in Fig. 2, the width of the minima is about 200 cm^{-1}, which is much broader than $\Delta\omega_{\rm res} \approx 30$ cm^{-1}. We also note that the measured excitation dynamics is influenced by the propagation effects leading to the relaxation of the populations. We attribute the observed dynamics to the development of a wave of superradiant decay of the excited transitions, which moves with a temporal delay with respect to the moment of level excitation.

In conclusion, we have proposed and demonstrated, here for the Rb doublet, that optical interference of identical frequency-chirped optical fields provides a mechanism of selectivity which allows coherent quantum control at an intensity level much lower than that used in the traditional (ARP) technique. The proposed method is very general and therefore applicable to a wide variety of quantum systems of different nature and level configuration. Finally, we point out that the modulation law ($\cos[(\omega\Delta\tau + \Delta\psi)/2]$) 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. This suggests that this technique can be extended to few cycle frequency-swept laser fields with extremely broad spectrum, thus opening the way to the coherent quantum control in the attosecond regime.

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