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Induction of the Maximum Raman Coherence in an Extended Medium through Fractional Adiabatic Passage

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Abstract—The effect of fractional stimulated Raman adiabatic passage in an optically dense medium of three-level Λ atoms is shown to result in the maximum coherence on the Raman transition on a length of the medium that considerably exceeds the length of linear resonant absorption. The general case of unequal oscillator strengths of the adjacent transitions is analyzed. The results are of interest for coherent anti-Stokes Raman spectroscopy with delayed pulses.

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1. The preparation of atoms in the form of a coherent superposition of quantum states is of considerable interest since atoms in such a state have unusual properties that open new possibilities in quantum and nonlinear optics [1–5]. Atoms in a coherent state are called coherently prepared atoms, or phaseonium [3]. In the case of equal (in magnitude) probability amplitudes of the states forming the superposition, the coherence induced on this transition reaches its maximum value; i.e., the off-diagonal element of the density matrix is equal to 1/2 (in absolute value). Media consisting of such inphase atoms are used, for example, for efficient conversion of laser radiation by way of nonlinear mixing [5–9], for generation of subfemtosecond pulses [10, 11], and in quantum and atomic optics [5]. Several methods for achieving the maximum coherence are known [2, 3, 10–15].

In [15], a method of induction of the maximum coherence on the Raman transition of a three-level Λ atom called the method of fractional stimulated Raman adiabatic passage (FStiRAP) was proposed and its specific features were studied for the case of a thin optical medium. This phenomenon is a modification of stimulated Raman adiabatic passage (StiRAP) [5] or adiabatic population transfer [16]. As in StiRAP, a control pulse couples the initially unpopulated levels $|2\rangle$ and $|3\rangle$ (Fig. 1a) and enters the medium earlier than a probe pulse (Fig. 1b). However, as distinct from StiRAP, where the control pulse is switched off somewhat earlier than the probe pulse, here, both pulses are switched off so that the ratio of their amplitudes is maintained constant. When this ratio is close to unity, only half the population is transferred from the lower state $|1\rangle$ to the excited state $|2\rangle$ and the atom remains in a coherent superposition of these states (a coherent state) after the coupling is switched off. This corresponds to the maximum coherence on the Raman transition. Recently [17], the above effect was used in a nonlinear optical experiment to increase the efficiency of anti-Stokes coherent Raman scattering.

In this paper, the effect of FStiRAP in an optically dense medium is considered. It is shown that, under certain conditions, this effect can lead to the maximum coherence in an extended medium whose length exceeds the length of linear absorption of the probe field by several orders of magnitude. On this length, the value of coherence remains constant (equal to 1/2 in absolute value) and then vanishes. The process is stable not only against a change in the time delay between the pulses but also against variations in the shape of the pulses inside the medium.

2. Let two laser pulses with envelopes E_p (probe) and E_c (control) propagate collinearly along the z axis in a medium consisting of three-level atoms (Fig. 1).

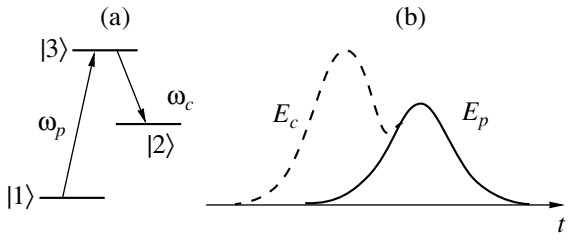

Fig. 1. (a) Diagram of atomic levels: $|1\rangle$ is the ground state, $|2\rangle$ is the metastable state, and $\omega_{p,c}$ are the carrier frequencies of the resonant pulses; (b) the shape and the sequence of switching on of the control (E_c) and probe (E_p) pulses at the entrance of the medium for observing FStiRAP.

The propagation of the pulses is described by the Schrödinger equation and the reduced wave equation for the Rabi frequencies (the Maxwell–Schrödinger equations), which should be solved self-consistently,

$$\frac{\partial}{\partial\tau}\begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} = \frac{i}{2}\begin{pmatrix} i\gamma_1 + \delta_1 & 0 & G_p^* \\ 0 & i\gamma_2 + \delta_2 & G_c^* \\ G_p & G_c & i\gamma_3 \end{pmatrix}\begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix}, \quad (1)$$

$$\frac{\partial}{\partial\zeta}\begin{pmatrix} G_p \\ G_c \end{pmatrix} = 2i\begin{pmatrix} K_p a_1^* a_3 \\ K_c a_2^* a_3 \end{pmatrix}.$$

Here, $\tau = t - z/c$ and $\zeta = z$ are, respectively, the time and the depth of the medium in the coordinate system moving with the velocity of light in vacuum; $a_{1,2,3}$ are the probability amplitudes of the atomic states; $\gamma_{1,2,3}$ are the relaxation constants of the atomic states; $\delta_{1,2}$ are the one-photon detunings of the fields from the frequencies of the resonant transitions; $2G_{p,c} = E_{p,c}d_{p,c}/\hbar$ are the Rabi frequencies of the fields; $E_{p,c}$ are the strengths of the probe and control fields (envelopes); $d_{p,c}$ are the electric dipole moments of the corresponding transitions in the atom; $\hbar$ is Planck's constant; $K_{p,c} = 2\pi N\omega_{p,c}|d_{p,c}|^2/\hbar c$ are the propagation coefficients; and N is the atomic concentration.

Initially, all the atoms are in the state $|1\rangle$: $a_{1,2,3}(\tau = -\infty, \varsigma) = 1; 0; 0$. At the entrance of the medium, the control field E_c is switched on earlier than the probe field E_p (Fig. 1b) and the pulse durations satisfy the relation $T_c > T_p$. The pulses are assumed to be short enough that we can further neglect the relaxation ($\gamma_{1,2,3}$) of the atomic subsystem. We also assume that $\delta_{1,2} = 0$, i.e., the fields are in one- and two-photon resonances.

In the adiabatic approximation [5], $G_p/G_c = -a_2/a_1$ and $|a_3| \ll 1$. Taking into account the above relations, it follows from (1) that the number of photons of the interacting pulses is conserved, $G_c^2/K_c + G_p^2/K_p = \text{const}$, and the equation for the Rabi frequencies [18] takes the form

$$\frac{\partial\vec{G}}{\partial\zeta} = -\hat{K}\frac{1}{\vec{G}^2}\frac{\partial\vec{G}}{\partial\tau}, \quad (2)$$

where $\vec{G} = (G_c, G_p)$.

It is convenient to change to a new variable called the mixing angle, which is defined as $\tan\theta = G_p/G_c$,

$$\frac{\partial\theta}{\partial\zeta} + \frac{K^2(\theta)}{K_pG_c^2 + K_cG_p^2}\frac{\partial\theta}{\partial\tau} = 0, \quad (3)$$

where $K(\theta) = (K_pG_c^2 + K_cG_p^2)/\vec{G}^2 = K_p\cos^2\theta + K_c\sin^2\theta$. The mixing angle is constant along the characteristic curves

$$\zeta(\tau, \tau_0) = K^{-2}(\theta(\tau_0, \zeta = 0))\int_{\tau_0}^{\tau}(K_pG_c^2 + K_cG_p^2)d\tau', \quad (4)$$

where τ_0 is the instant of entrance of the field corresponding to this characteristic into the medium. The physical quantities can be expressed in terms of the mixing angle $\theta(\tau, \zeta)$ as

$$\begin{pmatrix} G_p \\ G_c \end{pmatrix} = 2\sqrt{\frac{(K_pG_c^2 + K_cG_p^2)\big|_{\zeta=0}}{K(\theta)}}\begin{pmatrix} \sin\theta \\ \cos\theta \end{pmatrix},$$

$$\begin{pmatrix} a_1 \\ a_2 \\ a_3 \end{pmatrix} = \begin{pmatrix} \cos\theta \\ \sin\theta \\ -\left|\partial\vec{G}/\vec{G}^2\partial\tau\right| \end{pmatrix}. \quad (5)$$

The numerical analysis of (5) shows that FStiRAP is realized when the trailing edge of the control pulse duplicates the shape of the probe pulse, as is shown in Fig. 1b. As in the case of StiRAP, the control pulse is switched on earlier than the probe pulse but both pulses are switched off simultaneously. Therefore, after the passage of the pulses, the system remains in a coherent superposition of the states $|1\rangle$ and $|2\rangle$ with the maximum coherence (Fig. 2). We emphasize that FStiRAP requires the exact matching of the trailing edges of the control and probe pulses. Such matched switching off can be achieved, for example, if, simultaneously with the probe pulse, a pulse of the same shape but with the carrier frequency of the control pulse is fed to the medium. Methods for producing identical-in-shape pulses at different frequencies were proposed in [19–21]. In the case of degenerate transitions, when the lower levels $|1\rangle$ and $|2\rangle$ differ only in spin, it is possible to represent a linearly polarized pulse as a superposition of two circularly polarized pulses. Then the control of the pulse polarization ensures the boundary conditions (Fig. 1b) [15].

Figure 2 demonstrates the dependences of the normalized Rabi frequencies and the off-diagonal element of the density matrix $|\rho_{21}|$ on the time and the length of the medium in the case of two equal propagation coefficients $K(\theta) \equiv K = K_c = K_p$ (equal oscillator strengths of the adjacent transitions). One can see that, under the FStiRAP conditions, pulses can propagate in a resonant medium over a distance considerably exceeding the linear length of resonance absorption (as in the case of StiRAP [22, 23]). But, eventually, the probe pulse is completely transferred into the control pulse. The maximum coherence on the Raman transition is induced in the form of a "step." The higher the pulse intensities, the larger the length on which the maximum coherence

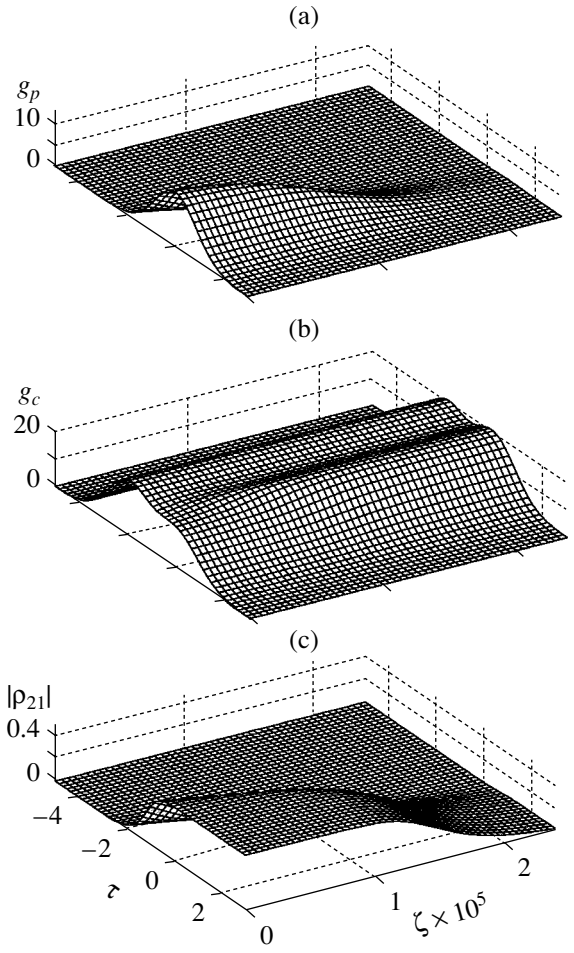

Fig. 2. Spatiotemporal evolution of the Rabi frequencies of the (a) probe $g_p = G_1 T_p$ ($g_{p\max} = 15$) and (b) control $g_c = G_c T_p$ ($g_{c\max} = 15\sqrt{2}$) pulses and (c) Raman coherence (the off-diagonal element of the density matrix on the Raman transition $|1\rangle$–$|2\rangle$). The quantity ζ is measured in units of the linear absorption length of the probe field, and the time τ is measured in units of the pulse duration T.

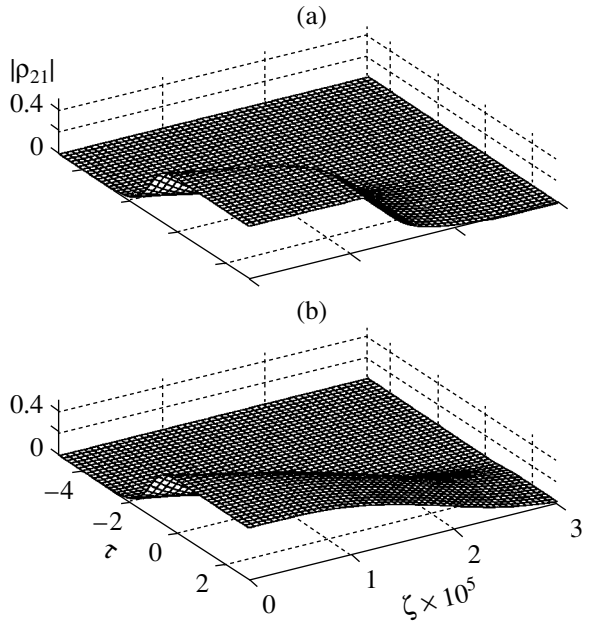

Fig. 3. Spatiotemporal evolution of the off-diagonal element of the density matrix in the case of unequal oscillator strengths of the adjacent transitions. (a) $K_p = 2K_c$; (b) $K_p = K_c/2$. The values of the other parameters are the same as in Fig. 2.

is induced. With the probe pulse delay $\Delta T = 2T_p$, where T_p is the half-width of the probe pulse at half maximum, the numerical solution of Eqs. (1) agrees well with the analytical solution (5). The length of the medium on which the maximum coherence takes place is of the order of $T_p G^2/K$. The coherence lifetime is governed by the time of its relaxation. At a delay between the pulses $\Delta T = 3T_p$ or $\Delta T = T_p$, a distortion in the form of small nonadiabatic oscillations appears in the coherence profile. For the mentioned sequence and configuration of the pulses, the effect is stable with respect to variations in the time delay and the pulse intensities.

Figure 3 demonstrates the spatiotemporal dependence of the off-diagonal element of the density matrix $|\rho_{21}|$ for $K_p > K_c$ and $K_p < K_c$. In the first case, the trailing spatial edge of the coherence is more pronounced than in the second case.

3. It was shown in this paper that, in an optically dense medium, in the process of FStiRAP, the maximum Raman coherence is induced in the form of a step whose length can considerably exceed the length of linear absorption of the probe field. In the region of adiabaticity, the results of the analytical calculations agree well with the direct numerical calculation. The induced profile of the coherence is stable against variations in the field intensities and a delay between the pulses within rather wide limits. The results obtained may be of interest for coherent anti-Stokes Raman spectroscopy with delayed pulses [24].

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