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# Stark-Chirped Rapid Adiabatic Passage: Propagation of Laser Pulses and Spacetime Evolution of Populations and of Two-Photon Coherence

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**Abstract**—On the basis of a vector model, the propagation of laser pulses under the conditions of a two-photon quasiresonance in the case of Stark-chirped rapid adiabatic passage through the resonance is studied with allowance for a diabatic character of the interaction. It is shown that the shape of a pulse propagating in a medium changes, the sweeping of its carrier frequency occurring concurrently. Special features of the spacetime evolution of the population difference in a two-photon transition and of the two-photon coherence during pulse propagation are analyzed. It is established that a complete population inversion and a maximum coherence may exist over a long length of the medium if the corresponding conditions are satisfied at the boundary. A new possibility for achieving a high coherence (close to a maximum value) is proposed. *© 2003 MAIK "Nauka/Interperiodica".*

## 1. INTRODUCTION

In recent years, new concepts based on atomic coherence and effects of quantum interference have given a strong impetus to the development of the resonance nonlinear optics of gaseous media. Various effects in these realms, such as induced transparency [1], the coherent trapping of populations [2, 3], and the adiabatic transfer of populations [4] (or stimulated Raman adiabatic passage [5]), have found widespread use. The above phenomena make it possible to control linear and nonlinear optical properties of matter. Concurrently, laws that govern the propagation of laser pulses under resonance conditions change significantly (there arise matched pulses [1], adiabatons [6], a decrease in the group velocity of a pulse by a factor of $10^7$ to $10^8$ [7], and some other effects [8–10]). New possibilities open up here, including that for a high-efficiency implementation of nonlinear-optics interaction even for very weak light fields [11, 12] and for the generation of subfemtosecond light pulses [13].

The new effect of Stark-chirped rapid adiabatic passage (referred to in the following as SCRAP) was recently demonstrated in [14]. It is closely related to a rapid adiabatic passage due to sweeping the pumping-laser frequency (see, for example, [15]) or due to the intrinsic Stark shift of levels in two-photon interaction [16]. In contrast to what occurs in the phenomena described in those studies, the passage through a resonance in the case of SCRAP is accomplished owing to the dynamic Stark shift caused by an additional strong laser pulse that is delayed in time with respect to the pumping-laser pulse. Specific conditions must be satisfied for the observation of SCRAP to be possible. The SCRAP phenomenon also differs from the coherent two-quantum interaction of ultrashort pulses of light under the conditions of self-induced transparency [17].

SCRAP makes it possible to coherently control both populations and coherence in atomic and molecular systems. It enables one to obtain the inversion of populations between high-lying levels and the ground-state level [14] or to prepare atoms in states that are characterized by a maximum two-photon coherence [18–20]. It was proposed that the latter be employed to ensure an efficient generation of coherently tuned radiation—in the vacuum-ultraviolet region inclusive—in the process of nonlinear-optics mixing of short pulses [19, 20]. In all probability, this effect may be used to control chemical reactions and to treat quantum information; also, it may find applications in atomic optics.

Only the time dynamics of population inversion between levels and of the maximum two-photon coherence in the SCRAP process was investigated in [14, 18] in terms of adiabatic states. In the present study, we examine the propagation of a pumping-laser pulse under the SCRAP conditions and analyze the spacetime dynamics of level population and of two-photon coherence with allowance for the propagation of this pulse. The analysis is performed on the basis of a vector model that is quite clear and which makes it possible to obtain analytic expressions both for populations and for the off-diagonal density-matrix element describing atomic coherence with allowance for the first nonadiabatic correction. Below, we show that this correction

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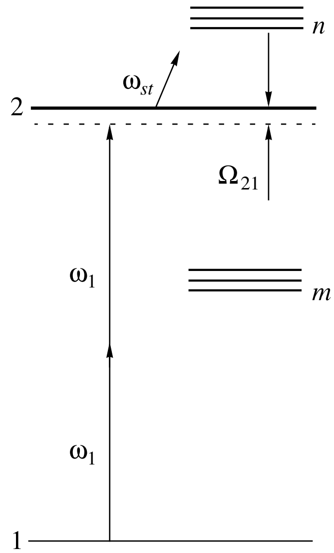


**Fig. 1.** Simplified diagram of energy levels for Stark-chirped rapid adiabatic passage.

must be taken into account in considering the propagation of a pulse in a medium. We also show that, under the SCRAP conditions, population inversion in a resonance transition and the induced maximum coherence may occur over a long distance in a medium.

This article is organized as follows. In Section 2, we discuss the SCRAP phenomenon on the basis of a vector model, disregarding pulse propagation. There, we present basic equations and their solutions, describe the pattern of the SCRAP effect in terms of the vector model, and discuss conditions necessary for its observation. In Section 3, we derive equations that describe the propagation of a pumping-laser pulse under the SCRAP conditions, give their solution, and discuss the spacetime dynamics of population inversion and atomic coherence with allowance for pulse propagation. In the Conclusions, we consider the possible applications of our results.

## 2. EFFECT OF RAPID ADIABATIC PASSAGE THROUGH A TWO-PHOTON RESONANCE DUE TO A LASER-INDUCED STARK SHIFT: VECTOR REPRESENTATION

A simplified energy-level diagram corresponding to the process under consideration is shown in Fig. 1. In order to observe the SCRAP effect, use is made of two laser pulses following each other with a specific time delay. One of them (triggering or pumping pulse) interacts with the *1–2* two-photon transition (see Fig. 1), its frequency being initially detuned from the two-photon resonance; the other, strong, off-resonance (Stark), pulse leads to a change in the two-photon frequency, inducing a time-dependent dynamic Stark shift. As the two-photon detuning changes from large negative to large positive values (or vice versa), the population of the ground-state (lower) level (*1*) can be transferred almost entirely, by the end of the triggering pulse, to the excited (upper) state (level *2*); that is, a complete population inversion may arise in the *1–2* transition, the lifetime of this inversion being determined by the relaxation time for state *2*. The pulse durations are assumed here to be shorter than all atomic relaxation times.

This process is usually explained in terms of dressed (adiabatic) states [14], but it can also be described in terms of the vector model proposed in [16]. Within this model not only can one visualize the time dynamics of the process, but also obtain analytic expressions for population inversion and for two-photon coherence.

### *2.1. Equations for Interacting Fields and for the Density Matrix in the Approximation of a Generalized Two-Level System*

We will first consider the time evolution of the population inversion $\rho_2$–$\rho_1$ in the *1–2* two-photon transition and of the off-diagonal density-matrix element $\rho_{21}$, which describes two-photon coherence under the SCRAP conditions. For $\rho_{21}$, we hereafter use the term “two-photon coherence” or merely “coherence.” The triggering pulse $\mathscr{E}_1(z, t)$ and the Stark pulse $\mathscr{E}_s(z, t)$ propagate along the $z$ axis in an isotropic medium,

$$\mathscr{E}_{1,s}(z,t) = \frac{1}{2}E_{1,s}(z,t)\exp[-i(\omega_{1,s}t - k_{1,s}z)] + \text{c.c.}, \quad (1)$$

where $k_{1,s} = \omega_{1,s}/c$ is the wave number at the frequency $\omega_{1,s}$, $c$ is the speed of light, and $E_{1,s}(z, t)$ stands for complex-valued slowly varying amplitudes (envelopes). Since the Stark pulse undergoes off-resonance interaction with the medium, we assume that it is preset in the process of propagation and that the amplitude is a real-valued quantity.

The propagation of the triggering pulse is described by the reduced wave equation

$$\frac{\partial E_1(z,t)}{\partial z} + \frac{1}{c}\frac{\partial E_1(z,t)}{\partial t} = -i2\pi k_1 P_1(z,t), \quad (2)$$

where $P_1(z, t)$ is the complex-valued amplitude that characterizes polarization induced by the triggering pulse. It can be calculated on the basis of the equation for the density matrix,

$$P_1(\omega_1) = N\sum_m (d_{1m}\rho_{m1} + d_{m2}\rho_{2m}), \quad (3)$$

where $N$ is the density of atoms, $d_{ij}$ stands for the transition electric dipole moments, and $\rho_{ij}$ are the components of the density matrix.

In the following, we assume, for the frequency of the two-photon quasiresonance, that

$$\Omega_{21} = 2\omega_1 - \omega_{21} \ll \omega_{ij},$$

we also assume that the one-photon detuning satisfies the conditions

$$\Omega_{ji} = \omega_k - \omega_{ji} \gg |G_{ji}| \gg T_k^{-1}, \qquad (4)$$

where $G_{ji} = E_k d_{ji}/2\hbar$ is the Rabi frequency and $T_k$ is the pulse duration. In the approximation specified by Eq. (4), one can pass from a multilevel system to a generalized two-level scheme (see, for example, [21]). Level *1* is the ground-state level; the group of levels labeled with *m* makes a leading contribution to the two-photon quantum transition and to the Stark shift due to the triggering field; and the group of levels labeled with *n* contributes to the Stark shift due to the Stark field.

If use is made of the rotating-wave approximation, the equations describing the density matrix for the generalized two-level system in the interaction representation have the form

$$\begin{gathered}\frac{\partial(\rho_2 - \rho_1)}{\partial t} = -2qA_1^2 \mathrm{Im}\rho_{21}, \\ \frac{\partial \rho_{21}}{\partial t} - i(\Omega'_{21} - \Omega_s)\rho_{21} = \frac{i}{2}qA_1^2(\rho_2 - \rho_1).\end{gathered} \qquad (5)$$

In Eq. (5), we introduced the following notation:

$$E_1 = A_1 \exp(-i\varphi_1),$$

where $A_1$ and $\varphi_1$ are, respectively, the real-valued amplitude and phase, which are functions of coordinates and time;

$$q = \frac{1}{2\hbar^2}\sum_m \frac{d_{2m}d_{m1}}{\Omega_{m1}}, \quad \Omega_{m1} = \omega_1 - \omega_{m1},$$

$$\Omega'_{21} = \Omega_{21} + \frac{2\partial\varphi_1}{\partial t} = 2\omega'_1 - \omega_{21}, \quad \Omega_{21} = 2\omega_1 - \omega_{21}$$

is the initial (static) detuning, with $\omega_{21}$ and $\omega'_1 = \omega_1 + \partial\varphi_1/\partial t$ being, respectively, the resonance transition frequency and the instantaneous frequency; and $\Omega_s = S_1 + S$ is the Stark shift of the two-photon resonance due to the triggering ($S_1 = a_1 A_1^2$) and the Stark ($S = a_s E_s^2$) pulse, with

$$\begin{gathered}a_1 = \frac{1}{4\hbar^2}\sum_m \left(\frac{|d_{m1}|^2}{\omega_1 - \omega_{m1}} + \frac{|d_{2m}|^2}{\omega_1 - \omega_{2m}}\right), \\ a_s = \frac{1}{4\hbar^2}\sum_n \frac{|d_{n1}|^2}{\omega_s - \omega_{n1}}\end{gathered} \qquad (6)$$

being parameters that describe the Stark shifts.

In the above approximation, the polarization given by Eq. (3) can be represented in the form

$$\begin{gathered}P_1(\omega_1) = [\chi_1(\omega_1)\rho_1 - \chi_2(\omega_1)\rho_2]NA_1\exp(i\varphi_1) \\ -2N\hbar qA_1\rho_{21}\exp(i\varphi_1),\end{gathered} \qquad (7)$$

where $\chi_{1,2}(\omega_1)$ are linear susceptibilities,

$$\chi_i(\omega_1) = \frac{1}{2\hbar}\sum_m \frac{|d_{mi}|^2}{\omega_1 - \omega_{mi}}, \quad i = 1, 2. \qquad (8)$$

### *2.2. Vector Representation*

It is convenient to rewrite Eq. (5) by using the vector model proposed in [15, 16] and by introducing the notation

$$r_1 = \rho_{21} + \rho_{12}, \quad r_2 = i(\rho_{21} - \rho_{12}), \quad r_3 = \rho_2 - \rho_1.$$

The quantities $r_j$ ($j$ = 1, 2, 3) can be treated as the components of the vector

$$\mathbf{r} = \mathbf{e}_1 r_1 + \mathbf{e}_2 r_2 + \mathbf{e}_3 r_3$$

($\mathbf{e}_{1,2,3}$ are unit vectors) in some vector space (vector $\mathbf{r}$ is sometimes referred to as a pseudospin). The pseudospin satisfies the equation of motion

$$\frac{\partial \mathbf{r}}{\partial t} = \boldsymbol{\gamma} \times \mathbf{r}, \qquad (9)$$

where $\boldsymbol{\gamma}$ is a vector whose components are given by

$$\gamma_1 = qA_1^2, \quad \gamma_2 = 0, \quad \gamma_3 = (\Omega'_{21} - \Omega_s).$$

These parameters have a clear physical meaning: $\gamma_1$ is the effective two-photon Rabi frequency and $\gamma_3$ is the instantaneous detuning away the two-photon resonance.

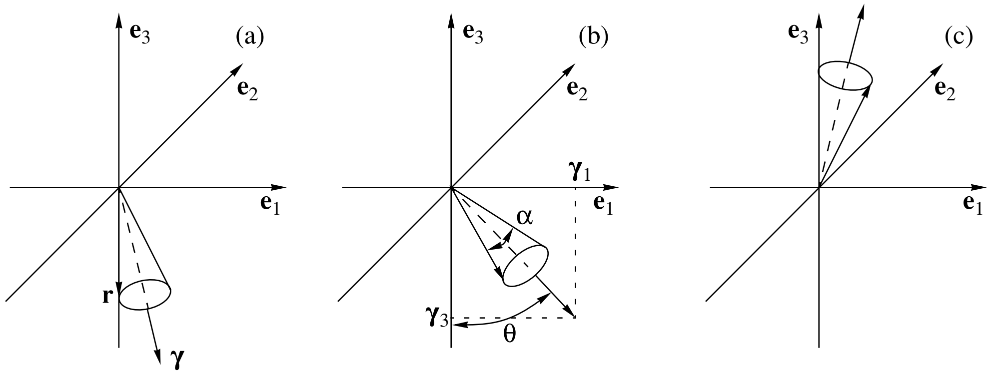


**Fig. 2.** Evolution of vector **r** during Stark-chirped rapid adiabatic passage: (a) the case where the two-photon frequency of the triggering pulse is less than the resonance frequency and where vector **r** precesses over the cone surface about vector $\boldsymbol{\gamma}$; (b) the case where, as the sweeping of the frequency of the passage through the resonance due to the Stark shift occurs, the angle θ between the vector $\boldsymbol{\gamma}$ and the negative direction of the $e_3$ axis increases, while vector **r** continues precessing about the vector $\boldsymbol{\gamma}$; and (c) the case where the doubled frequency of the triggering pulse is greater than the resonance transition frequency and where, upon the completion of motion, vector $\boldsymbol{\gamma}$ is oriented in a direction close to the positive direction of the $e_3$ axis.

The equations of motion for the components of vector **r** have the form

$$\frac{\partial r_1}{\partial t} = \gamma_3 r_2, \quad \frac{\partial r_2}{\partial t} = -\gamma_3 r_1 + \gamma_1 r_3, \quad \frac{\partial r_3}{\partial t} = -\gamma_1 r_2. \qquad (10)$$

In this approximation, the solution to Eqs. (10) with the allowance for the first nonadiabatic correction has the form (see, for example, [15])

$$r_1 = \mp\frac{\gamma_1}{\gamma}, \quad r_3 = \mp\frac{\gamma_3}{\gamma}, \quad r_2 = -\frac{1}{\gamma_3}\frac{\partial r_1}{\partial t}, \qquad (11)$$

where

$$\gamma = |\gamma| = \sqrt{\gamma_1^2 + \gamma_3^2}.$$

In expressions (11) and in those that follow, the upper (lower) signs are chosen for the case of $\gamma_3(z = 0, t \longrightarrow -\infty) < 0$ [$\gamma_3(z = 0, t \longrightarrow -\infty) > 0$].

We note that, upon the inclusion of the nonadiabatic correction, the coherence

$$\rho_{21} = \frac{r_1 - ir_2}{2}$$

becomes complex-valued. As will be shown below, this must be taken into account in considering the propagation of a pulse in a medium.

It is convenient to introduce the parameter $\theta = \arctan(\gamma_1/\gamma_3)$, which is the angle between vector $\boldsymbol{\gamma}$ and the negative direction of the $e_3$ axis aligned with unit vector $\mathbf{e}_3$ (see Fig. 2). In this notation, we have $r_1 = \pm\sin\theta$ and $r_3 = \mp\cos\theta$.

The adiabaticity condition implies that the angular velocity $\partial\theta/\partial t$ of vector $\boldsymbol{\gamma}$ is much less than the frequency $\gamma$ of precession of vector **r**; that is,

$$\left|\frac{\partial\theta}{\partial t}\right| = |\dot{\theta}| \ll \gamma = \sqrt{\gamma_1^2 + \gamma_3^2}. \qquad (12)$$

By using expression (12), it can easily be shown that $r_2 = -\dot{\theta}/\gamma$. This means that the angle between **r** and $\boldsymbol{\gamma}$ is much smaller than the angle between $\boldsymbol{\gamma}$ and $\mathbf{e}_3$ (see Fig. 2); that is, $|r_2| \ll |r_1|$.

Adiabaticity condition (12) can be represented in the form

$$\left|\frac{\dot{\gamma}_1\gamma_3 - \dot{\gamma}_3\gamma_1}{(\gamma_1^2 + \gamma_3^2)^{3/2}}\right| \ll 1. \qquad (13)$$

This condition is similar to that obtained in [5] in terms of adiabatic states.

Let us show that solution (11), together with condition (13), describes the SCRAP phenomenon well. We consider the case where the two-photon Rabi frequency $\gamma_1(t)$ and the Stark shift $S(t)$ of the frequency of the transition induced by the second pulse have Gaussian shape; that is,

$$\gamma_1(t) = \gamma_{10}\exp\left(-\frac{t^2}{T_1^2}\right), \quad S(t) = S_0\exp\left[-\frac{(t-\delta t)^2}{T_s^2}\right], \qquad (14)$$

where $T_1$ and $T_s$ are the durations of, respectively, the triggering and the Stark pulse and $\delta t$ is the time delay between them. In the ensuing analysis, we disregard, for the sake of simplicity, the shift due to the triggering pulse. In order to achieve SCRAP, it is necessary to meet certain requirements. Fulfillment of the adiabaticity condition (13) can be ensured if the initial detuning $\Omega_{21}$, the maximum Rabi frequency $\gamma_{10}$, and the delay time $\delta t$ satisfy a specific relation. For passage through a two-photon resonance to occur, it is necessary, first of all, that the maximum Stark shift $S_0$ exceed the initial detuning $\Omega_{21}$ ($|S_0| > |\Omega_{21}|$) and that the two quantities in question have the same sign. It is obvious that, under these conditions, the resonance is swept twice, at the instants of time

$$t_{1,2} = \delta t \mp T_s\sqrt{\ln\frac{S_0}{\Omega_{21}}}.$$

In this case, the triggering pulse must be rather strong in order to ensure an adiabatic transition during the first passage through the resonance, but it must be at the same time rather weak in order to prevent an adiabatic transition during the second passage. Under the assumption that the triggering pulse attains a maximum at the instant of time $t_1 = 0$, these requirements lead to the conditions [14]

$$1 \ll \frac{\gamma_{10}^2 T_s^2}{\Omega_{21}\delta t} \ll \exp\frac{8\delta t^2}{T_1^2}. \tag{15}$$

Relation (15) yields an upper and a lower limit on the detuning $\Omega_{21}$ and the maximum value $\gamma_{10}$ of the Rabi frequency. From the formula for $r_3$ in (11), it follows that, for the population transfer to be maximal at the end of the triggering pulse, fulfillment of the condition $\gamma_1 \ll |\gamma_3|$ must be ensured, which is most easily achieved at the instant $t = \delta t$, when the Stark shift $S$ is maximal. This condition can be recast in the form

$$\gamma_{10}\exp\left(-\frac{\delta t^2}{T_1^2}\right) \ll |\Omega_{21} - S_0|. \tag{16}$$

At a specific instant of time when the population difference vanishes, $r_3 = 0$, the two-photon coherence attains the maximum value of $|\rho_{21}| = 1/2$. These conditions can be satisfied in a rather wide region.

For numerical illustrations, we have chosen the diagram that represents transitions in the Kr atom and which was used in experiments aimed at generating vacuum-ultraviolet radiation under the conditions of electromagnetically induced transparency [22]: there, a triggering pulse of a wavelength of $\lambda_1 \approx 212.55$ nm undergoes two-photon interaction with the $4p^{6\,1}S$–$4p^5 5p$ transition. Since relaxation times characteristic of the transitions being considered are about a few tens of nanoseconds, the duration of the laser pulses used must not exceed a few nanoseconds.

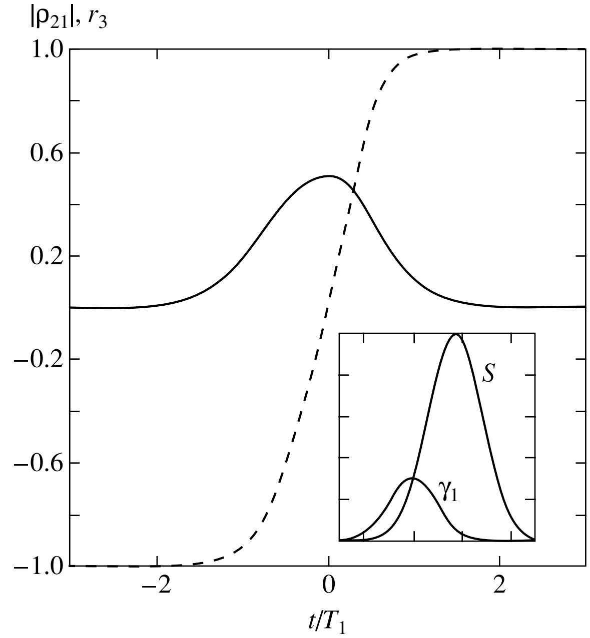


**Fig. 3.** Time dependences of (solid curve) the two-photon coherence $|\rho_{21}|$ and (dashed curve) the population difference $r_3 = \rho_2 - \rho_1$ for the pulse sequence shown in the inset (the triggering pulse precedes the Stark pulse). The following parameter values were used here: $\delta t/T_1 = 1.7$ for the delay between the pulses; $T_s/T_1 = 1.6$ for the duration of the Stark pulse; $S_0T_1 = 50$ for the maximum Stark shift; $\gamma_{10}T_1 = 15$ for the amplitude of the two-photon Rabi frequency; and $\Omega_{21}T_1 = -16$ for the initial detuning.

The population difference $r_3$ and the modulus of the atomic coherence, $|\rho_{21}| = |(r_1 - ir_2)/2|$, versus time are shown in Fig. 3, whence it can be seen that the population inversion and the maximum coherence in the two-photon transition are realized under the above conditions.

In terms of the vector model, the SCRAP process can be visualized as follows. Suppose that, at the initial instant of time ($t \longrightarrow -\infty$), all atoms are in the ground state *1* ($r_3 = -1$) and that the double frequency of the triggering pulse is tuned rather far away from the resonance and is, for definiteness, less than the resonance frequency of the *1–2* transition. This means that the vector $\boldsymbol{\gamma}$ is directed nearly along the negative direction of the $e_3$ axis and is virtually parallel to vector $\mathbf{r}$ (that is, $\theta \approx 0$). In this case, vector $\mathbf{r}$ moves over the surface of a cone (precesses about vector $\boldsymbol{\gamma}$) having a small apex angle proportional to $r_2$ (see Fig. 2). As a Stark pulse is applied, the sweeping of the frequency toward the resonance begins, which is accompanied by the growth of

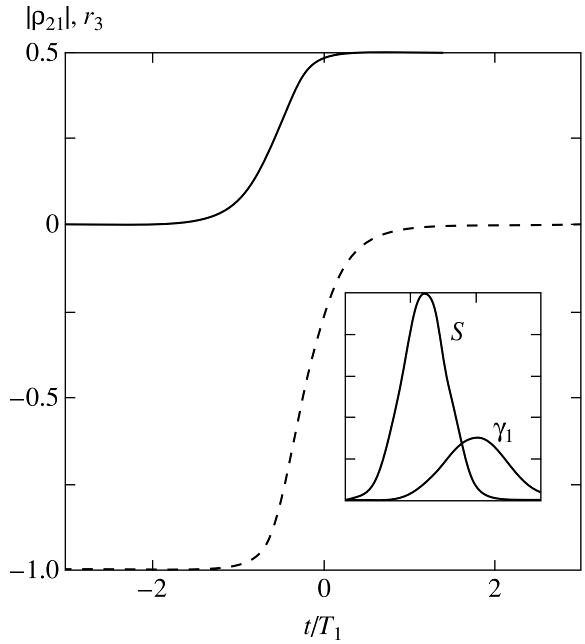


**Fig. 4.** Time dependences of (solid curve) the two-photon coherence $|\rho_{21}|$ and (dashed curve) the population difference $r_3 = \rho_2 - \rho_1$ for the case where the Stark pulse precedes the triggering pulse, as is shown in the inset. The following parameter values were used here: $\delta t/T_1 = -1.6$, $T_s/T_1 = 1$, $S_0T_1 = 50$, $\gamma_{10}T_1 = 15$, and $\Omega_{21} = 0$.

angle θ. If angle θ varies more slowly than the precession frequency γ [condition (13) is satisfied], vector **r** will follow the vector **γ** as the angle θ varies, its precession persisting. The amplitude of the Stark pulse must be such that, upon passage through the resonance, the frequency of the triggering pulse appears to be considerably higher than the resonance transition frequency (see Fig. 2). At the end of the pulse, vector **r** will form a small angle with the positive direction of the $e_3$ axis. Thus, the inversion of the level populations is realized owing to sweeping through the resonance. The inclusion of $r_2$ does not change the pattern qualitatively, but one can show that, in this case, the precession of vector **r** occurs about vector $\boldsymbol{\gamma} + \dot{\boldsymbol{\theta}}$, where $\boldsymbol{\gamma} = (0, 0, -\gamma_1)$ and $\dot{\boldsymbol{\theta}} = (0, -\dot{\theta}, 0)$ are the vectors written in the doubly rotating coordinate frame (see, for example, [15]).

By using this technique, one can prepare atoms in a coherent superposition of states *1* and *2* that is characterized by a maximum coherence [18]. This effect, known as the half-SCRAP effect [18], can also be described in terms of the vector model. In contrast to SCRAP, it arises under different conditions. From the formula for $r_1$ in (11), it follows that $r_1 \longrightarrow \mp 1$ if $\gamma_1 \gg |\Omega_{21} - S|$ (near the instant of time at which the triggering field takes a maximum value). At the initial instant of time, we have $r_3 = -1$; therefore, it is obvious that the inequality $\gamma_1 \ll |\Omega_{21} - S|$ must hold at this instant. These conditions are satisfied most readily at $\Omega_{21} = 0$. Moreover, they must be consistent with adiabaticity condition (13). Analysis shows that these conditions may be satisfied for the sequence of pulses shown in the inset to Fig. 4, in which case the Stark pulse is switched on earlier than the triggering pulse. The case of the opposite sequence of pulses—that is, the case in which the triggering pulse is switched on earlier than the Stark pulse [18]—is not considered in the present approximation.

Figure 4 shows the time dependences of $r_3$ and $|\rho_{21}| = |(r_1 - ir_2)/2|$ for the chosen sequence of pulses. It can be seen that, in contrast to what occurs in the case of SCRAP, there arises here a plateau of maximum coherence. As the Stark and the triggering field change, vector **r** initially oriented nearly along the negative direction of the $e_3$ axis then rotates through angle π/2, becoming aligned with the $e_1$ axis.

## 3. PROPAGATION OF TRIGGERING PULSE AND SPACE DYNAMICS OF THE POPULATION INVERSION AND OF THE COHERENCE

Let us now consider the propagation of a triggering pulse under the SCRAP conditions. We express the complex-valued polarization $P_1(\omega_1)$, which is defined by formula (7), in terms of quantities $r_{1,2,3}$ as

$$P_1(\omega_1) = \frac{1}{2}\{[\chi_1(1-r_3) - \chi_2(1+r_3)]NA_1\exp(i\varphi_1)\} - \hbar q(r_1 - ir_2)NA_1\exp(i\varphi_1) = (U+iV)\exp(i\varphi_1), \quad (17)$$

where $N$ is the density of atoms. It should be emphasized that the imaginary part $V$ of the polarization owes its existence to the nonadiabatic correction $r_2$ and leads, despite its smallness, to a change in the shape of a pulse as it propagates in a medium.

Substituting expression (17) into Eq. (2), we obtain equations for the real-valued amplitude and phase; that is,

$$\frac{\partial A_1}{\partial z} + \frac{1}{c}\frac{\partial A_1}{\partial t} = \pm 2\pi k_1 V, \quad (18)$$

$$\frac{\partial \varphi_1}{\partial z} + \frac{1}{c}\frac{\partial \varphi_1}{\partial t} = \mp 2\pi k_1 \frac{U}{A_1}, \quad (19)$$

where

$$U = \frac{1}{2}\left[\chi_1(\omega_1)\left(1 - \frac{\gamma_3}{\gamma}\right) - \chi_2(\omega_1)\left(1 + \frac{\gamma_3}{\gamma}\right)\right]NA_1 - \frac{\hbar q^2 N A_1^3}{\gamma}, \qquad (20)$$

$$V = -\frac{\hbar q^2 N A_1}{\gamma_3}\frac{\partial}{\partial t}\frac{A_1^2}{\gamma}.$$

Here, the upper (lower) signs correspond to a negative (positive) initial detuning away the two-photon resonance, $\Omega_{21} = 2\omega_1 - \omega_{21} < 0$).

If we discard the nonadiabatic correction ($V = 0$ corresponds to the ideal adiabatic limit), the right-hand side of Eq. (18) vanishes, in which case one can see that the pulse propagates in a medium without any distortion of its shape. As to the phase of the pulse, it will change anyway according to Eq. (19)—that is, sweeping of the instantaneous frequency occurs as the pulse propagates. Upon inclusion of the nonadiabatic correction, the two-photon coherence becomes complex-valued, with the result that the medium-induced macroscopic polarization develops an imaginary part proportional to the derivative $\partial(A_1^2/\gamma)/\partial t$. Its sign may be either positive (static detuning is negative, $\Omega_{21} < 0$) or negative; that is, the generated field can either enhance or suppress a pulse propagating in a medium. Below, we will show that this is determined by the sign of the derivative of the instantaneous detuning away from the two-photon resonance with allowance for the Stark shift.

Evaluating the derivative $\partial(A_1^2/\gamma)/\partial t$ and substituting the expression for $V$ into Eq. (18), we find that the real-valued amplitude obeys the equation

$$\frac{\partial A_1}{\partial z} + \frac{1}{c}\frac{\partial A_1}{\partial t} = \pm\frac{2\pi\omega_1\hbar q^2 N}{c\gamma^3}\left(-A_1^3\frac{\partial \Delta}{\partial t} + 2\Delta A_1^2\frac{\partial A_1}{\partial t}\right), \qquad (21)$$

where

$$\Delta = 2\omega_1' - \omega_{21} - \Omega_s^{(2)}$$

is the instantaneous detuning away from the two-photon resonance with allowance for the shift induced by the external Stark pulse.

It is convenient to pass from the equation for the phase to the equation for the instantaneous frequency $\omega' = \omega + \partial\varphi/\partial t$; that is,

$$\frac{\partial \omega_1'}{\partial z} + \frac{1}{c}\frac{\partial \omega_1'}{\partial t} = \mp\frac{2\pi\omega_1\hbar q^2 N}{c\gamma^3}\left(-A_1^3\frac{\partial \Delta}{\partial t} + 2\Delta A_1^2\frac{\partial A_1}{\partial t}\right)\frac{\Delta}{A_1}. \qquad (22)$$

Equations (21) and (22) describe the propagation of a light pulse under the SCRAP conditions as long as adiabaticity condition (13) is satisfied. One can see that the right-hand sides of these equations differ only by the factor $\Delta/A_1$. This indicates that the changes in the frequency and in the pulse shape occur simultaneously and that there is a relation between them. The first terms on the right-hand sides of the equations in question are due to the high-frequency Kerr effect, which leads both to a change in the shape of a pulse as it propagates in a medium and to phase self-modulation during the propagation process. The second terms lead to a change in the velocity of pulse propagation.

We note that, if there occurs a passage through the resonance ($\Delta = 0$) in scanning the frequency, the sign of the derivative does not change, as in the SCRAP case, since $\Delta$ is either an increasing (for a negative initial detuning, $\Omega_{21} < 0$) or a decreasing ($\Omega_{21} > 0$) function of time. The field generated by the imaginary part of the polarization is then in antiphase with the incident pulse, suppressing it. The situation is different if the scanned frequency does not reach the resonance. The derivative will be positive within one part of the pulse and negative within the remaining part, so that one part of the pulse will be suppressed, while the remaining part will be enhanced.

Going over to the coordinates $\xi = z$ and $\tau = t - z/c$ and taking into account Eq. (21), one can recast Eq. (22) in the form

$$\frac{\partial \omega_1'}{\partial \xi} = -\frac{\Delta(\tau, \xi)}{A_1(\tau, 0)}\frac{\partial A_1}{\partial \xi}. \qquad (23)$$

Integrating Eq. (23), we obtain

$$\frac{\Delta(\tau, \xi)}{\Delta(\tau, 0)} = \frac{A_1^2(\tau, 0)}{A_1^2(\tau, \xi)}. \qquad (24)$$

This is a general relation that is valid in the adiabatic approximation. It reflects the interplay of the change in the instantaneous frequency of a pulse and the change in its envelope during the propagation of the pulse in a medium under the conditions of a two-photon resonance. Relation (24) also shows that the effects of the change in the pulse shape and of self-modulation are related to each other and cannot be considered separately.

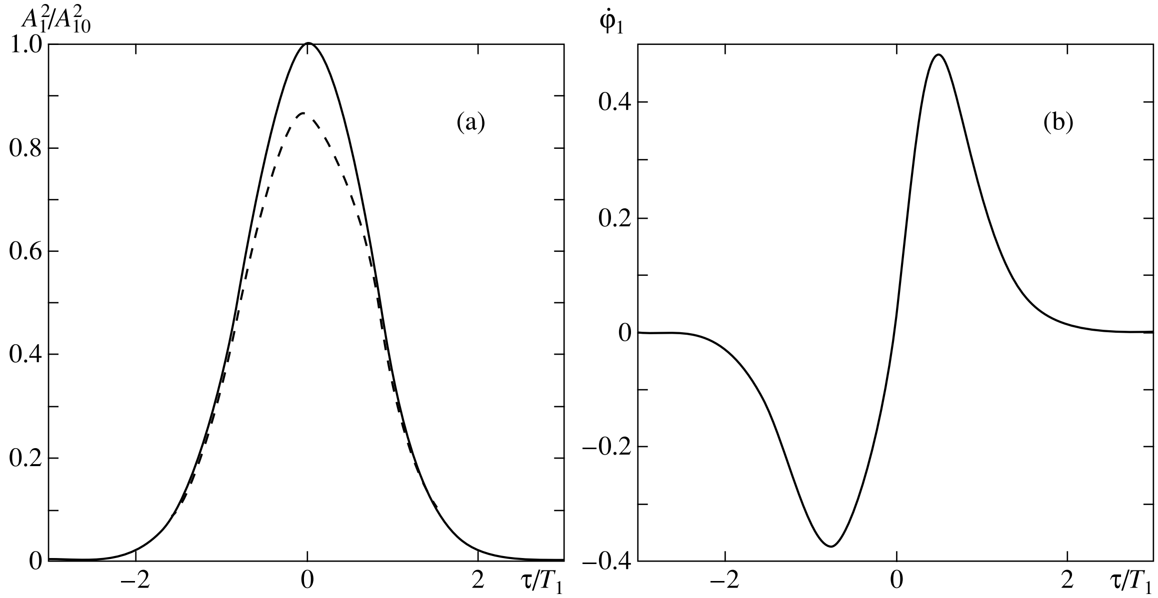


**Fig. 5.** Spacetime evolution of a pulse under the SCRAP conditions: (a) envelope of the pulse as a function of time at $Z$ = (solid curve) 0 and (dashed curve) 10; (b) instantaneous frequency $\dot{\varphi}$ of the triggering pulse as a function of time at $Z = 10$ (it is normalized to $\alpha_0 = 8\pi k_1 N\chi_1(\omega_1)$). The parameters of the pulse were set to the values identical to those used for Fig. 3.

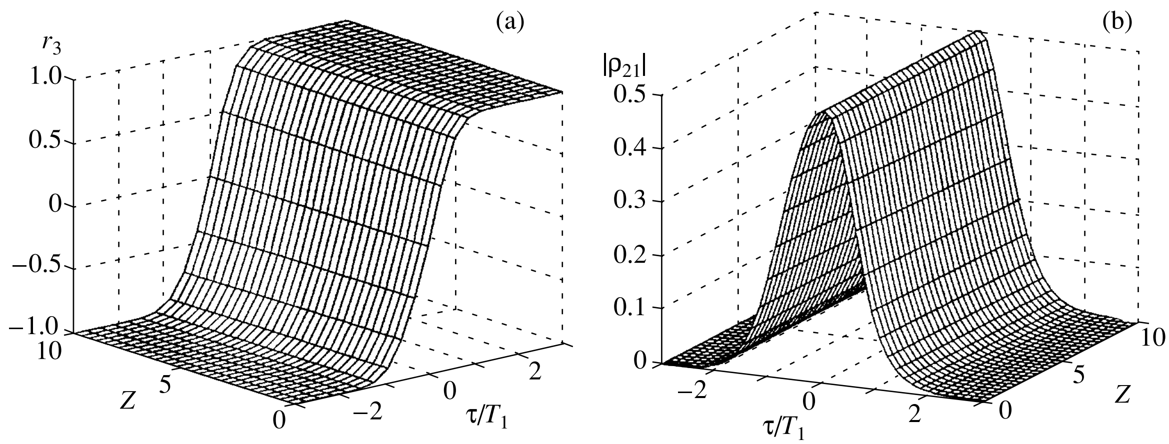


**Fig. 6.** Spacetime evolution of (a) the population difference $r_3 = \rho_2 - \rho_1$ and (b) the two-photon coherence $|\rho_{21}|$ under the SCRAP conditions. The pulse parameters were set to the values identical to those used for Fig. 3.

Let us rewrite Eq. (21) as

$$\frac{\partial A_1}{\partial z} + \frac{1}{v_p}\frac{\partial A_1}{\partial t} = \pm\frac{2\pi\omega_1\hbar q^2 N}{c\gamma^3}A_1^3\frac{\partial\Delta}{\partial t}, \qquad (25)$$

where

$$\frac{1}{v_p} = \frac{1}{c}\left(1 \pm \frac{4\pi\omega_1\hbar q^2 N}{\gamma^3}\Delta A_1^2\right). \qquad (26)$$

The results obtained by numerically solving Eq. (25) are illustrated in Fig. 5, where the time dynamics of the normalized intensity of the triggering pulse is shown at the input and output of the medium. At the input, the triggering and the Stark pulse have a Gaussian shape, their parameters and the time of delay between them being chosen in such a way as to ensure fulfillment of conditions (15) and (16), which are necessary for implementing SCRAP and a complete population transfer. One can see that the amplitude of a propagating pulse decreases smoothly, while its shape changes slowly. We can identify two effects that are responsible for the change in the pulse shape: (i) that which is due to a diabatic character of interaction [formula (26)] and (ii) that which is due to phase self-mod-

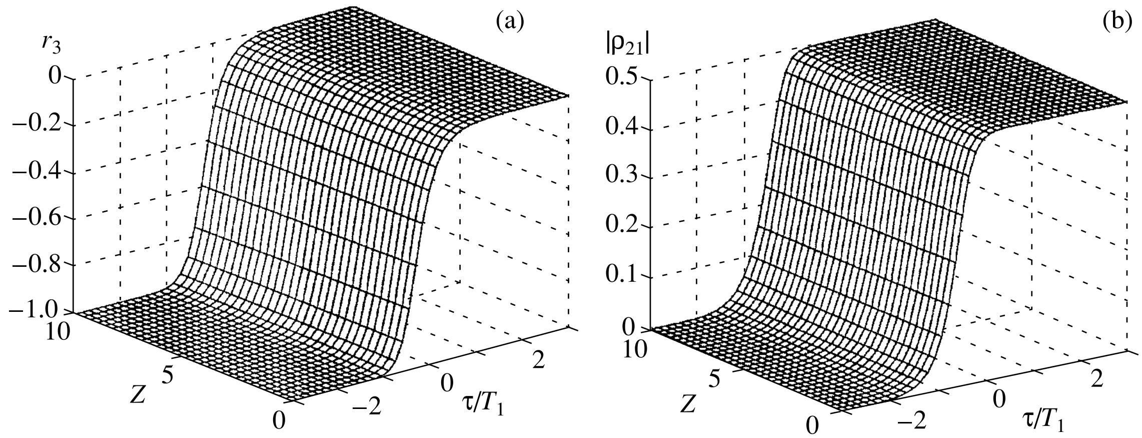


**Fig. 7.** Spacetime evolution of (a) the population difference $r_3 = \rho_2 - \rho_1$ and (b) the two-photon coherence $|\rho_{21}|$ under the half-SCRAP conditions. The pulse parameters were set to the values identical to those used for Fig. 4.

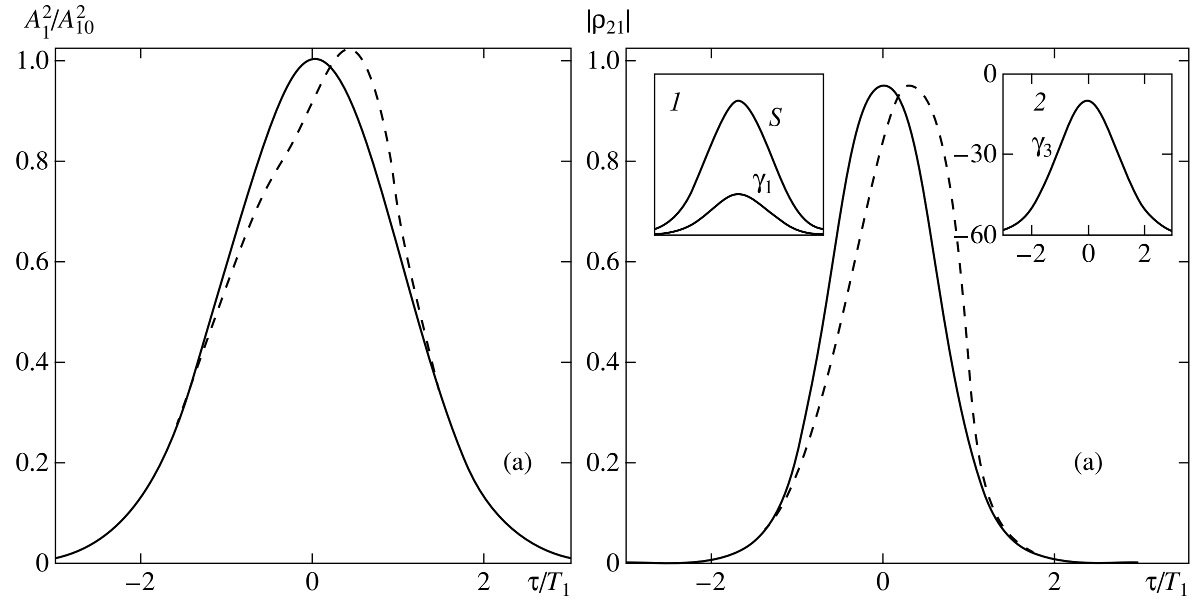


**Fig. 8.** Time evolution of the triggering pulse and the two-photon coherence $|\rho_{21}|$ in the case where the sweeping of the frequency does not lead to passage through the resonance: (a) envelope of the pulse at $Z$ = (solid curve) 0 and (dashed curve) 70; (b) two-photon coherence $|\rho_{21}|$ (the notation for the curves is identical to that in Fig. 8a). The insets display (*1*) the timing of the triggering and the Stark pulse and (*2*) the instantaneous detuning $\gamma_3$ away the two-photon resonance. The following parameter values were used here: $\delta t = 0$, $\Omega_{21}T_1 = -60$, $T_s/T_1 = 1.6$, $S_0T_1 = 50$, and $\gamma_{10}T_1 = 15$.

ulation because of the high-frequency Kerr effect. The instantaneous frequency of the pulse varies with time as is shown in Fig. 5b. In order to calculate $\dot{\varphi}$, we used Eq. (24).

Figures 6 and 7 show the spacetime evolution of the population difference and of the two-photon coherence under the SCRAP and half-SCRAP conditions, respectively. We can see that the population inversion and the maximum coherence induced in the two-photon transition persist over a long length of the medium.

We would like to emphasize that, in order to achieve a high two-photon coherence (close to a maximum value), it is not necessary to pass through the resonance. As can be seen from the formula for $r_1$ in (11), fulfillment of condition $|\gamma_1| \gg |\gamma_3|$, along with adiabaticity condition (13), is sufficient for this. This condition means that the two-photon Rabi frequency must exceed the detuning away the resonance at the instant of time when the difference of the populations is close to zero, $\rho_2 - \rho_1 = r_3 \approx 0$. Figure 8a shows how the envelope of the triggering pulse that propagates in a medium varies in this case. A moderate increase in the pulse amplitude is associated with the redistribution of energy in the pulse due to the nonadiabatic correction, which leads to the weakening of the forward front of the pulse and to the strengthening of the backward front [see discussion after formula (22)], the total pulse energy (the area under the intensity curve) remaining virtually unchanged. Figure 8b shows that, under the quasiresonance conditions, a two-photon coherence close to a maximum value is induced over a long length of the medium. This can be used to implement an efficient generation of the third harmonic in the short-wavelength region of the spectrum.

## 4. CONCLUSIONS

On the basis of the vector model for a two-photon resonance, the SCRAP phenomenon, which can lead to a complete population transfer and a maximum coherence in the resonance transition, has been considered in the two-level approximation. This approach has enabled us to derive analytic expressions for populations and two-photon coherence with allowance for a diabatic character of the interaction, to analyze them over a broad region of parameter values, and to specify the adiabaticity conditions, as well as to develop a simple geometric interpretation of the effect. On this basis, we have investigated special features of the propagation of short laser pulses under various conditions. We have also proposed a new possibility for achieving a high coherence (close to a maximum value).

It has been shown that, because of a diabatic character of the interaction, the off-diagonal density-matrix element describing the two-photon coherence is complex-valued. Its imaginary part induces a second-order nonlinear Kerr polarization, which is responsible for the change in energy transfer between the propagating pulse and the medium, while the real part of the Kerr polarization leads to phase self-modulation. As the pulse propagates, these factors lead to a decrease in its amplitude and to a change in its shape (over small propagation lengths, the pulse shape remains virtually unchanged). In addition, the carrier frequency of the pulse changes with time, the region of linear sweeping existing over a large part of the pulse duration. We note that the change in the pulse frequency is related to the change in the pulse shape since they are both due to self-interaction via the high-frequency Kerr effect in a time-dependent laser field. It seems that the duration of such a pulse can be reduced by transmitting it through a dispersive delay line by using the compression effect [23].

We have also investigated the spacetime evolution of the population difference and of the two-photon coherence during pulse propagation. We have shown that the population inversion and the maximum coherence persist over a long medium length under specific conditions at the boundary. This method makes it possible to obtain a nearly complete inversion between the ground state and a high-lying excited level or to force a medium into a coherent state by using visible-range lasers that produce pulses of duration shorter than the relaxation time of the excited state. In view of this, the proposed method can find various applications in creating new sources of pulsed coherent radiation. By way of example, we indicate that, on the basis of the anti-Stokes Raman scattering, a medium inverted in this way can be used to generate, in the short-wavelength region of the spectrum, including the vacuum-ultraviolet region, short pulses tunable with respect to the wavelength. In all probability, cooperative anti-Stokes scattering can also be investigated by using this scheme. We would also like to note the possibility of generating the third harmonic in the short-wavelength region of the spectrum under the conditions of maximum coherence.

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