

Self-induced transparency in a heterogeneous isotopic mixture

S.V. Sazonov

Abstract. Self-induced transparency is theoretically studied in the system of quantum transitions whose frequencies differ due to the isotope shift caused by differences in the mass, shape, and shell structure of atomic nuclei. In the case of good mutual resolution of the spectral lines of different isotopes, only one component of the mixture is in resonance with the pulse field, while the rest of the components interact with the pulse under quasi-resonance conditions. This circumstance determines the specific features of the effect in heterogeneous media. The conditions for realisation of self-focusing and quasi-channeling are found taking into account transverse perturbations. The latter regime is characterised by a change in the shape of the propagating pulse whose transverse size remains invariable.

Keywords: self-induced transparency, soliton, isotope shift.

1. Introduction

The resonance effect of self-induced transparency (SIT) has been attracting the attention of researchers from the time of its discovery [1, 2] till now [3–5]. The SIT is very sensitive to the detuning $\Delta = \omega_0 - \omega$ of the carrier frequency ω of a light pulse from the central frequency ω_0 of a resonance atomic transition. This is especially pronounced in media with a small inhomogeneous broadening when the corresponding relaxation time T_2^* greatly exceeds the optical pulse duration τ_p . If $|\Delta| > 1/T_2^*$ and $1/\tau_p$, atoms in the medium are excited weaker and slowing down of the pulse velocity occurs weaker than in the opposite case. In addition, the tuning value and sign play an important role in the transverse dynamics of the pulse [6]. Thus, when $\Delta < 0$, the pulse propagation in an equilibrium medium is accompanied by its self-focusing, whereas for $\Delta > 0$, the pulse defocusing occurs [7, 8].

The SIT effect has been studied in detail both in homogeneous media and media with resonance quantum transitions, which differ from each other in some parameters. Thus, the propagation of laser pulses in the system of quantum transitions with equal frequencies but different

matrix elements of dipole moments was studied in papers [9, 10] where many nonlinear regimes of the SIT development, in particular, chaotic regimes were observed. The passage from one regime to another was achieved by changing the relative concentrations of different components of the mixture.

The heterogeneous nature of resonance media introduces considerable specific features in collective emission at initially inverted quantum transitions [11]. This concerns to a great extent the situation when atoms of different types are inhomogeneously spatially distributed [12]. A rigid correlation between the type of spatial distribution of atoms and the degree of their initial excitation, on the one hand, and parameters of the emitted pulses, on the other, allows the control of the collective emission process.

In a gas mixture of isotopes emitting narrow spectral lines, only atoms of one of the isotopes can be exactly resonant with the laser pulse field, which is caused by the isotope shift [13–15]. This shift is especially large for the atoms of light (the nuclear mass number $A \sim 10$) and heavy ($A \sim 100$) elements. In the first case, the isotope shift is caused by the difference in nuclear masses of the corresponding isotopes, while in the second one – by the difference in their size and distribution of intranuclear charges [13–15]. The relative isotope shifts $|\Delta\omega_0/\omega_0|$ achieve $\sim 10^{-5} - 10^{-4}$ [13]. By taking $\omega_0 \sim 10^{15}$ for the visible range, we obtain $\Delta\omega_0 \sim 10^{11} \text{ s}^{-1}$. For $\tau_p \sim 0.1 \text{ ns}$ and $T_2^* \sim 1 \text{ ns}$, this value of $\Delta\omega_0$ is sufficient for selective excitation of atoms of various isotopes.

This selectivity should efficiently affect the differences in the delay times upon the propagation of pulses resonantly tuned to the atomic frequencies of various isotopes, which can give information, for example, on the partial concentrations of isotopes in the isotopic mixture. In addition, as pointed out above, the type of the transverse dynamics of SIT pulses should depend on these tunings and relative concentrations of isotopes. As a result, a competition between self-focusing and defocusing can appear because the signs of the resonance detuning for different isotopes can be different. Here, the situation is completely opposite to that considered in [9, 10] in the sense that quantum transitions for different components of the mixture differ only in frequency but have almost identical matrix elements of the dipole moments (see below).

Note that, generally speaking, the isotope scatter of quantum transition frequencies cannot be taken into account by considering an arbitrary inhomogeneously broadened absorption line. The inhomogeneous broadening caused by the Doppler effect has the statistical nature,

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Received 25 April 2006; revision received 6 August 2006

Kvantovaya Elektronika 37 (1) 29–35 (2007)

Translated by M.N. Sapozhnikov

whereas the isotope frequency scatter is regular, i.e. in the latter case the absorption frequency at each transition (the Doppler effect apart) remains fixed during the interaction of this transition with the light pulse field. In this case, each isotopic component of the mixture has its own absorption line, which can be inhomogeneously broadened due to the Doppler effect. The difference between the regular and statistical frequency scatters caused by the isotope shift and Doppler effect, respectively, can be especially pronounced in the case of inhomogeneous spatial distributions of atoms of different isotopic components.

In this connection it is interesting to study the features of the longitudinal and transverse dynamics of SIT pulses propagating in an isotopic mixture. The results of this study are presented below.

2. Equations of the pulse and medium dynamics

Consider a laser pulse propagating parallel to the z axis in the N -component isotopic mixture. The pulse frequency is assumed close to quantum transition frequencies, which are different due to different isotope shifts for isotopic components. We describe all the isotopes by the model of two-level atoms. In this case, the pulse and medium dynamics is described the Maxwell–Bloch equations [16, 17]

$$\frac{\partial \Omega}{\partial z} + \frac{n_0}{c} \frac{\partial \Omega}{\partial t} = -i\beta \sum_{j=1}^N \alpha_j R_j - i \frac{c}{2n_0\omega} \Delta_{\perp} \Omega, \quad (1)$$

$$\frac{\partial R_j}{\partial t} = iA_j R_j + i\Omega W_j, \quad (2)$$

$$\frac{\partial W_j}{\partial t} = \frac{i}{2} (\Omega^* R - \Omega R^*), \quad (3)$$

where $i = 1, \dots, N$; $\Omega = 2d\psi/\hbar$ is the complex Rabi frequency of the optical pulse; d is the dipole moment of resonance transitions; \hbar is Planck's constant; ψ is the slowly varying complex envelope of the electric field E of the pulse determined by the expression $E(\mathbf{r}, t) = \psi(\mathbf{r}, t) \exp[i\omega(t - n_0 z/c)] + \text{c. c.}$; c is the speed of light in vacuum; n_0 is the refractive index of the mixture caused by nonresonance quantum transitions, which differ from transitions described by constitutive equations (2) and (3); $\beta = \omega_c \omega / (cn_0)$; $\omega_c = 4\pi d^2 n / \hbar$ is the collective frequency [18]; n is the concentration of atoms in the isotopic mixture; $\alpha_j = n_j / n$; n_j is the partial concentration of the j th isotope; A_{\perp} is the transverse Laplacian; $A_j = \omega_0^{(j)} - \omega$ and $\omega_0^{(j)}$ are the resonance detuning and the central absorption frequency for the j th isotope, respectively; R_j is the Bloch variable determining the nonstationary dipole moment and related to the element $\rho_{12}^{(j)}$ of the density matrix of the $1 \leftrightarrow 2$ resonance transition in the j th isotopic component by the expression $\rho_{12}^{(j)} = R_j \exp[-i\omega(t - n_0 z/c)]$; and $W_j = (\rho_{22}^{(j)} - \rho_{11}^{(j)})/2$ is the population inversion for resonance transitions.

We neglected here the difference in the transition dipole moments for different isotopes as the value of the higher smallness order compared to the isotope shift. Indeed, the isotope shift is the first-order effect of the quantum-mechanical perturbation theory, which is determined by the corresponding addition to the atomic Hamiltonian at invariable wave functions, whereas the perturbation of

matrix elements $d \equiv d_{21}$ differs from zero only in the second order because it is characterised by the deformation of wave functions. Another argument in favour of the neglect of this difference will be presented at the beginning of the next section.

The homogeneous and inhomogeneous widths of the resonance absorption lines of isotopic components, characterised by the relaxation times T_2 and T_2^* , respectively, are neglected in (1)–(3). This assumes that the pulse duration is small compared to these relaxation times and the important condition $\Delta\omega_0^{(j)} \gg 1/T_2^*$, $1/T_2$ is fulfilled, which means that the resonance spectral lines of different isotopes can be well resolved. Under such conditions, only one of the isotopic components can be exactly resonant with the pulse field, which we will denote below by the subscript r . By taking this into account, the condition of selective excitation has the form

$$A_j T_2^* \gg A_j \tau_p \gg 1. \quad (4)$$

The typical value of T_2 in gases is $\sim 10^{-8}$ s, while the inhomogeneous broadening is caused by the Doppler effect. Then, $T_2^* \sim (c/\omega_0)[M/(k_B T)]^{1/2}$, where M is atom mass; T is the mixture temperature; and k_B is the Boltzmann constant. By taking the value $\omega_0 \simeq 3 \times 10^{15} \text{ s}^{-1}$ for the visible range, we obtain $T_2^* \sim (A/T)^{1/2}$, where T_2^* is measured in nanoseconds and temperature T in kelvins.

For $A_j \sim 10^{11} \text{ s}^{-1}$ and $\tau_p \sim 0.1$ ns, both of the sides of inequality (4) can be satisfied if $T_2^* \sim 1$ ns. Then, $T \sim 10$ K for light isotopes and $\sim 10^2$ K for heavy isotopes. Thus, in the first case a gas mixture should be preliminary cooled, which can be achieved by various laser methods [19]. In the second case, the conditions of selective excitation can be obtained at room temperature.

It is convenient to exclude material variables in (1)–(3), thereby reducing the study to the analysis of the nonlinear wave equation.

For $j = r$ (in the case of the zero detuning from the resonance), we rewrite system (2), (3) in the matrix form

$$\frac{\partial \mathbf{S}}{\partial t} = i\hat{\Omega} \mathbf{S}, \quad (5)$$

where $\mathbf{S} = (R_r, R_r^*, W_r)^T$;

$$\hat{\Omega} = \frac{1}{2} \begin{pmatrix} 0 & 0 & 2\Omega \\ 0 & 0 & -2\Omega^* \\ \Omega^* & -\Omega & 0 \end{pmatrix}. \quad (6)$$

Due to the self-consistency of the problem, the elements of matrix $\hat{\Omega}$ depend on the coordinate and time. Generally speaking, this matrix is not self-commutable at different instants. However, because condition (4) is violated for $j = r$ ($A_r = 0$), the pulse can be considered so short that the commutator $|\hat{\Omega}(t), \hat{\Omega}(t + \tau_p)|$ can be neglected with good accuracy [20, 21]. Then,

$$\mathbf{S}(t) = \hat{U}(t, t_0) \mathbf{S}(t_0), \quad (7)$$

where t_0 is the onset time of the pulse action, and the evolution operator can be written approximately in the form

$$\hat{U}(t, t_0) = \exp(i\hat{\theta})_{t \rightarrow t_0, |\Omega| \rightarrow \infty}; \quad \hat{\theta} = \int_{t_0}^t \hat{\Omega} dt'.$$

From the definition of $\hat{\theta}$ and the form of the matrix $\hat{\Omega}$, it is easy to obtain the relations $\hat{\theta}^{2k+1} = |\theta|^{2k} \hat{\theta}$ and $\hat{\theta}^{2(k+1)} = |\theta|^{2k} \hat{\theta}^2$ for any natural k , where $|\theta| = |\int_{t_0}^t \Omega dt'|$. By expanding the operator exponential $\exp(i\hat{\theta})$ in a Maclaurin series and summing it taking into account these relations, we obtain

$$\exp(i\hat{\theta}) = I + i \frac{\hat{\theta}}{|\theta|} \sin |\theta| + 2 \frac{\hat{\theta}^2}{|\theta|^2} \sin^2 \frac{|\theta|}{2}.$$

By passing in this expression to the limit $t \rightarrow t_0, |\Omega| \rightarrow \infty$ and using the L'Hospital rule, we find the evolution operator

$$\hat{U} = I + i \frac{\hat{\Omega}}{|\Omega|} \sin \theta + 2 \frac{\hat{\Omega}^2}{|\Omega|^2} \sin^2 \frac{\theta}{2}, \quad (8)$$

where I is the unit operator; $\theta = \int_{-\infty}^t |\Omega| dt$, and the onset time of the pulse action is formally assigned to $-\infty$. By assuming that $\hat{S}(t_0 \rightarrow \infty) = (0, 0, W_{r\infty})^T$ in (7), which corresponds to the absence of initial atomic coherence, we obtain

$$R_r = iW_{r\infty} \frac{\Omega}{|\Omega|} \sin \theta, \quad W_r = W_{r\infty} \cos \theta. \quad (9)$$

Here, $W_{j\infty}$ is the initial inversion of j th quantum transition (for all j , including $j = r$).

For other isotopes, inequalities (4) are fulfilled, of which the second one is called the quasi-resonance condition [22, 23]. In this case, for $j \neq r$ the left side of Eqn (2) is proportional to the small parameter $\sim (A_j \tau_p)^{-1}$. Then, the solution of system (2), (3) can be found in the form of the expansion in this parameter, which corresponds in fact to the Crisp expansion [24]. As a result, we obtain in the third order

$$R_j = -\frac{\Omega}{A_j} W_j + \frac{i}{A_j^2} \frac{\partial}{\partial t} (\Omega W_j) + \frac{W_{j\infty}}{A_j^3} \frac{\partial^2 \Omega}{\partial t^2} - i \frac{W_{j\infty}}{A_j^4} \frac{\partial^3 \Omega}{\partial t^3}. \quad (10)$$

Here, the replacement $W_j \rightarrow W_{j\infty}$ was made in the two last terms because atoms quasi-resonant with the pulse field are weakly excited.

By substituting (10) into (3) and taking into account only the first two terms in (3), we obtain

$$\begin{aligned} \frac{\partial W_j}{\partial t} &= -\frac{1}{2A_j^2} \left[\Omega^* \frac{\partial}{\partial t} (\Omega W_j) + \Omega \frac{\partial}{\partial t} (\Omega^* W_j) \right] \\ &\approx -\frac{W_{j\infty}}{2A_j^2} \frac{\partial}{\partial t} (|\Omega|^2), \end{aligned}$$

which gives after integration

$$W_j = W_{j\infty} \left(1 - \frac{|\Omega|^2}{2A_j^2} \right). \quad (11)$$

From (10) and (11), we obtain

$$R_j = -\frac{W_{j\infty}}{A_j} \Omega + \frac{W_{j\infty}}{2A_j^3} |\Omega|^2 \Omega - \frac{iW_{j\infty}}{2A_j^4} \frac{\partial}{\partial t} (|\Omega|^2 \Omega) +$$

$$+ \frac{iW_{j\infty}}{A_j^2} \frac{\partial \Omega}{\partial t} + \frac{W_{j\infty}}{A_j^3} \frac{\partial^2 \Omega}{\partial t^2} - i \frac{W_{j\infty}}{A_j^4} \frac{\partial^3 \Omega}{\partial t^3}. \quad (12)$$

By substituting (9) and (12) into (1), we obtain the equation

$$\begin{aligned} \frac{\partial \Omega}{\partial z} + \frac{1}{v_g} \frac{\partial \Omega}{\partial t} &= -\sigma \frac{\Omega}{|\Omega|} \sin \theta - i\gamma \Omega + i \frac{b}{2} |\Omega|^2 \Omega \\ &+ \frac{g}{2} \frac{\partial}{\partial t} (|\Omega|^2 \Omega) + ib \frac{\partial^2 \Omega}{\partial t^2} + g \frac{\partial^3 \Omega}{\partial t^3} - i \frac{c}{2n_0 \omega} A_{\perp} \Omega, \end{aligned} \quad (13)$$

where v_g is the linear group velocity taking into account the contribution of quasi-resonance transitions, which is determined by the relation $1/v_g = n_0/c - \beta \sum_{j \neq r} W_{j\infty} \alpha_j / A_j^2$; where $b = -\beta \sum_{j \neq r} W_{j\infty} \alpha_j / A_j^3$ and $g = -\beta \sum_{j \neq r} W_{j\infty} \alpha_j / A_j^4$ are the group velocity dispersion parameters of the first and second orders, respectively; $\sigma = -\beta \alpha_r W_{r\infty}$ and $\gamma = -\beta \sum_{j \neq r} W_{j\infty} \alpha_j / A_j$.

Let us represent the complex Rabi frequency in the form

$$\Omega = |\Omega| \exp(i\Phi), \quad (14)$$

where the correction Φ to the light pulse phase (or its eikonal) is, generally speaking, the function of coordinates and time.

If only the first term is retained in the right-hand side of (13), we will have $\Phi = \text{const}$ under the condition that the input pulse is not phase-modulated. This agrees with the conclusion obtained earlier in [25, 26]. Thus, the nontrivial dependence of Φ on coordinates and time can be caused by all the terms in the right-hand side of (13) except the first one. These terms, except the last one, represent expansions in powers of small parameters $(A_j \tau_p)^{-1}$. Therefore, the dependence $\Phi(r, t)$ is weak. For this reason, by substituting (14) into (13), we can neglect the derivatives of Φ with respect to time in the right-hand side of (13) as the values of the higher order of smallness relative to the powers of parameters $(A_j \tau_p)^{-1}$.

By separating the real and imaginary parts, we obtain from (13) and (14) the nonlinear system

$$\begin{aligned} \frac{\partial^2 \theta}{\partial z \partial \tau} + \sigma \sin \theta - \frac{3g}{2} \left(\frac{\partial \theta}{\partial \tau} \right)^2 \frac{\partial^2 \theta}{\partial \tau^2} - g \frac{\partial^4 \theta}{\partial \tau^4} \\ = \frac{c}{2n_0 \omega} \left[(A_{\perp} \Phi) \frac{\partial \theta}{\partial \tau} + 2(\nabla_{\perp} \Phi) \left(\frac{\partial}{\partial \tau} \nabla_{\perp} \theta \right) \right], \end{aligned} \quad (15)$$

$$\begin{aligned} \frac{\partial \theta}{\partial \tau} \left(\gamma + \frac{\partial \Phi}{\partial z} \right) - \frac{b}{2} \left(\frac{\partial \theta}{\partial \tau} \right)^3 - b \frac{\partial^3 \theta}{\partial \tau^3} \\ = \frac{c}{2n_0 \omega} \left[(\nabla_{\perp} \Phi)^2 \frac{\partial \theta}{\partial \tau} - \frac{\partial}{\partial \tau} A_{\perp} \theta \right], \end{aligned} \quad (16)$$

where $\tau = t - z/v_g$ and it is taken into account that $|\Omega| = \partial \theta / \partial \tau$.

System (15), (16) describes the nonlinear propagation of a laser pulse in a quasi-resonance isotopic mixture taking into account transverse perturbations.

3. One-dimensional SIT solitons

Before studying the effect of transverse perturbations on light pulses, we consider their one-dimensional propagation

when the right-hand sides in (15) and (16) can be neglected. In this case, this system can be split with respect to variables θ and Φ , and (15) takes the form of the Konno–Kameyama–Sanuki equation, which can be integrated by the method of inverse scattering problem [27] and generalises the sine-Gordon equation describing SIT in a one-component resonance medium. It is important to note here that the necessary condition of integrability is that the ratio of the nonlinearity coefficient produced by quasi-resonance atoms to the corresponding dispersion coefficient should be equal to $3/2$ [27]. One can see from (15) that this condition is fulfilled, which is a direct consequence of the neglect of the difference in the transition dipole moments for different isotopic components. Taking into account that the third and fourth terms in the left-hand sides of (15) are of the expansion type, this difference is vanishingly small. Therefore, its consideration would contradict the approximations adopted in this paper.

The one-soliton solution of (15) for the zero right-hand side has the form

$$\theta = 4 \arctan \left[\exp \left(\frac{t - z/v}{\tau_p} \right) \right], \quad (17)$$

$$|\Omega| = \frac{2}{\tau_p} \operatorname{sech} \left(\frac{t - z/v}{\tau_p} \right), \quad (18)$$

where the soliton velocity v in the laboratory coordinate system is related to the soliton duration τ_p by the expression

$$\frac{1}{v} = \frac{1}{v_g} + \sigma \tau_p^2 - \frac{g}{\tau_p^2}. \quad (19)$$

By substituting (17) into (16) in the absence of the right-hand side, we obtain after integration

$$\Phi = - \left(\gamma - \frac{b}{\tau_p^2} \right) z, \quad (20)$$

where the integration constant is set equal to zero (simply by displacing the coordinate system).

The population differences for the resonance and quasi-resonance components found from (17), (18), (9), and (11) are

$$W_r = W_{r\infty} \left[1 - 2 \operatorname{sech}^2 \left(\frac{t - z/v}{\tau_p} \right) \right], \quad (21)$$

$$W_j = W_{j\infty} \left[1 - \frac{2}{(\Delta_j \tau_p)^2} \operatorname{sech}^2 \left(\frac{t - z/v}{\tau_p} \right) \right], \quad (22)$$

respectively. We assume below that all the atoms are in the ground state before pulsed excitation, i.e. $W_j = -1/2$ for all j , including $j = r$.

One can see from (21) that the resonance component of the isotopic mixture experiences a complete inversion during pulse propagation and then returns to the initial state. As for quasi-resonance components, they are excited insignificantly, as follows from (22) and the second inequality in (4). As a result, the resonance and quasi-resonance components differently affect the pulse propagation velocity.

The expression for the linear group velocity can be rewritten in the form

$$v_g = \frac{c}{n_0(1 + \kappa)},$$

where $\kappa = \eta(\omega/\bar{\Delta})^2/(2n_0^2)$; $\eta = \omega_c/\omega$; and $1/\bar{\Delta}^2 = \sum_{j \neq r} \alpha_j / \Delta_j^2$.

Consider, as an example, a mixture consisting of samarium vapour in which two isotopes ^{150}Sm and ^{152}Sm are present. The frequency of the spectral lines of the second isotope in the visible region exceeds the characteristic frequency of the lines of the first isotope by $\Delta \sim 10^{11} \text{ s}^{-1}$ [9]. Let $\alpha_1 = 0.9$ and $\alpha_2 = 0.1$. By assuming also that $n_0 \sim 1$, $n \sim 10^{13} \text{ cm}^{-3}$, $d \sim 10^{-18} \text{ esu}$, and $\omega \sim 10^{15} \text{ s}^{-1}$ [7, 8], we obtain $\omega_c \sim 10^5 \text{ s}^{-1}$, $\eta \sim 10^{-10}$, and $\kappa \sim 10^{-2}$. Thus, quasi-resonance transitions reduce the linear group velocity of an optical pulse by a few percent. As for the nonlinear addition to the inverse velocity of these transitions, which is determined by the last term in (19), its ratio to the linear addition considered above is $\sim (\Delta \tau_p)^{-2} \ll 1$.

In turn, the resonance transitions reduce the propagation velocity of pulses essentially nonlinearly, which is expressed by the second term in the right-hand side of (19). The corresponding dimensionless addition to the inverse velocity is of the order of the value $c\sigma\tau_p^2/n_0 \sim \alpha_r\eta(\omega\tau_p)^2 \sim \alpha_r$, which we determined by using the numerical estimates presented above. Therefore, the pulse velocity considerably depends on the percentage of the resonance component in the isotopic mixture.

By tuning the laser pulse frequency to resonances with different components and determining the corresponding time delays of the pulse emerging from the medium, we can find the percent composition of different isotopes.

It is important to note that the threshold value of the input pulse area

$$A_p = \int_{-\infty}^{+\infty} |\Omega(z=0, t)| dt \geq \pi$$

is independent of the resonance to which the laser pulse is tuned in the mixture.

It follows from (9), (12), (17), (18), and (20) that expressions for the dipole moments $D_j = d(\rho_{21}^{(j)} + \rho_{12}^{(j)}) = 2d \operatorname{Re}\{R_j \exp[i\omega(t - n_0 z/c)]\}$ induced by the light pulse have the form

$$D_r = -2d \tanh \left(\frac{2 - z/v}{\tau_p} \right) \operatorname{sech} \left(\frac{t - z/v}{\tau_p} \right) \sin[\omega(t - z/v_{\text{ph}})] \quad (23)$$

for resonance components and

$$D_j = \frac{2d}{\Delta_j \tau_p} \operatorname{sech} \left(\frac{t - z/v}{\tau_p} \right) \cos[\omega(t - z/v_{\text{ph}})] \quad (24)$$

for quasi-resonance components. Here, v_{ph} is the phase velocity of a soliton determined by the expression

$$\frac{1}{v_{\text{ph}}} \equiv \frac{n_s}{c} = \frac{n_0}{c} + \frac{\gamma}{\omega} \left(1 - \frac{b}{\gamma \tau_p^2} \right), \quad (25)$$

where $n_s \equiv n_{\text{lin}} + n_2 |\psi|_{\text{lm}}^2$ is the soliton refractive index; $n_{\text{lin}} = n_0 + c\gamma/\omega$ is its linear part; $|\psi|_{\text{lm}} = 2d/(\hbar\tau_p)$ is the amplitude of the 2π soliton; and

$$n_2 = - \frac{\pi d^4 n}{2\hbar^3 n_0} \sum_{j \neq r} \frac{\alpha_j}{\Delta_j^3} \quad (26)$$

is the nonlinear refractive index.

As expected, the ratio $|D_j/D_r| \sim |A_j\tau_p| \ll 1$. On the other hand, the dipole response of resonance atoms is shifted by $\pi/2$ with respect to the light pulse phase and its envelope has the two-polar shape, as in the case of SIT in a homogeneous medium [3]. The response of quasi-resonance atoms is one-polar and virtually coincides in phase with the pulse field.

Note that the correction Φ to the phase of a one-dimensional soliton is independent of time. Therefore, one-soliton solution (17)–(20) is the exact solution of Eqn (13) without the assumption that the derivatives of Φ with respect to time are small because here they are rigorously zero. Thus, the frequency of the produced 2π soliton is rigorously equal to the input pulse frequency. Analysis of the multi-soliton solution of system (15), (16) in the one-dimensional approximation shows that during collisions between solitons, the phase modulation appears for each of them. However, this modulation disappears after their elastic interaction and the phase of each of the solitons recovers its simple addition in form (20).

4. Effect of transverse perturbations

To take into account the effect of transverse perturbations on the propagation of a SIT soliton in the isotopic mixture, we will assume that its eikonal Φ and duration $\tau_p = 1/\rho$ are now the functions of all the three coordinates. Note first that, by using (19) and (20), we can write the argument of one-dimensional soliton (18) in the form

$$\frac{t - z/v}{\tau_p} = \rho[\tau + F(\rho)\Phi], \quad (27)$$

where

$$F(\rho) = \frac{\sigma - g\rho^4}{\rho^2(\gamma - b\rho^2)}. \quad (28)$$

Taking into account transverse perturbations by the adiabatic method [28], we will assume below that expressions (27) and (28) remain also valid for non-one-dimensional solitons. According to this, we will seek the solution of system (15), (16) for θ in the form (17) taking into account the substitution (27), where now $\rho = \rho(\mathbf{r})$ and $\Phi = \Phi(\mathbf{r})$. Because $\rho = \text{const}$ and $\Phi \sim z$ in the one-dimensional case, the variable ρ is assumed a ‘slow’ function of coordinates and Φ is assumed a ‘fast’ function in the consideration of transverse perturbations [28]. Therefore, we have $\nabla_\perp(F(\rho)\Phi) \approx F(\rho)\nabla_\perp\Phi$. The dependence on τ can be excluded on average by multiplying (15) and (16) by $|\Omega|$ and then integrating over τ taking (17), (18), and (27) into account. As a result, we obtain the system

$$\frac{\partial\rho}{\partial z} + \nabla_\perp(\rho\nabla_\perp\varphi) = 0, \quad (29)$$

$$\begin{aligned} \frac{\partial\varphi}{\partial z} + \left[1 + \frac{1}{3}\rho^2 F^2(\rho)\right] \frac{(\nabla_\perp\varphi)^2}{2} - \frac{c}{n_0\omega}(\gamma - b\rho^2) \\ = \left(\frac{c}{2n_0\omega}\right)^2 \left[\frac{A_\perp\rho}{\rho} - \frac{1}{3}\left(\frac{\pi^2}{6} + 2\right) \frac{(\nabla_\perp\rho)^2}{\rho^2}\right], \end{aligned} \quad (30)$$

where $\varphi = -c\Phi/n_0\omega$.

Transverse perturbations in the left-hand side of (30) are taken into account in the geometric optics approximation

[28], which corresponds to nonlinear refraction, while the right-hand side takes diffraction effects into account [29].

Before proceeding to the general investigation of system (29), (30), note that the solutions of this system in the one-dimensional case ($\nabla_\perp = A_\perp = 0$) have the form $\rho = 1/\tau_p = \text{const}$ and $\Phi = -n_0\omega\varphi/c = -(\gamma - b/\tau_p^2)z$, which exactly coincide with solutions for a one-dimensional SIT soliton. This circumstance is an important argument in favour of the approach proposed here.

According to [29, 30], we will seek the solution of system (29), (30) for ρ in the self-simulating axially symmetric form

$$\rho = \frac{1}{\tau_0} \left(\frac{R_0}{R}\right)^2 \exp\left(-\frac{r^2}{R^2}\right). \quad (31)$$

Here, τ_0 and $2R_0$ are the input pulse duration at the centre of its cross section and its aperture, respectively; $2R = 2R(z)$ is the pulse aperture in the medium; and r is the radial component of the cylindrical coordinate system.

We represent the expression for φ in the form [29]

$$\varphi = f_1(z) + \frac{1}{2}f_2(z)r^2, \quad (32)$$

where f_1 and f_2 are the required functions of the coordinate z .

The first term in (32) corresponds to the one-dimensional approximation, while the second one takes into account the transverse structure of the wave fronts of the pulse (their bending).

By substituting (31) and (32) into (29), we obtain the relation

$$f_2 = \frac{R'}{R}. \quad (33)$$

Hereafter, the prime denotes the derivative with respect to z .

By substituting (31) and (32) into (30), expanding in powers of $\varepsilon = (r/R)^2 \ll 1$ (the axial approximation [29–32]), and equating expressions with the zero and second powers of ε in the right- and left-hand sides, we obtain, by using (33),

$$f_1' = \frac{c}{n_0\omega} \left(\gamma - \frac{b}{\tau_p^2} \frac{R_0^4}{R^4}\right) - \left(\frac{c}{n_0\omega}\right)^2 \frac{1}{R^2}, \quad (34)$$

$$R'' = -\frac{a}{R^3} + \frac{g}{R^5} - \mu R'^2 R^3, \quad (35)$$

where $a = 0.5(\pi^2/6 - 1)[c/(n_0\omega)]^2$; $g = cbR_0^4/(n_0\omega\tau_p^2)$; and $\mu = \sigma^2\tau_p^2/(3\gamma^2R_0^4)$. Expression (35) was obtained by assuming, taking into account the second inequality in (4), that $F(\rho) \approx \sigma/(\gamma\rho^2)$. This circumstance does not affect principally the following conclusions.

The first term in the right-hand side of (35) corresponds to diffraction effects in the transverse dynamics of the pulse, while the second and third ones – to the nonlinear transverse refraction produced by the quasi-resonance and resonance isotopic components, respectively.

In the absence of quasi-resonance components ($g = 0$), it follows from (35) that a resonance SIT soliton is stable with respect to self-focusing [16]. The same conclusion is also valid in the presence of quasi-resonance components of the

mixture if $g < 0$. One can see from expressions for g and b that in this case the components with frequencies lower than the pulse carrier frequency dominate in the mixture. This also follows from expression (26) for the nonlinear refractive index, because $n_2 > 0$ in this case, which provides self-focusing. A similar conclusion was made in [7, 8], but in the presence of atoms of only one type.

Of special interest is a situation when components with transition frequencies exceeding the pulse carrier frequency dominate in the isotopic mixture. In this case, $g > 0$ and, as follows from (35), a competition appears between the resonance and quasi-resonance components which favour self-focusing and defocusing, respectively. Let us find the conditions under which these trends can be mutually compensated and elucidate the type of pulse propagation.

Equation (35) has the first integral of the type

$$\frac{R'^2}{2} + U(R) = 0, \quad (36)$$

where

$$U(R) = -\frac{R_0'^2}{2} \exp\left[-\frac{\mu}{2}(R^4 - R_0^4)\right] + \int_{R_0}^R \exp\left[-\frac{\mu}{2}(R^4 - \zeta^4)\right] \left(\frac{a}{\zeta^3} - \frac{g}{\zeta^5}\right) d\zeta. \quad (37)$$

Equation (36) coincides formally with the energy integral for a unit mass particle moving in a field with the potential energy $U(R)$. Therefore, conditions of a stable mutual compensation of self-focusing and defocusing have the form $\partial U/\partial R = 0$ and $\partial^2 U/\partial R^2 > 0$.

Let the wave fronts of the input pulse be plane. Then, it follows from (32) and (33) that $R_0' = 0$. In this case, the first compensation condition gives the solution $R = R_0 = \sqrt{g/a} = \text{const}$, which means, as follows from (33), that $f_2 = 0$. By taking the second derivative from (37) with respect to R , we find that $(\partial^2 U/\partial R^2)_{R=R_0} = 2a^3/g^2 > 0$. Thus, we conclude that the required compensation is possible in principle.

By using expressions for g and a , we find

$$R_0 = \frac{0.80}{\sqrt{\omega_c}} \frac{c\tau_p}{\omega} \left(\sum_{j \neq r} \frac{\alpha_j}{A_j^3} \right)^{-1/2}. \quad (38)$$

Expressions for the group and phase velocities of the pulse obtained from (27), (28), (32), and (34) taking into account the finite transverse size of the pulse have the form

$$\frac{1}{v} = \frac{n_0}{c} + \frac{\sigma/\rho^2 - g\rho^2}{1 - b\rho^2/\gamma} \left(1 - \frac{b}{\gamma\tau_p^2} - \delta \right), \quad (39)$$

$$\frac{1}{v_{\text{ph}}} = \frac{n_0}{c} + \frac{\gamma}{\omega} \left(1 - \frac{b}{\gamma\tau_p^2} - \delta \right), \quad (40)$$

where $\delta = c/(\gamma n_0 \omega R_0^2) \sim (\bar{\Delta}\tau_p)^{-2}$ is the correction to these velocities caused by the finite transverse size of the pulse. For $R_0 \rightarrow \infty$, these relations, as expected, transform to expressions (19) and (25), respectively, for a one-dimensional soliton. This demonstrated that the phase velocity of the soliton is independent of the transverse coordinate. Therefore, constant-phase surfaces in the medium remain plane. The group velocity, however, decreases from the

centre of the pulse cross section to pulse periphery with decreasing ρ . As a result, the central part of the pulse bends forward during pulse propagation, overtaking its periphery sites. As the pulse is elongated in such a way, its transverse size $2R_0$ remains invariable, although its shape changes as a whole, acquiring the form of an elongating hollow missile. According to these two features, we will call this propagation regime quasi-channeling.

If $R_0 \neq \sqrt{g/a}$ at the input to the medium, quasi-channeling will be accompanied by periodic pulsations of the pulse radius around this value in accordance with the stability of the process. In turn, this will produce pulsations of the pulse amplitude, duration, and phase and group velocities.

As an example, we determine the possible type of propagation of a one-dimensional SIT soliton in the two-component samarium mixture considered above. Let the frequency of a pulsed laser be initially tuned to a quantum transition in the ^{150}Sm isotope. In this case, $\Delta > 0$ and, therefore, the quasi-channeling regime is possible. Then, we find from (38) for numerical parameters presented in the previous section that $R_0 \sim 0.1$ cm. When the pulse frequency is tuned to the ^{152}Sm isotope, the value of Δ becomes negative and, therefore, self-focusing should occur.

Thus, the tuning of the pulse frequency to one or another component of the isotopic mixture not only affects the pulse velocity but can qualitatively change its transverse dynamics.

5. Conclusions

We have established the specific features of the SIT effect in a heterogeneous isotopic mixture, which are absent in homogeneous media. By studying the longitudinal and transverse dynamics in various pulse propagation regimes, it is possible to perform diagnostics of heterogeneous mixtures by measuring the percentage of various isotopes (first of all, of artificially enriched mixtures). In addition, media containing the specified amounts of various isotopes can be used in optical delay lines with properties variable by tuning the input pulse carrier frequency.

Acknowledgements. This work was supported by the Russian Foundation for Basic Research (Grant No. 05-02-16422a).

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