Influence of detuning from resonance on the instability of coherent light pulses in absorbing media

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An instability of coherent propagation of light pulses in a resonant medium, predicted earlier theoretically [L. A. Bol'shov, V. V. Likhanskiĭ, and A. P. Napartovich, Sov. Phys. JETP 45, 928 (1977)], is demonstrated to be in agreement with the published experimental data. It is shown that detuning from resonance by $\Delta\omega = \omega_0 - \omega_{12}$ (ω_0 is the frequency of light and ω_{12} is the frequency of a resonance transition) causes the growth rate of this instability to decrease by a factor of $\Delta\omega t_p$, where t_p is the pulse duration. For $|\Delta\omega t_p| \gg 1$ and $\Delta\omega > 0$, the effect reduces to the self-focusing instability of Bespalov and Talanov [JETP Lett. 3, 307 (1966)]. For $\Delta\omega < 0$, when there is no self-focusing in the noncoherent case, a coherent pulse is unstable. The growth rate of the instability is of the same order as in the $\Delta\omega > 0$ case but it is numerically slightly smaller.

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1. The phenomenon of self-induced transparency discovered by McCall and Hahn[1] has been investigated intensively for the last decade. This phenomenon represents lossless propagation of short sufficiently intense light pulses through resonantly absorbing media (see, for example, the reviews of Poluéktov *et al.*[2]). Attention has been concentrated on such characteristics of ultrashort pulses as the propagation velocity, shape, delay time, absorption, etc. In comparing the experimental results with the theory, use has been made of one-dimensional calculations allowing for inhomogeneous broadening of absorption lines, resonance transition degeneracy, etc. Under real experimental conditions a pulse is finite in the transverse direction but in the case of sufficiently large apertures the one-dimensional approximation is regarded as fully justified.[3] The influence of the transverse structure of pulses on their propagation in resonantly absorbing media has not yet been investigated in detail. Some years ago Mattar and Newstein[4] carried out numerical calculations of the propagation of coherent pulses of Gaussian shape in the transverse direction. These calculations indicated a tendency of the pulses to self-focus as a whole.

In an earlier theoretical paper[5] the present authors and Napartovich proved the existence of an instability of light pulses in respect of transverse perturbations during resonant coherent propagation. We have shown that the fastest to grow are the perturbations with a characteristic transverse size $\sim(\lambda l_p)^{1/2}$, which grow in a distance which is of the order of the length of a light pulse l_p in a medium (λ is the wavelength of light). Experimental investigations have been made[6,7] of coherent propagation of short resonant light pulses through sodium vapor and of the transverse distribution of the intensity of the transmitted pulses. Numerical calculations[7] indicate a factor of ~10 increase in the intensity on the beam axis in the case of sufficiently large transverse dimensions r_p of the input pulse, i.e., when $F \gg 1$ ($F = 4\pi r_p^2/\lambda l_p$ is the Fresnel number); this is due to focusing of the pulse as a whole. However, experimental attempts to increase the intensity on the axis by a factor exceeding 2 have resulted in transverse splitting of a large-diameter light beam into filaments whose transverse size is of the order of $(\lambda l_p)^{1/2}$. The appearance of "hot spots" in the transverse cross section of a pulse transmitted by an absorber was pointed out in the very first paper of McCall and Hahn.[1] The results of the experiments mentioned above were in agreement with the theoretical predictions[5] of the existence of a coherent pulse instability.

It would seem that the development of an instability should affect the propagation of pulses in sufficiently long absorbers [longer than $l_p \ln(\mathscr{E}_0/\tilde{\mathscr{E}})$, where $\mathscr{E}_0$ is the amplitude of a steady-state pulse and $\tilde{\mathscr{E}}$ is the amplitude of transverse perturbations]. In fact, a recent numerical experiment[8] showed that a 2π pulse breaks up after traversing a distance of about ten pulse lengths in a medium with homogeneous broadening. However, the majority of the experimental data agree more or less satisfactorily with one-dimensional calculations. This has been due to the fact that such experiments have been carried out mainly in absorbers of moderate length ($L/l_p \sim 5-7$). In those cases when the length of an absorber has exceeded the pulse length by a factor of 8–10, the observations have indicated a considerable influence of the transverse structure on the nature of pulse propagation.[1,3,6,7,9,10] Diels and Hahn[11] investigated the propagation of 2π pulses in ruby in the $L/l_p \gtrsim 20$ case. They found that a coherent pulse crosses an absorber only when the carrier frequency ω_0 is detuned sufficiently from the center of an absorption line ω_{12}. They also found experimentally that the transmission of a 2π pulse depends on the sign of the detuning $\Delta\omega = \omega_0 - \omega_{12}$ and that for $\Delta\omega > 0$ the absorption is considerably stronger than for $\Delta\omega < 0$ for the same absolute detuning.

2. We shall consider the influence of detuning of the frequency of light from the center of a resonance transition line on the stability of coherent propagation of light pulses in the presence of transverse perturbations. For simplicity, we shall confine ourselves to a two-level model without degeneracy and we shall ignore inhomogeneous broadening. In this case the reduced equations for the field E, polarization of the medium P, and difference between the populations of the lower and upper levels n are

$$\frac{\partial\mathscr{E}}{\partial x} + \frac{\eta}{c}\frac{\partial\mathscr{E}}{\partial t} + \frac{i}{2k}\Delta_\perp\mathscr{E} + i\left(\frac{\omega\eta}{c} - k\right)\mathscr{E} = -\frac{2\pi\omega N}{\eta c} iP; \qquad (1a)$$

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$$\left.\begin{aligned}&\partial P_1/\partial t=-(\Delta\omega+\partial\varphi/\partial t)P_2,\\&\frac{\partial P_2}{\partial t}=\left(\Delta\omega+\frac{\partial\varphi}{\partial t}\right)P_1+\frac{\mu^2}{\hbar}\mathscr{E}n,\\&\partial n/\partial t=-\mathscr{E}P_2/\hbar.\end{aligned}\right\}\tag{1b}$$

Here,

$$E(x,t)=\mathscr{E}(x,t)\exp[i(\omega_0t-kx)+i\varphi(x,t)],$$
$$P(x,t)=(P_1-iP_2)\exp[i(\omega_0t-kx)+i\varphi(x,t)],$$

η is the nonresonant refractive index, N is the density of resonant particles, and μ is the dipole moment of the transition.

The system (1) has a familiar steady-state solution in the form of a one-dimensional soliton (nonresonance 2π pulse):

$$\begin{aligned}&\mathscr{E}_0(x,t)=\frac{2\hbar}{\mu t_p}\operatorname{sech}\left(\frac{t-x/v}{t_p}\right),\\&\partial\varphi_0/\partial t=0,\quad(\omega\eta/c-k)=\Delta\omega(1/v-\eta/c).\end{aligned}\tag{2}$$

where v is the pulse velocity and t_p is the pulse duration, related by the expression

$$\frac{c}{\eta v}-1=\frac{2\pi\omega\mu^2N}{\hbar\eta^2}\frac{t_p^2}{1+(\Delta\omega t_p)^2}.\tag{3}$$

We shall now analyze the stability of the system (1) against transverse perturbations which we shall assume to be proportional to $\cos\varkappa\cdot\mathbf{r}_\perp$. Linearization of the material equations (1b) near the steady-state solution (2) gives

$$\left.\begin{aligned}&\frac{\partial\tilde P_1}{\partial t}=-\Delta\omega\tilde P_2-\frac{\partial\tilde\varphi}{\partial t}P_{20},\\&\frac{\partial\tilde P_2}{\partial t}=\Delta\omega\tilde P_1+\frac{\partial\tilde\varphi}{\partial t}P_{10}+\frac{\mu^2}{\hbar}(\tilde{\mathscr{E}}n_0+\mathscr{E}_0\tilde n),\\&\frac{\partial\tilde n}{\partial t}=-\frac{1}{\hbar}(\tilde{\mathscr{E}}P_{20}+\mathscr{E}_0\tilde P_2),\end{aligned}\right\}\tag{4}$$

where

$$P_{10}=-\frac{\Delta\omega t_p^2}{1+(\Delta\omega t_p)^2}\frac{\mu^2}{\hbar}\mathscr{E}_0,$$
$$P_{20}=\frac{t_p^2}{1+(\Delta\omega t_p)^2}\frac{\mu^2}{\hbar}\frac{\partial\mathscr{E}_0}{\partial t},$$
$$n_0=1-\frac{2}{1+(\Delta\omega t_p)^2}\operatorname{ch}^{-2}\left(\frac{t-x/v}{t_p}\right).$$

A tilde above the quantities n, P_1, P_2, φ, and $\mathscr{E}$ represents small perturbations. The equations for the perturbations of the amplitude and phase of the field are

$$\left.\begin{aligned}&\frac{\partial\tilde{\mathscr{E}}}{\partial t}+\frac{c}{\eta}\frac{\partial\tilde{\mathscr{E}}}{\partial x}+\frac{c\varkappa^2}{2\eta k}\mathscr{E}_0\tilde\varphi=-\frac{2\pi\omega N}{\eta^2}\tilde P_2,\\&\left(\frac{\partial\tilde\varphi}{\partial t}+\frac{c}{\eta}\frac{\partial\tilde\varphi}{\partial x}\right)\mathscr{E}_0+\Delta\omega\left(\frac{c}{v\eta}-1\right)\tilde{\mathscr{E}}-\frac{c\varkappa^2}{2\eta k}\tilde{\mathscr{E}}=-\frac{2\pi\omega N}{\eta^2}\tilde P_1.\end{aligned}\right\}\tag{5}$$

It is now convenient to adopt a coordinate system moving together with a pulse because all the coefficients in the above equations depend on the variable $\tau=(t-x/v)/t_p$. The dependence on the longitudinal coordinate x will be sought in the form $\exp(\Gamma x)$, where Γ is the growth rate of transverse perturbations. Differentiating Eq. (5) with respect to τ and using Eq. (4), we readily obtain the following system of equations in terms of dimensionless variables:

$$\left.\begin{aligned}&\frac{d^2V}{d\tau^2}+\left(-1+\frac{2}{\operatorname{ch}^2\tau}\right)V=\Delta\omega t_p(\gamma U+g^2V)+\gamma\frac{dV}{d\tau}-g^2\frac{dU}{d\tau},\\&\frac{d^2U}{d\tau^2}+\left(-1+\frac{6}{\operatorname{ch}^2\tau}\right)U=\Delta\omega t_p(g^2U-\gamma V)\\&+\gamma\frac{dU}{d\tau}+g^2\frac{dV}{d\tau}+\frac{4\gamma}{\operatorname{ch}\tau}\int_{-\infty}^{\tau}\frac{U}{\operatorname{ch}(\tau_1)}d\tau_1+\frac{4g^2}{\operatorname{ch}\tau}\int_{-\infty}^{\tau}\frac{V}{\operatorname{ch}\tau_1}d\tau_1.\end{aligned}\right\}\tag{6}$$

Here, we have adopted the notation

$$V=\mu t_p\tilde\varphi\mathscr{E}_0/\hbar,\quad U=\mu t_p\tilde{\mathscr{E}}/\hbar,$$
$$g^2=\frac{\varkappa^2t_p}{2k}\frac{v}{1-v\eta/c},\quad\gamma=\Gamma t_p\frac{v}{1-v\eta/c}.$$

Our task is to determine the eigenvalue spectrum $\gamma(g)$ of the system (6) and the eigenfunctions satisfying zero boundary conditions in the limit $\tau\to\pm\infty$. For $g=0$, we have $\operatorname{Re}\gamma\leq0$. The doubly degenerate eigenvalue $\gamma=0$ corresponds to the eigenfunctions

$$V_0=\frac{1}{\operatorname{ch}\tau},\quad U_0=0\text{ and }V_0=0,\quad U_0=\frac{d}{d\tau}\left(\frac{1}{\operatorname{ch}\tau}\right),$$

which in turn correspond to a small initial displacement of the pulse envelope and a small phase shift of the light pulse.

We shall find the dispersion relationship $\gamma(g)$ employing perturbation theory. For $g^2\ll1$ the eigenvalue is $|\gamma|\ll1$ but we shall show later that $|\gamma|\gg g^2$ and, therefore, to find the relationship between γ and g we have to calculate the eigenfunctions of the system (6) in the first order of the largest parameter γ. The eigenfunctions are then

$$\left.\begin{aligned}&V=\frac{c_1}{\operatorname{ch}\tau}+\frac{\gamma}{2}\frac{\tau}{\operatorname{ch}\tau}(c_1+\Delta\omega t_pc_2)+V_2,\\&U=c_2\frac{d}{d\tau}\left(\frac{1}{\operatorname{ch}\tau}\right)+\frac{\gamma}{2}\left[c_2\frac{d}{d\tau}\left(\frac{\tau}{\operatorname{ch}\tau}\right)-2\Delta\omega tc_1y(\tau)\right]+U_2.\end{aligned}\right\}\tag{7}$$

Here, c_1 and c_2 are the constants related by an expression which we shall find later; V_2 and U_2 are terms of the second order of smallness in respect of γ; $y(\tau)$ is the solution of an inhomogeneous differential equation

$$\frac{d^2y}{d\tau^2}+\left(-1+\frac{6}{\operatorname{ch}^2\tau}\right)y=\frac{1}{\operatorname{ch}\tau};$$

this $y(\tau)$ is an even function which decreases monotonically in the limit $\tau\to\pm\infty$.

The orthogonality of the right-hand sides of the equations in the system (6) to the solutions of the unperturbed equations (with $\gamma=g=0$) is the condition of solubility of the system (6) because the left-hand sides contain only the Hermitian operators. Multiplying the first equation by $1/\cosh\tau$ and the second by $\tanh\tau/\cosh\tau$ and then integrating with respect to τ, we obtain the following linear system of algebraic equations:

$$\left.\begin{aligned}&c_1[{}^1\!/_2(\Delta\omega t_p)^2\gamma^2-2\Delta\omega t_pg^2-{}^1\!/_2\gamma^2]-c_2[\Delta\omega t_p\gamma^2+{}^2\!/_3g^2]=0,\\&c_1[\Delta\omega t_p\gamma^2-2g^2]+c_2[{}^2\!/_3g^2\Delta\omega t_p+{}^1\!/_2(\Delta\omega t_p)^2\gamma^2-{}^1\!/_2\gamma^2]=0.\end{aligned}\right\}\tag{8}$$

The condition of existence of nonzero solutions of the system (8) gives the following dispersion relationship:

$$\gamma^2=\frac{4}{3}\frac{\Delta\omega t_p\pm[(2\Delta\omega t_p)^2+3]^{1/2}}{1+(\Delta\omega t_p)^2}.\tag{9}$$

Thus, the propagation of a 2π pulse is unstable against transverse perturbations for any detuning from resonance. The unstable perturbations correspond to the plus sign in front of the root in Eq. (9). For $|\Delta\omega t_p| \ll 1$ the results obtained above reduce to those found earlier.[5]

We shall now consider the case of large detuning: $|\Delta\omega t_p| \gg 1$. We then have to distinguish two possibilities: $\Delta\omega > 0$ and $\Delta\omega < 0$. If $\Delta\omega > 0$, the growth rate of the instability is given by

$$\Gamma = \frac{\varkappa}{(\Delta\omega t_p)^{1/2}} \frac{(1-v\eta/c)^{1/2}}{(kvt_p)^{1/2}}. \tag{10}$$

The envelope of the field and its phase are then

$$\left.\begin{aligned} \mathscr{E} &= \mathscr{E}_0\left(t-\frac{x}{v}\right) + \alpha\frac{\Gamma v t_p}{1-v\eta/c}\frac{\hbar}{\mu t_p}\Delta\omega t_p y\left(t-\frac{x}{v}\right)\cos\varkappa\mathbf{r}_\perp e^{\Gamma x}, \\ \varphi &= -\alpha\cos\varkappa\mathbf{r}_\perp e^{\Gamma x}. \end{aligned}\right\} \tag{11}$$

Hence, we can see that if $\Delta\omega t_p \gg 1$, transverse perturbations result in small-scale self-focusing of the light pulse (constrictions develop). We shall now assume that $\Delta\omega < 0$. Then, the propagation of coherent light pulses is once again unstable against transverse perturbations. Perturbations are now snake-like:

$$\left.\begin{aligned} \mathscr{E} &= \mathscr{E}_0\left(t-\frac{x}{v}\right) + 2\Delta\omega t_p\frac{d\mathscr{E}_0(t-x/v)}{dt} t_p\cos\varkappa\mathbf{r}_\perp e^{\Gamma x}\alpha, \\ \varphi &= \alpha\frac{\Gamma v t_p}{1-v\eta/c}\frac{(t-x/v)}{t_p}(\Delta\omega t_p)^2\cos\varkappa\mathbf{r}_\perp e^{\Gamma x}. \end{aligned}\right\} \tag{12}$$

The growth rate of such an instability is given by

$$\Gamma = \frac{1}{\sqrt{3}}\frac{\varkappa}{|\Delta\omega t_p|^{1/2}}\frac{(1-v\eta/c)^{1/2}}{(kvt_p)^{1/2}}. \tag{13}$$

The terms snake and constriction were applied to transverse perturbations of pulses by Zakharov and Rubenchik[12] in discussing instabilities of solitons in dispersive media. The range of validity of the results obtained is governed by the range of validity of perturbation theory in respect of g^2 in the system (6): $g^2|\Delta\omega t_p| \ll 1$. It is worth considering the stability of propagation of a coherent light pulse against transverse perturbations of smaller scale such that $g^2|\Delta\omega t_p| \gg 1$.

Introducing a large parameter $g^2\Delta\omega t_p$ and lowering the order of equations in the system (6), we can show—as in Ref. 5—that the eigenvalue is given by

$$\gamma(g) = \pm ig^2 + \gamma_1,$$

where

$$\operatorname{Im}\gamma_1 = \frac{A}{\Delta\omega t_p}, \quad \operatorname{Re}\gamma_1 = \frac{B}{(\Delta\omega t_p)^2 g^2};$$

A and B are numbers of the order of unity. Thus, the growth rate of transverse perturbations increases monotonically in the range $|g^2\Delta\omega t_p| < 1$ and then falls for perturbations of smaller scale $|g^2\Delta\omega t_p| > 1$. Consequently, the maximum growth rate corresponds to the limit of validity of perturbation theory where $g^2|\Delta\omega t_p| \sim 1$.

3. We shall now consider the results obtained. If $\Delta\omega = 0$, instability of coherent propagation of light pulses against transverse perturbations is due to the appearance of diffracted oblique waves which interfere with the field of the main wave and cause (because of the dependence of the velocity of a 2π pulse on the field amplitude) transverse perturbations to grow. If $|\Delta\omega t_p| \gg 1$, it follows from the adiabatic approximation theory[13,14] that the propagation of light can be described by the refractive index of a medium (if $\mu\mathscr{E}_0/\hbar \ll |\Delta\omega|$) and then the field broadening of a two-level transition results in a nonlinear dependence of the refractive index on the field intensity. According to the theory of Bespalov and Telanov,[15] this nonlinear refraction results for $\Delta\omega > 0$ in small-scale self-focusing of light. It should be noted that for $\Delta\omega t_p \gg 1$ Eq. (10) is identical with the response obtained by Zakharov and Rubenchik[12] if the nonlinear dependence of the refractive index is deduced from the adiabatic approximation.[13,14]

The condition of existence of small-scale self-focusing—which results in a considerable excess of the light power density above a critical value for a nonresonant 2π pulse—is equivalent to the inequality

$$F \gg (\Delta\omega t_p)^{1/2}(1-v\eta/c)^{-1}. \tag{14}$$

Consequently, if $|\Delta\omega t_p| \gg 1$ ($\Delta\omega > 0$), the diffraction instability changes to self-focusing described by the theory of Bespalov and Talanov. In this case the refraction of rays and the dependence of the refractive index on the intensity result in self-focusing of light and diffraction limits the transverse size of light filaments which then form.

An important difference from the theory of Bespalov and Talanov[15] appears in the case of negative detuning of the light pulse frequency from that of a two-level transition. According to Bespalov and Talanov, propagation of homogeneous light in the $\Delta\omega < 0$ case is stable against transverse perturbations. However, it is shown above that in this case the propagation of a coherent light pulse is unstable against snake-like transverse perturbations. In the case of large detuning this instability can be interpreted in a simple physical manner.

Let us consider a part of a pulse which is convex in the propagation direction. If $\Delta\omega < 0$, the refractive index decreases on increase of the intensity and, therefore, the trailing edge of the pulse acts as a converging lens and the leading edge as a diverging lens. The radiation passing through two consecutive positive and negative lenses is transformed in the same way as in a telescopic system and increases in intensity. Similarly, the intensity decreases in a concave part of a pulse. The group velocity of light increases on increase of the intensity and, therefore, the convex parts overtake an unperturbed pulse and concave parts lag behind, thus increasing further the distortions of the pulse shape. The condition for the growth of snake-like small-scale perturbations in the $\Delta\omega < 0$ case is similar to the condition for the existence of small-scale self-focusing given by Eq. (14). It should be pointed out that if $\Delta\omega = 0$, we can expect transverse perturbations which are superpositions of constrictions and snakes. A change from one type of perturbation to another occurs for detuning amounting to $\Delta\omega t_p \sim 1$.

The above analysis applies to the stability of propagation of coherent pulses with a wide radiation spectrum so that $t_p < T_2^*$, where $1/T_2^*$ is the inhomogeneous width of the transition. In the other limiting case when the inhomogeneous broadening of the absorption line is important, $T_2^* < t_p$, we can carry out a similar analysis representing the polarization of the medium as factorized in respect of the frequency:

$$P_2(\Delta\omega)=\chi(\Delta\omega)P_2(0),\quad \chi(\Delta\omega)=\frac{1}{1+(\Delta\omega t_p)^2}.$$

In all the above expressions one then has to replace the detuning parameter $\Delta\omega t_p$ with its effective value $(\Delta\omega t_p)_{eff}=\langle \Delta\omega\chi\rangle/\langle\chi\rangle$, where the averaging is carried out over the profile of an inhomogeneously broadened line. The effective detuning parameter is proportional to $\Delta\omega T_2^*$ and the coefficient of proportionality depends on the actual line profile and on $\Delta\omega$. Detuning of the frequency of light with a narrow spectrum from the center of an inhomogeneously broadened absorption line has the greatest influence on the diffraction instability for $|\Delta\omega T_2^*|\sim 1$. The asymmetric dependence of the growth rate of transverse perturbations on the sign of detuning [see Eq. (9)] is in agreement with the experimental results on the influence of detuning of the light frequency on the passage of coherent pulses through a resonantly absorbing medium[11] and on the transverse structure of the transmitted radiation.[6,7]

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Light generation by a moving active medium

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The influence of motion of the active medium between the mirrors of an open resonator on the interaction of the generated radiation with its ensuing periodic structure of the inverted population is investigated theoretically. It is shown that the distributed feedback of the opposing light waves decreases when the active medium moves; this leads to establishment of single-frequency stationary generation at velocities exceeding the calculated critical value (which agrees well with experiment).

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An electromagnetic analysis of a resonator filled with a moving medium[1] is carried out here for the purpose of studying the progagation and generation of light in a dispersive active medium that moves uniformly along the optical axis of an open resonator. The problem is both of independent interest and serves to reveal the role of the spatial-periodic structure which is produced in the active medium because of the inhomogeneous saturation of the inverted population,[2,3] since motion of the medium is one method of eliminating structure effects.[4]

It will be shown that the distributed feedback that is self-induced in the active medium decreases rapidly with increasing velocity of the active medium, both because of the smoothing of the periodic structure of the inverted population, and because it lags in phase the generating standing light wave. As a result, at sufficiently high velocities exceeding a certain critical value, stable stationary generation is produced in the medium and has been observed in a number of experiments.[4,5] The model considered here does not take into account the modulation that can occur in the generated radiation when the dielectric boundaries move parallel to the resonator mirrors[4-6] and does not occur, for example, when the end faces of the active element are cut at the

