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Instability of coherent propagation of light pulses in resonantly absorbing media

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We show that the standard form of 2π pulses (short and relatively powerful light pulses propagating in resonantly absorbing media without loss) is unstable with respect to transverse perturbations. A transverse structure develops when the pulse traverses a distance of the order of its length L in the medium. The characteristic scale length of the transverse structure arising is $\sim(\lambda L)^{1/2}$, where λ is the wavelength of the light.

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1. McCall and Hahn[1] showed experimentally and theoretically that a short light pulse with a sufficiently large energy can propagate in a resonantly absorbing medium without loss and retaining its shape (2π pulse). Subsequently this effect has been widely studied both theoretically and experimentally (see, for instance, the surveys by Poluéktov *et al.*[2]). Theoretically one usually considers the propagation of one-dimensional pulses which extend to infinity in the transverse direction. The evolution of the transverse structure of a 2π pulse connected with its transverse dimension being finite has been studied numerically (see, e.g.,[2–4]). In particular it was noted in[3,4] that a 2π pulse has a tendency for self-focusing. The problem of the evolution of a three-dimensional coherent light pulse is complicated for a complete analytical study. However, one can obtain a number of important conclusions by studying the stability of a one-dimensional 2π pulse with respect to transverse perturbations. The present paper is devoted to the solution of that problem.

2. To simplify the exposition we restrict ourselves to the two-level model of a medium without degeneracy, neglecting inhomogeneous broadening. In that case the reduced equations for the field E, the polarization P of the particles in the medium, and the difference n in the populations of the lower and upper levels have the form

$$\frac{\partial E_1}{\partial x}+\frac{\eta}{c}\frac{\partial E_1}{\partial t}+\frac{1}{2k}\nabla_\perp^2 E_2=-\frac{2\pi N\omega}{c\eta}P_1,$$
$$\frac{\partial E_2}{\partial x}+\frac{\eta}{c}\frac{\partial E_2}{\partial t}-\frac{1}{2k}\nabla_\perp^2 E_1=\frac{2\pi N\omega}{c\eta}P_2, \tag{1a}$$

$$\frac{\partial P_1}{\partial t}=\frac{\mu^2}{\hbar}E_1 n,\qquad \frac{\partial P_2}{\partial t}=-\frac{\mu^2}{\hbar}E_2 n,\qquad \frac{\partial n}{\partial t}=-\frac{1}{\hbar}(E_1P_1-E_2P_2). \tag{1b}$$

Here

$$E=E_1\cos(\omega t-kx)+E_2\sin(\omega t-kx),$$
$$P=P_1\sin(\omega t-kx)+P_2\cos(\omega t-kx),$$

η is the non-resonance refractive index, N the number of resonant particles, and μ the transition dipole moment. We assume that the light frequency is the same as the resonance frequency of the medium. Equations (1) have a well known stationary solution[1] in the form of a one-dimensional soliton (2π pulse):

$$E_1 = E_0 = \frac{2\hbar}{\mu t_p} \operatorname{sech} \frac{(t - x/v)}{t_p}, \quad E_2 = 0, \tag{2}$$

where t_p is the length of the pulse and v its velocity given by the expression

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

The high-frequency loading ($\propto \cos(\omega t - kx)$) describes photons which are emitted at the trailing front of the pulse and which propagate with a velocity $\omega/k = c/\eta > v$ in the medium, overtaking the pulse and being absorbed at the leading front. We note that as the field amplitude E_0 increases, the length of the pulse (2) decreases and, hence, its velocity increases according to (3).

Let us first of all study qualitatively what occurs when a one-dimensional 2π pulse is perturbed transversely. Let the surface of constant phase in the trailing part of the 2π pulse be modulated in the transverse direction with a period $l_\perp = 2\pi/\varkappa$. In that case, as in the case when a plane wave passes through a phase diffraction lattice, apart from the main wave there appear two oblique diffraction waves at an angle $\varkappa/k$ to the direction of propagation of the 2π pulse. The interference of these waves with the main wave leads to a modulation of the amplitude of the field with the same period $l_\perp$ in the transverse direction. We noted above that this means that parts where the field becomes larger overtake parts of the pulse with smaller field values. If this is taken into account the curvature of the wave front is amplified and this leads to a growth in the oblique waves, and so on. In order that the mechanism of the instability described here is realized it is necessary that the inequality $l_\perp > L\varkappa/k$, i.e., $l_\perp > (\lambda L)^{1/2}$ ($L = vt_p$, λ the wavelength of the radiation) is satisfied. In the opposite case the "oblique" photons emitted at the trailing front go through several maxima and minima of the field amplitude before being absorbed at the leading front and the feedback through the effect of the amplitude on the pulse speed will be made difficult.

There may arise a problem about the connection between the mechanism of the instability described above and the mechanism for the self-focussing of intense radiation in non-linear media, first suggested by Askaryan.[5] The well known self-focussing effect is caused by the non-linear refraction of the radiation when the radiation is confined to regions with a large refractive index. This occurs in media in which the refractive index increases with increasing light intensity. Self-focussing (a purely geometrical optics effect) is stabilized by the diffraction of the light when the radiation is constricted to sufficiently small transverse dimensions. In the case of the coherent interaction of the light with the medium considered in the present paper the situation is exactly the opposite. The effective refractive index (the ratio of the light velocity to the propagation velocity of the pulse) decreases with increasing intensity and diffraction causes instability. This is connected with the specific coherent interaction. Indeed, for a qualitative explanation of the instability of 2π pulses we used an analogy with a phase diffraction lattice. We emphasize that to first order in the phase perturbation the light intensity after passing through the phase lattice is unchanged. The specific feature of the coherent interaction consists in that the medium affects the field amplitude and not its intensity. The non-uniformity of the amplitude in the transverse direction which arises in first order also leads to instability. In its outward appearance—the development into filaments—this instability reminds us of the self-focussing instability of solitons in dispersive media[6] or of plane waves in non-linear media (the first results in that field are due to Bespalov and Talanov[7]).

3. We proceed to an exact analysis of the stability of the set (1) with respect to transverse perturbations, choosing them to be proportional to $\cos(\boldsymbol{\varkappa} \cdot \mathbf{r}_\perp)$. Linearizing the material Eqs. (1b) near the stationary solution (2) we have

$$\frac{\partial \tilde{P}_1}{\partial t} = \frac{\mu^2}{\hbar}(E_0 \tilde{n} + \tilde{E}_1 n_0), \quad \frac{\partial \tilde{P}_2}{\partial t} = -\frac{\mu^2}{\hbar} \tilde{E}_2 n_0,$$
$$\partial \tilde{n}/\partial t = -(E_0 \tilde{P}_1 + \tilde{E}_1 P_0)/\hbar. \tag{4}$$

Here $P_0 = \mu^2 t_p^2 \hbar^{-1} \partial E_0/\partial t$, $n_0 = t_p^2 E_0^{-1} \partial^2 E_0/\partial t^2$, E_0 is given by Eq. (2), and the tilde indicates the perturbations of the quantities P_1, P_2, E_1, E_2, and n. The equations for the field have the form

$$\begin{aligned} \frac{\partial \tilde{E}_1}{\partial x} + \frac{\eta}{c}\frac{\partial \tilde{E}_1}{\partial t} - \frac{\varkappa^2}{2k}\tilde{E}_2 &= -\frac{2\pi N\omega}{c\eta}\tilde{P}_1, \\ \frac{\partial \tilde{E}_2}{\partial x} + \frac{\eta}{c}\frac{\partial \tilde{E}_2}{\partial t} + \frac{\varkappa^2}{2k}\tilde{E}_1 &= \frac{2\pi N\omega}{c\eta}\tilde{P}_2. \end{aligned} \tag{5}$$

Here $\varkappa = |\boldsymbol{\varkappa}|$. For what follows it is useful to change to the "proper" time of the pulse, i.e., to a system of coordinates moving with the 2π pulse as all coefficients in Eqs. (4), (5) depend only on the quantity $\tau = (t - x/v)/t_p$. Equations (4), (5) take a simpler form in the dimensionless variables:

$$\xi = \frac{x}{vt_p}\left(1 - \frac{\eta v}{c}\right), \quad E_0 = \frac{\hbar}{\mu t_p}\varepsilon_0, \quad \tilde{E}_{1,2} = \frac{\hbar}{\mu t_p}\varepsilon_{1,2},$$

$P_0 = \mu \pi_0$, $\tilde{P}_{1,2} = \mu \pi_{1,2}$, viz.:

$$\begin{aligned} &\frac{\partial \varepsilon_1}{\partial \tau} - \frac{\partial \varepsilon_1}{\partial \xi} + q^2 \varepsilon_2 = \pi_1, \quad \frac{\partial \varepsilon_2}{\partial \tau} - \frac{\partial \varepsilon_2}{\partial \xi} - q^2 \varepsilon_1 = -\pi_2, \\ &\frac{\partial \pi_1}{\partial \tau} = \varepsilon_0 \tilde{n} + \varepsilon_1 n_0, \quad \frac{\partial \pi_2}{\partial \tau} = -\varepsilon_2 n_0, \quad \frac{\partial \tilde{n}}{\partial \tau} = -\varepsilon_0 \pi_1 - \varepsilon_1 \pi_0. \end{aligned} \tag{6}$$

Here

$$q^2 = \frac{\varkappa^2}{2k} vt_p \left(1 - \frac{\eta v}{c}\right)^{-1}, \quad n_0 = 1 - \frac{2}{\operatorname{ch}^2 \tau}.$$

As the coefficients in the set of Eqs. (6) is independent of ξ its solutions are proportional to $e^{\gamma\xi}$. It is possible by using the properties of the stationary solution to reduce the set of five ordinary equations to a set of two ordinary second-order differential equations for the quantities

$$u = e^{-\gamma\xi} \int_{-\infty}^{\tau} \varepsilon_1 \, d\tau, \quad w = e^{-\gamma\xi} \varepsilon_2,$$

namely:

$$\frac{d^2u}{d\tau^2}-n_0(\tau)u-\gamma\frac{du}{d\tau}=-q^2w,$$
$$\frac{d^2w}{d\tau^2}-n_0(\tau)w-\gamma\frac{dw}{d\tau}=q^2\frac{d^2u}{d\tau^2}. \quad (7)$$

The standard substitution $u=\varphi e^{\gamma\tau/2}$, $w=\psi e^{\gamma\tau/2}$ leads to the set of equations

$$\frac{d^2\varphi}{d\tau^2}-n_0(\tau)\varphi-\frac{\gamma^2}{4}\varphi=-q^2\psi,$$
$$\frac{d^2\psi}{d\tau^2}-n_0(\tau)\psi-\frac{\gamma^2}{4}\psi=q^2e^{-\gamma\tau/2}\frac{d^2}{d\tau^2}(e^{\gamma\tau/2}\varphi). \quad (8)$$

which differs from (7) in that the left-hand side of both equations is a self-adjoint operator. Our problems consists in determining the eigenvalue spectrum $\gamma(q)$ (dispersion curves) and the corresponding eigenfunctions satisfying the vanishing boundary conditions as $\tau\to\pm\infty$. We note that the set (8) for $q=0$ splits into two identical Schrödinger equations with a potential $n_0=1-2\,\mathrm{sech}^2\tau$. The eigenvalues of the energy in such a potential $-\gamma^2/4\geq 0$, [8] which corresponds to neutral stability of the 2π pulse with respect to longitudinal perturbations. The eigenfunctions corresponding to $\gamma=0$, $\varphi_0=\psi_0=\mathrm{sech}\tau$ (i.e., $\tilde{E}_1\propto dE_0/d\tau$, $\tilde{E}_2\propto E_0$) correspond to small shifts in the initial position of the envelope and a small phase shift of the high-frequency loading of the 2π pulse.

The fact just noted enables us to use perturbation theory to find $\gamma(q)$ when $\gamma\ll 1$ (or in terms of quantities with dimensions $l_\perp=2\pi/\varkappa\gg(\lambda L)^{1/2}$). The unperturbed state is two-fold degenerate ($\varphi=\varphi_0$, $\psi=0$ and $\varphi=0$, $\psi=\psi_0$). The solution of the corresponding secular equation has the form

$$\gamma^4=16q^4\int_{-\infty}^{\infty}\left(\frac{d\varepsilon_0}{d\tau}\right)^2d\tau\Big/\int_{-\infty}^{\infty}\varepsilon_0^2\,d\tau$$

or

$$\gamma=2q\sqrt[4]{1/3}. \quad (9)$$

We thus found that the 2π pulse is unstable with respect to transverse perturbations. The growth rate is then the larger the smaller the wavelength of the perturbation $l_\perp$. Changing to variables with dimensions we get ($\tilde{E}_{1,2}$, $\tilde{P}_{1,2}\propto e^{\Gamma x}$)

$$\Gamma=\left(\frac{4}{3}\right)^{1/4}\varkappa\left(\frac{1-\eta v/c}{kvt_p}\right)^{1/2}=\left[\frac{4}{3}\left(1-\frac{\eta v}{c}\right)^2\right]^{1/4}\varkappa\left(\frac{\lambda L}{2\pi}\right)^{1/2}\frac{1}{L}. \quad (10)$$

The field then has the form

$$E=E_0(t-x/v+\alpha t_pe^{\Gamma x}\cos(\varkappa\mathbf{r}_\perp))\cos(\omega t-kx)$$
$$+E_0(t-x/v)(1/3)^{1/4}\alpha e^{\Gamma x}\cos(\varkappa\mathbf{r}_\perp)\sin(\omega t-kx); \quad (11)$$

here $\alpha\ll 1$ is the initial amplitude of the perturbation.

It follows from (10) that when $\varkappa\sim(\lambda L)^{-1/2}$ the transverse structure of the 2π pulse develops over distances of the order of its length in the medium $L=vt_p$. The growth rate of the transverse instability reaches its maximum value at the boundary of the region of applicability of perturbation theory, $q\sim 1$. The problem arises of the stability of transverse modes with finer scale lengths. We note that Eqs. (1a) and (5) are applicable for $\varkappa\ll k$, i.e., $q^2\ll kL$. As $kL\gg 1$, the equations studied are applicable also when $q\gg 1$. It is more convenient for a study of the function $\gamma(q)$ to use in that case Eqs. (7). From the asymptotic behavior of their solutions as $\tau\to\pm\infty$ it follows that when $q\gg 1$ the eigenvalue γ can be written in the form $\gamma=iq^2+\gamma_1$ where γ_1 is a number of order of magnitude unity. In the limit of large wavenumbers of the perturbations we can reduce the order of the set of Eqs. (7) by splitting off one of the solutions corresponding to the fast oscillations of u and w. This can be done formally by isolating in (7) the operator

$$\hat{A}=\frac{d^2}{d\tau^2}-n_0(\tau)-\gamma_1\frac{d}{d\tau}$$

and reducing the set of the two Eqs. (7) to a single equation

$$\left(\frac{\hat{A}^2}{q^2}+i\hat{A}\frac{d}{d\tau}+i\frac{d}{d\tau}\hat{A}\right)u=0. \quad (12)$$

In the limit as $q\to\infty$ Eq. (12) reduces to an equation without parameters:

$$\left(\hat{A}\frac{d}{d\tau}+\frac{d}{d\tau}\hat{A}\right)u=0. \quad (13)$$

It was not possible to find the eigenvalues γ_1 of Eq. (13), but one can show that $\mathrm{Re}\,\gamma_1=0$. To prove this we multiply (13) by u^* and add it to the complex conjugate expression and integrate the equation obtained over the infinite range of values. After simple transformations we get

$$(\mathrm{Re}\,\gamma_1)\int_{-\infty}^{\infty}|u'|^2\,d\tau=0. \quad (14)$$

As $u\neq 0$ because of the boundary conditions, a nontrivial solution of (13) corresponds to an eigenvalue with $\mathrm{Re}\,\gamma_1=0$. We have thereby shown that $\mathrm{Re}\,\gamma_1\to 0$ as $q\to\infty$. As in the initial problem (7) there are no other parameters than q, it is clear that $\mathrm{Re}\,\gamma$ is maximal when $q\sim 1$ and then we have as to order of magnitude $\mathrm{Re}\,\gamma\sim 1$ ($\mathrm{Re}\,\Gamma\approx L^{-1}$).

The results obtained agree with the qualitative considerations given by us at the beginning of the paper.

4. We have thus shown that the transverse structure of a 2π pulse develops roughly during the same time as the one over which the 2π pulse is itself formed. The change in the transverse structure can then also be large provided a sufficiently wide pulse of radius $l_\perp\gg(\lambda L)^{1/2}$ is incident upon the medium. A study of the non-linear stage of the evolution of the instability considered is of great interest. Here, the problem remains unsolved of the existence of a stable three-dimensional 2π pulse which, as one may expect, has a radius $\sim(\lambda L)^{1/2}$. At the present moment this problem is studied numerically.

We note in conclusion that taking the inhomogeneous broadening into account can be done trivially in the linear theory of the stability of a 2π pulse; the answer is also given by Eq. (10).

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Note added in proof (*March* 31, 1977). Recently a paper by Gibbs *et al*. has been published (Phys. Rev. Lett. 37, 1743 (1976)) in which the observation of the instability predicted by us was reported for the case where a resonant light pulse traversed a cell with sodium vapor.

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Undisplaced resonant scattering line of a strong quasimonochromatic field

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The change in the shape of a narrow undisplaced resonant-scattering line of a strong quasimonochromatic field is investigated. At low field intensity it is known that the line duplicates the spectrum of the exciting radiation. The change in the shape and width of a narrow undisplaced line depends essentially on the statistical field of the exciting-radiation field. The dependence of the line width on the field intensity is investigated for typical radiation statistics. The physical phenomenon in question can, in particularly, serve as the basis of an investigation of the statistical properties of an electromagnetic field.

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1. INTRODUCTION

In view of the progress made in the development of tunable lasers, interest has greatly increased of late in investigation of resonant scattering (resonant fluorescence) of a strong laser field. In the last few years many theoretical papers[1–12] 1) have been devoted to the analysis of the spectrum of resonant scattering of intense laser radiation. The results of the first experimental investigations in this direction have also been published.[13–19] The physical concepts developed in[1–12] are based on simplified models, and it is therefore obvious that many more advances will be made in the nearest future in the study of the spectrum of resonant scattering, both theoretical and experimental.

The first theoretical analysis of the spectrum of the resonant scattering of a strong monochromatic field (analysis of the variation of the spectral density of spontaneous emission in the presence of a strong field) was carried out by Rautian and Sobel'man.[18] They have shown, in particular, that the resonant-scattering line splits into three components, corresponding to the splitting of the atomic levels into quasi-energy levels under the influence of a strong field.[19] The analysis in[18] pertained to scattering by atoms in excited states. In the "classical" formulation of the resonant-scattering problem one of the combining states is assumed to be the ground state.[20] Of course, in view of the fundamental character of the physical cause (level splitting), the multiplet structure of the spectrum (three components) and the position of the components on the frequency scale are valid also for the classical formulation. However, the participation of the ground state leads to a redistribution of the intensity between the components and to the development of a more complicated structure within the individual components, this being connected with the singularities of the relaxation processes under the given conditions. The problem of resonant scattering in which the ground state participates is therefore of independent interest.

The question of the spectrum of resonant scattering of a strong field in the classical formulation (the only one dealt with henceforth) was first raised by

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