NEAR RESONANCE RADIATION ATTENUATION IN A WEAKLY NONLINEAR GASEOUS DIELECTRIC

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A spectral approach to the three-dimensional problem of nonlinear phenomena which are associated with the radiation intensity is proposed. The approach is based on Maxwell's equations and the general expression for the polarization of the medium. The frequency-dependent component of the polarization is represented as a power series in the spectral components of the field intensity; the solution of the nonlinear problem is expressed in terms of the solution of the corresponding linear problem. The problem is reduced to a second-order nonlinear differential equation, whose solution is found. The problems associated with using the solutions to boundary-value problems are discussed. The main attention is devoted to the propagation of radiation near the spectral lines of a molecular atmosphere. The discussion is limited to media with cubic nonlinearity.

1. INTRODUCTION

In this paper the spectral approach to the problem of propagation of high-power radiation near the spectral lines of natural media is discussed. The difference from regimes of linear interaction appears owing to the nonlinear response of the medium to the light and has not been adequately studied, even for monochromatic radiation. At the same time the propagation of light pulses near the spectral lines of a medium is of practical interest. In this region certain peculiarities appear owing to the finite spectral composition of the incident radiation and the power of the pulses. In particular, the distortion produced in the pulse spectrum (shape) by the nonlinearity of the complex index of refraction of a molecular medium near spectral lines is of interest. The real and imaginary parts of the nonlinear dielectric constant of a medium are responsible for different nonlinear processes.[1] It is well known, for example, that the dependence on Reε (ε is the complex dielectric constant) significantly affects the evolution of the pulse shape. Here the behavior of the pulse shape as a function of the sign of the detuning near resonance is of special interest; this difference can be very significant.[2,3] In the works cited the method of amplitude envelopes was employed. This method is generally accepted and is very fruitful in nonlinear optics.

In addition to describing the field with the help of envelopes, there exists a different approach to the solution of such problems. This is the spectral approach. In what follows the spectral approach is used to give a spectral description of self-action; it hardly has any explicit advantages over the envelope method, but in some problems the spectral description of self-action is more convenient. For example, it is more convenient for solving problems of radiation transfer in nonlinear media or for problems in nonlinear diffraction.[4] In addition, the method makes it possible to obtain a solution in a final form from Maxwell's equations. We understand the so-called spectral method as follows.

Any physically realizable field can be expanded in a Fourier integral. The nonlinear field, prescribed at a time t at the point r (and produced by the nonlinear response of the medium) can be expressed as

$$E(r,\ t,\ \xi) = \int E(\omega,\ r,\ \xi)e^{-i\omega t}d\omega, \tag{1}$$

where $E(r,\ \omega,\ \xi)$ is the frequency component of the nonlinear field and ξ is the nonlinearity parameter. The condition that the nonlinear field must reduce to the linear field means that

$$E(\omega,\ r,\ \xi = 0) = \mathbf{E}(\omega,\ r), \tag{2}$$

where $\mathbf{E}(\omega,\ r)$ is the frequency component of the linear field.

If the representation

$$E(\omega,\ r,\ \xi) = \hat{\rho}(\mathbf{E}(\omega,\ r),\ \xi)\mathbf{E}(\omega,\ r), \tag{3}$$

is assumed, then the solution of the problem (3) can be employed in Eq. (1). In Eq. (3) we write $\hat{\rho}$, but it will become clear below that in simple cases this is simply a function ρ of $[\mathbf{E}(\omega,\ r)]$. In the case of an isotropic medium and linearly polarized radiation

$$\hat{\rho} \rightarrow \rho\big(|\mathbf{E}(\omega,\ r)|\big). \tag{4}$$

In what follows we shall confine our attention to this simple case.

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In the spectral approach it is assumed that' the solution of the linear problem is known. The peculiarity, however, lies in the fact that if a detailed solution of the linear problem is not available, then the qualitative behavior of the nonlinear solution can be judged only based on some general relations, which depend on the character of the nonlinear corrections to the linear problem. In addition, we are often interested precisely in the changes in the spectrum, so that it is not necessary to solve Eq. (1), i.e., to invert an integral.

2. POLARIZATION

We shall investigate the possibility of constructing the representation (3) from the time-dependent Maxwell's equations. In Maxwell's equations the expression for the nonlinear polarization can be represented in the form[5]

$$P(t) = \int_{-\infty}^{\infty}\kappa(t_1)E(t - t_1)dt_1 + \iiint_{-\infty}^{\infty}\kappa(t_1, t_2, t_3)E(t - t_1)E(t - t_2) \times E(t - t_3)dt_1dt_2dt_3 + \iint_{-\infty}^{\infty}\ldots\int\kappa(t_1, t_2,\ldots t_5) \times E(t - t_1)\ldots E(t - t_5)dt_1\ldots dt_5 +\ldots \quad (5)$$

Here all quantities are real and E is the nonlinear field. The field is linearly polarized. It is assumed that the light pulse is narrow: $\Delta\omega \ll \omega_0$, where $\Delta\omega$ is the spectral width of the pulse and ω_0 is the average frequency of the pulse. We are interested in the interactions associated with the intensity of the radiation at the frequency of the incident field (self-action). The medium is isotropic (a gas or liquid). The even terms in the expansion (5) vanish. The linear polarization of the field makes it possible to rewrite Eq. (5) in the following form:

$$P(t) = \int dt_1E(t - t_1)\Big\{\kappa(t_1) + \iint dt_2dt_3E(t - t_2) \times E(t - t_3)\Big[\kappa(t_1, t_2, t_3) + \iint dt_4dt_5E(t - t_4) \times E(t - t_5)\big[\kappa(t_1\ldots t_5) + \ldots\big]\Big]\Big\}. \quad (6)$$

We shall transfer in Eq. (6) to frequency components:

$$P(t) = \int d\omega P(\omega)e^{-i\omega t} = \int dt_1E(t - t_1)\{\kappa(t_1) + \ldots\} = \int d\omega E(\omega)e^{-i\omega t}\int dt_1K(t, t_1)e^{i\omega t_1}. \quad (7)$$

Here the expression in braces of Eq. (6) is denoted by $K(t, t_1)$. We further assume that the integral in Eq. (7)

$$\int dt_1K(t, t_1)\, e^{i\omega t_1} \equiv A_\omega = A_0 + A_1 + A_2 + \ldots$$

does not depend on t and must be a function only of the frequency ω not only in the linear approximation but also in the general nonlinear case:

$$A_\omega = A\big(\omega, |E_\omega|^2\big). \quad (8)$$

This assumption simplifies the calculation of the integrals in the expression (7). In the zeroth (linear) approximation we have

$$A_0 = \int dt_1\kappa(t_1)e^{i\omega t_1} = \alpha_0(\omega).$$

Consider the next term:

$$A_1 = \int_{-\infty}^{\infty}dt_1e^{i\omega t_1}\iint dt_2dt_3E(t - t_2) \times E(t - t_3)\kappa(t_1, t_2, t_3). \quad (*)$$

By assumption this expression must be a function only of the frequency (and it should not contain oscillating functions of the type $\exp(\pm i\omega t)$). We have

$$A_1 = \int d\omega'\int d\omega''\kappa(\omega, \omega', \omega'')e^{-i(\omega'+\omega'')t},$$

where

$$\kappa(\omega, \omega', \omega'') = \int dt_1dt_2dt_3\kappa(t_1, t_2, t_3) \times \exp(i\omega t_1 + i\omega' t_2 + i\omega'' t_3).$$

Using further the condition $\omega' + \omega'' = 0$, where ω' and ω'' differ from u by not more than the width $\Delta\omega$ of the signal spectrum ($\Delta\omega \ll \omega$), we can write approximately

$$A_1 \approx E_\omega E_{-\omega}\int d(\Delta\omega')\int d(\Delta\omega'')\kappa(\omega, \omega + \Delta\omega', -\omega - \Delta\omega'') = |E(\omega)|^2\alpha_1(\omega); \quad (**)$$

In writing the quantity $\alpha_1(\omega)$ the fact that the signs of ω' and ω'' can be interchanged is taken into account. The transition from (*) to (**) is thus made under the assumption that $\Delta\omega \ll \omega - \omega_0$, where ω_0 is the average frequency of the pulse (see Ref. 6, p. 95). This also means that mode interaction is neglected within the spectral width of the pulse; this approximation becomes better as the spectrum of the pulse becomes narrower. The result is exact when the pulse degenerates into a monochromatic field.

Proceeding in an analogous manner with the remaining terms in the series (6), we arrive at the following expression for the polarization:

$$P(t) = \int d\omega E(\omega)A(\omega,\ |E_\omega|^2)e^{-i\omega t},$$

$$P(\omega) = E(\omega)\left\{\alpha_0(\omega) + \alpha_1(\omega)|E_\omega|^2 + \alpha_2(\omega)|E_\omega|^4 + \dots\right\}. \qquad (9)$$

Here α_0, α_1, α_2, ... are function of the frequency; spatial dispersion is neglected. Using the relation between the induction and the polarization $D = E + 4\pi P$, we obtain for the dielectric constant

$$\varepsilon(\omega) = \varepsilon_L(\omega) + \varepsilon_{NL}(\omega) = \varepsilon_L(\omega) + \varepsilon_2(\omega)|E_\omega|^2 + \varepsilon_4(\omega)|E_\omega|^4 + \dots, \qquad (10)$$

where $\varepsilon_L = \varepsilon_0 + i\varepsilon_1$ is the standard (linear) dielectric constant. The specific form of the function α_i(or ε_i) can be found, as usual, from model representations.

3. CONSEQUENCES OF THE ASSUMPTION (3). EQUATION FOR THE FUNCTION ρ

The equations of the field in an isotropic nonmagnetic medium, in which the time dependence of the fields is of the form $e^{-i\omega t}$, satisfy time-independent relations:[7]

$$\mathrm{rot}\mathbf{E} - ik_0\mathbf{H} = 0; \qquad (11)$$

$$\mathrm{rot}\mathbf{H} + ik_0\varepsilon_L(\omega)\mathbf{E} = 0. \qquad (12)$$

where $k_0 = w/c$, and ε_L is the dielectric constant.

Because of linearity (the index L) the equations (11) and (12) are insensitive to the strength of the field. For sufficiently large **E** the measured quantities (quadratic in the field) do not, however, agree with Eqs. (11) and (12). To remedy this situation the response of the medium to the incident field must be taken into account, i.e., the dielectric constant ε_L must be supplemented by terms which depend on the intensity of the field. The corrected equations (11) and (12) will have the form

$$\mathrm{rot}E - ik_0 H = 0; \qquad (13)$$

$$\mathrm{rot}H + ik_0(\varepsilon_L + \varepsilon_{NL})E = 0. \qquad (14)$$

Here in order to distinguish the solutions of Eqs. (11) and (12) from the solutions of Eqs. (13) and (14) different fonts are used for the fields.) In what follows we shall study cubic media. Equation (14) assumes the form

$$\mathrm{rot}H + ik_0\left(\varepsilon_L + \varepsilon_2|E|^2\right)E = 0. \qquad (15)$$

The problem is to determine the relation between the fields E and **E** under the assumption that the solutions of these problems for each of the fields are known. Thus obtaining a relation between E and **E** makes it possible, inprinciple, to write the solution of the nonlinear problem in terms of the solution of the linear problem. We shall write out below the required relations. It is assumed that

$$E = \rho(S)\mathbf{E}, \qquad (16)$$

where $S = |\mathbf{E}_\omega|^2$ and ρ is a complex function. It is significant that this solution makes it possible to assume that the field is locally transverse. This fact makes it possible to identify the direction

$$\mathrm{grad}\rho = \frac{d\rho}{dS}\,\mathrm{grad}S, \quad S = |\mathbf{S}|, \qquad (17)$$

at each point with the direction of the vector S of the linear problem. Here[8]

$$\mathrm{grad}S = -2k_0\kappa\mathbf{S} \qquad (18)$$

$m = n + i\kappa$ is the complex index of refraction; $\sigma = 2k_0\kappa$ is the absorption coefficient: $\varepsilon_L = m^2$ is the dielectric constant

$$\mathbf{H} = m(n \times \mathbf{E}), \quad \mathbf{E} = -\frac{1}{m}(n \times H);$$

and $n = \mathbf{S}/S$ is the unit Poynting vector. Together with the definition of the electric vector (16), we define with the help of Eq. (13) the nonlinear magnetic vector

$$H = \rho\mathbf{H} + \frac{1}{ik_0}[\nabla\rho \times \mathbf{E}] = \left(\rho + i\frac{2\kappa}{m}\rho S\right)\mathbf{H} = R\mathbf{H}. \qquad (19)$$

Here (and below) a prime or a dot denotes a derivative with respect to the argument.

The equation for the function ρ can be derived by a number of methods. One method is the following. From Eqs. (13) and (14) and the definition (16) it follows that ($\mu = \varepsilon_2/\varepsilon_L$)

$$\mathrm{rot\ rot}\rho\mathbf{E} - k_0^2\varepsilon_L(1 + \mu|\rho|^2 S)\rho\mathbf{E} = 0. \qquad (A)$$

Here Eq. (14) in the form Eq. (15) was used.

From Eq. (13) we obtain, with the help of the definitions of E and H

$$\mathrm{rot\ rot}\rho\mathbf{E} + ik_0^2(i\varepsilon_L R - 2\kappa m S\dot{R})\mathbf{E} = 0. \qquad (B)$$

From Eqs. (A) and (B) we obtain

$$(\rho' S)'(2\kappa)^2 - 2im\rho'(2\kappa) + \varepsilon_2|\rho|^2\rho = 0. \qquad (C)$$

Thus the equation for the function p follows from the condition that Eqs. (13) and (14) of the nonlinear system of Maxwell's equations under the additional condition (16) be compatible. An equation for media whose nonlinearity is determined by an arbitrary dependence $\varepsilon_{NL}\left(|\varepsilon|^2\right)$ can be written down in an analogous manner. The method developed here does not cover the case when the propagation medium is completely transparent ($\kappa = 0$). This fact, of course, is not an obstacle to using Eq. (C), because, for example, molecular scattering is always present. Scattering of electromagnetic waves removes some electromagnetic energy from the total beam and this is equivalent to absorption of this energy. Moreover, it is impossible to imagine a transparent medium whose nonlinearity can have any physical meaning.

The following remarks are in order. The radius vector r does not appear explicitly in Eq. (C). The quantity $S = |\mathbf{E}_\omega(r)|^2$ can always be constructed from the solution of the linear problem. It can be shown that the formulation of Poynting's theorem for a nonlinear field has a very clear interpretation:

$$\operatorname{div}\mathbf{S}_{NL} = -\left(\sigma + 2k_0 \operatorname{Im}\varepsilon_{NL}\right)|\rho|^2 S.$$

where $\mathbf{S}_{NL}$ is the Poynting vector for nonlinear vectors; this fact is an additional argument in support of the reasonableness of the assumption (6). The meaning of the expression (16) thus consists of the following. If S has some distribution over amplitude and frequencies in a neighborhood of the radius vector r, then with the help of the function $\rho(S)$ it is possible to construct in a neighborhood of this point in space the field $E_\omega(r)$, which can then be integrated over frequencies according to (1). Of course, the linear field $\mathbf{E}_\omega(r)$ must be known.

In Eq. (C) the product of the parameter by the intensity S is the nonlinear correction to the dielectric constant ε_L. We shall study the simplest case of a cubic nonlinearity. Other cases associated with the representation (10) can be studied. For example, these can be multiphoton processes or processes with saturation, etc. (a long list of self-action effects is given, for example, in Ref. 10).

Finally, we make a remark regarding the boundary conditions for Eq. (C). These conditions follow from the definition of the fields (18) and (19):

$$\rho_0 = \rho(S = S_0) = 1, \quad \rho'_0 = \rho'(S = S_0) = 0. \tag{20}$$

where S_0 is the initial intensity of the field.

4. EXPLICIT FORM OF THE FUNCTION ρ

We shall start from Eq. (C) in the form

$$S\rho'' + a\rho' + b|\rho|^2\rho = 0; \tag{21}$$

$$a = 2 - \frac{n}{\kappa}, \qquad b = \frac{\varepsilon_2}{(2\kappa)^2}. \tag{22}$$

After introducing the new independent variable $x = S/S_0$ Eq. (21) assumes the form

$$x\rho'' + a\rho' + bS_0|\rho|^2\rho = 0, \qquad \rho(1) = 1,$$
$$\rho'(1) = 0. \tag{23}$$

Next, the substitution of variables

$$\rho(x) = Z(\xi)\exp\left(i\frac{m}{2\kappa}\xi\right), \qquad \xi = \ln x, \tag{24}$$

leads to the following equation for $Z(\xi)$:

$$\left[\frac{\dot Z}{Z}\right]' + \left[\frac{\dot Z}{Z}\right]^2 + \left[\frac{m}{2\kappa}\right]^2 + bS_0|Z|^2 = 0. \tag{25}$$

introducing the notation

$$\beta = n/\kappa, \tag{26}$$

the substitution of variables (24) assumes the form

$$\rho(x) = \frac{\exp\left[i\frac{\beta}{2}\xi\right]}{x^{1/2}} Z(\xi), \tag{27}$$

and Eq. (25) can be written in the form

$$\left[\frac{\dot Z}{Z}\right]' + \left[\frac{\dot Z}{Z}\right]^2 + \left[\frac{\beta + i}{2}\right]^2 + bS_0|Z|^2 = 0. \tag{28}$$

We shall now summarize the solution of Eq. (28). Dividing Eq. (28) by the real and imaginary parts, we have

$$Z(\xi) = R(\xi)\exp(i\Phi(\xi));$$

$$\left[\frac{\dot R}{R}\right]' + \left[\frac{\dot R}{R}\right]^2 + \frac{\beta^2 - 1}{4} + \gamma S_0 R^2 = \dot\Phi^2; \tag{29}$$

$$\ddot\Phi + 2\frac{\dot R}{R}\dot\Phi + \frac{\beta}{2} + \delta S_0 R^2 = 0; \tag{30}$$

$$\gamma = \operatorname{Re} b, \qquad \delta = \operatorname{Jm} b.$$

The solution of Eq. (29) can be sought, for example, in the form of an elliptic cosine

$$R(\xi) = A\,\mathrm{cn}(B\xi + C,\ k) = A\,\mathrm{cn}\vartheta, \tag{31}$$

where k is the modulus and C is an additive constant. It is easy to verify, however, that compatibility with Eq. (30) can be achieved only if the cosine-amplitude is degenerate

$$\mathrm{cn}\vartheta \longrightarrow \mathrm{sech}\vartheta, \qquad k = 1. \tag{32}$$

We shall study below four regions of the spectrum in which the constants γ and δ have different signs.[11]

a. $\delta < 0$, $\gamma < 0$. As the solutions of the system of equations (29) and (30) we study the system of functions

$$R = A\,\mathrm{sech}\vartheta, \qquad \dot\Phi = D\,\mathrm{th}\vartheta, \qquad \vartheta = B\xi + C, \tag{33}$$

where Φ is a particular solution of Eq. (30). Substituting Eq. (33) into the system of equations (29) and (30) leads to the following values of the constants in Eq. (33):

$$A^2 = \frac{\beta^2 - 2}{4\gamma S_0}, \quad B = \frac{1}{2}, \quad D = \frac{\beta}{2}, \quad \frac{\gamma}{\delta} = \frac{1}{3}\frac{\beta^2 - 2}{\beta}. \tag{34}$$

We call attention to the last relation in Eqs. (34). It relates the nonlinearity parameters with the parameters of linear absorption. For the phase we have

$$\Phi = \frac{D}{B}\ln\mathrm{ch}\vartheta + \Phi_0, \tag{35}$$

and the constant Φ_0 is determined from the conditions (20). Thus the solution has the form

$$E = \frac{A}{x^{1/2}}\mathrm{sech}(B\xi + C)\exp\left[i\left(\frac{\beta}{2}\xi + \frac{D}{B}\ln\mathrm{ch}\vartheta + \Phi_0\right)\right]E_L. \tag{36}$$

In order to normalize this solution we shall use the additive constant C. For $S = S_0$ we have $x = 1$ and then from Eq. (36)

$$R(\xi = 0) = A\,\mathrm{sech}(C) = 1,$$

hence

$$C = \mathrm{Arsech}\frac{1}{A} = \mathrm{Arch}\,A. \tag{37}$$

Next we must return to the starting independent variables:

$$\xi = \ln x = \ln\frac{S}{S}, \qquad x = \exp(-\tau)$$

for monochromatic radiation, i.e., $\xi = -\tau$, where τ is the optical thickness of the medium: $\tau = \sigma z$, where z is the distance (path).

Next we assume that the values of the initial intensity S_0 are such that the corresponding constant A is equal to one. Then $C = 0$, $\Phi_0 = 0$, and

$$E(z) = \mathrm{sech}\left(-\frac{\sigma}{2}z\right)\exp\left\{\frac{\sigma}{2}z + i\left[-\frac{n}{c}z + \beta\ln\mathrm{ch}\left(\frac{\sigma z}{2}\right)\right]\right\}E(z).$$

For the intensity we have

$$|E(z)|^2 = \mathrm{sech}^2\left(-\frac{\sigma}{2}z\right)e^{\sigma z}|E(z)|^2 = \mathrm{sech}^2\left(-\frac{\sigma}{2}z\right)|E(0)|^2. \tag{38}$$

Note that the nonlinear parameters γ and δ do not appear in the expression (38). For large optical thicknesses

$$\mathrm{sech}(\tau) \approx 2\exp(-\tau),$$

so that for large z from the source

$$|E(z)|^2 = 4\,e^{-\sigma z}|E_0|^2. \tag{39}$$

This result distinguishes Bouguer's law for a linear medium from a nonlinear medium of the cubic type for large τ. Here, by assumption, $A = 1$ and the initial intensity S_0 satisfies the relation (see Eqs. (34), (26), and (22))

$$1 = \frac{\beta^2 - 2}{4\gamma S_0}; \quad \beta = \frac{n}{\kappa}; \quad \gamma = \frac{\mathrm{Re}\varepsilon_2}{(2\kappa)^2}. \tag{40}$$

b. $\delta < 0$, $\gamma > 0$. The result is the same as above.

c. $\delta > 0$, $\gamma < 0$. The solution of the system of equations (29) and (30) can be written in the form

$$R = A\,\mathrm{cosech}\vartheta, \quad \dot{\Phi} = D\,\mathrm{cth}\vartheta, \quad \vartheta = B\xi + C; \tag{41}$$

$$E = \frac{A}{x^{1/2}}|\mathrm{cosech}\vartheta|\exp\left[i\left\{\frac{\beta}{2}\xi + \frac{D}{B}\ln|\mathrm{sh}\vartheta| + \Phi_0\right\}\right]E, \tag{42}$$

where the constants C and Φ_0 are defined in the usual manner. We write Eq. (42) under the assumption that $A = 1$:

$$E(z) = \left|\mathrm{cosech}\left(-\frac{\sigma}{2}z - \mathrm{Arcosech}\,1\right)\right| \times \exp\left\{\frac{\sigma}{2}z + i\left[-\frac{n}{c}z + \beta\ln|\mathrm{sh}\vartheta| + \Phi_0\right]\right\}E.$$

For the intensity we have

$$|E(z)|^2 = \mathrm{cosech}^2\left(-\frac{\sigma}{2}z - 0.881\right)|E(0)|^2. \tag{43}$$

This result differs from Eq. (38) for large values of z also. Experiments indicate that the effects indicated above can also appear in the far wings of spectral lines.[12]

d. $\delta > 0$, $\gamma > 0$. The result is the same as in the preceding case.

5. CONCLUSIONS

This investigation raises the problem of studying further nonlinear effects in the region of spectral lines of real media. It was found that in the case of cubic media the attenuation of the radiation depends on the sign of the imaginary nonlinear correction ($\delta > 0$ or $\delta < 0$) to the dielectric constant of linear optics. The formulas describing the generalized form of Bouquer's law for nonlinear media show the difference from the case of the usual fields. In addition, the ratio γ/δ as a function of the parameters of the linear theory is predicted – a result that follows from Maxwell's equations for cubic media with a mechanism for self-action of the waves. It is significant that the proposed model of spectral self-action makes it possible to obtain an exact solution of the problem posed in the general three-dimensional form. It should be noted that within certain limits the characteristics of the propagation of a radiation pulse as well as some problems of nonlinear diffraction of waves by particles can be studied. In particular, under certain restrictions on the shape of the initial pulse, it can be expected that soliton-like pulses will form in the medium.

The problem studied in this paper is one of the simplest examples illustrating the proposed method.

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