Nonlinear resonant optics of thin films: the inverse method

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The interaction of light with a thin film of a nonlinear resonant medium located at the interface between two linear media can be described by a set of nonlinear Maxwell–Bloch-like differential equations which effectively take the presence of the reflected wave into account. The set of equations obtained is completely integrable. Using this approach, we solve for the passage of a soliton-like pulse of light through a film with an arbitrary line profile for inhomogeneously broadened two-level systems in the film.

1. INTRODUCTION

The theory of light propagation in resonant media has been developed in large part through the use of the inverse scattering method (ISM)[1] to solve the Maxwell–Bloch (MB) nonlinear equations.[2–5]

Recent investigations of surface and thin-film spectroscopy have awakened interest in a new class of problem in which a nonlinear resonant medium is a thin film at the interface between linear media. In such systems, both the propagation of nonlinear surface waves along the film[6] and the passage of light pulses through the interface[7] have been examined.

The equations describing surface-wave propagation effectively reduce to the one-dimensional MB equations for an unbounded medium.[6] The surface-wave problem is thereby directly subsumed into the general scheme of the inverse method.[1–5] A qualitatively different situation occurs with the passage of light pulses through an interface, where the presence of a reflected wave prevents a direct application of the ISM (recall that the MB equations are only integrable for the class of solutions describing unidirectional propagation of light). In a previous paper,[7] we considered the simplest case of exact resonance, in which the problem reduces to the solution of an ordinary differential equation. The general case, with inhomogeneous broadening and off-resonant behavior taken into account, has remained unstudied.

In the present paper, we show that in general, the interaction of light with a thin film is described by an integrable set of Maxwell–Bloch-like differential equations which effectively take the presence of a reflected wave into account. The set of equations obtained allows for the use of the ISM. The passage of a light pulse of given shape through a nonlinear film may then be formulated and solved as a scattering problem for a point potential, without the usual formulation in terms of the Cauchy problem in the theory of nonlinear waves for an unbounded medium. As an example of our approach, we solve for the passage of a soliton through a film with an arbitrary inhomogeneous broadening profile.

2. BASIC EQUATIONS

Assume that the nonlinear resonant medium has the form of a thin layer (of thickness $d \ll \lambda_L$, where λ_L is a typical wavelength of light) at the interface ($z = 0$) between two linear media with dielectric constants $\varepsilon_1(z<0)$ and $\varepsilon_2(z>0)$. A pulse of light is incident on the interface from medium I. For simplicity, we deal here with a normally incident plane wave polarized along the X-axis.

The presence of a thin polarizing layer at the interface must be taken into account in the boundary conditions for the E_x and H_y components of the electromagnetic field:

$$E_x(0+,\ t)-E_x(0-,\ t)=0, \tag{1a}$$

$$H_y(0+,\ t)-H_y(0-,\ t)=-4\pi\partial P_x/\partial t, \tag{1b}$$

where $P_x(t)\delta(z)$ is the polarization of the film. The electric fields in media I and II and the polarization of the film take the form

$$E_x^{(1)}(z,t)= {}^1/_2[E_0(z,t)\exp[i(k_1z-\omega_Lt)] + E_r(z,t)\exp[-i(k_1z+\omega_Lt)]+\text{c.c.}], \tag{2a}$$

$$E_x^{(2)}(z,t)= {}^1/_2[E_t(z,t)\exp[i(k_2z-\omega_Lt)]+\text{c.c.}], \tag{2b}$$

$$P_x(t)={}^1/_2[P(t)\exp(-i\omega_Lt)+\text{c.c.}], \tag{2c}$$

where $E_{0,r,t}(z,t)$ are the smooth envelopes of the incident, reflected, and transmitted fields, with carrier frequency ω_L; $k_{1,2}=\omega_L(\varepsilon_{1,2})^{1/2}$.

Substituting the equations (2) into the boundary conditions (1) and, within the scope of the resonant approximation, neglecting derivatives of the smooth envelopes, we find at $z=0$

$$E_0(0,\ t)+E_r(0,\ t)=E_t(0,\ t), \tag{3a}$$

$$k_2E_t(0,\ t)-k_1[E_0(0,\ t)-E_r(0,\ t)]=4\pi i\omega_LP(t). \tag{3b}$$

By virtue of the continuity in (1a), the electric field in the film is the same as that of the transmitted wave, $E_t(0,t)$. Eliminating the field E_r from (3), we obtain

$$E_t(0,t)=\frac{2\varepsilon_1^{1/2}}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}E_0(0,t)+i\frac{4\pi\omega_L}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}P(t), \tag{4}$$

where in addition to the Fresnel term, there is a contribution proportional to the polarization of the film.

In a model with noninteracting two-level systems, the matter equations are the Bloch equations for the atomic polarization $\sigma(t)$ and population $\sigma_3(t)$:

$$i\frac{\partial}{\partial t}\sigma(\Delta,t)=\Delta\sigma(\Delta,t)+{}^1/_2\mu\sigma_3(\Delta,t)E_t(0,t), \tag{5a}$$

$$i\frac{\partial}{\partial t}\sigma_3(\Delta,t)=\mu\{\bar{E}_t(0,t)\sigma(\Delta,t)-E_t(0,t)\bar{\sigma}(\Delta,t)\}, \tag{5b}$$

where $\Delta=\omega_{12}-\omega_L$ is the frequency offset of the pulses from the transition frequency ω_{12} of the given atom, μ is the dipole moment of the transition, and the overbar denotes complex conjugation. The layer polarization is related to the function $\sigma(\Delta,t)$ by

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$$P(t)=\mu n\int d\Delta\, g(\Delta)\sigma(\Delta,t), \tag{6}$$

where n is the surface density of two-level systems, and $g(\Delta)$ is the distribution function for the atomic transition frequencies. Relaxation terms have been omitted from (5), since we are assuming that the incident pulse is much shorter than the characteristic relaxation time of a two-level atom in the layer.

The set of equations (4)–(6) completely describes the problem formulated above, determining the field E_t transmitted through a nonlinear film when the pulse shape of the incident field E_0 is given.

In the simplest case, with no inhomogeneous broadening and exact resonance ($\Delta=0$), and with a real field $E_t(t)$ (which in turn is assured by the field $E_0(t)$ being real), the Bloch equations (5) give a solution

$$\sigma(t)={}^1/_2 i\sin\Psi(t),\quad \Psi(t)=\mu\int_{-\infty}^{t} dt'\, E_t(t'). \tag{7}$$

Substituting (7) into (4), we obtain an ordinary differential equation for the function $\Psi(t)$:

$$\frac{d\Psi}{dt}+\frac{1}{\tau_0}\sin\Psi=\frac{2\varepsilon_1^{1/2}}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}\mu E_0(t), \tag{8a}$$

$$\tau_0^{-1}=4\pi\omega_L\mu^2 n/(\varepsilon_1^{1/2}+\varepsilon_2^{1/2}), \tag{8b}$$

solutions of which have been studied in a previous paper.[7]

The general case (4)–(6) can be investigated using the approach described below.

3. THE INVERSE METHOD

We transform Eqs. (4) and (5) to a form enabling us to use the ISM. In order to do so, instead of the local algebraic relation (4), we introduce for consideration a differential equation for the auxiliary field $E(z,\tau)$, which depends on the coordinate z and the cone variable $\tau=t-z$:

$$i\frac{\partial}{\partial z}E(z,\tau)=-2\frac{4\pi\omega_L}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}\delta(z)P(\tau), \tag{9}$$

with the necessary constraint

$$E(0,\tau)={}^1/_2[E(0+,\tau)+E(0-,\tau)] \tag{10}$$

because of the discontinuity in $E(z,\tau)$. Equation (9) describes the unidirectional propagation of the field $E(z,\tau)$, so that field can be specified arbitrarily for $z<0$. Assuming

$$E(z<0,\tau)=\frac{2\varepsilon_1^{1/2}}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}E_0(\tau)\equiv E_{in}(\tau), \tag{11}$$

we find the field for $z>0$ from Eq. (9):

$$E(z>0,\tau)=E_{in}(\tau)+2i\frac{4\pi\omega_L}{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}P(\tau)\equiv E_{out}(\tau). \tag{12}$$

With (11) and (12) taken into consideration, the right-hand side of Eq. (10) is obviously the same as the right-hand side of Eq. (4), and therefore the auxiliary field $E(z,\tau)$ at $z=0$ has the same value as the physical field $E_t(0,t)$ at the interface. Thus, we may formulate the problem of a light pulse passing through a nonlinear film as a scattering problem for a set of Maxwell–Block-like equations with unidirectional propagation:

$$i\frac{\partial}{\partial z}u(z,\tau)=-\delta(z)\int d\Delta\, g(\Delta)s(\Delta,\tau), \tag{13a}$$

$$i\frac{\partial}{\partial\tau}s(\Delta,\tau)=\Delta s(\Delta,\tau)+2s_3(\Delta,\tau)u(0,\tau), \tag{13b}$$

$$i\frac{\partial}{\partial\tau}s_3(\Delta,\tau)=\bar u(0,\tau)s(\Delta,\tau)-u(0,\tau)\bar s(\Delta,\tau), \tag{13c}$$

where for $z<0$, $u(z,\tau)$ is a given field $u_{in}(\tau)$, and the field $E_t(\tau)$ transmitted through the film is given by

$$\mu E_t(\tau)=[u_{in}(\tau)+u_{out}(\tau)], \tag{14}$$

where $u_{out}(\tau)$ is the solution of the system (13) for $z>0$. Here we have introduced the notation

$$s(\Delta,\tau)=\sigma(\Delta,\tau)/\tau_0,\quad s_3(\Delta,\tau)=\sigma_3(\Delta,\tau)/2\tau_0,$$
$$u(z,\tau)={}^1/_2\mu E(z,\tau).$$

The set of nonlinear equations (13) is completely integrable, and can be written as a consistency condition

$$\frac{\partial}{\partial z}U-\frac{\partial}{\partial\tau}V+[U,V]=0$$

for the overdetermined set of equations

$$\partial\varphi/\partial\tau=U\varphi, \tag{15a}$$

$$\partial\varphi/\partial z=V\varphi, \tag{15b}$$

where in the present case, the 2×2 matrices U and V are of the form

$$U=i\begin{bmatrix}\lambda/2, & \bar u(z,\tau)\\ u(z,\tau), & -\lambda/2\end{bmatrix},$$
$$V=i\delta(z)\int\frac{d\Delta g(\Delta)}{\lambda-\Delta+i0}\begin{bmatrix}-s_3(\Delta,\tau), & \bar s(\Delta,\tau)\\ s(\Delta,\tau), & s_3(\Delta,\tau)\end{bmatrix}. \tag{16}$$

Following the general ISM approach,[1] it is necessary to determine the scattering data $a_{in}(\lambda)$ and $b_{in}(\lambda)$, solving the Zakharov–Shabat spectral problem (15a) with the given field $u_{in}(\tau)$. The evolution of the scattering data along the z-axis is governed by Eq. (15b), and in a standard manner, this reduces to a solution of the set of equations

$$\frac{d}{dz}a(\lambda,z)=0, \tag{17a}$$

$$\frac{d}{dz}b(\lambda,z)=\frac{i}{\tau_0}b(\lambda,z)\delta(z)\int\frac{d\Delta g(\Delta)}{\lambda-\Delta+i0}, \tag{17b}$$

where by definition

$$b(\lambda,0)={}^1/_2[b(\lambda,0+)+b(\lambda,0-)]. \tag{17c}$$

The solution of (17) relates the scattering data $a_{out}(\lambda)\equiv a(\lambda,z>0)$ and $b_{out}(\lambda)\equiv b(\lambda,z>0)$ to their initial values $a_{in}(\lambda)$ and $b_{in}(\lambda)$:

$$a_{out}(\lambda)=a_{in}(\lambda), \tag{18a}$$

$$b_{out}(\lambda)=b_{in}(\lambda)[1+i\Gamma(\lambda)/2][1-i\Gamma(\lambda)/2]^{-1}, \tag{18b}$$

$$\Gamma(\lambda)=\frac{1}{\tau_0}\int\frac{d\Delta g(\Delta)}{\lambda-\Delta+i0}. \tag{18c}$$

In our case, the equations (18) are the analog of the Gardner–Greene–Kruskal–Miura evolution equations.[1] The field at $z>0$ is obtained by solving the Gel'fand–Levitan equations with scattering data $a_{out}(\lambda)$, $b_{out}(\lambda)$.

4. ONE-SOLITON SOLUTION

For the soliton aspect of the problem, the Gel'fand–Levitan equations are algebraic. Here we examine a one-soliton solution, which addresses the passage of an isolated pulse of light of duration η^{-1},

$$E_0(\tau)=\frac{1}{\mu}\frac{\varepsilon_1^{1/2}+\varepsilon_2^{1/2}}{\varepsilon_1^{1/2}}\frac{\eta}{\operatorname{ch}(\eta\tau)}, \qquad (19)$$

through a nonlinear film; up to the Fresnel factor, this is the same as the pulse generated by the self-induced transparency effect.[8]

In the potential

$$u_{in}(\tau)=\eta/\operatorname{ch}(\eta\tau), \qquad (20)$$

corresponding to the pulse (19), the coefficient $a_{\text{in}}(\lambda)$ takes the form

$$a_{in}(\lambda)=(\lambda-i\eta)/(\lambda+i\eta), \qquad (21a)$$

while $b_{\text{in}}(\lambda)=0$ when $\operatorname{Im}\lambda=0$, and

$$b_{in}(\lambda=i\eta)=i. \qquad (21b)$$

The fact that the spectral problem (15a) is identical to the Zakarov–Shabat problem enables us immediately to write out an explicit expression for the field $u_{\text{out}}(\tau)$:

$$u_{out}(\tau)=\eta e^{-i\varphi_0}/\operatorname{ch}[\eta(\tau-\tau_c)], \qquad (22)$$

where the phase shift φ_0 and the displacement τ_c of the center of the soliton are given in terms of the coefficient $b_{\text{out}}(\lambda)$ at $\lambda=i\eta$:

$$\varphi_0=\arg b_{out}(i\eta)=\arg\frac{1+i\Gamma(i\eta)/2}{1-i\Gamma(i\eta)/2}, \qquad (23a)$$

$$\tau_c=\frac{1}{\eta}\ln|b_{out}(i\eta)|=\frac{1}{\eta}\ln\left|\frac{1+i\Gamma(i\eta)/2}{1-i\Gamma(i\eta)/2}\right|; \qquad (23b)$$

here we have made use of (18b).

According to (14), the field transmitted through the film is given by

$$E_t(\tau)=\frac{\eta}{\mu}\left[\frac{1}{\operatorname{ch}(\eta\tau)}+\frac{e^{-i\varphi_0}}{\operatorname{ch}[\eta(\tau-\tau_c)]}\right], \qquad (24)$$

i.e., it is a sum of two solitons, with relative phase shift and center shift given by (23), and with amplitudes and durations the same as those for the soliton incident on the film.

Equations (18) and (23) hold for arbitrary inhomogeneous broadening profile $g(\Delta)$. For instance, for a Lorentzian profile

$$g(\Delta)=\frac{1}{\pi}\frac{\gamma}{(\Delta-\Delta_0)^2+\gamma^2}, \qquad (25)$$

where γ is the line width and Δ_0 is the offset of its maximum from the frequency of the incident field ω_L, Eqs. (23) take the form

$$\varphi_0=-\operatorname{arctg}\frac{\Delta_0/\tau_0}{\Delta_0^2+[\eta+\gamma-1/2\tau_0]^2}, \qquad (26a)$$

$$\tau_c=\frac{1}{2\eta}\ln\frac{\Delta_0^2+[\eta+\gamma+1/2\tau_0]^2}{\Delta_0^2+[\eta+\gamma-1/2\tau_0]^2}. \qquad (26b)$$

5. CONCLUSION

In this paper, we have considered the passage of a light pulse through a nonlinear film at the interface between two linear media. The approach developed here can also be used to investigate other phenomena in the optics of nonlinear resonant films, such as photon echoes, radiative dissociation of excited atoms in the film, and so forth. Furthermore, the suggested approach can be directly extended (by analogy with Refs. 7, 9, and 10) to arbitrary angles of incidence and variable polarization of the incident field.

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[1]V. E. Zakharov, S. V. Manakov, S. P. Novikov, and L. P. Pitaevskii, *Teoriya solitonov: method obratnoi zadachi (Soliton Theory: the Inverse Method)*, Nauka, Moscow (1980)

[2]G. L. Lamb, *Vvedenie v teoriyu solitonov*, Mir, Moscow (1983) [*Elements of Soliton Theory*, John Wiley and Sons, New York (1980)].

[3]M. J. Ablowitz, D. J. Kaup, and A. C. Newell, J. Mathematical Phys. **15**, 1852 (1974).

[4]S. V. Manakov, Zh. Eksp. Teor. Fiz. **83**, 68 (1982) [Sov. Phys. JETP **56**, 37 (1982)].

[5]I. R. Gabitov, V. E. Zakharov, and A. V. Mikhailov, Zh. Eksp. Teor. Fiz. **86**, 1204 (1984) [Sov. Phys. JETP **59**, 703 (1984)]; TMF **63**, 11 (1985).

[6]V. M. Agranovich, V. I. Rupasov, and V. Ya. Chernyak, Pis'ma Zh. Eksp. Teor. Fiz. **33**, 196 (1981) [JETP Lett. **33**, 185 (1981)]; Fiz. Tverd. Tela **24**, 2992 (1982) [Sov. Phys. Solid State **24**, 1693 (1982)].

[7]V. I. Rupasov and V. I. Yudson, Kvantovaya Elektron. (Moscow) **9**, 2179 (1982) [Sov. J. Quantum Electron. **12**, 415 (1982)].

[8]S. L. McCall and E. L. Hahn, Phys. Rev. **183**, 457 (1984)

[9]A. M. Basharov and A. I. Maimistov, Zh. Eksp. Teor. Fiz. **87**, 1594 (1984) [Sov. Phys. JETP **60**, 913 (1984)].

[10]V. Ya. Chernyak and V. I. Rupasov, Phys. Lett. **108A**, 434 (1985).

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