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Steady propagation of a coherent light pulse in a dielectric medium I†

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Abstract. A systematic method to treat the steady propagation of a coherent light pulse in a dielectric medium is developed, paying special attention to the effect of polariton formation. The non-linear optical Bloch equation and the Maxwell equation are solved simultaneously by means of the method of series expansion in powers of a small parameter which is related to the pulse width. It is shown that, besides the usual self-induced transparency pulse, steady pulse solutions exist also in the case where the pulse width is much longer than the reciprocal of the polariton gap frequency. This result suggests that steady propagation may be realized also in dense dielectric media such as crystals in which the polariton effect is remarkable.

1. Introduction

Since the pioneering work by McCall and Hahn (1967, 1969), the phenomenon of self-induced transparency (SIT) has attracted much attention both experimentally and theoretically (for example, Gibbs and Slusher 1970, Lamb 1971, Courtens 1972, Allen and Eberly 1975, and references cited therein). A dilute absorbing medium such as gaseous atoms or impurity ions in solids absorbs the usual light of resonant frequency. However, if the incoming light is a sufficiently intense, coherent pulse and if its duration is shorter than the atomic relaxation times, the medium can become transparent and the pulse propagates without change of its shape and velocity. The front part of such a pulse excites the atoms in the medium coherently up to the state of complete inversion. The macroscopic polarization formed in this process then interacts with the remaining part of the pulse and emits coherent light again, which joins the back part of the pulse. If the atoms return to the ground state after this stimulation process, steady propagation is realized.

Self-induced transparency is described by solving simultaneously the Maxwell equation and the so called optical Bloch equation which governs the motion of the electric dipole of a two-level atom in an oscillating electric field. Unlike the case of a harmonic oscillator, the dipole of a two-level atom and the electric field do not couple directly to each other: they couple to each other parametrically through population inversion. It is the non-linearity inherent in such a coupling that plays an essential role in

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the formation of a steady pulse. The self-induced transparency which is accompanied by complete inversion of the atoms is a *strongly non-linear* phenomenon.

In dense dielectric media such as crystals, on the other hand, complete inversion of the atoms can hardly be achieved even by an intense light; instead of this, the formation of polaritons is significant (Hopfield 1958). The polariton, a mixed mode of the electromagnetic field and the polarization wave in matter, has a dispersion which varies steeply near the resonant frequency accompanied by, in the case of no spatial dispersion, a gap in which any mode of real wavevector does not exist (the polariton gap). In gases, such a polariton effect is very small and is smeared out by the inhomogeneous dipole–dipole interaction between randomly arranged atoms.

The purpose of the present paper is to develop a systematic method which unifies the two complementary concepts, the SIT and the polariton, and to show that a pulse of polaritons which is *weakly non-linear* can propagate steadily in a dielectric medium.

The light pulses are categorized more explicitly as follows. A pulse which has a finite time width contains a spread of Fourier components of frequency. If this spread covers the polariton gap completely, the pulse cannot feel the existence of the gap. We call such a pulse *short*†. In the opposite case where the spread of Fourier components of frequency lies completely inside or completely outside the polariton gap, we call the pulse *long*. A long pulse will show different behaviours outside and inside the gap.

The polariton gap frequency is given by $8\pi N d^2/\hbar$, where N is the dipolar density and d the dipole matrix element. The orders of magnitude of the reciprocal of this frequency are roughly estimated to be microsecond in gases and picosecond in crystals. This means that a nanosecond pulse of nearly resonant frequency, for example, is short for gases, but long for crystals. This is the reason why we are interested in a long pulse, which has never been discussed before.

The plan of the paper is as follows. In § 2 a steady pulse is defined as a product of a slowly varying pulse envelope and a rapidly oscillating carrier wave, and the fundamental equations which describe its propagation are discussed. As a special solution of these equations, the plane wave solution for non-linear polaritons is derived in § 3. This solution is instructive because it shows that a sufficiently intense electromagnetic field modifies the dispersion relation of the polariton and becomes able to propagate even inside the polariton gap. In § 4, the dispersion relation of the carrier wave and the velocity of the pulse envelope are determined from the linearized equations in a self-consistent manner in connection with the pulse width. On the basis of these results, the coupled non-linear equations are solved in § 5. The method of solution is a series expansion in powers of a small parameter which is related to the pulse width. The expansion parameter is chosen in different ways depending on the kind of pulse. By this method the coupled equations are reduced to solvable forms in which the non-linearity is taken into account in a well balanced way. It will be shown that a long pulse whose frequency lies outside the polariton gap propagates steadily as a solitary wave of polariton, and that a long pulse inside the gap also exists and propagates very slowly. Finally in § 6, some physical interpretations and discussions are given.

2. Steady pulse and fundamental equations

The starting point of our theory is the same as that of existing theories (for example,

† A short pulse is usually defined as a pulse whose width is shorter than the atomic relaxation times. In the present paper, however, we do not use this definition, assuming all the relaxation times are infinite.

McCall and Hahn 1969, Lamb 1971). The dielectric medium is represented by a continuum which consists of non-interacting two-level atoms and the coherent electromagnetic field is treated classically. The steady pulse solution to be found is of the form

$$\boldsymbol{E}(t, z) = \hat{E}(t - z/V)\{\mathbf{1}\}, \tag{2.1}$$

$$\boldsymbol{P}(t, z) = \tfrac{1}{2}N\hbar\kappa[u(t - z/V)\{\mathbf{1}\} + v(t - z/V)\{\mathbf{2}\}], \tag{2.2}$$

where

$$\begin{aligned} &\{\mathbf{1}\} = \boldsymbol{x}\cos\theta + \boldsymbol{y}\sin\theta, \qquad \{\mathbf{2}\} = -\boldsymbol{x}\sin\theta + \boldsymbol{y}\sin\theta, \\ &\theta = \omega t - KZ + \phi(t - z/V). \end{aligned} \tag{2.3}$$

The electric field $\boldsymbol{E}$ is factorized into the slowly varying pulse envelope $\hat{E}$ and the rapidly oscillating carrier wave $\{\mathbf{1}\}$, in which $\boldsymbol{x}$ and $\boldsymbol{y}$ are the unit vectors of polarization. Steadiness of the propagation is expressed by letting the envelope be a function of only $t - z/V$, where V is the velocity of the pulse envelope propagating in the z direction. In the carrier wave, the frequency ω and the wavenumber K are those defined at the pulse tail in the medium; the relation between them is to be determined in a self-consistent manner. Possible phase modulation ϕ ($\dot{\phi} \to 0$ for $t \to \pm\infty$) is also taken into account. The macroscopic polarization per unit volume, $\boldsymbol{P}$, is the sum of the in-phase (dispersive) component and the out-of-phase (absorptive) one; each is also factorized in the same way as for $\boldsymbol{E}$. In (2.2), κ is the dipole matrix element divided by $\hbar/2$, i.e. $\kappa = 2d/\hbar$.

The electric field vector $\boldsymbol{E}$ obeys the Maxwell wave equation in one dimension:

$$\left(\frac{\partial^2}{\partial z^2} - \frac{1}{c^2}\frac{\partial^2}{\partial t^2}\right)\boldsymbol{E}(t, z) = \frac{4\pi}{c^2}\frac{\partial^2}{\partial t^2}\boldsymbol{P}(t, z). \tag{2.4}$$

Substituting (2.1) and (2.2) into (2.4) and taking its components along $\{\mathbf{1}\}$ and $\{\mathbf{2}\}$ separately, we have coupled differential equations of the second order for $\hat{E}$ and $\dot{\phi}$:

$$\begin{aligned} &\left(\frac{1}{V^2} - \frac{1}{c^2}\right)\ddot{\hat{E}} - \left[(K^2 - k^2) + 2\left(\frac{K}{V} - \frac{k}{c}\right)\dot{\phi} + \left(\frac{1}{V^2} - \frac{1}{c^2}\right)\dot{\phi}^2\right]\hat{E} \\ &\qquad = \frac{2\pi N\hbar\kappa}{c^2}[\ddot{u} - (\omega + \dot{\phi})^2 u - \ddot{\phi}v - 2(\omega + \dot{\phi})\dot{v}], \end{aligned} \tag{2.5}$$

$$\begin{aligned} &\left(\frac{1}{V^2} - \frac{1}{c^2}\right)\ddot{\phi}\hat{E} + 2\left[\frac{K}{V} - \frac{k}{c} + \left(\frac{1}{V^2} - \frac{1}{c^2}\right)\dot{\phi}\right]\dot{\hat{E}} \\ &\qquad = \frac{2\pi N\hbar\kappa}{c^2}[\ddot{v} - (\omega + \dot{\phi})^2 v + \ddot{\phi}u + 2(\omega + \dot{\phi})\dot{u}], \end{aligned} \tag{2.6}$$

where the dots denote differentiation with respect to $\zeta = t - z/V$, and k is the wavenumber of light in vacuum, i.e. $k = \omega/c$.

The dimensionless amplitudes of the electric dipole of an atom, u and v, together with the population difference between the ground and excited states, w, form a classical pseudo-spin vector in the rotating frame; the motion of this vector is governed by the optical Bloch equation:

$$\dot{u} = (\omega - \omega_0 + \dot{\phi})v, \tag{2.7}$$

$$\dot{v} = -(\omega - \omega_0 + \dot{\phi})u + \kappa\hat{E}w, \tag{2.8}$$

$$\dot{w} = -\kappa\hat{E}v, \tag{2.9}$$

where ω_0 is the resonant frequency of the medium. All the relaxation times have been assumed to be infinite. It can be seen from (2.7)–(2.9) that the three components of the pseudo-spin vector satisfy the conservation law

$$u^2+v^2+w^2=1. \tag{2.10}$$

Our problem is to solve equations (2.7)–(2.9) together with (2.5) and (2.6) simultaneously under the boundary condition that in the limit of $\zeta\to\pm\infty$, the electric field vanishes and the crystal is in the ground state, i.e. $\hat{E}=0$ and $w=-1$.

To make those coupled non-linear equations manageable, some simplifications have been made since McCall and Hahn (1967, 1969). In their theory, the phase modulation $\dot{\phi}$ has been ignored and all but the lowest derivative terms on each side of (2.5) and (2.6) have been discarded. This approximation is based on the consideration that the envelopes $\hat{E}$, u and v vary sufficiently slowly in space and time as compared with the carrier wave parts, i.e.

$$|\partial\hat{E}/\partial t|\ll\omega|\hat{E}| \qquad \text{and} \qquad |\partial\hat{E}/\partial z|\ll K|\hat{E}| \quad \text{etc,} \tag{2.11}$$

and is called the slowly varying envelope approximation. The equations thus simplified can easily be solved and yield the pulse solution. The electric field $\hat{E}$ takes the well known hyperbolic secant shape and its area is normalized to 2π independent of the pulse width τ, which can be chosen arbitrarily. At the same time, the wavenumber K and the pulse velocity V are determined as functions of ω and τ. As τ increases, V shows a remarkable slowing down from the light velocity c; this is one of the characteristics of the SIT.

When τ is sufficiently large and ω lies near or inside the polariton gap, however, the slowly varying envelope approximation breaks down for the following reason. In such a case, the dispersion relation $K(\omega)$ indeed approaches that of the polariton, but the pulse velocity becomes much smaller than c, so that $\partial\hat{E}/\partial z\cong -V^{-1}\dot{\hat{E}}$ cannot be neglected in comparison with $K\hat{E}$. That is, among the higher-derivative terms in (2.5) and (2.6), those which come from the spatial derivative in the original Maxwell equation can no longer be discarded. Moreover, as will be shown later, a long pulse should be obtained on a balance of small quantities including $\dot{\phi}$, so that we cannot neglect $\dot{\phi}$. When τ is small or ω is far off-resonant, on the other hand, V remains close to c and the slowly varying envelope approximation may be valid.

It has also been pointed out that the McCall–Hahn theory requires corrections for a very short pulse whose envelope is no longer slowly varying. Marth *et al* (1974) considered that an improvement is appropriate for a pulse such that $\tau\ll\tau_c$ where $\tau_c=(\frac{1}{2}\pi N\hbar\omega_0\kappa^2)^{-1/2}$, and developed a new method of approximation based on a series expansion in powers of a small parameter which is related to the field strength. By this method, they succeeded in introducing the phase modulation $\dot{\phi}$ into the theory.

However, their method is inapplicable to a long pulse such that $\tau\gtrsim\omega\tau_c^2$, because their power series does not show good convergence in such a case. Marth *et al* left such a long pulse out of consideration for the reason that the coherence of a pulse in gases will be destroyed in a dephasing time $\omega\tau_c^2$ due to the inhomogeneous dipole–dipole interaction between atoms. This is not the case in crystals, however, in which the atoms are regularly arranged on the lattice.

After Marth *et al*, Bialynicka-Birula (1974) proposed another method of power-series expansion and obtained corrections to the McCall–Hahn solution, but her interest was still restricted to a short pulse such that $\tau\approx\tau_c$. Eilbeck *et al* (1973) developed a method of simplification by which they reduced the second-order Maxwell equation into a first-order form. However, their interest was also in describing the SIT pulse more accurately, and the limitation of the approximation is not very clear.

3. Plane wave solution—non-linear polariton

If all the derivative terms in (2.5)–(2.9) are set equal to zero, these equations are reduced to those which describe the plane wave solution. Their non-trivial solutions satisfy the relations

$$u=\left[\left(\frac{cK}{\omega}\right)^2-1\right]\frac{\kappa\hat{E}}{2\pi N\hbar\kappa^2},\qquad v=0,\qquad w=\left[\left(\frac{cK}{\omega}\right)^2-1\right]\frac{\omega-\omega_0}{2\pi N\hbar\kappa^2}. \tag{3.1}$$

In order for the conservation law (2.10) to hold, these relations demand the field-dependent dispersion relation

$$\left(\frac{cK}{\omega}\right)^2=1+\frac{2\pi N\hbar\kappa^2}{\omega_0-\omega}\left(\frac{(\omega-\omega_0)^2}{(\omega-\omega_0)^2+(\kappa\hat{E})^2}\right)^{1/2} \tag{3.2}$$

for an absorber ($w<0$), which also defines the field-dependent dielectric function $\tilde{\epsilon}(\omega,\hat{E})$.

In the limit of $\hat{E}\to 0$, (3.2) is reduced to the dispersion relation of the usual (linear) polariton, which has the frequency gap $\omega_0<\omega<\omega_0+2\pi N\hbar\kappa^2$ in which K is purely imaginary and any plane wave cannot propagate.

For general $\hat{E}$, on the other hand, (3.2) gives the dispersion relation of the *non-linear* polariton. The frequency gap in which K is purely imaginary now becomes narrower depending on $\hat{E}$ like

$$\omega_0<\omega<\omega_0+[(2\pi N\hbar\kappa^2)^2-(\kappa\hat{E})^2]^{1/2}, \tag{3.3}$$

and vanishes if $\hat{E}\geqslant 2\pi N\hbar\kappa$. This fact means that the medium can become transparent even for an electromagnetic plane wave whose frequency lies inside the polariton gap, if its amplitude is large enough. Such a field-dependent effect may be explained as follows. In the linear limit, u should be proportional to $\hat{E}$; the proportional coefficient has a fixed value determined by the dipole matrix element. When $\hat{E}$ increases, however, u cannot increase to any extent proportional to $\hat{E}$, because it is bound to v and w through (2.10). In other words, the proportional coefficient itself decreases with the increase of $\hat{E}$, as if the dipole matrix element decreased. The field dependence in (3.2) reflects this apparent reduction of the dipole matrix element.

4. Behaviour of pulse tail

Before solving our non-linear equations, let us determine the dispersion relation of the carrier wave and the pulse velocity. Some authors assumed the dispersion relation equal to those of the photon (Haken and Schenzle 1973) and of the polariton (Hanamura 1974, Inoue 1974) according to the case of the strong field and of the weak field they were concerned with, respectively. However, the dispersion relation should necessarily be determined in a self-consistent manner in connection with the pulse solution to be found. The situation is similar to that, the dispersion relation of the usual polariton is so determined that it can give a non-vanishing plane wave solution.

We have defined ω and K at the pulse tail. In order to see the behaviour of the pulse tail where the excitation is very weak, it is sufficient to consider the linearized version of our equations. If one solves the linearized equations, which are given by setting $\dot{\phi}=0$ and $w=-1$ in (2.5)–(2.9) corresponding to the atomic ground state, one can obtain exponential solutions $E\propto\exp[\pm(t-z/V)/\tau]$ in general, besides the plane wave solution which corresponds to the usual polariton. Such a divergent solution as it stands is physically meaningless in infinite media and is usually left out of consideration. However, the *tail* of a steadily propagating pulse is in fact not the plane wave but a growing (or decaying) wave, so that its behaviour should be described just by such an exponential solution at least locally. When the non-linearity is taken into account, the divergence of the linear solution will be suppressed and a pulse will be formed; at the same time, the dispersion relation around the pulse peak will be modulated through non-zero $\dot{\phi}$. The growth (or decay) rate τ, which is an integral constant, then gives the measure of the pulse width, because the pulse shape is reasonably supposed to be a slowly varying function in space and time. We call τ the pulse width hereafter.

The wavenumber K and the pulse velocity V determined as functions of ω and τ from the linearized equations are as follows:

$$\left(\frac{K}{k}\right)^2=\frac{1}{2}\left[(1-s^2)-\frac{(1-s^2)\Delta+2s\Lambda}{\Delta^2+\Lambda^2}+(1+s^2)\left(\frac{(\Delta-1)^2+\Lambda^2}{\Delta^2+\Lambda^2}\right)^{1/2}\right], \tag{4.1}$$

$$\left(\frac{c}{V}\right)^2=\frac{s^{-2}}{2}\left[-(1-s^2)+\frac{(1-s^2)\Delta+2s\Lambda}{\Delta^2+\Lambda^2}+(1+s^2)\left(\frac{(\Delta-1)^2+\Lambda^2}{\Delta^2+\Lambda^2}\right)^{1/2}\right], \tag{4.2}$$

where Δ and Λ are the frequency difference and the reciprocal pulse width, respectively, both of which are scaled by the polariton gap frequency, i.e.

$$\Delta=\frac{\omega-\omega_0}{2\pi N\hbar\kappa^2}, \qquad \Lambda=\frac{\tau^{-1}}{2\pi N\hbar\kappa^2}, \tag{4.3}$$

and s is defined as

$$s=(\omega\tau)^{-1}, \tag{4.4}$$

which is assumed to be always smaller than unity. The polariton gap corresponds to the range $0<\Delta<1$.

The τ-dependent dispersion relation (as we tentatively call it) given by (4.1) is plotted in figure 1. For Λ sufficiently large as compared with unity, this dispersion relation is close to that of the photon. As Λ decreases, the dispersion relation outside the gap approaches that of the polariton. Note that K remains real also inside the gap and tends to zero like $K\simeq k\Lambda[4\Delta^3(1-\Delta)]^{-1/2}$ in the limit of $\Lambda\to 0$. This fact means that the growing (or decaying) wave shows real propagation also inside the gap, though the plane wave does not.

The pulse velocity V is plotted in figure 2. For Λ sufficiently large so that $s\Lambda\gg 1$ is satisfied, V is approximately given by the relation $(c/V)^2=1+(s\Lambda+\frac{1}{4})/(s\Lambda)^2$, and tends to c in the limit of $s\Lambda\to\infty$. As Λ decreases, V outside the gap approaches $c(s/\Lambda)[4\Delta^3(1-\Delta)]^{1/2}$, which is equal to the group velocity $\mathrm{d}\omega/\mathrm{d}K$ calculated from the dispersion relation of the polariton. Here s/Λ is equal to $2\pi N\hbar\kappa^2/\omega$, i.e. approximately the ratio of the polariton gap frequency to the resonant frequency, and is regarded as a material constant. The pulse velocity is real also inside the gap and tends to zero like $c\Lambda(s/\Lambda)[\Delta/(1-\Delta)]^{1/2}$ in the limit of $\Lambda\to 0$. Figure 2 shows that the pulse velocity near the gap varies steeply depending on Δ and Λ. In contrast with the dispersion relation,

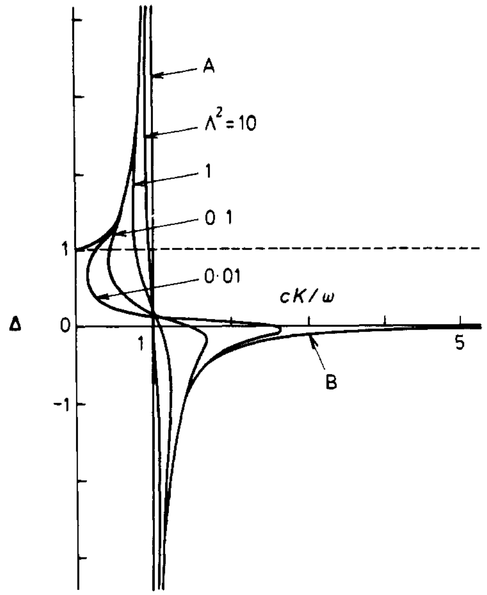

Figure 1. Dispersion relation of the carrier wave at the pulse tail. The ordinate Δ and the parameter Λ denote the frequency difference $\omega - \omega_0$ and the reciprocal of the pulse width τ^{-1}, respectively, scaled by the polariton gap frequency $2\pi N\hbar\kappa^2$. The dispersion relation does not depend strongly on the material constant $2\pi N\hbar\kappa^2/\omega_0$, which has been approximately set equal to zero. Curve A, photon; curve B, polariton.

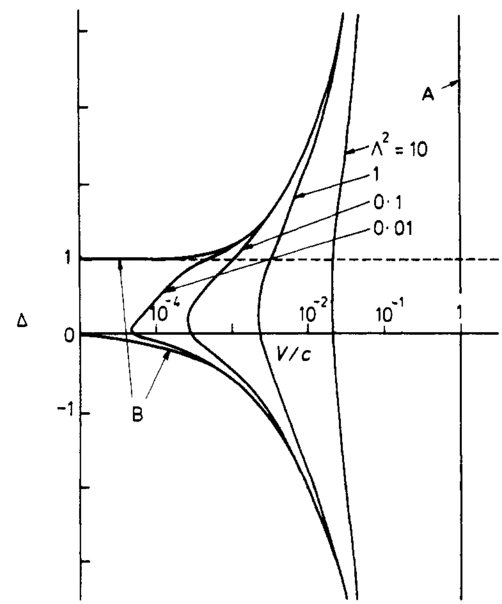

Figure 2. Propagation velocity of the pulse envelope as a function of Δ and Λ. The behaviour of the pulse velocity strongly depends on the material constant $2\pi N\hbar\kappa^2/\omega_0$, which has been chosen to be 10^{-3}. Curve A, photon; curves B, polariton.

the pulse velocity depends strongly on the material constant $2\pi N\hbar\kappa^2/\omega_0$, which has been chosen to be 10^{-3} in figure 2.

The relations (4.1) and (4.2) form the basis of our theory which unifies the two complementary concepts, the SIT and the polariton. Note that the differential coefficient $d\omega/dK$ calculated from (4.1) does not have the meaning of group velocity of a wave packet, except in the limit of $\tau \to \infty$. The envelope of a growing (or decaying) wave has by itself a definite velocity, which is coupled to the dispersion relation parametrically through τ.

5. Pulse solutions

Let us now return to the non-linear equations. By introducing a dimensionless field amplitude

$$E = \hat{E}/2\pi N\hbar\kappa, \tag{5.1}$$

and choosing a dimensionless time

$$\xi = \zeta/\tau = (t - z/V)/\tau \tag{5.2}$$

as the variable, equations (2.5)–(2.9) are reduced to the following coupled differential equations for E, ϕ, u, v and w:

$$\gamma\ddot{E} - (\alpha + \beta\dot{\phi} + \gamma\dot{\phi}^2)E = s^2\ddot{u} - (1 + s\dot{\phi})^2 u - s^2\ddot{\phi}v - 2(1 + s\dot{\phi})s\dot{v}, \tag{5.3}$$

$$\gamma\ddot{\phi}E + (\beta + 2\gamma\dot{\phi})\dot{E} = s^2\ddot{v} - (1 + s\dot{\phi})^2 v + s^2\ddot{\phi}u + 2(1 + s\dot{\phi})s\dot{u}, \tag{5.4}$$

$$\Lambda\dot{u} = (\Delta + \Lambda\dot{\phi})v, \tag{5.5}$$

$$\Lambda\dot{v} = -(\Delta + \Lambda\dot{\phi})u + Ew, \tag{5.6}$$

$$\Lambda\dot{w} = -Ev, \tag{5.7}$$

where the dots denote differentiation with respect to ξ and the coefficients α, β and γ are defined as

$$\alpha = \left(\frac{K}{k}\right)^2 - 1, \qquad \beta = 2s\left(\frac{cK}{Vk} - 1\right), \qquad \gamma = s^2\left[\left(\frac{c}{V}\right)^2 - 1\right], \tag{5.8}$$

which are functions of three dimensionless parameters Δ, Λ and s. The method of solution is a power-series expansion in which all the coefficients and the functions to be solved are expanded in powers of a small parameter which is related to the pulse width. The expansion parameter is chosen depending on the kind of the pulse, as will be discussed separately below.

5.1. A long pulse outside the polariton gap

A long pulse is defined as a pulse which satisfies the inequalities

$$\Lambda \ll |\Delta| \qquad \text{and} \qquad \Lambda \ll |\Delta - 1|. \tag{5.9}$$

Such a pulse will be sensitive to Δ and show different behaviours outside and inside the polariton gap. Let us begin with a pulse outside the gap.

Setting

$$\Lambda = \epsilon\Delta, \qquad \text{i.e.} \qquad \epsilon = [(\omega - \omega_0)\tau]^{-1}, \tag{5.10}$$

we transform parameters Λ and Δ into a new set ϵ and Δ, and choose $\epsilon(|\epsilon| \ll 1)$ as the expansion parameter. Unless the case of far off-resonance is considered, parameter s is much smaller than ϵ, so that all the terms involving s in the right-hand sides of (5.3) and (5.4) can safely be neglected as far as higher powers of ϵ do not come into question. Let all the functions in (5.3)–(5.7) be expanded in powers of ϵ as

$$E = \epsilon^{\nu}(E_0 + \epsilon E_1 + \epsilon^2 E_2 + \ldots) \qquad \text{etc.} \tag{5.11}$$

The explicit forms of coefficients α, β and γ are already known; they are expanded as

$$\begin{aligned} \alpha &= -\frac{1}{\Delta}\left(1 - \frac{4\Delta - 3}{4(\Delta - 1)}\epsilon^2 + \ldots\right), \\ \beta &= \frac{1}{\Delta}(\epsilon - \epsilon^3 + \ldots), \\ \gamma &= \frac{1}{\Delta}\left(\frac{1}{4(\Delta - 1)}\epsilon^2 + \ldots\right). \end{aligned} \tag{5.12}$$

All terms involving s have also been neglected here. These expansions are valid for any value of Δ except in the vicinity of $\Delta = 1$ such that $|\Delta - 1| \ll \epsilon$. By inserting these expansions together with (5.11) into (5.3)–(5.7) and comparing terms of the same power of ϵ with each other, a sequence of differential equations is obtained, which can be solved successively under a suitable boundary condition (see appendix). Note that no pulse solution can be obtained until the contribution from the second term in the expansion of α is taken into account; the first term gives only the dispersion relation of the plane wave polariton†.

Explicit forms of the lowest-order pulse solutions are

$$\begin{aligned} E &= \epsilon\Delta\left(\frac{4\Delta - 3}{\Delta - 1}\right)^{1/2} \operatorname{sech} \xi, \qquad & \dot{\phi} &= -\frac{3\epsilon}{2(4\Delta - 3)}\operatorname{sech}^2 \xi, \\ u &= -\epsilon\left(\frac{4\Delta - 3}{\Delta - 1}\right)^{1/2} \operatorname{sech} \xi, \qquad & v &= \epsilon^2\left(\frac{4\Delta - 3}{\Delta - 1}\right)^{1/2} \operatorname{sech} \xi \tanh \xi, \\ w &= -1 + \epsilon^2 \frac{4\Delta - 3}{2(\Delta - 1)}\operatorname{sech}^2 \xi. \end{aligned} \tag{5.13}$$

The field envelope has a hyperbolic secant shape like the SIT pulse, but its area is no longer equal to 2π. As the pulse width becomes longer, both the field envelope and the population inversion tend to zero. Contrary to the case of SIT, a principal part of the polarization is given by the in-phase (dispersive) component u; i.e. $u \gg v$. In view of this fact and that the dispersion relation and the pulse velocity are very close to those of the polariton, we call the pulse obtained above a polariton-soliton.

5.2. A long pulse inside the polariton gap

A long pulse can exist also inside the polariton gap. By choosing the expansion

† This is the reason why previous authors (Hanamura 1974, Inoue 1974), who assumed the dispersion relation of the carrier wave equal to that of the polariton, could not obtain any pulse solution.

parameter in the same way as outside the gap, coefficients α and γ are expanded as

$$\alpha=-1-\frac{1}{4\Delta(\Delta-1)}\epsilon^2+\ldots,$$
$$\gamma=-1+\frac{1}{\Delta}\left(1-\frac{4\Delta-3}{4(\Delta-1)}\epsilon^2+\ldots\right), \tag{5.14}$$

while β is the same as in (5.12). Equations (5.3)–(5.7) then lead to, in the zeroth order of ϵ, a differential equation for E:

$$(\Delta^{-1}-1)\dot{E}^2=-E^2+2[(\Delta^2+E^2)^{1/2}-\Delta]. \tag{5.15}$$

The solution of this equation is given by the reciprocal function of

$$\xi=\frac{1-\Delta}{\Delta}\cos^{-1}[(\Delta^2+E^2)^{1/2}+\Delta-1]+\mathrm{sech}^{-1}\left(\frac{(\Delta^2+E^2)^{1/2}-\Delta}{(1-\Delta)^{1/2}E}\right). \tag{5.16}$$

Near the upper edge of the polariton gap $\Delta=1$, $E(\xi)$ is approximately a hyperbolic secant: $2(1-\Delta)^{1/2}\,\mathrm{sech}\,\xi$; while near the lower edge $\Delta=0$, it is approximately a period of cosine $1+\cos(\Delta^{1/2}\xi)$ $(-\pi<\Delta^{1/2}\xi<\pi)$ accompanied by a tail which decays like $4\exp(-|\xi|)$ as $|\xi|\to\infty$. The peak value of E is given by

$$E_{\max}=2(1-\Delta)^{1/2}, \tag{5.17}$$

or, restoring the dimension,

$$\hat{E}_{\max}=\frac{2}{\kappa}[2\pi N\hbar\kappa^2(\omega_0+2\pi N\hbar\kappa^2-\omega)]^{1/2}, \tag{5.18}$$

which does not vanish even in the limit of $\tau\to\infty$. The other functions are related to E as

$$\dot{\phi}=\epsilon\frac{\Delta^2}{1-\Delta}\left(\frac{1}{(\Delta^2+E^2)^{1/2}[\Delta+(\Delta^2+E^2)^{1/2}]}-\frac{1}{2\Delta^2}\right)$$
$$u=-\frac{E}{(\Delta^2+E^2)^{1/2}},\qquad v=-\epsilon\frac{\Delta^2\dot{E}}{(\Delta^2+E^2)^{3/2}}, \tag{5.19}$$
$$w=-\frac{\Delta}{(\Delta^2+E^2)^{1/2}}.$$

As can be seen from (5.14), the wavenumber and the pulse velocity tend to zero in the limit of $\tau\to\infty$; this means that the pulse shows neither spatial oscillation nor propagation. Nevertheless, the *spatial* width remains finite and is measured by the quantity

$$V\tau=\frac{c}{\omega}\left(\frac{\omega-\omega_0}{\omega_0+2\pi N\hbar\kappa^2-\omega}\right)^{1/2}, \tag{5.20}$$

which also characterizes the spatial shape of the pulse tail. We can see how the pulse solution obtained above is related to the linear solution. If the linearized equations are solved by assuming constant E inside the gap, an exponentially decaying (or growing) solution without spatial oscillation or propagation will be obtained, because the wavenumber K_i there is purely imaginary. Such a solution can exist inside the surface of the medium irradiated by light. The tail of the pulse solution obtained above continues to this type of linear solution, because $V\tau$ in (5.20) is just equal to $|K_i|^{-1}$.

5.3. A long pulse with $\Delta = 1$

For $\Delta = 1$, i.e. $\omega = \omega_0 + 2\pi N\hbar\kappa^2$, the expansions (5.12) and (5.14) break down. In this case, however, we can expand α and γ in powers of $\epsilon = \Lambda$ after setting $\Delta = 1$ in (4.1) and (4.2). The results are

$$\begin{aligned} \alpha &= -1 + \tfrac{1}{2}\epsilon + \tfrac{1}{2}\epsilon^2 + \ldots, \\ \gamma &= \tfrac{1}{2}\epsilon - \tfrac{1}{2}\epsilon^2 + \ldots. \end{aligned} \tag{5.21}$$

By using these expansions, E and $\dot{\phi}$ are solved as

$$\begin{aligned} E &= 2^{1/2}\epsilon^{1/2} \operatorname{sech} \xi, \\ \dot{\phi} &= -\tfrac{3}{2}\epsilon^{1/2} \operatorname{sech}^2 \xi. \end{aligned} \tag{5.22}$$

Relations between E and each of u,v and w are the same as in (5.13).

5.4. A short pulse

A short pulse satisfies the condition

$$\Lambda \gg 1 \qquad \text{and} \qquad \Lambda \gg |\Delta|. \tag{5.23}$$

Such a pulse will not be sensitive to the existence of the gap. By choosing

$$\epsilon = 1/\Lambda \tag{5.24}$$

as the expansion parameter, coefficients α, β and γ are expanded as

$$\begin{aligned} \alpha &= -s\epsilon - \frac{4\Delta - 1 - s^2}{4}\epsilon^2 + \ldots, \\ \beta &= (1 - s^2)\epsilon - 2\Delta s\epsilon^2 + \ldots, \\ \gamma &= s\epsilon + \frac{1 - (4\Delta - 1)s^2}{4}\epsilon^2 + \ldots. \end{aligned} \tag{5.25}$$

Note that parameter s can become smaller or larger than ϵ in this case, in contrast with the case of a long pulse where s is always smaller than $\epsilon = \Lambda/\Delta$.

When $s \ll \epsilon$, i.e. $\tau^{-1} \ll (2\pi N\hbar\kappa^2\omega)^{1/2}$, all the terms involving s in the right-hand sides of (5.3) and (5.25) can be neglected as far as the lowest-order solutions are concerned. The solutions are given as

$$\begin{aligned} E &= 2\epsilon^{-1} \operatorname{sech} \xi, & \dot{\phi} &= -\tfrac{3}{8}\epsilon \operatorname{sech}^2 \xi, \\ u &= -2\epsilon(\Delta - \tfrac{1}{8}\operatorname{sech}^2 \xi) \operatorname{sech} \xi, & v &= 2 \operatorname{sech} \xi \tanh \xi, \\ w &= -1 + 2 \operatorname{sech}^2 \xi. \end{aligned} \tag{5.26}$$

These are the same as the SIT solutions derived by McCall and Hahn, except that non-zero $\dot{\phi}$ and the additional term in u (the second term in the parentheses) have been obtained here as corrections. The pulse area of $\hat{E}$ is equal to 2π and the population of atoms is completely inverted at the pulse peak.

When the pulse width is so short that $s \gtrsim \epsilon$, i.e. $\tau^{-1} \gtrsim (2\pi N\hbar\kappa^2\omega)^{1/2}$, the terms involving s can no longer be neglected even for the lowest-order solution. We show here only the results for $s \approx \epsilon$. In this case, the additional term $\tfrac{3}{2}s \operatorname{sech}^2 \xi$ is added to

$\dot{\phi}$ in (5.26) and the term $s\ \mathrm{sech}^3\ \xi$ is added to u. The new expression for $\dot{\phi}$ thus obtained is the same as the one derived by Marth *et al* (1974); it shows that $\dot{\phi}_{\max}$ vanishes if $\tau = \tau_c$ but can be large enough if $\tau \ll \tau_c$.

5.5. A pulse of intermediate width

Among the cases other than those given in the preceding subsections, only the case where $\Delta^2 + \Lambda^2 \gg 1$ can be treated analytically. In this case, by transforming parameters Δ and Λ into a new set

$$\epsilon = (\Delta^2 + \Lambda^2)^{-1/2}, \qquad \delta = \Delta/\Lambda \sim O(1), \tag{5.27}$$

and choosing ϵ as the expansion parameter, pulse solutions are obtained as

$$E = \frac{2}{(1+\delta^2)^{1/2}}\epsilon^{-1}\ \mathrm{sech}\ \xi, \qquad \dot{\phi} = -\frac{3}{8}\frac{1}{(1+\delta^2)^{1/2}}\epsilon\ \mathrm{sech}^2\ \xi,$$

$$u = -\frac{2\delta}{1+\delta^2}\mathrm{sech}\ \xi, \qquad v = \frac{2}{1+\delta^2}\mathrm{sech}\ \xi\ \tanh\ \xi, \tag{5.28}$$

$$w = -1 + \frac{2}{1+\delta^2}\mathrm{sech}^2\xi.$$

These expressions except $\dot{\phi}$ have already been given in the framework of the slowly varying envelope approximation (Courtens 1972). If $\delta \gg 1$ and $\delta \ll 1$ are assumed in (5.28), the expressions *for large* Δ of (5.13) and (5.26) are obtained, respectively. Equations (5.28) thus connect the solutions for a short pulse and for a long pulse with each other in their leading terms.

6. Discussions

All the pulse solutions obtained in § 5 are summarized in figure 3. The usual polariton corresponds to the line $\tau^{-1} = 0$. Above the straight line $\tau^{-1} = |\omega - \omega_0|$, the out-of-phase

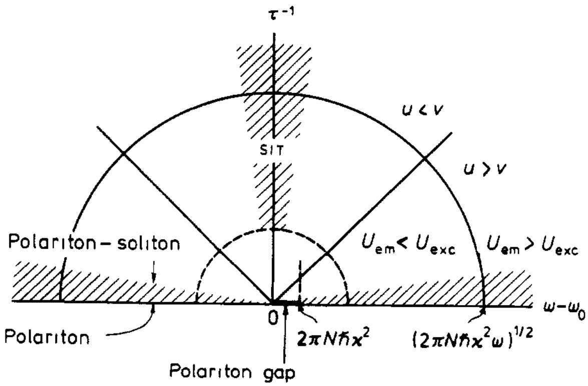

Figure 3. Classification of the pulse solutions. The solutions have been obtained: in the hatched regions, inside the polariton gap near $\tau^{-1} = 0$, and outside the broken semicircle. The energy density of electromagnetic field $\hat{E}^2/4\pi$ and that of excitation $N\hbar\omega(w+1)$ are denoted by U_{em} and U_{exc}, respectively.

component v of the dipole moment is larger than the in-phase one u; pulses in this region are SIT-like. Below this line, the in-phase component is larger; pulses are polariton-like. The SIT and the polariton-soliton are indicated by the hatched regions. Steady pulse solutions have been obtained also inside the polariton gap near $\tau^{-1}=0$ and outside the broken semicircle. Inside the full semicircle, the energy density of excitation $N\hbar\omega(w+1)$ is larger than that of the electromagnetic field $\hat{E}^2/4\pi$, and the dispersion relation differs considerably from that of the photon. Outside the circle, on the other hand, the energy density of electromagnetic field is larger, and the dispersion relation is almost photon-like.

Figure 3 implies that the case of a long pulse can also be realized in gases, if an off-resonant pulse is used. For example, a nanosecond pulse of resonant frequency is a short pulse in gases, but it becomes a long pulse if the frequency shifts by $\Delta\omega/\omega \gtrsim 10^{-4}$. Grishkowsky and his coworkers (Grishkowsky 1973, Grishkowsky *et al* 1973) studied the propagation of such off-resonant pulses using a dilute rubidium vapour cell and observed that the velocities of the delayed output pulses were in good agreement with the values $d\omega/dK$ obtained from the linear dispersion (polariton) theory. In their experiments, however, no steady propagation was observed; instead of this, pulses were steepened after passage through the cell and complicated asymmetric pulse envelopes developed.

Grishkowsky *et al* (1973) explained this self-steepening using the adiabatic-following model. When the condition $(\omega-\omega_0)\tau \gg 1$ is satisfied, the dipole moment follows adiabatically the electric field, i.e. $u \gg v$. In this case, by setting $\dot{v}=\dot{w}=0$ in (2.7)–(2.10), u is approximately given as a functional of $\hat{E}$. This expression for u is then inserted, together with $v=(\omega-\omega_0+\dot{\phi})^{-1}\dot{u}$, into the Maxwell equation, which leads to two coupled differential equations for $\hat{E}$ and $\dot{\phi}$. These equations contain the dispersion relation, the group velocity and the phase modulation in field-dependent forms, which describe the initial stage of the self-steepening.

The equations thus obtained, however, do not admit any steady solution of $\hat{E}$ and $\dot{\phi}$. This is due to the fact that small quantities of the order of $[(\omega-\omega_0)\tau]^{-1}$ including $\dot{\phi}$ are not taken into account carefully in the adiabatic-following model. A pulse should be formed on a balance of such small quantities, as has been shown in § 5. Of course, the adiabatic-following model is sufficient to describe the initial stage of the self-steepening. Grishkowsky *et al* confined themselves to such a stage, because the cell they used was too short to realize a steady propagation. Further experiments using a longer cell are strongly desirable.

Inside the polariton gap, on the other hand, the adiabatic-following model can give a steady pulse solution. A pulse is obtained by retaining only the lowest-order quantities and it also exists in the limit of $\tau \to \infty$.

It is not surprising that a long pulse can propagate inside the polariton gap where the dielectric function is negative. As has been seen in § 3, even the plane wave can exist as a steady solution inside the gap if the amplitude is large enough. The relation of the pulse solution to the non-linear polariton is as follows. The electric field given by (5.16) takes the form

$$\hat{E} \propto 1-\frac{\Delta}{2(2-\Delta)}\xi^2+\ldots \approx \cos\left(\frac{\omega}{c}(\tilde{\epsilon}(\omega,\hat{E}_{\max}))^{1/2}z\right) \qquad (6.1)$$

near the pulse peak. Here we have used the fact that the variable ξ becomes only spatial-like as $\xi \approx -z/V\tau$ for large τ, and equations (3.2) and (5.20). Since $\tilde{\epsilon}(\omega,\hat{E}_{\max})$ is the field-dependent dielectric function and is positive, (6.1) expresses a standing wave of non-linear polariton. Unlike the linear case, however, this standing wave cannot exist as a whole. This is because, if one goes away from the peak of the cosine, one can no longer use $\tilde{\epsilon}(\omega, \hat{E})$ with $\hat{E} = \hat{E}_{\max}$. In fact, the tail of $\hat{E}$ decays like

$$\hat{E} \propto \exp(-|\xi|) \approx \exp\left(-\frac{\omega}{c}(-\tilde{\epsilon}(\omega, 0))^{1/2}|z|\right), \tag{6.2}$$

which is just the linear solution with negative dielectric function $\tilde{\epsilon}(\omega, 0)$. Thus, a long pulse inside the polariton gap may be interpreted as a sort of standing wave of non-linear polariton which is bounded on both sides by exponentially decaying tails and propagates very slowly.

Gurovich and his coworkers (Gurovich and Karpman 1969, Gurovich *et al* 1969) treated a problem similar to ours for liquids and plasmas with negative dielectric constant, and pointed out that a non-linear solitary wave of low amplitude shows a stable propagation as a consequence of the instability of a non-linear plane wave. By analogy with their results, it may be expected that the plane wave is in fact unstable also in our case, while the single pulse is stable. The same may be said of the solutions outside the polariton gap.

In conclusion, we have developed a systematic method of treating the non-linear propagation of a coherent light pulse in a dielectric medium. It has been shown that, besides the usual SIT, steady pulse solutions exist also in the case where the pulse width is much longer than the reciprocal of the polariton gap frequency. A long pulse outside the gap behaves as a polariton-soliton, and a pulse inside the gap propagates very slowly as a sort of standing wave of non-linear polariton. Although our conclusions have been obtained for a simplified model without taking into consideration realistic conditions such as spatial dispersion, relaxation times, surface effects and so on, they suggest that the steady propagation of a light pulse may also be observed in crystals. A pulse longer than the order of a picosecond then behaves as a solitary wave of polariton, while a shorter pulse will realize the SIT. A steady propagation of a polariton-soliton may be observed also in gases, if an off-resonant pulse and a sufficiently long cell are used.

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Appendix

We explain here in some detail how to solve our coupled non-linear equations. Only the case of a long pulse outside the polariton gap is presented, since the other cases can be treated in a similar way. By neglecting all the terms involving s and using (5.10), equations (5.3)–(5.7) are rewritten as follows:

$$\gamma \ddot{E} - (\alpha + \beta\dot{\phi} + \gamma\dot{\phi}^2)E = -u, \tag{A.1}$$

$$\gamma\ddot{\phi}E + (\beta + 2\gamma\dot{\phi})\dot{E} = -v, \tag{A.2}$$

$$\epsilon \dot{u} = (1+\epsilon\dot{\phi})v, \tag{A.3}$$

$$\epsilon \dot{v} = -(1+\epsilon\dot{\phi})u + \Delta^{-1}Ew, \tag{A.4}$$

$$\epsilon \dot{w} = -\Delta^{-1}Ev. \tag{A.5}$$

Observing the orders of magnitude $\alpha \sim O(1)$, $\beta \sim O(\epsilon)$ and $\gamma \sim O(\epsilon^2)$, which are seen from (5.12), let us expand the functions in the above equations in the following form:

$$\begin{aligned} E &= \epsilon^{\nu}(E_0+\epsilon^2E_2+\ldots), & \dot{\phi} &= \epsilon\theta_1+\epsilon^3\theta_3+\ldots, \\ u &= \epsilon^{\nu}(u_0+\epsilon^2u_2+\ldots), & v &= \epsilon^{\nu}(\epsilon v_1+\epsilon^2 v_3+\ldots), \\ w &= w_0+\epsilon^2w_2+\ldots, \end{aligned} \tag{A.6}$$

where $E_0 \neq 0$ is assumed. By inserting these expansions and (5.12) into (A.1)–(A.4) and retaining only the lowest-order terms in each equation, the following simple equations are obtained:

$$\begin{aligned} -\alpha_0 E_0 &= -u_0, & \beta_1\dot{E}_0 &= -v_1, \\ \dot{u}_0 &= v_1, & 0 &= -u_0+\Delta^{-1}E_0w_0, \end{aligned} \tag{A.7}$$

where $\alpha_0 = -\Delta^{-1}$ and $\beta_1 = \Delta^{-1}$. Elimination of u_0 from the first and fourth equations leads to

$$E_0(w_0+1) = 0, \qquad \text{thus} \qquad w_0 = -1. \tag{A.8}$$

Use of this result in (A.5) gives

$$\dot{w}_2 = -\epsilon^{2\nu-2}\Delta^{-1}E_0v_1, \tag{A.9}$$

from which $\nu = 1$ is determined. Independent relations hitherto derived are

$$u_0 = -\Delta^{-1}E_0, \qquad v_1 = -\Delta^{-1}\dot{E}_0, \qquad w_2 = \tfrac{1}{2}\Delta^{-2}E_0^2; \tag{A.10}$$

the last relation has been obtained by integrating (A.9). The next order terms in (A.1)–(A.4) are

$$\gamma_2\ddot{E}_0 - (\alpha_2+\beta_1\theta_1)E_0 - \alpha_0E_2 = -u_2, \tag{A.11}$$

$$\gamma_2(\dot{\theta}_1E_0 + 2\theta_1\dot{E}_0) + \beta_1\dot{E}_2 + \beta_3\dot{E}_0 = -v_3, \tag{A.12}$$

$$\dot{u}_2 = v_3 + \theta_1v_1, \tag{A.13}$$

$$\dot{v}_1 = -u_2 - \theta_1u_0 + \Delta^{-1}(E_0w_2 - E_2), \tag{A.14}$$

where

$$\alpha_2 = \frac{4\Delta-3}{4\Delta(\Delta-1)}, \qquad \beta_3 = -\frac{1}{\Delta}, \qquad \gamma_2 = \frac{1}{4\Delta(\Delta-1)}. \tag{A.15}$$

By inserting (A.10) into (A.11) and (A.14), and eliminating $u_2 - \Delta^{-1}(\theta_1E_0 - E_2)$ from them, the differential equation

$$\ddot{E}_0 = E_0 - \frac{2(\Delta-1)}{\Delta^2(4\Delta-3)}E_0^3 \tag{A.16}$$

is finally derived; integration of which gives the solution in (5.13). The other functions u_0, v_1 and w_2 are then obtained from (A.10). In order to obtain θ_1, we only have to eliminate first v_3 from (A.12) and (A.13) and then $\dot{u}_2 + \Delta^{-1}\dot{E}_2$ using the differential form of (A.14).

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