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Steady propagation of a coherent light pulse in a dielectric medium: II The effect of spatial dispersion†

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Abstract. The effect of spatial dispersion or exciton formation on the steady propagation of a coherent light pulse in a dielectric medium is studied. The optical Bloch equation for a lattice of atomic dipoles coupled to one another by an exchange-type interaction is solved simultaneously with the Maxwell equation by the method of power-series expansion developed in the preceding paper. Two kinds of pulse solutions are obtained in general: one propagating by the medium of the radiation field and the other by excitation transfer between atomic dipoles. The former solution is SIT-like in the short pulse limit, but a polariton–soliton or a standing wave of non-linear polariton if the pulse width is long. The latter is an exciton–soliton containing little photon component. When the pulse width is extremely long, there appears a polariton–soliton of a new type in which the two propagation mechanisms are mixed. It is found that the spatial dispersion prevents the polariton–soliton solutions from existing in a certain frequency range. This effect is explained on the basis of the functional behaviour of the dispersion relation of the non-linear polariton.

1. Introduction

With the aim of investigating how the steady propagation of a coherent light pulse would appear in crystals, although it has been extensively studied for gases, we have developed in a previous paper (Akimoto and Ikeda 1977, referred to as I hereafter) a systematic method of treating such a phenomenon in a dielectric medium of non-interacting two-level atoms, paying special attention to the effect of polariton formation. In the absence of spatial dispersion the effect of polariton formation is measured by the magnitude of the polariton gap, i.e. the frequency range in which any plane wave of low amplitude cannot propagate. This magnitude is proportional to the density of atomic dipoles in the medium and amounts to about 10^{12} Hz in crystals. In the case when the spread of Fourier components of frequency contained in a light pulse covers the polariton gap completely, we call the pulse *short*. In the opposite case where this spread lies completely inside or completely outside the gap, we call the pulse *long*. The pulse width boundary between these two cases lies in the microsecond range in gases and in the picosecond range in crystals. It has been shown in I that, besides a short pulse

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which is merely the usual pulse of self-induced transparency (SIT) (McCall and Hahn 1969, Lamb 1971), a long pulse can also propagate steadily in the medium as a polariton–soliton.

In crystals in which the atomic dipoles are dense and regularly arranged on the lattice, however, a long pulse should necessarily be affected not only by polariton formation but also by broadening of the resonant frequency due to interaction between dipoles, that is by exciton formation. The exciton can propagate in the medium by means of its own transfer mechanism even if it does not couple to the electromagnetic field. Such coupling forms a polariton. Unlike the case where the exciton effect is absent, however, the dispersion relation of the polariton in this case has no clear-cut gap and shows the existence of two modes having different wavenumbers for one frequency in a certain frequency range. For higher frequencies, these two modes correspond to the two types of propagation; one is almost photon-like and the other exciton-like. Near the resonant frequency of the exciton with $\boldsymbol{K}=0$, on the other hand, both components of photon and exciton are mixed in a (or each) mode. The exciton formation is sometimes referred to as the effect of spatial dispersion, because it brings about non-local dielectric behaviour of the medium (Hopfield and Thomas 1963).

In the present paper we discuss how the effect of exciton formation or spatial dispersion is reflected on the steady propagation of a coherent light pulse. For this purpose we consider a model crystal in which the atomic dipoles couple to one another through an exchange-type interaction, and treat this crystal as a continuous dielectric medium. This model may be somewhat *ad hoc* and not fully explain real crystals in all their details. Nevertheless, as a standard model for studying spatial dispersion in the simplest form (see e.g. Anderson 1963), it is sufficiently relevant to our purpose. The present paper is therefore confined to the analysis of the various possible pulse solutions of this model.

The paper is organised as follows: In § 2 the optical Bloch equation which governs the motion of the macroscopic polarisation in such a medium is derived as an extension of the case of non-interacting atoms. After the homogeneous solution of this equation, coupled to the Maxwell equation, is discussed in § 3, pulse solutions are sought in the following two sections by the method of power-series expansion developed in I. Two kinds of solutions propagating by different mechanisms will be obtained in general: one propagating by the medium of the radiation field and the other by excitation transfer between atomic dipoles. The former solution is almost SIT-like in the short pulse limit, but a polariton–soliton or a standing wave of non-linear polariton if the pulse width is long. The latter solution, on the other hand, contains little photon component and behaves as a pulse of non-linear exciton. When the pulse width is extremely long, there appears near the resonant frequency of the exciton a polariton–soliton of a new type in which the two propagation mechanisms are mixed. A remarkable result is that the spatial dispersion prevents the polariton–soliton solutions from existing in a certain frequency range. This result will be explained in § 6 by showing that the behaviour of such a long pulse obeys a non-linear Schrödinger equation whose potential is closely related to the dispersion relation of non-linear polariton.

2. Hamiltonian and fundamental equations

Consider a crystal lattice of equivalent atoms each of which has a single excited state and couples to other atoms through an exchange-type dipole–dipole interaction. The

Hamiltonian of this crystal is written as

$$H_0 = \sum_l \tfrac{1}{2}\hbar\omega_0\sigma_l^{(3)} - \tfrac{1}{2}\sum_{l,m} J(\boldsymbol{r}_{lm})(\boldsymbol{d}_l \cdot \boldsymbol{d}_m)/d^2, \tag{2.1}$$

where ω_0 is the resonant frequency of the atom, $\boldsymbol{d}_l$ the dipole moment operator of atom l, d its magnitude, $J(\boldsymbol{r}_{lm})$ the interaction energy between atoms l and m, which is assumed to be non-negative, and $\sigma_l^{(i)}$ the Pauli matrix of atom l. The operator $\boldsymbol{d}_l$ can be regarded as representing two components of a pseudo-spin operator, the third component of which is the population difference between the ground and excited states of the atom, so that $\boldsymbol{d}_l$ is related to the Pauli matrices as $d_l^{(i)} = d\sigma_l^{(i)}$ $(i = 1, 2)$. The excitation spectrum of the crystal in which one atom is excited is given by

$$\hbar\omega_{\boldsymbol{K}} = \hbar\omega_0 - 2\sum_l J(\boldsymbol{r}_l)\exp(\mathrm{i}\boldsymbol{K} \cdot \boldsymbol{r}_l)$$

$$\equiv \hbar\omega_0 + J(\boldsymbol{K}), \tag{2.2}$$

which represents in our case the exciton dispersion relation.

The Hamiltonian for the interaction between the crystal and the radiation field is, in the dipole approximation, written as

$$H_1 = -\sum_l \boldsymbol{d}_l \cdot \boldsymbol{E}(\boldsymbol{r}_l), \tag{2.3}$$

where $\boldsymbol{E}(\boldsymbol{r})$ is the operator of oscillating electric field at the point $\boldsymbol{r}$.

The Heisenberg equation of motion for $\sigma_l^{(+)} = \sigma_l^{(1)} + \mathrm{i}\sigma_l^{(2)}$, driven by $H_0 + H_1$, is

$$\mathrm{i}\dot{U}(t, \boldsymbol{r}_l) = (\omega - \omega_0)U(t, \boldsymbol{r}_l) - (2d/\hbar)E(t, \boldsymbol{r}_l)w(t, \boldsymbol{r}_l)$$

$$-\Big(2\sum_{l' \neq l} J(\boldsymbol{r}_{l'l})\exp[-\mathrm{i}\boldsymbol{K} \cdot (\boldsymbol{r}_{l'} - \boldsymbol{r}_l)]U(t, \boldsymbol{r}_{l'})\Big)\, w(t, \boldsymbol{r}_l), \tag{2.4}$$

where rapidly oscillating factors have been separated by setting

$$E^{(+)} = E(t, \boldsymbol{r})\exp[\mathrm{i}(\omega t - \boldsymbol{K} \cdot \boldsymbol{r})], \tag{2.5}$$

$$\sigma_l^{(+)} = U(t, \boldsymbol{r}_l)\exp[\mathrm{i}(\omega t - \boldsymbol{K} \cdot \boldsymbol{r}_l)], \qquad \sigma_l^{(3)} = w(t, \boldsymbol{r}_l). \tag{2.6}$$

Since we are concerned with a coherent phenomenon, operators E, U and w are assumed to vary sufficiently slowly over the lattice spacing. Thus we can decorrelate the expectation values of both sides of (2.4). Let us hereafter denote the expectation values by the same notation as for the operators themselves and regard them as continuous functions of $\boldsymbol{r}$, dropping the suffix l. Using the expansion

$$U(t, \boldsymbol{r}') = \exp[(\boldsymbol{r}' - \boldsymbol{r}) \cdot \boldsymbol{\nabla}]U(t, \boldsymbol{r}), \tag{2.7}$$

the summation over l' in (2.4) is expressed as $-J(\boldsymbol{K} + \mathrm{i}\boldsymbol{\nabla})U(t, \boldsymbol{r}_l)$; the equation of motion for $U(t, \boldsymbol{r})$ thus becomes

$$\mathrm{i}\dot{U}(t, \boldsymbol{r}) = (\omega - \omega_0)U(t, \boldsymbol{r}) - [\kappa E(t, \boldsymbol{r}) - J(\boldsymbol{K} + \mathrm{i}\boldsymbol{\nabla})U(t, \boldsymbol{r})]w(t, \boldsymbol{r}), \tag{2.8}$$

where $\kappa = 2d/\hbar$. The equation of motion for $w(t, \boldsymbol{r})$ is also derived in the same way. This set of equations forms the optical Bloch equation in the presence of spatial dispersion.

We adopt here the effective mass approximation in which $J(\boldsymbol{K})$ is replaced by a quadratic function of $\boldsymbol{K}$, i.e.

$$\hbar^{-1}J(\boldsymbol{K}) = -j_0 + j_2\boldsymbol{K}^2, \tag{2.9}$$

confining ourselves to an isotropic medium. The meaning of j_0 and j_2 in real crystals will be mentioned in § 6. We further set

$$E(t, \boldsymbol{r}) = \hat{E}(t, \boldsymbol{r}) \exp[\mathrm{i}\phi(t, \boldsymbol{r})], \tag{2.10}$$

$$U(t, \boldsymbol{r}) = [u(t, \boldsymbol{r}) + \mathrm{i}v(t, \boldsymbol{r})] \exp[\mathrm{i}\phi(t, \boldsymbol{r})], \tag{2.11}$$

where $\hat{E}$ is the slowly varying envelope of the electric field and ϕ denotes the phase modulation depending on the position of the envelope. It can be seen that u and v are the components of dipole moment which oscillate in phase and $\pi/2$ out of phase with the electric field, respectively. The conservation law

$$[u(t, \boldsymbol{r})]^2 + [v(t, \boldsymbol{r})]^2 + [w(t, \boldsymbol{r})]^2 = 1 \tag{2.12}$$

can be proved to hold.

Since we are interested in one-dimensional steady propagation, all the envelopes E, ϕ, u, v and w are assumed to be functions of only $t - z/V$, where z is the direction of propagation and V its velocity. By this assumption, the optical Bloch equation is further reduced to

$$\dot{u} = (\omega - \omega_0 - j_0 w + \dot{\phi})v - (j_2/V^2)[\ddot{v} + 2(KV + \dot{\phi})\dot{u} - (KV + \dot{\phi})^2 v + \ddot{\phi}u]w, \tag{2.13}$$

$$\dot{v} = -(\omega - \omega_0 - j_0 w + \dot{\phi})u + (j_2/V^2)[\ddot{u} - 2(KV + \dot{\phi})\dot{v} - (KV + \dot{\phi})^2 u - \ddot{\phi}v]w + \kappa\hat{E}w, \tag{2.14}$$

$$\dot{w} = (j_2/V^2)[u\ddot{v} - \ddot{u}v + 2(KV + \dot{\phi})(u\dot{u} + v\dot{v}) + \ddot{\phi}(u^2 + v^2)] - \kappa\hat{E}v, \tag{2.15}$$

where the dots denote differentiation with respect to $\zeta = t - z/V$, and $K \equiv |\boldsymbol{K}|$. Under the same assumption, the Maxwell equation for $\hat{E}$ takes the following form:

$$\left(\frac{1}{V^2} - \frac{1}{c^2}\right)\ddot{\hat{E}} - \left[(K^2 - k^2) + 2\left(\frac{K}{V} - \frac{k}{c}\right)\dot{\phi} + \left(\frac{1}{V^2} - \frac{1}{c^2}\right)\dot{\phi}^2\right]\hat{E}$$
$$= \frac{2\pi N\hbar\kappa}{c^2}[\ddot{u} - 2(\omega + \dot{\phi})\dot{v} - (\omega + \dot{\phi})^2 u - \ddot{\phi}v], \tag{2.16}$$

$$\left(\frac{1}{V^2} - \frac{1}{c^2}\right)\ddot{\phi}\hat{E} + 2\left[\frac{K}{V} - \frac{k}{c} + \left(\frac{1}{V^2} - \frac{1}{c^2}\right)\dot{\phi}\right]\dot{\hat{E}}$$
$$= \frac{2\pi N\hbar\kappa}{c^2}[\ddot{v} + 2(\omega + \dot{\phi})\dot{u} - (\omega + \dot{\phi})^2 v + \ddot{\phi}u], \tag{2.17}$$

where k is the wavenumber of light in vacuum, i.e. $k = \omega/c$. These two equations have been derived from the Maxwell equation in vector form by separating the components which oscillate in phase and $\pi/2$ out of phase with $\boldsymbol{E}$, as has been done in I. Our problem is now to solve (2.13)–(2.17) 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 $w = -1$. It is also assumed that the phase modulation $\dot{\phi}$ vanishes in this limit, so that the frequency ω and the wavenumber K are those defined at the pulse tail.

3. Non-linear polariton involving the effect of spatial dispersion

Let us first discuss the homogeneous solution of (2.13)–(2.17). By setting all the derivative terms equal to zero, these equations are reduced to

$$u=\left[\left(\frac{cK}{\omega}\right)^2-1\right]\frac{\kappa\hat{E}}{2\pi N\hbar\kappa^2},\qquad v=0,\qquad w=\frac{(\omega-\omega_0)u}{\kappa\hat{E}+(j_0-j_2K^2)u}. \tag{3.1}$$

Elimination of u from these equations gives the dispersion relation

$$\left(\frac{cK}{\omega}\right)^2=1+\frac{2\pi N\hbar\kappa^2\,w}{\omega-\omega_0-(j_0-j_2K^2)w}. \tag{3.2}$$

This expression involves the effect of spatial dispersion, i.e. the K-dependence of the denominator in the right-hand side, and further depends on the population inversion w.

In the limit of weak excitation, $w=-1$, (3.2) is reduced to the dispersion relation of the usual (linear) polariton, which is a mixed mode of the photon $\omega=cK$ and the exciton whose dispersion is given by

$$\omega=\omega_0-j_0+j_2K^2. \tag{3.3}$$

The frequency difference between the polariton and the exciton at $K=0$ is $2\pi N\hbar\kappa^2$; we call this frequency the polariton gap frequency in the present paper also and take it as a measure of the effect of polariton formation, although there no longer exists a clear-cut gap as in the case where the spatial dispersion is absent.

The w-dependence of the right hand side of (3.2) can be interpreted as a superposition of the following two effects. First, the dispersion relation of the exciton changes depending on w as

$$\omega=\omega_0+(j_0-j_2K^2)w. \tag{3.4}$$

Owing to this change, the dispersion curve of the polariton for $K<K_0\equiv(j_0/j_2)^{1/2}$ is pushed up towards the high-frequency side as the excitation increases, while that for $K>K_0$ is pushed down towards the low-frequency side. Secondly, the coupling of exciton and photon decreases as if the dipole matrix element itself decreased from κ to $\kappa|w|^{1/2}$. This decrease reduces the polariton gap frequency to $2\pi N\hbar\kappa^2|w|$ and brings the two branches of the dispersion curve closer to each other. These two effects act in opposite directions in some cases and in the same direction in other cases. Direct calculation of the differential coefficient $\partial\omega/\partial w$ for fixed K, however, shows that the resultant shift of the dispersion curve due to these two effects is as follows: for $\omega<\omega_0$, both the upper and lower branches shift towards the high-frequency side with the increase of excitation, at least when the excitation is weak, while for $\omega>\omega_0$, they shift towards the low-frequency side.

The population inversion w is related to the field strength $\hat{E}$ through the relation

$$(\kappa\hat{E})^2=w^{-2}(1-w^2)[\omega-\omega_0-(j_0-j_2K^2)w]^2, \tag{3.5}$$

which is derived from the third equation of (3.1) by making use of the conservation law (2.12). Elimination of w from (3.2) and (3.5) completes the field-dependent dispersion relation $\omega(K,\hat{E})$ of the non-linear polariton, although its explicit expression is too complicated to be given here. Such a field dependence has been pointed out also by Haken and Schenzle (1973) and by Inoue (1974)†. We should only note here that both branches of the dispersion curve shift, also with the increase of $\hat{E}$, upwards for $\omega<\omega_0$ and downwards for $\omega>\omega_0$. This can be proved by showing that $|w|$ is inversely

† It is pointed out by Armstrong (1975) that there is a similar (unpublished) discussion by R K Bullough.

proportional to $\hat{E}$ for fixed K, at least when $\hat{E}$ is small, or more directly, by calculating the differential coefficient $\partial\omega/\partial(\hat{E}^2)$. It will be seen in § 6 that this upward or downward shift plays an essential role in the existence of a steady pulse of long width.

4. Dispersion relation of a growing wave

Following the procedure developed in I, we first consider the behaviour of the pulse tail and then solve the coupled non-linear equations to obtain the overall shape of the pulse. The pulse tail where the excitation is very weak can be sufficiently described by the linearised equations, which are obtained by setting $\dot{\phi}=0$ and $w=-1$ in (2.13)–(2.17). Those linearised equations admit exponentially growing (or decaying) solutions $\hat{E}\propto\exp[\pm(t-z/V)/\tau]$ in general, besides the plane wave solution which corresponds to the usual polariton. The growth (or decay) rate τ, which is an integral constant, characterises the shape of the tail and will also give the measure of the pulse width, when a slowly varying pulse solution can be obtained after taking into account the non-linearity. The linearised equations also determine the dispersion relation of the carrier wave and the pulse velocity as functions of the parameter τ. We call τ the pulse width hereafter.

Before proceeding further, let us rewrite our fundamental equations (2.13)–(2.17) in non-dimensional forms to make them easier to manipulate. By introducing a dimensionless field amplitude

$$E=\hat{E}/2\pi N\hbar\kappa, \tag{4.1}$$

and choosing a dimensionless time

$$\xi=\zeta/\tau=(t-z/V)/\tau \tag{4.2}$$

as the variable, equations (2.16)–(2.17) and (2.13)–(2.15) lead to

$$\gamma\ddot{E}-(\alpha+\beta\dot{\phi}+\gamma\dot{\phi}^2)E=-u, \tag{4.3}$$

$$\gamma\ddot{\phi}E+(\beta+2\gamma\dot{\phi})\dot{E}=-v, \tag{4.4}$$

$$\Lambda\dot{u}=[\Delta-j'(w+1)+\Lambda\dot{\phi}]v-j[\gamma\ddot{v}+(\beta+\gamma\dot{\phi})\dot{u}-(1+\alpha+\beta\dot{\phi}+\gamma\dot{\phi}^2)v+\gamma\ddot{\phi}u]w, \tag{4.5}$$

$$\Lambda\dot{v}=-[\Delta-j'(w+1)+\Lambda\dot{\phi}]u+j[\gamma\ddot{u}-(\beta+\gamma\dot{\phi})\dot{v}-(1+\alpha+\beta\dot{\phi}+\gamma\dot{\phi}^2)u-\gamma\ddot{\phi}v]w+Ew, \tag{4.6}$$

$$\Lambda\dot{w}=j[\gamma(u\ddot{v}-\ddot{u}v)+(\beta+2\gamma\dot{\phi})(u\dot{u}+v\dot{v})+\gamma\ddot{\phi}(u^2+v^2)]-Ev. \tag{4.7}$$

Here the dots denote differentiation with respect to ξ, and Δ and Λ are the frequency measured from ω_0-j_0 and the reciprocal pulse width, respectively, both of which are scaled by the polariton gap frequency as follows:

$$\Delta=(\omega-\omega_0+j_0)/2\pi N\hbar\kappa^2, \qquad \Lambda=\tau^{-1}/2\pi N\hbar\kappa^2. \tag{4.8}$$

The interaction constants have also been scaled as

$$j'=j_0/2\pi N\hbar\kappa^2, \qquad j=j_2k^2/2\pi N\hbar\kappa^2, \tag{4.9}$$

where k is assumed to be a material constant $(\omega_0-j_0)/c$, because we are concerned with a phenomenon near the resonant frequency of the exciton. Generally, j' is of the order of unity or larger, while j is much smaller ($\sim10^{-4}$). It should be noted that, in spite of its smallness, j cannot be discarded at the present stage, because it has an essential role of introducing a new branch of the polariton dispersion and the corresponding non-linear pulse solution. The coefficients α, β and γ are defined as

$$\alpha = (K/k)^2 - 1, \qquad \beta = 2(\omega\tau)^{-1}(cK/Vk), \qquad \gamma = (\omega\tau)^{-2}(c/V)^2, \tag{4.10}$$

and are connected with each other by the relation

$$4(\alpha+1)\gamma = \beta^2. \tag{4.11}$$

Note that all terms involving the small parameter $(\omega\tau)^{-1}$ have been neglected in (4.3)–(4.7) except those which involve it implicitly through (4.10). As has been seen in I, this procedure of approximation is justifiable in our treatment except in the ultra-short pulse case such that $\tau^{-1} \gtrsim (2\pi N\hbar\kappa^2\omega)^{1/2}$, as far as we are concerned with the lowest-order solutions.

Now let us return to the pulse tail. If we set

$$E = E_0 \exp(\pm\xi), \qquad u = u_0 \exp(\pm\xi), \qquad v = v_0 \exp(\pm\xi) \tag{4.12}$$

in those equations which are obtained by linearising (4.3)–(4.7), we have a set of simultaneous linear equations for E_0, u_0 and v_0. From the condition that these quantities should be non-zero, coefficients α, β and γ are determined as functions of Δ and Λ in such a way that β is first given by the two real roots of the quartic equation

$$4j\beta^2(j\beta-\Lambda)^2 + [(\Delta-j)^2 + \Lambda^2 + 4j]\beta(j\beta-\Lambda) + \Lambda^2 = 0, \tag{4.13}$$

and that $\alpha - \gamma$ is then obtained from the relation

$$(2j\beta-\Lambda)(\alpha-\gamma) = (\Delta-j)\beta. \tag{4.14}$$

α and γ are finally determined by (4.11).

In figure 1 the normalised wavenumbers $K/k = (\alpha+1)^{1/2}$ of a growing wave are plotted as functions of Δ for various values of the parameter Λ. The following characteristics can be seen: (i) For any set of values of Δ and Λ, there exist two real wavenumbers. (ii) When Λ is large, i.e. τ is small, one branch (branch A) is almost photon-like, while the other (branch B) has a large wavenumber, its asymptotes in the limit of high and low frequencies being the dispersion curve of the exciton and the Δ axis, respectively. These two branches cross each other at a certain frequency. (iii) When $\Lambda = 2j^{1/2}$, the two branches coalesce at $\Delta = j$. As Λ changes across $2j^{1/2}$, the branches in the frequency range $\Delta > j$ exchange their partners in the range $\Delta < j$, so that when $\Lambda < 2j^{1/2}$, the asymptotes in the low-frequency limit become the Δ axis for branch A and $cK/\omega = 1$ for branch B, respectively. (iv) When ζ becomes smaller, branch A approaches the dispersion curve of the upper polariton, but it still has a real wavenumber also in the frequency range $\Delta < 1$. Branch B, on the other hand, approaches the dispersion curve of the lower polariton.

5. Pulse solutions

In this section we solve the coupled non-linear equations (4.3)–(4.7) by the method of power-series expansion developed in I. As has been shown in the preceding section, the two branches of the dispersion relation of the carrier wave show different behaviour according as $\Lambda \ll 2j^{1/2}$ or $\Lambda \gg 2j^{1/2}$. It is therefore reasonable to separate the cases first according to this criterion before classifying short and long pulses.

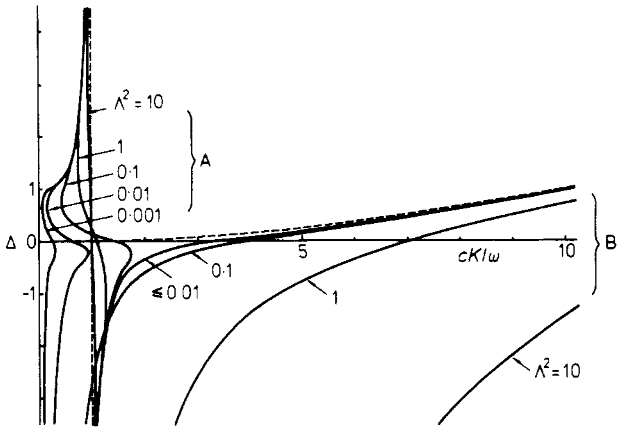

Figure 1. Dispersion relation of the carrier wave at the pulse tail (a growing wave). The ordinate Δ and the parameter Λ denote the frequency measured from $\omega_0 - j_0$ and the reciprocal of the pulse width (the growth rate), respectively, scaled by $2\pi N\hbar\kappa^2$. The dispersion relation does not depend strongly on the material constant $2\pi N\hbar\kappa^2(\omega_0 - j_0)^{-1}$, which has been approximately set equal to zero. The parameter j which measures the degree of exciton dispersion has been chosen to be 10^{-2}. We call the dispersion curves which tend to that of the photon in the limit of high frequency 'branch A', and those which tend to that of the exciton 'branch B'. The dispersion curves of the photon and the exciton are indicated by the broken curves.

5.1. $\Lambda \gg 2j^{1/2}$; *branch A*

Coefficients α, β and γ in this case are the same as those obtained in I, except that the definition of Δ is different from that in I by j'. (For explicit expressions of α, β and γ, see (5.12), (5.14) and (5.25) in I.) The introduction of j', however, gives rise not only to the shift of Δ but also to a drastic change in the pulse solutions through the term $j'(w+1)$ in (4.5) and (4.6). The effect of j, on the other hand, does not appear to the lowest order; this means that the excitation transfer between atomic dipoles has in the present case no effect on the pulse propagation. Three cases are discussed separately below.

(i) *Short pulse.* To the lowest order of the small parameter $\epsilon = \Lambda^{-1}$, the solutions of E, v and w are the same as the SIT solutions (given in (5.26) in I) and are not affected by j'; this is for the same reason as why they are not affected by the existence of the polariton gap. The effect of j' appears first in the leading terms of u and ϕ, which are of the order of ϵ, and in higher-order terms of E, v and w. Haken and Schenzle (1973), who treated essentially the short pulse case, have obtained an expression of E involving j'. The terms obtained by expanding their expression with respect to Λ^{-1} and some additional terms which could not be involved in their expression owing to the adopted approximation will appear in our higher-order terms.

(ii) *Long pulse outside the polariton gap.* Equations (4.3)–(4.7) are solved in the same way as has been done in the appendix in I, and give the *polariton–soliton* solution

$$E = \left(\frac{(4\Delta - 3)\Delta}{(\Delta - 1)(\Delta - j')}\right)^{1/2} \Lambda \operatorname{sech} \xi \tag{5.1}$$

to the lowest order of $\epsilon = \Lambda/\Delta$. An important effect of j' is that if $j' > 1$, no pulse solution does exist in the frequency range $1 < \Delta < j'$.

(iii) *Long pulse inside the polariton gap.* To the lowest order of $\epsilon = \Lambda/\Delta$, equations (4.3)–(4.7) are reduced to

$$\gamma \ddot{E} - \alpha E = -u, \qquad v = \dot{\phi} = 0, \qquad [\Delta - j'(w+1)]u - Ew = 0. \tag{5.2}$$

Although it is difficult to obtain E by solving these equations directly, the existence of a pulse solution in the case of $\Delta > j'$ can easily be proved by the following simple consideration†. Note at first that the third equation of (5.2) can be written as

$$u = [\tilde{\epsilon}(\omega, E) - 1]E, \tag{5.3}$$

where $\tilde{\epsilon}(\omega, E)$ is the field-dependent dielectric function which is obtained by eliminating w from (3.2) and (3.5). Equation (5.3) is derived by using the fact that the K dependence in $\tilde{\epsilon}(\omega, E)$ can be discarded in the present case where $\alpha \simeq -1$. Use of (5.3) in the first equation of (5.2) then leads to

$$(-1 + \Delta^{-1})\dot{E}^2 = -2\int_0^E \tilde{\epsilon}(\omega, E')E' \, \mathrm{d}E'. \tag{5.4}$$

When $E \simeq 0$, i.e. $w \simeq -1$, it follows from (3.2) that $\tilde{\epsilon}(\omega, E) \simeq 1 - \Delta^{-1}$, which gives $\dot{E}^2 \simeq E^2 > 0$. In the limit $E \to \infty$, i.e. $w \simeq 0$, on the other hand, it is obvious that $\tilde{\epsilon}(\omega, E) \simeq 1$, which gives $\dot{E}^2 \simeq (1 - \Delta^{-1})^{-1}E^2 < 0$. Therefore $\dot{E}^2$ is positive for $0 < E < E_{\max}$ ($E_{\max}$ finite), so that E takes a pulse shape with peak value $E_{\max}$.

It is found from (5.4) that the form of E near the peak is

$$E \simeq E_{\max} \cos[(\omega/c)(\tilde{\epsilon}(\omega, E_{\max}))^{1/2} z]. \tag{5.5}$$

Here $\gamma = (c/V\omega\tau)^2 \simeq -1 + \Delta^{-1}$ has been used in $\xi \simeq -z/V\tau$ for large τ. Expression (5.5) indicates that, in the same way as the long pulse inside the polariton gap discussed in I, the pulse in the present case can also be interpreted as a sort of *standing wave of non-linear polariton*, which is bounded on both sides by the tails

$$E \propto \exp[-(\omega/c)(-\tilde{\epsilon}(\omega, 0))^{1/2}|z|], \tag{5.6}$$

and propagates very slowly. Especially, the solution of E for $\Delta \simeq 1$ is simplified as

$$E \simeq 2\left(\frac{1-\Delta}{1-j'}\right)^{1/2} \operatorname{sech} \xi. \tag{5.7}$$

5.2. $\Lambda \gg 2j^{1/2}$; *branch* B

This is a new branch of large wavenumber which appears as an effect of non-zero j. The coefficients α, β and γ are given as

$$\begin{aligned} \alpha &= (2j)^{-1}[(\Delta^2 + \Lambda^2)^{1/2} + \Delta] + \mathrm{O}(1), \\ \beta &= (2j)^{-1}\Lambda + \mathrm{O}(1), \\ \gamma &= (2j)^{-1}[(\Delta^2 + \Lambda^2)^{1/2} - \Delta] + \mathrm{O}(1). \end{aligned} \tag{5.8}$$

By using these expansions in (4.3) and (4.4), it is found that E is of the order of j, so that the coupling terms Ew and Ev in the optical Bloch equation are neglected to the lowest order. This means that the motion of the Bloch vector is no longer affected by the

† In the case of $\Delta < j'$, E is proved to be a double-peaked pulse which is singular at the peaks. The appearance of such a pulse corresponds to the fact that in this case, $\tilde{\epsilon}(\omega, E)$ in (5.3) becomes a two-valued function of E. The physical meaning of such a pulse is not clear.

electric field E. The optical Bloch equation without coupling to the electromagnetic field is solved in the appendix, and it is shown that the population inversion w has a pulse solution for any value of the pulse width. In contrast to the pulses discussed in § 5.1, such a pulse propagates entirely by means of the excitation transfer inherent in interacting atomic dipoles and may be called a *non-linear solitary wave of exciton* or an *exciton–soliton*. Propagation of this exciton–soliton in turn produces a weak electric field, which is obtained by using (A.11) in the Maxwell equation of the lowest order:

$$\ddot{E}-(\nu+\dot{\phi})^2E=-2j\nu\Lambda^{-1}u,$$
$$\ddot{\phi}E+2(\nu+\dot{\phi})\dot{E}=-2j\nu\Lambda^{-1}v, \tag{5.9}$$

or, in integral form,

$$E\exp[\mathrm{i}(\phi+\nu\xi)]=-2j\nu\Lambda^{-1}\int^{\xi}\mathrm{d}\xi'\int^{\xi'}\mathrm{d}\xi''(u+\mathrm{i}v)\exp[\mathrm{i}(\phi+\nu\xi'')], \tag{5.10}$$

where $\nu=[(\Delta^2+\Lambda^2)^{1/2}+\Delta]/\Lambda$.

5.3. $\Lambda\ll 2j^{1/2}$; *branch* A ($\Delta>1$) *and branch* B

In this case the dispersion relation of the carrier wave is very close to that of the linear polariton, as can be seen in figure 1. Pulses are necessarily *polariton–solitons* of extremely large width, since j is in general sufficiently small. By choosing $\epsilon=\Lambda$ as the expansion parameter, coefficients α, β and γ are expanded as

$$\alpha=(2j)^{-1}[\tilde{\Delta}\mp(\tilde{\Delta}^2+4j)^{1/2}]+\mathrm{O}(\epsilon^2)$$
$$\beta=(2j)^{-1}[1\mp\tilde{\Delta}/(\tilde{\Delta}^2+4j)^{1/2}]\epsilon+\mathrm{O}(\epsilon^3) \tag{5.11}$$
$$\gamma=\mathrm{O}(\epsilon^2),$$

where $\tilde{\Delta}\equiv\Delta-j$. The minus and plus signs in the parentheses correspond to branches A and B, respectively. By observing that the leading terms in (5.11) are of the same order as those in the case of long pulse outside the gap studied in I, equations (4.3)–(4.7) are solved in the same way as has been done there. Instead of (A.16) in I, however, the following differential equation is finally derived:

$$\ddot{E}_0=E_0+\frac{(\Delta-j')(\tilde{\Delta}^2+4j)[\tilde{\Delta}\mp(\tilde{\Delta}^2+4j)^{1/2}][\tilde{\Delta}+2j\mp(\tilde{\Delta}^2+4j)^{1/2}]}{4j^2[\tilde{\Delta}^2-\tilde{\Delta}+2j\pm(\tilde{\Delta}-\frac{1}{2})(\tilde{\Delta}^2+4j)^{1/2}]}E_0^3, \tag{5.12}$$

where ϵE_0 gives the leading term of E. For a pulse solution to exist, the coefficient of E_0^3 must be negative. This condition is satisfied only in the limited range of frequency: $\Delta>j'$ for branch A and $\Delta<\Delta_c$ or $\Delta>j'$ for branch B, where Δ_c is the root of

$$\tilde{\Delta}^2-\tilde{\Delta}+2j-(\tilde{\Delta}-\tfrac{1}{2})(\tilde{\Delta}^2+4j)^{1/2}=0, \tag{5.13}$$

and is approximately given by $\Delta_c\simeq(4j/3)^{1/2}$ for $j\ll 1$. This result is remarkable because the existence of a steady pulse in the frequency range $\Delta_c<\Delta<j'$ is prevented by the spatial dispersion. When a pulse exists, it has a hyperbolic secant shape $E=E_{\max}\operatorname{sech}\xi$, where $E_{\max}$ is approximately given as follows:

(i) For branch A ($\Delta>1$ and $\Delta>j'$) and branch B ($\Delta<0$ and $|\Delta|\gg j^{1/2}$), $E_{\max}$ takes the same form as (5.1).

(ii) For branch B ($|\Delta|\ll j^{1/2}$),

$$E_{\max}=j^{1/4}\Lambda/(2j')^{1/2}. \tag{5.14}$$

This is a polariton–soliton of a new type in which two propagation mechanisms, photon-like and exciton-like, are mixed almost in equal proportions.

(iii) For branch B ($\Delta > j'$),

$$E_{\max} = j\Lambda/[\Delta^3(\Delta - j')]^{1/2}. \tag{5.15}$$

This pulse corresponds to the weak electric field produced by an exciton–soliton of extremely long width.

5.4. $\Lambda \ll 2j^{1/2}$; *branch* A ($\Delta < 1$)

In terms of the expansion parameter $\epsilon = \Lambda$, coefficients α and γ are expanded as

$$\begin{aligned} \alpha &= -1 + O(\epsilon^2), \\ \gamma &= -1 - (2j)^{-1}[\tilde{\Delta} - (\Delta^2 + 4j)^{1/2}] \end{aligned} \tag{5.16}$$

β being the same as in (5.11). The lowest-order equations are now obtained by discarding all terms involving Λ and β in (4.3)–(4.7). Since it is still difficult to solve these equations, let us further simplify them by assuming $j \ll 1$. For the frequency range $0 < \Delta < 1$, thus simplified equations become the same as (5.2), and the discussions in § 5.1 are applicable also to the present case. For the frequency range $\Delta < 0$, on the other hand, the equations are different from (5.2), because γ diverges as $-\Delta/j$ for $j \ll 1$. In this case, E is of the order of j and the coupling terms Ew and Ev in the optical Bloch equation are neglected as small quantities. The two quantities $U \equiv (u + iv)\exp(i\phi)$ and $W \equiv w + 1$ are then obtained as functions of ξ from (A.9) and (A.11). Because $W_{\max} = 2 - O(\epsilon^2)$, however, the phase modulation of U, i.e. $d(\arg U)/d\xi$, diverges like ϵ^{-1} at the peak of W, while it is of the order of ϵ at the tail. Furthermore, $|\dot{U}| = [(d|U|/d\xi)^2 + |U|^2(d(\arg U)/d\xi)^2]^{1/2}$ consequently becomes of the order of ϵ^0 at the peak of W, while it is of the order of ϵ at the tail. This fact indicates that our method of ϵ-expansion cannot be applied to the present case, because our method assumes that the orders of magnitude of both the quantities of interest and their derivatives are estimated on the basis of their behaviour at the pulse tail and do not change over the whole range of ξ. This does not mean, however, that no pulse solution does exist. It cannot eliminate the possibility of a pulse with two or more scaling constants of time depending on its position.

6. Summary and discussion

In the preceding sections we have obtained steady solutions of a light pulse propagating in a dielectric medium with spatial dispersion. It has been shown that the linearised Maxwell and optical Bloch equations admit two growing-wave solutions which have different wavenumbers for any set of values of the carrier-wave frequency and the growth rate. Such a growing wave determines the behaviour of the tail of a pulse which may be formed by introducing the non-linearity. When the pulse width is not very long, i.e. $\Lambda \gg 2j^{1/2}$, there exist two types of pulse solutions propagating by different mechanisms: one propagates by the medium of the radiation field and is not much affected by the spatial dispersion, while the other is a pulse of non-linear exciton which contains little photon component and propagates by means of excitation transfer between atomic dipoles. In the short-pulse limit, the former solution becomes SIT like, but it is a polariton–soliton or a standing wave of non-linear polariton if the pulse width is long. When the pulse width is extremely long, i.e. $\Lambda \ll 2j^{1/2}$, on the other hand, there appears near the resonant frequency of the exciton, a polariton–soliton of a new type in which both propagation mechanisms are mixed. A remarkable result is that the polariton–soliton solutions cannot exist in a certain frequency range. We briefly show below that this fact can be explained from another viewpoint, relating to the functional behaviour of the dispersion relation of the non-linear (plane-wave) polariton.

The dispersion relation of the non-linear polariton depends on the degree of excitation, or equivalently, on the strength of the electromagnetic field $\mathscr{E}$ ($\equiv E\exp(\mathrm{i}\phi)$). Let us first expand this field-dependent dispersion relation $\omega(K, |\mathscr{E}|^2)$ in a power series around K_0 in the weak-field limit:

$$\omega(K, |\mathscr{E}|^2) \simeq \omega(K_0) + \omega'(K_0)(K-K_0) + \tfrac{1}{2}\omega''(K_0)(K-K_0)^2 + Q|\mathscr{E}|^2, \quad (6.1)$$

where

$$\omega'(K_0) = \frac{\partial\omega(K, |\mathscr{E}|^2)}{\partial K}\bigg|_{K=K_0, \mathscr{E}=0}, \qquad Q = \frac{\partial\omega(K, |\mathscr{E}|^2)}{\partial|\mathscr{E}|^2}\bigg|_{K=K_0, \mathscr{E}=0}, \quad (6.2)$$

and prepare a plane wave with ω and K connected with each other through (6.1):

$$\exp[\mathrm{i}(\omega t - Kz)] = \exp[\mathrm{i}(\omega(K_0)t - K_0 z)]\exp[\mathrm{i}(\omega'(K_0)t - z)(K-K_0)] \times \exp\{\mathrm{i}[\tfrac{1}{2}\omega''(K_0)(K-K_0)^2 + Q|\mathscr{E}|^2]t\}. \quad (6.3)$$

Here, the first exponential function on the right hand side oscillates rapidly and the latter two vary slowly. If the pulse is sufficiently weak, it is practically a superposition of these plane waves, so that its envelope $\mathscr{E}$ can be expressed as

$$\mathscr{E}(t, z') = \int \mathrm{d}(K-K_0)\exp[-\mathrm{i}(K-K_0)z']F(K_0, K-K_0) \times \exp\{\mathrm{i}[\tfrac{1}{2}\omega''(K_0)(K-K_0)^2 + Q|\mathscr{E}|^2]t\}, \quad (6.4)$$

where $F(K_0, K-K_0)$ is a weighting function and $z' = z - \omega'(K_0)t$ is the space coordinate moving with velocity $\omega'(K_0)$. It is then found that this pulse envelope satisfies the following non-linear Schrödinger equation

$$-\mathrm{i}\frac{\partial}{\partial t}\mathscr{E}(t, z') = \left(-\frac{\omega''(K_0)}{2}\frac{\partial^2}{\partial z'^2} + Q|\mathscr{E}(t, z')|^2\right)\mathscr{E}(t, z'). \quad (6.5)$$

If and only if $\omega''(K_0)Q < 0$, equation (6.5) has a stationary bound solution in z' space, i.e. a steady propagating pulse solution in (t, z) space (see e.g. Taniuti 1974). We have seen in § 3 that the dispersion curve of the polariton shifts upwards for $\omega < \omega_0$ and downwards for $\omega > \omega_0$ as the excitation increases. These shifts are indicated by the sign of Q. On the other hand, the sign of $\omega''(K)$ indicates the concavity and convexity of the dispersion curve; it is obvious that $\omega''(K) < 0$ for $\omega < \omega_c$ and $\omega''(K) > 0$ for $\omega > \omega_c$, where the inflexion point at which $\omega''(K) = 0$ is approximately given by $\omega_c = (4j/3)^{1/2}\, 2\pi N\hbar\kappa^2$. The frequency range in which a pulse solution exists is thus determined by combining the signs of Q and $\omega''(K_0)$, and agrees with the results obtained in § 5.1.

The above discussion implies that the problem of a long pulse in a dielectric medium can be treated by means of the reductive perturbation method which has successfully been applied to various problems of non-linear wave propagation (Taniuti 1974). A similar treatment may also be possible for the standing wave of non-linear polariton studied in § 5.2, when its peak intensity is sufficiently small. A detailed discussion of such a treatment will be given in a forthcoming paper. Our method in the present paper, however, has the merit that it covers the cases to which such a treatment is not applicable.

Next, we mention briefly the applicability of our results to real crystals. In molecular crystals to which the Frenkel exciton model applies well, j_0 in equation (2.9) arises from the Lorentz field correction and is equal to one-third of the longitudinal-transverse splitting, i.e. of the polariton gap. For such small j_0, the polariton–soliton will be observed for both branches of the upper and lower polaritons over a wide range of frequency. The standing wave of non-linear polariton is also observable inside the gap, because j_0 is smaller than the gap frequency. In ionic or semiconducting crystals, to which the Wannier model is applicable, on the other hand, the exciton dispersion has its origin in Coulomb and exchange interactions between valence electrons. Insofar as the excitation is weak, which is the case of a polariton–soliton, we can see that our results are also effectively applicable to such crystals if we formally regard the quantities in our fundamental equations as follows: regard $\hbar\omega_0$ as the gap energy between the conduction and valence bands, $\hbar j_0$ as the binding energy of the exciton, $\hbar j_2$ as $\hbar^2/2M$, where M is the exciton mass, i.e. the sum of the effective masses of the conduction and valence electrons, and N as $(\pi a_0^3)^{-1}$, were a_0 is the Bohr radius of the exciton. In those crystals, j_0 ($\sim$10–100 meV) is generally larger than the polariton gap ($\sim$1–10 meV), so that there clearly appears a frequency range in which the polariton–soliton is not observable. Our results will also be applicable to some magnetic insulators in which Frenkel excitons are observed whose excitation transfer is due to superexchange interaction between magnetic ions rather than to dipole–dipole interaction (e.g. Loudon 1968). Because of weak coupling to the radiation field, such magnetic excitons will show only a small polariton effect and be appropriate for studying short pulses.

We have also derived the solution of an exciton–soliton, i.e. a pulse of non-linear exciton which does not couple to the radiation field. This exciton–soliton is an intrinsic one in the sense that it results entirely from the non-linearity inherent in atomic dipoles, in contrast to the solution derived by Davydov and Kislukha (1973) and by Weidlich and Heudorfer (1974), who have introduced the non-linearity through the variation of interaction constants due to lattice deformation. Explicit forms of the solution in the framework of our model are obtained by integrating (A.9) or (A.14), whatever the pulse width may be. It will also be possible to extend a similar treatment for longitudinal or triplet excitons as well as for magnetic excitons. Since these excitons do not couple strongly to the radiation field, the exciton–soliton becomes a more realistic one in such cases.

Finally, we remark that because of the formal identity of the Hamiltonians, all our results on the exciton–soliton can be applied to non-linear excitation of spin waves in ferromagnets placed in an external magnetic field (Nakamura and Sasada 1974, Tyon and Wright 1977).

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Appendix

In order to solve the optical Bloch equation without coupling to the electromagnetic field, it is convenient to rewrite (4.5) and (4.6) without $\kappa\hat{E}w$ in a complex form:

$$\Lambda\dot{U} = -\mathrm{i}(\Delta - j'W)U + j[\mathrm{i}\gamma\ddot{U} - \beta\dot{U} - \mathrm{i}(1+\alpha)U](W-1), \tag{A.1}$$

where

$$U = (u + \mathrm{i}v)\exp(\mathrm{i}\phi), \qquad W = w + 1. \tag{A.2}$$

For the linearised version of equation (A.1), which is obtained by setting $W = 0$, to admit non-trivial solution of the form $U = U_0 \exp(\pm\xi)$ $(U_0 \neq 0)$,

$$\pm\Lambda = -\mathrm{i}\Delta - j[\mathrm{i}\gamma \mp \beta - \mathrm{i}(1+\alpha)] \tag{A.3}$$

must be satisfied. The real and imaginary parts of this equation, with the use of (4.11), give

$$1+\alpha = (2j)^{-1}[\Delta + (\Delta^2 + 4j)^{1/2}] \qquad \beta = j^{-1}\Lambda \qquad \gamma = (2j)^{-1}[-\Delta + (\Delta^2 + 4j)^{1/2}]. \tag{A.4}$$

Multiplying (A.1) by U^* and integrating its real part, one obtains

$$\dot{U}U^* - U\dot{U}^* = \mathrm{i}(j\gamma)^{-1}[2\Lambda + j\beta(W-2)]W. \tag{A.5}$$

Multiplying (A.1) by $\dot{U}^*$ and integrating its imaginary part, on the other hand, one obtains

$$|\dot{U}|^2 = -(j\gamma)^{-1}[2\Delta - j'W + (1+\alpha)(W-2)]W. \tag{A.6}$$

The conservation law

$$|U|^2 + (W-2)W = 0 \tag{A.7}$$

has been used in deriving these two relations. By inserting (A.5), (A.6) and (A.7) into the relation

$$(\dot{U}U^* - U\dot{U}^*)^2 + 4|U|^2|\dot{U}|^2 = (\dot{U}U^* + U\dot{U}^*)^2, \tag{A.8}$$

the differential equation is finally derived:

$$\dot{W}^2 = 4W^2(W-a)(W-b)/ab(W-1)^2, \tag{A.9}$$

where

$$a + b = (2/j')[j' - (\Delta^2 + \Lambda^2)^{1/2}], \qquad ab = (2/j')[\Delta - (\Delta^2 + \Lambda^2)^{1/2}], \tag{A.10}$$

and $a > b$ is assumed. Since $\dot{W}^2$ is positive only for $0 < W < a$, it is obvious that W takes a pulse shape whose peak value is given by $W = a$. By using this W, U is expressed as

$$U = [W(2-W)]^{1/2} \exp\left(\mathrm{i}\frac{\Lambda}{2j\gamma}\int \frac{W}{2-W}\,\mathrm{d}\xi\right), \tag{A.11}$$

which is derived from (A.5) and (A.7).

In the short pulse limit, a and b are approximated by $a \simeq 1$ and $b \simeq -2\Lambda/j'$, respectively; W is then given by the reciprocal function of

$$|\xi| = -(1-W)^{1/2} + \frac{1}{2} \ln\left(\frac{1+(1-W)^{1/2}}{1-(1-W)^{1/2}}\right). \tag{A.12}$$

The pulse shape is singular as $W \simeq 1-(3\xi)^{2/3}$ at the peak, while the tail is approximated by $W \simeq \operatorname{sech}^2 \xi$. In the long-pulse limit, on the other hand, the pulse shape depends strongly on the reduced frequency Δ. (i) When $\Delta > j'$, then $a \simeq \Lambda^2/[2\Delta(\Delta - j')]$ and $b \simeq 2[1-(\Delta/j')]$; the pulse is given as $W \simeq a \operatorname{sech}^2 \xi$ and vanishes in the limit $\Lambda \to 0$. (ii) When $0 < \Delta < j'$, then $a \simeq 2[1-(\Delta/j')]$ and $b \simeq -\Lambda^2/[\Delta(j'-\Delta)]$; the pulse does not vanish in the limit $\Lambda \to 0$ and, if $W_{\max} = a > 1$, it takes an inverted bell shape with infinite differential coefficient at $W = 1$. The pulse tail is also approximately $W \simeq a \operatorname{sech}^2 \xi$ in this case. (iii) When $\Delta < 0$, then $a \simeq 2$ and $b \simeq 2\Delta/j'$; the pulse shape is similar to that in the case (ii) except that the population of atoms is completely inverted at the peak, i.e. $W_{\max} \simeq 2$. These solutions give the envelopes of non-linear solitary waves of exciton in various cases, in which no coupling to the electromagnetic field is assumed.

We can easily extend our results to take the effect of exciton–exciton interaction into account. This effect may be introduced by adding a new term

$$-\frac{1}{2} \sum_{l,m} J_{\parallel}(\boldsymbol{r}_{lm}) \sigma_l^{(3)} \cdot \sigma_m^{(3)} \tag{A.13}$$

to our Hamiltonian (2.1). The optical Bloch equation is then modified in such a way that $\eta(j_0 w + j_2 \nabla^2 w)v$ should be added to the right-hand side of (2.13) and $-\eta(j_0 w + j_2 \nabla^2 w)u$ to the right-hand side of (2.14), respectively, where $\eta \equiv J_{\parallel}/J$. As a result, the differential equation for W becomes

$$\dot{W}^2 = \frac{4W^2(W-a)(W-b)}{ab[(1-\eta)(W-1)^2+\eta]}, \tag{A.14}$$

where

$$a+b = 2\left(1 - \frac{(\Delta^2+\Lambda^2)^{1/2}}{j'(1-\eta)}\right), \qquad ab = \frac{2[\Delta - (\Delta^2+\Lambda^2)^{1/2}]}{j'(1-\eta)}. \tag{A.15}$$

The exciton–exciton interaction removes the singularities in the pulse shape as long as $\eta > 0$, because the denominator of the right-hand side in (A.14) has then no real zero. When $\eta = 1$, especially, W is reduced to a simple form

$$W = \left(1 - \frac{\Delta}{(\Delta^2+\Lambda^2)^{1/2}}\right) \operatorname{sech}^2 \xi. \tag{A.16}$$

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