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# Steady propagation of a coherent light pulse in a dielectric medium: III Dynamical behaviour of a long pulse

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Abstract. The dynamical process by which the steady propagation of a coherent light pulse of long width takes place in a dielectric medium is studied. In the absence of direct interaction between atomic dipoles, the nonlinear polariton is unstable against a small perturbation and develops self-modulation of its envelope. Nonlinear Schrödinger equations describing this self-modulation are derived for the two cases where the carrier wave frequency lies outside and inside the polariton gap. It is shown that an arbitrary incoming pulse of long width outside the polariton gap evolves as composite pulse of multiple peak structure, which is regarded as a bound state of the steady pulses obtained in a previous paper. The evolution process of a pulse inside the polariton gap and the effect of direct interaction between atoms are also discussed.

# 1. Introduction

In previous papers (Akimoto and Ikeda 1977, Ikeda and Akimoto 1979, referred to as I and II respectively), a systematic method has been developed to treat the steady propagation of a coherent light pulse in a dielectric medium, paying special attention to the effect of polariton formation. It has been shown in I that the behaviours of steadily propagating pulses are determined by the relative magnitude of three quantities: the time width of the pulse  $\tau$ , the polariton gap frequency  $\omega_{\rm G}$  which is proportional to the density of atomic dipoles in the medium, and the difference  $\Delta \omega$  between the carrier wave frequency of the pulse and the resonant frequency of the medium. It has also been shown that there exist two types of pulses having qualitatively different characters, that is, the short pulse  $(\tau^{-1} \gg |\Delta \omega|, |\Delta \omega - \omega_{\rm G}|)$  and the long pulse  $(\tau^{-1} \ll |\Delta \omega|, |\Delta \omega - \omega_{\rm G}|)$ . In II, the effect of direct interaction between atomic dipoles has been studied. It has been found there that such an interaction brings about, besides the usual pulse propagating in the form of the radiation field (optical pulse), another type of pulse which contains little photon component and propagates by means of the excitation transfer inherent in interacting dipoles (exciton-like pulse), and that a long pulse of the optical character, which we call the polariton-soliton, can exist only in a limited range of frequency.

The short pulse is nothing but the well known pulse of self-induced transparency (SIT), which has a hyperbolic secant shape and pulse area of  $2\pi$  (McCall and Hahn 1969). As has been studied in detail by Lamb (1971, 1974), the SIT pulse can be described by a stable soliton solution of a sine-Gordon equation which is derived as a simplified version of the Maxwell and the optical Bloch equations. More generally, any incoming pulse of short width obeys such a sine-Gordon equation and decomposes into

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several steady pulses with the increase of propagation distance. (For various properties of the sine-Gordon equation, see e.g. Ablowitz *et al* 1974; for behaviours of the SIT pulse as a soliton, see also Eilbeck *et al* 1973.) As for the long pulse, on the other hand, such a study has not been tried so far. The purpose of the present paper is to discuss the stability of the long pulse and its dynamical process through which the pulse tends to steady propagation. Since the long pulse shows quite different behaviours outside and inside the polariton gap, reflecting the anomalous dispersion near the resonant frequency, our discussions will be given separately on these two cases.

Our starting equations are the Maxwell equation

$$[(\partial/\partial Z - iQ)^2 - (i + \mu \partial/\partial T)^2]E = (i + \mu \partial/\partial T)^2U$$
(1.1)

and the optical Bloch equation derived in II

$$i \partial U/\partial T = [\Delta - j'(w+1) + jw(Q+i \partial/\partial Z)^2]U - Ew$$
(1.2)

in which the interaction between dipoles is also taken into account. The notation is as follows: E and U are the slowly varying envelopes of the electric field and of the macroscopic polarisation per unit volume respectively; the rapidly oscillating carrier waves  $\exp[i(\omega t - Kz)]$  have been separated out from them. The quantities E and U are in general complex and have been made dimensionless by scaling them by  $2\pi N\hbar\kappa$  and  $N\hbar\kappa/2$  respectively, where N is the dipolar density and  $\kappa$  the dipole matrix element divided by  $\hbar/2$ . The quantity w is the population difference between the ground and excited states and is related to U through

$$|U|^2 + w^2 = 1. (1.3)$$

Time T and distance Z as the independent variables in equations (1.1) and (1.2) have been scaled by  $(2\pi N\hbar\kappa^2)^{-1}$  and  $c/\omega$  respectively, so that they are also dimensionless. The frequency and the wavenumber of the carrier wave are denoted by  $\Delta$  and Q respectively, where  $\Delta$  is measured from the resonant frequency of the exciton with K = 0 and is scaled by the polariton gap frequency, while Q is scaled by  $\omega/c$ , i.e.

$$\Delta = (\omega - \omega_0)/2\pi N\hbar\kappa^2 + j', \qquad Q = cK/\omega.$$
(1.4)

The dimensionless parameters j and j', which have been introduced in II, characterise the direct interaction between dipoles; the exciton dispersion due to this interaction is given, in the limit of  $E \rightarrow 0$ , as

$$\omega_Q = \omega_0 + 2\pi N \hbar \kappa^2 (-j' + jQ^2). \tag{1.5}$$

Finally,  $\mu$  is a small parameter defined by  $\mu \equiv 2\pi N\hbar\kappa^2/\omega$  and will safely be set equal to zero in the following discussions.

The plan of the present paper is as follows. First, we discuss in § 2 the stability of a nonlinear polariton which is described by the homogeneous solution of equations (1.1) and (1.2). We do this from the consideration that the electromagnetic wave produced in the medium by an arbitrary but sufficiently long incoming wave can approximately be regarded as a nonlinear plane wave. By superposing a spatially modulated wave of small amplitude as a perturbation on the nonlinear plane wave and observing its time development, it will be found that the nonlinear polariton is in fact unstable against a perturbation and amplifies the initial modulation. The space and time scales (wavenumber, propagation velocity and growth rate) of the most rapidly growing modulation wave will suggest that the nonlinear polariton tends to self-decompose into an assembly of pulses, each of which is simply the steady pulse obtained in I. On the

basis of these space and time scales, we apply in §§ 3 and 4 the method of reductive perturbation expansion to our problem and show that the motion of a wave packet of sufficiently long width obeys a nonlinear Schrödinger equation. This equation has different forms outside and inside the polariton gap and their soliton solutions coincide with the steady solutions obtained in I. Applying the initial value problem of the nonlinear Schrödinger equation studied by Zakharov and Shabat (1971) to our equations, we discuss semiquantitatively in § 5 by which process an arbitrary incoming pulse develops self-modulation and comes to propagate as an assembly of several steady pulses. In §§ 2–5, we neglect for simplicity the effect of spatial dispersion and set j and j' in equation (1.2) equal to zero. More general discussions involving this effect will be given in § 6.

#### 2. Instability of the nonlinear polariton

Equations (1.1) and (1.2) admit the homogeneous solution  $E = E_0$  and  $U = U_0$ , which describes the nonlinear polariton, if the dispersion relation

$$Q^{2} = 1 - \Delta^{-1} [1 + (|E_{0}|/\Delta)^{2}]^{-1/2}$$
(2.1)

is satisfied. The right-hand side of equation (2.1) defines the field-dependent dielectric function  $\tilde{\epsilon}(\Delta, |E_0|)$ , which relates  $U_0$  to  $E_0$  through

$$U_0 = [\tilde{\boldsymbol{\epsilon}}(\Delta, |\boldsymbol{E}_0|) - 1] \boldsymbol{E}_0. \tag{2.2}$$

In the limit of  $E_0 \rightarrow 0$ , equation (2.1) is reduced to the dispersion relation of the usual (linear) polariton

$$Q^2 = 1 - \Delta^{-1} \tag{2.3}$$

which has the frequency gap  $0 < \Delta < 1$  in which no plane wave can propagate because of negative  $\tilde{\epsilon}(\Delta, 0)$ . We call this frequency gap the polariton gap. For a plane wave of sufficiently large amplitude satisfying

$$|E_0| > E_{\rm th} = (1 - \Delta^2)^{1/2} \tag{2.4}$$

however,  $\tilde{\epsilon}(\Delta, |E_0|)$  becomes positive even for  $0 < \Delta < 1$  and enables the wave to propagate in the medium.

Suppose that a spatially modulated wave of small amplitude, described by  $\delta E \exp[i(\bar{\omega}T - \bar{Q}Z)]$  and  $\delta U \exp[i(\bar{\omega}T - \bar{Q}Z)]$ , is superposed<sup>†</sup> as a perturbation on the plane wave with  $E_0$  and  $\Delta$ ; if this perturbation wave grows with time, the plane wave is unstable. Substituting the superposed wave for E and U in equations (1.1) and (1.2) and linearising these equations with respect to the coefficients  $\delta E$  and  $\delta U$ , we obtain  $\bar{\omega}$  as a function of  $\bar{Q}$  as follows:

$$\bar{\omega}(\bar{Q}) = \frac{2\Delta Q (1-Q^2)^2 \bar{Q} \pm [(1+3Q^2-\bar{Q}^2)f(\bar{Q})]^{1/2} \bar{Q}}{(1-Q^2)[(1-Q^2-\bar{Q}^2)^2-4Q^2\bar{Q}^2]}$$
(2.5)

where

$$f(\bar{Q}) = (1 - Q^2)^3 (1 - Q^2 - \bar{Q}^2) \Delta^2 - (1 - Q^2 - \bar{Q}^2)^2 + 4Q^2 \bar{Q}^2$$
(2.6)

 $\dagger$  By this notation, we mean that each of the real and imaginary parts of E and U is subject to a sinusoidal modulation. These modulations, or equivalently the modulations of amplitude and phase, couple to each other through the Maxwell-Bloch equations. This coupling is essential for inducing the instability.

and Q is related to  $E_0$  and  $\Delta$  through equation (2.1). It can be seen from equations (2.5) and (2.6) that  $\bar{\omega}(\bar{Q})$  has an imaginary part  $\bar{\omega}_i(\bar{Q})$  for small  $\bar{Q}$ . This means that the modulation wave of long wavelength grows with time; that is to say, the nonlinear polariton is unstable against such a perturbation. At the same time, this modulation wave propagates with the group velocity  $d\bar{\omega}_r(\bar{Q})/d\bar{Q} (\equiv V_p)$ .

Let us estimate the growth rate of the most rapidly growing modulation wave; such a modulation wave will determine the gross features of the unstabilised nonlinear plane wave. For simplicity, we confine ourselves to the case of  $|E_0/\Delta| \ll 1$ , i.e. a weakly nonlinear case. Inside the polariton gap, this condition is meaningful only near the upper edge of the gap  $\Delta \approx 1$  because the nonlinear plane wave itself is allowed to exist only when  $|E_0| > (1-\Delta^2)^{1/2}$ . Under this condition, the range of  $\bar{Q}$  in which  $\bar{\omega}(\bar{Q})$  becomes complex is obtained as

$$|\bar{Q}| < \bar{Q}_{c} = \Delta^{-2} (4 - 3/\Delta)^{-1/2} |E_{0}|.$$
(2.7)

The wavenumber  $\bar{Q}_{m}$  which maximises the growth rate  $\bar{\omega}_{i}(\bar{Q})$  is given by  $\bar{Q}_{m} = \bar{Q}_{c}/\sqrt{2}$ ; the maximum growth rate is

$$\bar{\omega}_{\rm i}(\bar{Q}_{\rm m}) = |E_0|^2 / 2|\Delta|.$$
 (2.8)

This means that the nonlinear plane wave develops self-modulation and decomposes, after the characteristic time  $T_c = (\bar{\omega}_i(\bar{Q}_m))^{-1}$ , into an assembly of wave domains whose spatial width is given by  $l_c \equiv \bar{Q}_m^{-1} = \sqrt{2}/\bar{Q}_c$ . The explicit expressions of  $l_c$  are given as follows: For  $\Delta > 1$  or  $\Delta < 0$  (outside the polariton gap),

$$l_{\rm c} \sim \sqrt{2} \, \Delta^2 (4 - 3/\Delta)^{1/2} |E_0|^{-1}. \tag{2.9}$$

For  $0 < \Delta < 1$  (inside the gap),

$$l_{\rm c} \sim \sqrt{2} |E_0|^{-1} \leq \sqrt{2} E_{\rm th}^{-1} \simeq (1-\Delta)^{-1/2}.$$
 (2.10)

These relations agree, apart from numerical factors, with the corresponding ones for the steady pulses derived in I, i.e. the relations between the spatial width and the amplitude of the pulse. This fact suggests that each of the wave domains into which the nonlinear plane wave decomposes is simply the steady pulse obtained in I.

For frequencies inside the polariton gap but not very close to its upper edge, the above results (2.7)-(2.10) derived for  $|E_0| \ll 1$  are no longer valid, because for such frequencies  $E_{\rm th}$  is of the order of unity and therefore  $|E_0| \ge 1$  is necessary for the electromagnetic field to propagate in the medium. Also in this case, however, the fact remains that the nonlinear polariton is unstable. For instance, it can easily be seen from equations (2.5) and (2.6) that the nonlinear polariton just satisfying  $|E_0| = E_{\rm th}$ , i.e. Q = 0, is in fact unstable against a modulation whose  $\overline{Q}$  is given by

$$|\bar{Q}| < \bar{Q}_{c} = (1 - \Delta^{2})^{1/2}$$
 (2.11)

where  $\bar{Q}_{c}$  is of the order of unity.

Instability of the system described by the Maxwell-Bloch equations has been studied also by Zel'dovich and Sobel'man (1971), Courtens (1974) and Armstrong (1975). Their results are somewhat different from ours and may be expressed in our language as follows: The nonlinear plane wave is unstable against a perturbation of any wavenumber  $\bar{Q}$ , making  $\bar{Q}_c$  infinite, and has a growth rate proportional to  $\bar{Q}$ . Consequently, any incoming wave acquires a singularity in its shape as it propagates (self-steepening; Armstrong 1975). These results are attributable to the fact that, in their treatment, the second-derivative terms in the Maxwell-Bloch equations are partly

neglected, although a balance of those terms plays the essential role of relaxing the self-steepening and making the appearance of a steady pulse possible. Of course, provided the discussions are confined to the initial stage of the self-steepening, their treatment is reasonable enough.

Before concluding the present section, we note that Gurovich and his co-workers (Gurovich and Karpman 1969, Gurovich *et al* 1969) treated a problem similar to ours but for liquids and plasmas with negative dielectric constant. They pointed out that, in these media, a nonlinear solitary wave of low amplitude shows steady propagation as a consequence of the instability of a nonlinear plane wave.

## 3. Nonlinear Schrödinger equation describing self-modulation of the nonlinear polariton—outside the polariton gap

The linear theory developed in the preceding section is valid only when the amplitude of the modulation wave superposed on the nonlinear polariton remains sufficiently small. In this and in subsequent sections, we derive equations which describe the whole process of evolution of the self-modulation, assuming that the initial amplitude of the electric field of the nonlinear polariton is small.

Outside the polariton gap, a modulation wave generated on a weakly nonlinear plane wave propagates with nonzero group velocity

$$V_{\rm p} = 2\Delta^2 (1 - \Delta^{-1})^{1/2} \tag{3.1}$$

as can be seen from equations (2.6) and (2.11). This leads us to choose Z and  $T - Z/V_p$  as the independent variables<sup>†</sup>. The most unstable modulation wave then evolves like

$$\exp[\mathrm{i}(\bar{\omega}(\bar{Q}_{\mathrm{m}})T - \bar{Q}_{\mathrm{m}}Z)] = \exp[\bar{\omega}_{\mathrm{i}}(\bar{Q}_{\mathrm{m}})V_{\mathrm{p}}^{-1}Z] \exp[(\mathrm{i}\bar{Q}_{\mathrm{m}}V_{\mathrm{p}} + \bar{\omega}_{\mathrm{i}}(\bar{Q}_{\mathrm{m}}))(T - Z/V_{\mathrm{p}})].$$

The characteristic constants of this evolution are given by  $(\bar{\omega}_i(\bar{Q}_m))^{-1}V_p$  and  $\min\{\bar{Q}_m^{-1}V_p^{-1}, (\bar{\omega}_i(\bar{Q}_m))^{-1}\}$ , or in other expressions,  $T_cV_p$  and  $\min\{l_cV_p^{-1}, T_c\}$ .

Considering that a weakly nonlinear wave is characterised by the condition  $E/\Delta \sim U \ll 1,\ddagger$  let us introduce here a small parameter  $\epsilon$  of the same order as these quantities and scale all the variables by this parameter. Observing the orders of magnitude  $T_c V_p \sim O(\epsilon^{-2}\Delta), l_c V_p^{-1} \sim O(\epsilon^{-1}\Delta^{-1})$  and  $T_c \sim O(\epsilon^{-2}\Delta^{-1})$ , we transform the independent variables into a new set

$$\eta = \epsilon^2 \Delta^{-1} Z$$

$$j = \epsilon \Delta (T - Z/V_p)$$
(3.2)

and expand the electric field and the polarisation as

$$E/\Delta = \epsilon \psi^{(1)}(\xi, \eta) + \epsilon^2 \psi^{(2)}(\xi, \eta) + \dots$$
  

$$U = \epsilon \phi^{(1)}(\xi, \eta) + \epsilon^2 \phi^{(2)}(\xi, \eta) + \dots$$
(3.3)

so that  $\psi^{(n)}(\xi, \eta)$  and  $\phi^{(n)}(\xi, \eta)$  may be of the order of unity and that their variation may become first noticeable when  $\xi$  and  $\eta$  vary to the extent of unity. This idea has been

<sup>&</sup>lt;sup>†</sup> For the purpose of discussing the boundary value problem, we have chosen the independent variables as in the text. It is also possible to make another choice,  $Z - V_p T$  and T. This choice is appropriate for discussing the initial value problem.

<sup>&</sup>lt;sup>‡</sup> The steady solution of a long pulse also satisfies this condition, as can be seen from equation (5.13) in I.

<sup>§</sup> These order estimations involving  $\Delta$  are valid for any value of  $\Delta$  except in the vicinity of  $\Delta = 0$  or 1.

taken from the reductive perturbation method for strongly dispersive media developed by Taniuti and Yajima (Taniuti and Yajima 1973, Taniuti 1974).

By inserting equations (3.2) and (3.3) in our starting equations (1.1) and (1.2) (in which j, j' and  $\mu$  are set equal to zero) and comparing terms of the same order with respect to  $\epsilon$ , the following sets of balance equations are obtained. To the order of  $\epsilon$ ,

$$\Delta (1 - Q^2) \psi^{(1)} + \phi^{(1)} = 0$$
  
$$\psi^{(1)} + \phi^{(1)} = 0.$$
 (3.4)

To the order of  $\epsilon^2$ ,

$$\Delta (1 - Q^2) \psi^{(2)} + \phi^{(2)} = -2iQ\Delta^2 V_{\rm p}^{-1} (\partial/\partial\xi) \psi^{(1)}$$
  
$$\psi^{(2)} + \phi^{(2)} = i(\partial/\partial\xi) \phi^{(1)}.$$
 (3.5)

To the order of  $\epsilon^3$ ,

$$\Delta (1 - Q^{2})\psi^{(3)} + \phi^{(3)} = -2iQ\Delta^{2}V_{p}^{-1}(\partial/\partial\xi)\psi^{(2)} - [\Delta^{3}V_{p}^{-2}(\partial^{2}/\partial\xi^{2}) - 2iQ(\partial/\partial\eta)]\psi^{(1)}$$
  

$$\psi^{(3)} + \phi^{(3)} = i(\partial/\partial\xi)\phi^{(2)} + \frac{1}{2}|\phi^{(1)}|^{2}\psi^{(1)}.$$
(3.6)

The conservation law (1.3) has been used in deriving these equations. In order for the two equations (3.4) to be compatible,  $\Delta$  and Q must satisfy equation (2.3), the dispersion relation of the linear polariton. In order for the two equations (3.5) to be compatible,  $V_p$  must satisfy relation (3.1). Note that this  $V_p$  is equal to the group velocity of the linear polariton,  $d\Delta/dQ$ . By eliminating  $\psi^{(3)} + \phi^{(3)}$  from the two equations in (3.6) and using (2.3), (3.1), (3.4)

By eliminating  $\psi^{(3)} + \phi^{(3)}$  from the two equations in (3.6) and using (2.3), (3.1), (3.4) and (3.5), the following nonlinear Schrödinger equation for  $\psi^{(1)}$  is obtained:

$$i\frac{\partial\psi^{(1)}}{\partial\eta} = \frac{1}{8}\left(\frac{\Delta}{\Delta-1}\right)^{1/2}\left(\frac{4\Delta-3}{\Delta-1}\frac{\partial^2}{\partial\xi^2} + 2|\psi^{(1)}|^2\right)\psi^{(1)}$$
(3.7)

or in the original coordinates,

$$i\frac{\partial E}{\partial Z} = \frac{1}{8\Delta^3} \left(\frac{\Delta}{\Delta - 1}\right)^{1/2} \left(\frac{4\Delta - 3}{\Delta - 1} \frac{\partial^2}{\partial (T - Z/V_p)^2} + 2|E|^2\right) E.$$
(3.8)

It is well known that this type of equation admits a stable soliton solution if the coefficients of the second-derivative term and of the self-potential term have the same sign (Taniuti 1974). This condition is in fact always satisfied outside the polariton gap  $(\Delta < 0 \text{ or } \Delta > 1)$  and the soliton solution is given as<sup>†</sup>

$$E = \frac{1}{\tau} \left(\frac{4\Delta - 3}{\Delta - 1}\right)^{1/2} \exp(-i\kappa Z) \operatorname{sech}\left(\frac{T - Z/V_{p}}{\tau}\right)$$
(3.9)

where  $\tau$  is an arbitrary constant which characterises the pulse width and  $\kappa = (4\Delta - 3)[8\Delta^{5/2}(\Delta - 1)^{3/2}\tau^2]^{-1}$  gives a wavenumber shift of the carrier wave. Expression (3.9) exactly reproduces the lowest-order pulse solution derived in I, except that some physically interesting terms of higher orders, such as phase modulation which gives the shift of instantaneous frequency (chirping), are missing.

<sup>&</sup>lt;sup>†</sup> The general form of the soliton solution of equation (3.8) should involve an additional phase factor  $\exp(i\Delta' T)$ , where  $\Delta'$  is arbitrary; in accordance with this,  $\kappa$  and  $V_p$  in equation (3.9) should also undergo a certain shift depending on  $\Delta'$ . Inclusion of this phase factor, however, only brings about a trivial shift of the frequency in the final expression of the electric field multiplied by the carrier wave factor. In order to fix this frequency at  $\Delta$ , we have chosen  $\Delta' = 0$  in equation (3.9).

If the fact is once known that E obeys a nonlinear Schrödinger equation, as is the case in equation (3.8), it is possible to represent this equation in a more general form, expressing its coefficients in terms of the dielectric function. This can be performed as follows. We note first that the nonlinear Schrödinger equation of the form (3.8) has, besides the soliton solution, a plane wave solution  $E_0 \exp[i(\Delta' T - Q'Z)]$ , where  $\Delta'$  and Q' are related to the coefficients in the equation as

$$Q' = \Delta' / V_{\rm p} - a \Delta'^2 + c |E_0|^2 \tag{3.10}$$

a and c denoting the coefficient of the second-derivative term and that of the self-potential term respectively. The above solution, when multiplied by the carrier wave  $\exp[i(\Delta T - QZ)]$ , on the other hand, must be the nonlinear plane wave solution of the original Maxwell-Bloch equations, so that  $\Delta + \Delta'$  and Q + Q' satisfy the nonlinear dispersion relation

$$(Q+Q')^2 = \tilde{\epsilon}(\Delta+\Delta', |E_0|) \tag{3.11}$$

while the linear dispersion relation

$$Q^2 = \tilde{\epsilon}(\Delta, 0) \tag{3.12}$$

holds between  $\Delta$  and Q. From these two relations, we obtain

$$Q' = [(\tilde{\epsilon}_0(\Delta))^{1/2}]' \Delta'^3 + \frac{1}{2} [(\tilde{\epsilon}_0(\Delta))^{1/2}]'' \Delta'^2 + \frac{\tilde{\epsilon}_2(\Delta)}{2(\tilde{\epsilon}_0(\Delta))^{1/2}} |E_0|^2$$
(3.13)

where we have expanded the field-dependent dielectric function as

$$\tilde{\epsilon}(\Delta, |E|^2) = \tilde{\epsilon}_0(\Delta) + \tilde{\epsilon}_2(\Delta)|E|^2 + \dots$$
(3.14)

and have denoted  $d[(\tilde{\epsilon}_0(\Delta))^{1/2}]/d\Delta$  by  $[(\tilde{\epsilon}_0(\Delta))^{1/2}]'$ , etc. By comparing equations (3.10) and (3.13), *a* and *c* are expressed in terms of the dielectric function and its derivatives. The general form of (3.8) is thus

$$i\frac{\partial E}{\partial Z} = \left\{-\frac{1}{2}\left[\left(\tilde{\epsilon}_{0}(\Delta)\right)^{1/2}\right]''\frac{\partial^{2}}{\partial(T-Z/V_{p})^{2}} + \frac{\tilde{\epsilon}_{2}(\Delta)}{2(\tilde{\epsilon}_{0}(\Delta))^{1/2}}|E|^{2}\right\}E$$
(3.15)

where

$$V_{\rm p} = 1/[(\tilde{\epsilon}_0(\Delta))^{1/2}]' = 2\tilde{\epsilon}_0'(\Delta)/(\tilde{\epsilon}_0(\Delta))^{1/2}.$$
(3.16)

This expression is useful because it holds also in the presence of spatial dispersion, i.e. in the case where a direct interaction between atomic dipoles is taken into account. In § 6, discussions of dynamical properties will be given on the basis of this expression. A similar nonlinear Schrödinger equation whose coefficients are expressed in terms of the dielectric function has already been used in the problem of self-focusing (Kadomtsev and Karpman 1971).

# 4. Nonlinear Schrödinger equation describing self-modulation of the nonlinear polariton—inside the polariton gap

Inside the polariton gap, a weak electromagnetic field such that  $|E| < E_{th} = (1 - \Delta^2)^{1/2}$  cannot propagate in the medium because it makes  $\tilde{\epsilon}(\Delta, |E|)$  negative. Therefore the usual reductive perturbation method for strongly dispersive media, as that used in § 3, can no longer be applied to the present case. As will be shown below, however, a similar

perturbational treatment is possible also in the present case, by making a special choice of the expansion parameter.

Let us first assume  $|E| \ge E_{\text{th}}$ . Confining ourselves to the vicinity of the upper edge of the polariton gap ( $\Delta \simeq 1$ ), where  $E_{\text{th}} \ll 1$  is satisfied, we introduce a small parameter

$$\boldsymbol{\epsilon} = (1 - \Delta)^{1/2} \sim E_{\rm th} / \sqrt{2}. \tag{4.1}$$

Since  $V_p$  is now a small quantity of the order of  $\epsilon$ , it is no longer meaningful to re-define the independent variables as has been done in § 3. Observing that the characteristic constants of the most unstable modulation wave  $\exp[i(\bar{\omega}(\bar{Q}_m)T - \bar{Q}_mZ)]$  are estimated as  $T_c \sim O(\epsilon^{-2})$  and  $l_c \sim O(\epsilon^{-1})$ , we introduce the stretch transformation

$$\eta = \epsilon^2 T$$

$$\xi = \epsilon Z$$
(4.2)

and expand E and U as

$$E = \epsilon \psi^{(1)}(\xi, \eta) + \epsilon^2 \psi^{(2)}(\xi, \eta) + \dots$$

$$U = \epsilon \phi^{(1)}(\xi, \eta) + \epsilon^2 \phi^{(2)}(\xi, \eta) + \dots$$
(4.3)

The balance equations for (1.1) and (1.2) are obtained as follows. To the order of  $\epsilon$ ,

$$(1 - Q^{2})\psi^{(1)} + \phi^{(1)} = 0$$
  
$$\psi^{(1)} + \phi^{(1)} = 0.$$
 (4.4)

To the order of  $\epsilon^2$ ,

$$(1 - Q^{2})\psi^{(2)} + \phi^{(2)} = 2iQ(\partial/\partial\xi)\psi^{(1)}$$
  
$$\psi^{(2)} + \phi^{(2)} = 0.$$
 (4.5)

To the order of  $\epsilon^3$ ,

$$(1 - Q^{2})\psi^{(3)} + \phi^{(3)} = -(\partial^{2}/\partial\xi^{2})\psi^{(1)}$$
  
$$\psi^{(3)} + \phi^{(3)} = i(\partial/\partial\eta)\phi^{(1)} + \phi^{(1)} + \frac{1}{2}|\phi^{(1)}|^{2}\psi^{(1)}.$$
(4.6)

We note that  $\Delta = 1 - \epsilon^2$  has been used in deriving these equations. In order for the two equations in (4.4) to be compatible, Q = 0 is required. This indicates the fact that the carrier wavenumber of a pulse inside the gap tends to zero in the limit of long pulse, as has been shown in I. With the condition Q = 0, the two equations in (4.5) are identical. By eliminating  $\psi^{(3)} + \phi^{(3)}$  from the two equations in (4.6) and using (4.4), the nonlinear Schrödinger equation for  $\psi^{(1)}$  is obtained<sup>†</sup>:

$$i \,\partial\psi^{(1)}/\partial\eta = (\partial^2/\partial\xi^2 - 1 + \frac{1}{2}|\psi^{(1)}|^2)\psi^{(1)} \tag{4.7}$$

or in the original coordinates,

$$i \partial E / \partial T = (\partial^2 / \partial Z^2 - 1 + \Delta + \frac{1}{2} |E|^2) E.$$
(4.8)

<sup>†</sup> In the text,  $|E| \sim E_{\text{th}} \ll 1$  has been assumed in deriving equation (4.7). However, even if |E| is much larger than  $E_{\text{th}}$ , this equation is valid as far as  $|E| \ll 1$  is satisfied. For |E|, the order of magnitude of which is given by  $|E| \sim (E_{\text{th}})^{\delta}$  ( $0 < \delta < 1$ ), in fact, we need only replace the definition of  $\epsilon$  by  $\epsilon \equiv (1-\Delta)^{\delta/2}$ ; this leads to the equation

$$\mathbf{i} \,\partial \boldsymbol{\psi}^{(1)}/\partial \boldsymbol{\eta} = (\partial^2/\partial \boldsymbol{\xi}^2 + \frac{1}{2}|\boldsymbol{\psi}^{(1)}|^2)\boldsymbol{\psi}^{(1)}$$

which is the same one as equation (4.7) if  $\psi^{(1)} \gg 1$ , i.e.  $|E| \gg E_{\text{th}}$ , is assumed.

The soliton solution of equation (4.8) is given as

$$E = 2(1 - \Delta + \kappa^2)^{1/2} \exp(-i\kappa Z) \operatorname{sech}[(1 + \Delta + \kappa^2)^{1/2}(Z - 2\kappa T)]$$
(4.9)

where  $\kappa$  is an arbitrary parameter. In the limit of  $\kappa \to 0$ , equation (4.9) reproduces the expression which lill be obtained by setting  $\Delta \approx 1$  in (5.16) in I, i.e. the steady pulse solution inside the gap. If  $\Delta = 1$  is assumed in equation (4.9), it reproduces equation (5.22) in I, i.e. the steady pulse solution at the upper edge of the gap. Note that, when equation (4.9) expresses a propagating pulse, the peak value of |E| is larger than  $2(1-\Delta)^{1/2} = \sqrt{2} E_{th}$ ; this reflects one of the characteristics of the nonlinear polariton itself inside the gap.

In the same way as has been done in § 3, equation (4.8) can also be expressed by using the field-dependent dielectric function as follows:

$$i\frac{\partial E}{\partial T} = \frac{1}{\tilde{\epsilon}_0'(1)} \left( \frac{\partial^2}{\partial Z^2} + \tilde{\epsilon}_0'(1)(\Delta - 1) + \tilde{\epsilon}_2(1) |E|^2 \right) E$$
(4.10)

where

$$\tilde{\epsilon}_0'(1) = \mathrm{d}\tilde{\epsilon}_0(\Delta)/\mathrm{d}\Delta\big|_{\Delta=1}$$

For frequencies inside the polariton gap but not very close to its upper edge, equation (4.10) can no longer describe the process of self-modulation satisfactorily. Instead of equation (4.10), let us use its generalised form

$$i\frac{\partial E}{\partial T} = \frac{1}{\tilde{\epsilon}'(\Delta, |E|)} \left( \frac{\partial^2}{\partial Z^2} + \tilde{\epsilon}(\Delta, |E|) \right) E$$
(4.11)

and see that this equation is appropriate for the present case. Here,  $\tilde{\epsilon}'_0(1)$  and  $\tilde{\epsilon}'_0(1)(\Delta-1)+\tilde{\epsilon}_2(1)|E|^2$  in equation (4.10) are replaced by  $\tilde{\epsilon}'(\Delta,|E|)$  and  $\tilde{\epsilon}(\Delta,|E|)$  respectively, by seeing that the former are power-series expansions of the latter for  $\Delta \simeq 1$  and  $|E| \simeq 0$ . (Note that  $\tilde{\epsilon}_0(1) = 0$ .) It can easily be shown that equation (4.11) has a steady pulse solution of the form

$$E = f(Z - VT) \exp[i\phi(Z - VT) - i\kappa Z]$$

involving a phase modulation, and that in the limit of V = 0, this solution exactly reproduces the previously derived solution of long pulse inside the gap, i.e. equations (5.16)-(5.19) in I. It is also possible to justify equation (4.11) on the basis of the original Maxwell-Bloch equations if we confine ourselves to the case where the time evolution of their solution is sufficiently slow and the polarisation can adiabatically follow the electric field. In reality, however, the characteristic time  $T_c$  introduced in § 2 is not very long in the present case, so that there appears a stage in which the time evolution is not so slow. Therefore, it is not obvious whether equation (4.11) can well describe the *whole* process of self-modulation of an arbitrary incoming pulse of sufficiently long width.

## 5. Dynamical properties of self-modulation

The initial value problem of the nonlinear Schrödinger equation such as equations (3.8) and (4.8) has been solved by Zakharov and Shabat (1971), who resorted to the inverse scattering method developed by Gardner *et al* (1967). On the basis of their results, we discuss in the present section the dynamical behaviours of an arbitrary incoming pulse.

Suppose that a field u defined in space-time  $(\rho, \sigma)$  obeys a nonlinear Schrödinger equation

$$i \,\partial u/\partial \sigma = [a \,\partial^2/\partial \rho^2 + (b+c|u|^2)]u \tag{5.1}$$

where a, b and c are real constants and satisfy ac > 0. As has been shown by Zakharov and Shabat, equation (5.1) has a family of steady solutions

$$u_n = \left(\frac{8a}{c}\right)^{1/2} \beta_n \operatorname{sech}[2\beta_n(\rho - 4\alpha_n a\sigma - \rho_0)] \exp\{i[-b\sigma + 4a(\alpha_n^2 - \beta_n^2)\sigma - 2\alpha_n\rho + \theta_0]\}$$
(5.2)

each of which describes a soliton characterised by parameters  $\alpha_n$  and  $\beta_n$  as well as arbitrary constants  $\rho_0$  and  $\theta_0$ . An arbitrary field  $u(\rho, \sigma)$  comes to behave, asymptotically with the increase of  $\sigma$ , as an assembly of these solitons, the parameters of which are determined by the initial form of u, i.e.  $u(\rho, 0) \equiv u_0(\rho)$  (we assume  $u_0(\pm \infty) = 0$ ). These parameters are in fact given as the real and imaginary parts respectively of the complex discrete eigenvalues  $\gamma_n = \alpha_n + i\beta_n$  of the two-component equation

$$\begin{pmatrix} \partial/\partial\rho + i\gamma & -i(c/2a)^{1/2}u(\rho) \\ -i(c/2a)^{1/2}u^*(\rho) & \partial/\partial\rho - i\gamma \end{pmatrix} \begin{pmatrix} v_1 \\ v_2 \end{pmatrix} = 0$$
(5.3)

with the boundary condition  $v_i(\pm \infty) = 0^{\dagger}$ . Especially, if  $u_0(\rho)$  is a real function of  $\rho$  except a constant phase factor, one has  $\alpha_n = 0$ . Consequently the group velocities  $4\alpha_n a$  of all solitons become zero. This means that, starting from such a  $u_0(\rho)$ , the field  $u(\rho, \sigma)$  does not split into individual solitons but evolves as a bound state of them. If  $u_0(\rho)$  is also a slowly varying function of  $\rho$ , equation (5.3) can be approximated by a Schrödinger equation

$$(\partial^2 / \partial \rho^2 + \gamma^2 + (c/2a) |u(\rho)|^2) v_1(\rho) = 0$$
(5.4)

whose discrete eigenvalues  $\gamma_n$  are purely imaginary. These eigenvalues are approximately obtained by using the Bohr–Sommerfeld quantisation condition

$$\oint \left[ (c/2a) |u(\rho)|^2 - \beta_n^2 \right]^{1/2} d\rho = (2n+1)\pi$$
(5.5)

where  $\beta_n^2 = -\gamma_n^2$ .

Let us apply the above results to equation (3.8), the nonlinear Schrödinger equation describing self-modulation of the nonlinear polariton outside the polariton gap. As variables  $T - Z/V_p$  and Z in equation (3.8) correspond to  $\rho$  and  $\sigma$  respectively, the boundary value problem of equations (3.8) is equivalent to the initial value problem in space-time ( $\rho$ ,  $\sigma$ ). Assume a non-chirped, symmetric and slowly varying pulse incident at the boundary Z = 0 and express its envelope as

$$E(T, Z = 0) = E_0 f(T/T_0)$$
(5.6)

where  $T_0$  is the time width of the pulse, f(x) a real even function satisfying f(0) = 1 and  $f(\pm \infty) = 0$ , and  $E_0$  a complex constant whose absolute value represents the pulse amplitude. Such a pulse makes  $\gamma_n$ , the eigenvalues of equation (5.3), purely imaginary, so that it evolves as a bound state of solitons propagating with the same group velocity

 $<sup>\</sup>dagger$  The general solution of equation (5.1) involves, besides the nonlinear (soliton) components, linear components which correspond to the continuous eigenvalues of equation (5.3). However, contribution of the latter to a whole pulse vanishes with time in accordance with the diffusion of linear Schrödinger wave packets.

 $V_{\rm p}$ . This behaviour is quite different from that of the SIT pulse, which splits into solitons propagating with different velocities.

The solitons in a bound state have different wavenumber shifts  $4|a|\beta_n^2 (\equiv \kappa_n)$ , where the  $i\beta_n$  are given as the eigenvalues of equation (5.4). By approximating f(x) near x = 0by a quadratic function and making use of the quantisation condition (5.5), they are calculated as

$$\beta_n^2 = \beta_0^2 - (n + \frac{1}{2})\Gamma \qquad (n = 0, 1, 2, ...)$$
(5.7)

where

$$\beta_0^2 = [(\Delta - 1)/(4\Delta - 3)]|E_0|^2$$
  

$$\Gamma = 2[(\Delta - 1)/(4\Delta - 3)]^{1/2}|f''(0)|^{1/2}(|E_0|/T_0).$$
(5.8)

The approximation used is good enough for small *n*. Due to these different wavenumber shifts the phase differences between the solitons accumulate with the increase of propagation distance, so that the incoming pulse (5.6) will change its shape. The distance  $Z_c$  traversed by the pulse before its gross shape changes to a considerable extent is measured by  $\pi \kappa_{\max}^{-1}$ , where  $\kappa_{\max}$  is the maximum value of  $\kappa_n$ . By using (5.8),  $Z_c$  is estimated as

$$Z_{\rm c} \sim \pi \kappa_{\rm max}^{-1} \sim 2\pi |\Delta|^3 (1 - \Delta^{-1})^{1/2} |E_0|^{-2}.$$
 (5.9)

The time required for this change is given by  $Z_c/V_p$ , which coincides with  $T_c$  derived in § 2 apart from a numerical factor. As the pulse propagates further, the phase differences between two constituent solitons successively approach  $\pi$  and the envelope E comes to appear as a composite pulse of multiple-peak structure. The distance  $Z_b$  required for this process is measured by  $\pi(\kappa_{n+1}-\kappa_n)^{-1}$ , i.e. the reciprocal of the difference of two adjacent  $\kappa_n$ , and is estimated as

$$Z_{\rm b} \sim \pi (\kappa_{n+1} - \kappa_n)^{-1} \sim \pi \Delta^2 |\Delta - 1| ((4 - 3\Delta^{-1}) |f''(0)|)^{-1/2} (T_0/|E_0|).$$
(5.10)

The maximum amplitude of this multiple pulse is roughly estimated by the amplitude of the constituent soliton with n = 0, which is given by  $(\sqrt{8a/c})\beta_n$ ; this is equal to  $2|E_0|$ , i.e. twice the amplitude of the electric field of the incoming pulse.

Armstrong (1975), who discussed self-steepening of a light pulse in gaseous media, introduced a critical distance  $Z_{crit}$  as the distance traversed by the pulse before its shape becomes singular. It is notable that for  $\Delta \gg 1$ , our  $Z_b$  is nearly equal to his  $Z_{crit}$ , although the pulse never becomes singular in our treatment. The appearance of singularity in Armstrong's theory should be attributable to the approximation used there, which is equivalent to the partial neglect of the second-derivative terms in equation (3.7).

So far we have assumed that the incoming pulse is non-chirped. If it is chirped, then the eigenvalues of equation (5.3) are generally complex, i.e.  $\alpha_n \neq 0$ , so that the solitons (5.2) propagate with different group velocities, and thus the incoming pulse splits into them. If we define the effective pulse area by

$$A = 2 \int_{-\infty}^{\infty} \left(\frac{c}{2a} |u_0(\xi)|^2\right)^{1/2} d\xi = 2\left(\frac{\Delta - 1}{4\Delta - 3}\right)^{1/2} \int_{-\infty}^{\infty} |E(T, Z = 0)| dT$$
(5.11)

the number of the solitons appearing after this split, which is equal to the number of the discrete eigenvalues of equation (5.3), is given by  $A/2\pi$ , as can be calculated by using equation (5.5). It is interesting to see that for  $\Delta \gg 1$ , A in equation (5.11) becomes equal to the pulse area defined by McCall and Hahn (1969), although their area was originally introduced for short pulses with  $\Delta = 0$ .

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Let us now proceed to equation (4.8), the nonlinear Schrödinger equation describing self-modulation of the nonlinear polariton inside the gap. In this case, variables Z and T correspond to  $\rho$  and  $\sigma$  respectively, in contrast to the case of 'outside the gap', so that the initial-value problem in space-time ( $\rho$ ,  $\sigma$ ) is equivalent to the initial-value problem of equation (4.8): not the boundary problem. Therefore, if an initial pulse is once introduced into the medium, it is straightforward to see how this pulse behaves hereafter; it will develop self-modulation and come to behave as an assembly of solitons (4.9). The questions are how a pulse incident on the boundary of the medium gets into it and what kind of initial pulse is formed in the medium. This may roughly be considered as follows. If the electric field of the incident pulse is weak enough, the pulse is totally reflected on the boundary of the medium. An incident pulse whose amplitude  $E_0$ exceeds  $E_{\rm th}$ , on the other hand, can propagate in the medium. If its time width  $T_0$  is sufficiently long, it is approximately regarded as a nonlinear polariton with wavenumber

$$Q_0 \sim (\tilde{\epsilon}(\Delta, |E_0|))^{1/2} \sim [(E_0^2 - E_{\rm th}^2)/2]^{1/2}$$

and group velocity

$$\partial \tilde{\boldsymbol{\epsilon}}(\Delta, |\boldsymbol{E}_0|) / \partial \Delta \sim 2 \boldsymbol{Q}_0.$$

It is therefore reasonable to assume an initial pulse with wavenumber  $Q_0$  and spatial width  $l_0 \sim 2Q_0T_0$  in the medium. Self-modulation of such an initial pulse can then be described by solving the initial-value problem of equation (4.8); our arguments go parallel to those in the case of 'outside the gap', except that the eigenvalues  $\gamma_n$  have a real part  $\alpha_n$  of the order of  $Q_0/2$ . The existence of  $\alpha_n$  means that the pulse propagates with field-dependent velocity  $V_p = 4\alpha_n a \sim [2(E_0^2 - E_{th}^2)]^{1/2}$ , if the velocity differences between the constituent solitons are neglected. From the same consideration as that in the case of 'outside the gap', it is known that the gross shape of the pulse changes after a time  $T_c \sim |E_0|^{-2}$ , and that a multiple-peak structure appears in the pulse after  $T_b \sim l_0/|E_0|$ . In contrast to the case of 'outside the gap', however, it may be probable that, after the time  $T_b$ , the pulse will no longer remain a bound state of constituent solitons, but will have already split into them as a consequence of their different velocities  $4\alpha_n a$ . To make a more detailed description of such a behaviour, however, it is required to solve the boundary problem of equation (4.8) exactly; this is still open for further investigations.

### 6. The effect of interaction between atomic dipoles

In the preceding sections, we neglected the effect of direct interaction between atomic dipoles, setting parameters j and j' in equation (1.2) equal to zero. As has been shown in II, this effect brings about two kinds of steady pulse solutions: one propagating in the form of the radiation field (optical solution) and the other by means of the excitation transfer between atoms (exciton-like solution). Let us briefly discuss here how this effect is reflected in the dynamical properties of a long pulse, confining ourselves to the optical solution. (The optical solution is the solution which corresponds to that obtained in I, i.e. in the case of no direct interaction.)

It has been shown in II that the parameter  $j \ll 1$ , which gives rise to the *K*-dependence of the dielectric function, does not appear in the final expression of the long pulse solution of the optical character, while  $j' (\sim O(1))$  makes a drastic change of the solution, affecting the existence of the steady solution itself, besides a trivial shift of the resonant frequency as seen in equation (1.4). The same thing is expected in the discussion of dynamical properties. It is straightforward to extend the results in the preceding sections so as to take j' into account. Namely, we should only regard the dielectric function  $\tilde{\epsilon}(\Delta, |E|)$  in equations (3.15) and (4.10) as the j'-dependent one<sup>†</sup>:

$$\tilde{\epsilon}(\Delta, |E|) = 1 - 1/\Delta + [(\Delta - j')/2\Delta^4]|E|^2 + \dots$$
(6.1)

Equations corresponding to (3.8) and (4.8) are then obtained by setting  $\tilde{\epsilon}_2(\Delta)$  in equations (3.15) and (4.10) equal to  $(\Delta - j')/2\Delta^4$  instead of  $1/2\Delta^3$ . The result produced by such a simple replacement is by no means trivial. In fact, the coefficient of the second-derivative term and that of the self-potential term in equations (3.8) and (4.8) have the same sign only when

$$\Delta > j'$$
 or  $\Delta < 0$  (6.2)

so that the soliton solution of those equations can exist only in this limited range of frequency. The amplitude of the soliton solutions which correspond to equations (3.8) and (4.8), if they exist, should be multiplied by a factor  $(1-j'/\Delta)^{1/2}$ . Furthermore, it can be shown that the frequency range in which the nonlinear polariton becomes unstable in accordance with the existence of soliton solutions is also limited to equation (6.2).

The effect of j' on the nonlinear polariton inside the polariton gap is especially remarkable. In dielectric media with j' > 1, not only a soliton but also the nonlinear plane wave of the optical character, however intense it may be, cannot propagate in the frequency range  $0 < \Delta < 1$ . It is because the threshold frequency below which the upper branch of the dispersion curve of the nonlinear polariton disappears is given by

$$\Delta_{\rm th} = j' - (j' - 1)(1 - |E|^2)^{1/2}. \tag{6.3}$$

If j' > 1, it increases from  $\Delta_{th} = 1$  with the increase of |E|, although it decreases if j' < 1. A typical example of such a medium with large j' is ionic or semiconducting crystals in which the Wannier exciton model is well applicable.

### 7. Concluding remarks

We have studied the dynamical process through which the steady propagation of a coherent light pulse of long width takes place in a dielectric medium. The results are summarised as follows. In the absence of direct interaction between atomic dipoles, the nonlinear plane wave of polariton is unstable against a small perturbation and develops self-modulation of its envelope. This also holds true for any incoming pulse of sufficiently long width. The evolution process of self-modulation is described by the nonlinear Schrödinger equations (3.8) and (4.8) corresponding to the two cases: respectively the case where the carrier wave frequency lies outside the polariton gap and the case where it lies in the vicinity of the upper edge inside the gap. The soliton solutions of these equations coincide with the lowest-order expressions of steady pulse solutions obtained in I. Also for frequencies inside the gap but not very close to its upper edge, a nonlinear Schrödinger equation whose soliton solution coincides with the steady solution obtained in I has been derived as a natural generalisation of equation (4.8). Introduction of direct interaction between atomic dipoles, on the other hand,

<sup>&</sup>lt;sup>†</sup> The j'-dependent dielectric function can be obtained by eliminating w from equations (3.2) and (3.5) in II. As its closed expression is too complicated, however, we present here only a form of expansion in a power series of |E|.

stabilises the nonlinear plane wave of polariton in a certain range of frequency near the resonance, so that any steady pulse cannot exist in this range. For other frequencies, however, the main results remain unchanged irrespective of such an interaction.

In order that the whole evolution process of self-modulation can be described by a nonlinear Schrödinger equation, it is necessary that the amplitude of the incoming pulse is sufficiently small, i.e.  $|E/\Delta| \ll 1$ , and that its time width is sufficiently long, i.e.  $\tau^{-1} \ll |\Delta \omega|$ . Such a pulse is in striking contrast to the *short* pulse which satisfies  $\tau^{-1} \gg |\Delta \omega|, |\Delta \omega - \omega_{\rm G}|$  and whose evolution obeys a sine-Gordon equation. By applying the initial-value problem of the nonlinear Schrödinger equation studied by Zakharov and Shabat to our problem, it has been shown that a long incoming pulse whose carrier wave frequency lies outside the polariton gap changes its shape with time and becomes a composite pulse of multiple-peak structure, which is regarded as a bound state of the steady pulses obtained in I. A pulse whose frequency lies inside the gap, on the other hand, cannot enter the medium, unless its amplitude reaches a certain threshold value. If it has once come into the medium, it also evolves as an assembly of the steady pulses. We have not studied in detail the boundary problem of such a pulse: that is, how a pulse gets over the interface of the medium. This problem, as well as the problem of evolution of a pulse whose frequency lies inside the gap but not very close to its upper edge, is still open for further investigations.

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