COHERENT OPTICAL PULSES IN CRYSTALS

Okikazu Akimoto

The Institute for Solid State Physics,
The University of Tokyo,
Roppongi, Tokyo 106,
Japan

and

Kensuke Ikeda

Department of Physics,
Kyoto University
Kyoto 606,
Japan

ABSTRACT

Polariton effects on the steady propagation of an optical pulse are studied. It is shown that pulse solutions exist even in the case where the pulse width is sufficiently long compared with the reciprocal of the polariton gap frequency. This suggests that the steady propagation may be realized also in crystals.

I. INTRODUCTION

In the field of quantum optics, a phenomenon named self-induced transparency (SIT) is well known.[1] Consider a dilute absorbing medium such as gaseous atoms or impurity ions in solids. If usual light comes into the medium at resonant frequency, it is absorbed. However, if the incoming light pulse is coherent and sufficiently intense, and if its duration is shorter than the atomic relaxation times, the medium becomes transparent and the pulse propagates without change of its shape. This phenomenon has its origin in the nonlinearity of the medium: The strongly excited atoms do not behave like a harmonic oscillator.

The purpose of this paper is to show theoretically that such a propagation may be realized also in crystals and to propose a new concept *polariton-soliton* which is complementary to SIT.

The concept of polariton is well known. It is a mixed mode of the radiation field and the polarization wave in matter. In dilute media, this effect of mixing is very small and is smeared out by the inhomogeneous dipole-dipole interaction between atoms. In crystals, however, this effect becomes quite significant due to the high density of electric dipoles. The dispersion relation of the polariton differs markedly from that of the photon, giving rise to a gap in which the mixed mode cannot exist. Moreover, when the polarization wave has a dispersion, two modes having different wave numbers appear for one frequency. These two facts, the polariton formation and the spatial dispersion, must be reflected on the optical pulse progagation. In this paper, confining ourselves to the case of no spatial dispersion, we study the effect of polariton formation on the steady propagation of an optical pulse.

An optical pulse has a finite time width, so that it contains a spread of Fourier components of frequency. If this spread covers the polariton gap completely, the pulse cannot feel the existence of the gap. We call such a pulse *short*. In the opposite case, where the spread of Fourier components of frequency is completely inside or completely outside the polariton gap, we call the pulse *long*.

The orders of magnitude of the reciprocal of the polariton gap frequency are roughly microsecond in gases and picosecond in crystals. This tells us that a nanosecond pulse having nearly resonant frequency, for example, is short for gases, but long for crystals. The existing theories of SIT[1~4] applies only to the case of a short pulse, so it is desirable to construct a unified theory which covers both cases.

II. STEADY PULSE AND FUNDAMENTAL EQUATIONS

The starting point of our theory is the same as that of the existing theories. The crystal is represented by a continuous dielectric medium and the radiation field is treated classically. The steady pulse solution we wish to find out is of the form:

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

$$\vec{P}(t,z) = \frac{1}{2}\ N\hbar\varkappa\ [\ u(t-z/V)\ \{\vec{1}\} + v(t-z/V)\{\vec{2}\}\ ]\ , \tag{2}$$

where

$$\{\vec{1}\} = \vec{x}\cos\theta + \vec{y}\sin\theta\ , \quad \{\vec{2}\} = -\vec{x}\sin\theta + \vec{y}\cos\theta$$

$$\theta = \omega t - Kz + \phi(t-z/V).$$

The electric field $\vec{E}$ is factorized into two parts: the slowly varying pulse envelope $\hat{E}$ and the rapidly oscillating carrier wave $\{\vec{1}\}$. Steadiness of the propagation is expressed by letting the envelope be a function of only t-z/V, where V is the pulse velocity. In the carrier wave, ω is the frequency and K is the wave number at the pulse tail; the relation between them is to be determined in a self-consistent manner. Possible phase modulation ($\dot{\phi}\to 0$ for $t\to\pm\infty$) is also taken into account. The macroscopic polarization $\vec{P}$ is the sum of the in-phase component and the out-of-phase component; each component is also factorized in the same way. N is the dipolar density and $\varkappa$ is the dipole matrix element divided by $\hbar/2$.

The two components of the electric dipole, u and v, and the population inversion w, form a classical pseudo-spin vector, the motion of which is governed by a set of equations:

$$\left.\begin{aligned} \dot{u} &= (\omega-\omega_0+\dot{\phi})\ v\ , \\ \dot{v} &= -\ (\omega-\omega_0+\dot{\phi})\ u + \varkappa\hat{E}\ w\ , \\ \dot{w} &= -\ \varkappa\hat{E}\ v\ , \end{aligned}\right\} \tag{3}$$

which is called the optical Bloch equations. Here, ω_0 is the resonant frequency of the medium and all the relaxation times have been assumed to be infinity. Our task is to solve this set of equations together with the Maxwell equation for $\hat{E}$ and ϕ simultaneously. The method of solution is a power-series expansion in which all the quantities in the fundamental equations are expanded in terms of a small parameter related to the pulse width. The small parameter will be chosen in different ways depending on what kind of pulse we treat, a short pulse or a long pulse.

III. BEHAVIOR OF PULSE TAIL

Before solving our equations, let us determine the dispersion relation at the pulse tail. In order to see the behavior of the pulse tail where the excitation is very weak, it is sufficient to consider linearized version of our equations. If one solves the linearized equations, which are given by setting w equal to -1 corresponding to the atomic ground state, one can obtain exponential solutions $E \propto \exp \pm(t-z/V)/\tau$ in general, besides the plane wave solution which corresponds to the usual polariton. Such a divergent solution is physically meaningless as itself in infinite media, and is usually left out of consideration. However, the tail of a steadily propagating pulse is not a plane wave but a growing (or decaying) wave, so that its behavior should be described at least locally by such an exponential solution. When the nonlinearity is taken into account, the divergence of this solution is suppressed and a pulse is formed; at the same time, the dispersion relation around the pulse peak is modulated through non-zero $\dot{\phi}$. The growth (or decay) constant τ, which is an integral constant, then gives a measure of the pulse width, so we call it the pulse width hereafter. The wave number K and the pulse velocity V determined as functions of ω and τ from the linearized equations are as follows:

$$\left(\frac{cK}{\omega}\right)^2 = \frac{1}{2}\left\{ 1 - \frac{\Delta}{\Delta^2 + \lambda^2} + \sqrt{\frac{(\Delta-1)^2+\lambda^2}{\Delta^2+\lambda^2}} \right\} , \qquad (4)$$

$$\left(\frac{c}{V}\right)^2 = \frac{(\omega\tau)^2}{2}\left\{-1 + \frac{\Delta}{\Delta^2+\lambda^2} + \sqrt{\frac{(\Delta-1)^2+\lambda^2}{\Delta^2+\lambda^2}}\right\}, \tag{5}$$

where Δ and λ are the frequency difference and the reciprocal pulse width scaled by the polariton gap frequency, *i.e.*

$$\Delta = \frac{\omega-\omega_0}{2\pi N\hbar\varkappa^2}, \qquad \lambda = \frac{\tau^{-1}}{2\pi N\hbar\varkappa^2}. \tag{6}$$

In eqs.(4) and (5), $\lambda \ll \omega\tau$ has been assumed.

The τ-dependent dispersion relation given by eq.(4) is plotted in Fig.1. When λ is large, namely in the case of a short pulse, the dispersion relation is close to that of the photon. When λ becomes small, *i.e.* when the pulse width becomes long, the dispersion relation approaches that of the polariton. An important fact seen in Fig.1 is that K remains real even inside the polariton gap. This fact means that a growing (or decaying) wave can propagate inside the gap, although a plane wave cannot. It suggests that a long pulse having an exponential tail can also propagate inside the gap.

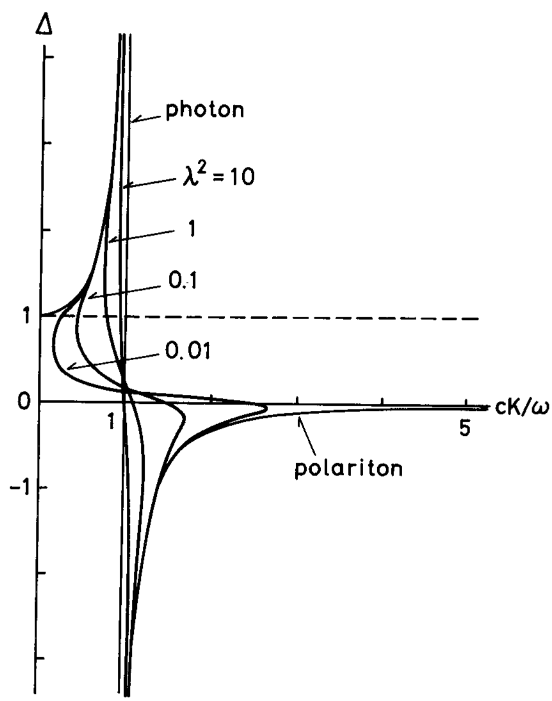

Fig.1. Dispersion relation at the pulse tail. Δ and λ are the frequency difference $\omega-\omega_0$ and the reciprocal of the pulse width τ^{-1}, respectively, scaled by the polariton gap frequency. The polariton gap corresponds to $0<\Delta<1$.

IV. PULSE SOLUTIONS

Now, we return to our nonlinear equations. By choosing a dimensionless time $(t-z/V)/\tau$ as the variable and using the inequality $(\omega\tau)^{-1} \ll 1$, our equations are reduced to five ordinary differential equations for $\hat{E}/2\pi N\hbar\varkappa$, ϕ, u, v and w, the coefficients of which are functions of λ and Δ. We expand all the coefficients and the functions to be determined in power-series of λ/Δ or $1/\lambda$, according to the long pulse ($\lambda \ll |\Delta|$) or the short pulse ($\lambda \gg 1$ and $\lambda \gg |\Delta|$), respectively. By this expansion, the equations are reduced to simpler forms, which can be solved easily.

The results are summarized as follows. Behaviors of the pulse strongly depend on ω and τ. The short pulse is the same as the usual SIT pulse, and is not affected by the existence of the polariton gap. The pulse envelope has a hyperbolic secant shape, and the population is completely inverted at the pulse peak; *i.e.*

$$\left.\begin{aligned} \varkappa\hat{E} &= 2\tau^{-1}\,\mathrm{sech}\left[(t-z/V)/\tau\right], \\ w &= -1 + 2\,\mathrm{sech}^2\left[(t-z/V)/\tau\right]. \end{aligned}\right\} \qquad (7)$$

The wave number K is close to that of the photon, and the group velocity V is given by $c/V \sim 1+(\omega\tau/2\lambda)$ and is smaller than the light velocity. A principal part of the polarization $\vec{P}$ is given by the out-of-phase (absorptive) component v of the dipole moment; that is, the inequality $u \ll v$ holds in this case.

The long pulse, on the other hand, shows different behaviors outside and inside the gap. Outside the gap ($\Delta > 1$ or $\Delta < 0$), the pulse has a hyperbolic secant shape, but as the pulse width becomes longer, both the envelope and the population inversion tend to zero; their explicit forms are

$$\left.\begin{aligned} \varkappa\hat{E} &= \sqrt{\frac{4\Delta-3}{\Delta-1}}\ \tau^{-1}\ \mathrm{sech}[\ (t-z/V)/\tau\ ], \\ w &= -1 + \frac{1}{2(\omega-\omega_0)^2}\ (\varkappa\hat{E})^2 \end{aligned}\right\} \qquad (8)$$

When $\tau\to\infty$, the wave number and the pulse velocity approach those of the polariton. Furthermore, in contrast with the case of a short pulse, a principal part of the polarization is given by the in-phase (dispersive) component u; that is $u \gg v$. This means that the electric dipoles adiabatically follow the electric field. Because of these facts, the pulse may be called a polariton-soliton.

The long pulse can propagate with real K and V also inside the gap. In this case, however, the envelope has no longer a hyperbolic secant shape, and does not vanish even in the limit of $\tau\to\infty$;

$$\left.\begin{aligned} \varkappa\hat{E} &= f(t-z/V;\ \omega-\omega_0)\cdot 2\pi N\hbar\varkappa^2\ , \\ w &= -(\omega-\omega_0)\ [\ (\varkappa E)^2 + (\omega-\omega_0)^2\ ]^{-1/2}\ . \end{aligned}\right\} \qquad (9)$$

Here, f is a dimensionless real function of the order of unity. Near the upper edge of the polariton gap, f is approximated by a hyperbolic secant, *i.e.* $f \sim 2\sqrt{1-\Delta}\ \mathrm{sech}\,[\ (t-z/V)/\tau\ ]$, while near $\omega=\omega_0$, it is close to a period of cosine $\{1+\cos[\sqrt{\Delta}\ (t-z/V)/\tau]\}$ $(-\pi < \sqrt{\Delta}(t-z/V)/\tau < \pi)$ accompanied with an exponential tail. In the limit of $\tau\to\infty$, the pulse velocity and the wave number tend to zero, while the spatial width of the pulse V remains finite. This means that the pulse becomes a sort of standing wave without spatial oscillation in this limit. If we solve the linearized equations by assuming constant $\hat{E}$ inside the gap, we can obtain an exponentially decaying (or growing) solution without spatial oscillation, because the wave number is purely imaginary there. Such a solution can appear just inside the surface of the medium irradiated by light. The tail of the pulse solution we have obtained above continues to this type of linear solutions.

In the case of intermediate pulse width, our equations are not solved analytically, so they have to be treated numerically.

All the pulse solutions are classified in Fig.2. In this figure,

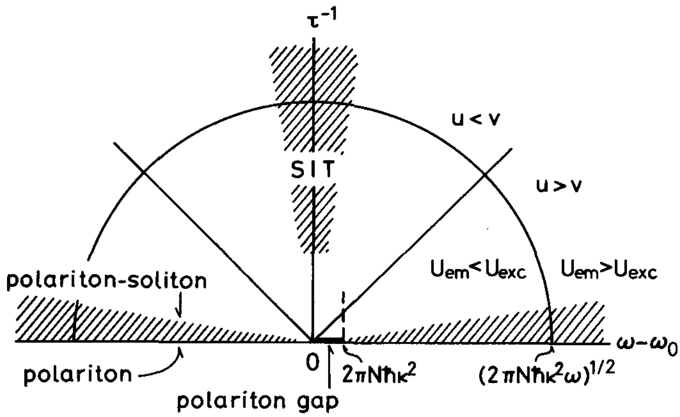

Fig.2. Classification of the pulse solutions. See the text.

the abscissa is the optical frequency measured from the resonant frequency, and the ordinate the reciprocal of the pulse width. The usual polariton corresponds to the line $\tau^{-1}=0$. Above the straight line $\tau^{-1} = |\omega-\omega_0|$, the out-of-phase component v of the dipole moment is larger than the in-phase component u; the pulses in this region may be called SIT-like. Below this line, the in-phase component is larger; the pulses may be called polariton-like. The usual SIT and the polariton-soliton we have studied above are indicated by the hatched regions. Outside the circle shown in Fig.2, the energy density of electromagnetic field $E^2/4\pi$, which is denoted by U_{em} in Fig.2, is larger than the energy density of excitation $N\hbar\omega(w+1)$, which is denoted by U_{exc} in the figure; inside the circle, it is reversed.

In conclusion, we have shown that, besides the usual SIT pulse, steady pulse solutions exist also in the case where the pulse width is sufficiently long compared with the reciprocal of the polariton gap frequency, and that such a long pulse having frequency outside the gap behaves as a polariton-soliton. Our conclusion suggests that the steady pulse propagation may be realized also in crystals, though it has been obtained by using a simplified model without taking into consideration realistic conditions such as spatial dispersion, relaxation times, surface effects *etc.* In view of recent remarkable development of picosecond pulse technique, it is hoped to observe such a propagation in near future.

REFERENCES

1) S. L. McCall and E. L. Hahn: Phys. Rev. 183 (1969) 457.
2) G. L. Lamb, Jr.: Rev. mod. Phys. 43 (1971) 99.
3) H. Haken and A. Schenzle: Z. Physik 258 (1973) 231.
4) R. A. Marth, D. A. Holmes and J. H. Eberly: Phys. Rev. A9 (1974) 2733; Z. Bialynicka-Birula, Phys. Rev. A10 (1974) 999.