Time-Dependent Optical Spectroscopy of Polariton Systems Having a Minimum Group Velocity

David Linton Johnson
Department of Physics, Northeastern University, Boston, Massachusetts 02115
(Received 12 September 1977)

If a pulse of light is transmitted through a slab of material whose polariton dispersion curve has a minimum value for the group velocity [$\omega(q)$ has an inflection point], then the intensity of emergent light will oscillate before decaying, in accordance with the properties of the Airy function. The effect is linear in the applied field of the incident light. Application is made to the A ($n = 1$) exciton-polariton of CdS, with no adjustable parameters.

Recent interest has emerged in systems exhibiting spatial dispersion—the dielectric function depends on wave vector (q) as well as frequency (ω)—in an exciton-polariton mode. Indeed, the dispersion curves $\omega(q)$ have been measured for both polariton branches in GaAs and in CdS by resonant Brillouin scattering,[1] confirming ideas based on simple models such as Eqs. (5) and (6) below. To date there has been very little interest in the time-dependent spectroscopy (TDS) of such systems, although TDS has been used to investigate a fascinating variety of phenomena such as self-induced transparency, optical nutation, etc.[2] In this Letter, it is demonstrated that when a pulse of light is transmitted through a slab of material whose polariton dispersion curve has a minimum value of the group velocity, the intensity of the trailing edge oscillates before damping out; the envelope function (the amplitude) is describable by an Airy function and, unlike self-induced transparency, this is a linear effect.

Frankel and Birman[3] (FB) previously showed that the leading edge of the pulse should exhibit similar behavior due to the classical Sommerfeld and Brillouin precursors[4] plus a new exciton precursor (EP). However, these precursors are extremely weak as will be discussed below. The Sommerfeld and Brillouin precursors have not been detected in real optical systems although they were predicted over 60 years ago; the exciton precursor is apparently intermediate in strength between the two.[3]

Consider any linear one-dimensional wave phenomenon exhibiting dispersion

$$A(x,t) = \int \hat{a}(\omega) \exp\{i[q(\omega)x - \omega t]\} d\omega. \qquad (1)$$

In the presence of damping, $q(\omega)$ may be complex. If $A(x,t)$ in a finite pulse of a single plane wave [$\hat{a}(\omega)$ is peaked at some ω_0] and there is no damping, then simple considerations[5] indicate that the single plane wave is modulated by an envelope function which propagates with the pertinent group velocity $v_g = d\omega/dq$ and which broadens as it propagates if $dv_g/dq \neq 0$. The more rigorous derivation[6,7] shows that if x,t are large and $x/t = v$ (a constant), one "sees" only that wave whose group velocity is v. If the dispersion relation is such that $v_g(q)$ takes on a minimum value v_g^* [i.e., $dv_g(q^*)/dq = d^2\omega(q^*)/dq^2 = 0$], then there is a relatively *sharp* boundary between an unperturbed region $x < v_g^* t$ and a perturbed one $x > v_g^* t$, at least asymptotically.[7,8] To handle the transition region, one keeps the cubic term in the expansion[9]:

$$q(\omega) = q^* + (v_g^*)^{-1}(\omega - \omega^*) - \xi(v_g^*)^{-4}(\omega - \omega^*)^3, \qquad (2)$$

where $\xi = \frac{1}{6}[d^3\omega/dq^3]_{q^*} > 0$. Equation (1) now becomes

$$A(x,t) = \exp[i(q^* x - \omega^* t)] \int \hat{a}(\omega) \exp\{i[(\omega - \omega^*)(x/v_g^* - t) - \xi(v_g^*)^{-4}(\omega - \omega^*)^3 x]\} d\omega. \qquad (3)$$

Equation (3) is an example of a situation where the standard asymptotic results[6] based on the method of steepest descent and/or stationary phase do not apply, basically because there is no $(\omega - \omega^*)^2$ term in Eq. (2), and because we are interested only in the region $|x - v_g^* t| \ll x$. Considered as a function of ω, the exponential is highly oscillatory for $\omega \neq \omega^*$ and becomes more so as the wave progresses, because of the cubic. Hence $\hat{a}(\omega)$, a relatively smoothly varying function {at least compared to $\exp[-i\xi \times (v_g^*)^{-4}(\omega - \omega^*)^3 x]$}, does not contribute for $\omega \neq \omega^*$, in the asymptotic limit $x \to \infty$. It may therefore be assumed constant and be taken outside the integral; the remainder is an Airy function[10]:

$$A(x,t) = 2\pi \hat{a}(\omega^*) v_g^* \exp[i(q^* x - \omega^* t)](v_g^*/3\xi x)^{1/3} \mathrm{Ai}[(v_g^* t - x)(v_g^*/3\xi x)^{1/3}]. \qquad (4)$$

More rigorous derivations are given in Ref. 7 and especially Ref. 8. Roughly speaking, for $v_g \gtrsim v_g^*$ there are two waves of differing wavelengths which alternately interfere constructively and destructively.

The derivation of Eq. (4) is still correct in the presence of damping; all parameters are complex and there are no poles or branch cuts between ω^* and the real axis.[3] Use of the complex v_g^* has obviated the necessity of introducing the signal velocity or the energy velocity.[4]

The clearest example of the applicability of these concepts is waves on the surface of a liquid,[7] in which restoring forces due to gravity and to surface tension combine to give a minimum v_g. For water waves ($\rho = 1$ g/cm^3, $T = 72$ dyn/cm), this minimum group velocity is $v_g^* = 18$ cm/sec with a corresponding wavelength $2\pi/q^* = 4.3$ cm. The asymptotic approximation is valid for distances greater than $(3\xi/v_g^*)^{1/2}$ which is 0.4 cm for water waves, much less than one wavelength. The thickness of the transition region is $[x/(0.4\ \text{cm})]^{1/3}(0.4\ \text{cm})$ which is only 4.0 cm, less than a wavelength, after the disturbance has propagated a distance of 4 m—a very sharp transition indeed. Hence if a stone is dropped in a deep pool of still water, "the most conspicuous feature is the ring of waves with the length corresponding to the minimum group velocity surrounding a circle of smooth water."[7] This theoretical treatment has actually been experimentally verified in detail for a maximum group velocity which occurs in water of a finite depth.[11]

In solids of high symmetry the dispersion relations for transverse polariton modes are given

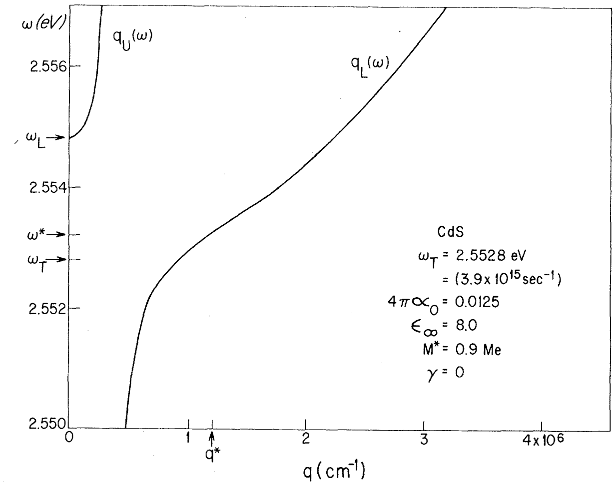

FIG. 1. Dispersion relations for the two transverse branches of the $A(n=1)$ exciton-polariton in CdS as determined from Eqs. (5) and (6). The input data were taken from Ref. 13 except that $\gamma = 0$ was used. The group velocity of the lower branch is a minimum at the inflection point, ω^*. There is also a longitudinal branch which is not shown.

by[12]

$$\omega^2 \epsilon_t(q,\omega) = c^2 q^2, \tag{5}$$

where ϵ_t is the transverse dielectric function. Near a sharp exciton line it is given approximately by[12]

$$\epsilon_t(q,\omega) = \epsilon_\infty + \frac{4\pi\alpha_0 \omega_T^{\,2}}{\omega_T^{\,2} + (\hbar\omega_T/M)q^2 - \omega^2 - i\gamma\omega}. \tag{6}$$

Equations (5) and (6) together yield an upper (U) and a lower (L) polariton branch which are plotted in Fig. 1 for the $A(n=1)$ exciton of CdS.[13] Be-

TABLE I. Numerically determined values of quantities associated with the inflection point of Fig. 1, except that $\gamma = 10^{-4}$ eV (Ref. 13) has been used.[19] If $\gamma = 0$, the imaginary parts vanish, the real are unchanged (vice versa for q_U^*). A slab thickness $a = 10^{-4}$ cm was assumed for the last two entries. Values for $M = 0.4m_e$ are provided for comparison only.

	$M = 0.9m_e$	$M = 0.4m_e$
ω^*	$(2.5533,\ -5.\times 10^{-5})$ eV	$(2.5536,\ -5.\times 10^{-5})$ eV
q_L^*	$(1.26\times 10^{6},\ -14.)$ cm^{-1}	$(1.06\times 10^{6},\ -12.)$ cm^{-1}
q_U^*	$(8.,\ 5.5\times 10^{5})$ cm^{-1}	$(6.,\ 3.8\times 10^{5})$ cm^{-1}
v_g^*	$(2.1\times 10^{6},\ -22.)$ cm/sec	$(3.9\times 10^{6},\ -43.)$ cm/sec
dv_g/dq_L	0	0
$d^2v_g/dq_L^2 = 6\xi$	$(4.6\times 10^{-6},\ -1.4\times 10^{-10})$ cm^3/sec	$(1.3\times 10^{-5},\ -5.\times 10^{-10})$ cm^3/sec
$\omega_M = [(v_g^*)^3/3\xi]^{1/2}$	$(2.0\times 10^{12},\ -1.6\times 10^{6})$ sec^{-1}	$(3.1\times 10^{12},\ 6.\times 10^{6})$ sec^{-1}
$T_0 = a/v_g^*$	$(48.,\ 5.\times 10^{-4})$ psec	$(25.,\ 3.\times 10^{-4})$ psec
$\alpha = (\omega_M^2/T_0)^{1/3} = [(v_g^*)^4/3\xi a]^{1/3}$	$(4.4\times 10^{11},\ -1.8\times 10^{6})$ sec^{-1}	$(7.1\times 10^{11},\ -1.6\times 10^{6})$ sec^{-1}

low ω_L the wave vector of the upper polariton branch, $q_U(\omega)$, is pure imaginary (and large near ω_T). The group velocity of the lower branch has a minimum at ω^*; some pertinent quantities, derived numerically, are listed in Table I, where the effect of damping [$\gamma = 10^{-4}$ eV (Ref. 13)] has been included.

It is also clear that the group velocity takes on relative *maxima* at the extremal limits of the branches of Fig. 1. The Sommerfeld precursor[4] is governed by the high-frequency limit of $q_U(\omega) \to \epsilon_\infty^{1/2}\omega/c$, the Brillouin precursor[4] by the low-frequency limit of $q_L(\omega) \to (\epsilon_\infty + 4\pi\alpha_0)^{1/2}\omega/c$, and the new exciton precursor[3] by the high-frequency limit of $q_L(\omega)$ [$\to (M/\hbar\omega_T)^{1/2}\omega$ if Eq. (6) is taken seriously in this limit].

If the carrier frequency of the pulse is "near" ω_T and if the duration of the pulse is larger than $1/\omega_T \sim 10^{-15}$ sec, the fourier spectrum $a(\omega)$ in Eq. (1) falls off rapidly in either the high-frequency or low-frequency limit. Hence all three precursors discussed by Frankel and Birman[3] have small amplitudes as can be seen from their Table I. (They also neglected damping and used a large propagation distance in a semi-infinite sample.) By contrast, if the carrier frequency is tuned to ω^* so that $\hat{a}(\omega)$ is largest at ω^*, the amplitude of the emergent pulse is largest for $x \approx v_g^* t$ for which Eq. (4) applies. If $\hat{a}(\omega)$ is detuned from ω^*, most of the wave propagates faster than v_g^* and Eq. (4), while true, has a small amplitude compared to the main pulse.

To be more concrete, let us consider the specific model of the nonlocal electromagnetic properties of a slab introduced by Pekar.[14] Light is normally incident on a slab occupying the region $0 < z < a$.[15] In this model, each frequency component of the incident field

$$E_I(z,t) = \int \hat{\epsilon}(\omega) \exp[i(\omega z/c - \omega t)] d\omega, \tag{7}$$

couples to a reflected field, a transmitted field, and a right-going and a left-going wave for each of the two polariton modes of that frequency. The approximate susceptibility function is specified uniquely by stipulating the additional boundary conditions (ABC), $P_x(\text{exciton}) = 0$ on the two surfaces $z = 0, a$; this is equivalent to the value $U = -1$ in the susceptibility function of Johnson and Bimbey.[16] The transmitted field (for $a < z$) is

$$E_T(z,t) = \int D(\omega)\hat{\epsilon}(\omega) \exp[i(\omega z/c - \omega t)] d\omega, \tag{8}$$

where $D(\omega)$ is given explicitly by formulas (25), (60), and (61) of Ref. 14. If the thickness of the slab is a > 200 Å, then $\exp[iq_U(\omega)a] \ll 1$ for $\omega \approx \omega^*$ and, in this case, $D(\omega)$ reduces to

$$D(\omega) = \frac{p(\omega)\exp\{i[q_L - (\omega/c)]a\}}{1 - s(\omega)\exp(2iq_L a)}, \tag{9}$$

where p and s do *not* depend on a. The transmitted field, evaluated as $z \to a+$ is [from (8) and (9)]

$$E_T(a,t) = \sum_{n=0}^{\infty} \int \hat{\epsilon}(\omega)p(\omega)s^n(\omega)\exp\{i[q_L(\omega)(2n+1)a - \omega t]\}\, d\omega. \tag{10}$$

The index n in (10) is the number of round trips the wave makes before emerging from the slab. It is clear that each term has the general property of Eq. (4) for $t \approx (2n+1)a/v_g^*$ [$\hat{a}(\omega) = \hat{\epsilon}(\omega)p(\omega)s^n(\omega)$]. In particular, the intensity of light due to the first term can be written, for $t \approx T_0 = a/v_g^*$, as

$$I(t) \propto e^{-\beta a} a^{-2/3} |\mathrm{Ai}[\alpha(t - T_0)]|^2, \tag{11}$$

where $\alpha = [(v_g^*)^4/3\xi a]^{1/3}$ and T_0 are given in Table I for a slab of thickness $a = 1\,\mu$m, which was chosen because it is a convenient thickness for such transmission experiments[17]; the upper branch has completely damped out and the lower is essentially undamped. The thickness must also be large enough for the asymptotic approximation to be valid i.e., $a \gg (3\xi/v_g^*)^{1/2}$ which is indeed the case. The time scale of Eq. (11) is $1/\alpha$ ($\simeq$2.3 psec) which is only weakly thickness dependent ($\sim a^{1/3}$). Such picosecond resolution has already been achieved in a number of systems.[18]

The prefactor in (11) does not depend on a or t; it does depend on the detailed nature of the surface (e.g., on the ABC) but it is clear that (11) will hold, regardless of ABC, as long as $\exp(iq_U a) \ll 1$ and the asymptotic expansion is valid. That is, each Fourier component of the incident field couples to both polariton branches in amounts depending on ABC; the upper branch quickly damps out, whereas the lower acquires a phase $\exp[iq_L(\omega)a]$ when it reaches the second surface. Therefore, the intensity of light passing through the slab will pulsate (in time) before decaying exponentially, in accordance with the properties of the Airy function.

The imaginary parts of α, T_0 are first order in the damping parameter γ [Eq. (6) depends on $i\gamma$] and are extremely small. We may therefore expand Eq. (11) in a Taylor's series about $\gamma = 0$. Using the notation $\alpha = \alpha' + i\alpha''$, etc., and keeping only the lowest-order terms in γ, one finds

$$|\mathrm{Ai}[\alpha(t-T_0)]|^2 = \{\mathrm{Ai}[\alpha'(t-T_0')]\}^2 + \{\alpha''(t-T_0') - \alpha' T_0''\}^2 \{\mathrm{Ai}'[\alpha'(t-T_0')]\}^2.$$

Since the maxima of $[d\mathrm{Ai}(z)/dz]^2$ coincide with the zeroes of $\mathrm{Ai}^2(z)$ (and vice verse), we see that damping tends to blur structure in the intensity, in addition to the exponential thickness dependence. For the parameters of Ref. 13, as evinced by Table I, $\{\alpha''(t-T_0'') - \alpha' T_0''\}$ is less than 6×10^{-8} for all values of t such that $|\alpha'(t-T_0')| < 10$. This includes the last seven zeros of the Airy function. Therefore, even in the presence of damping, the ratios of relative maxima to minima in the intensity of the trailing edge seem to be quite large. Kiselev *et al.* have concluded that $\gamma \approx 10^{-5}$ eV in some of their samples.[17]

By performing this experiment, one ought to be able to determine ω^*, v_g^*, and d^2v_g/dq^2 (but not q^*) purely by optical means and, more importantly, the interpretation is independent of assumed ABC's (a drawback present in more conventional techniques).[16]

I would like to acknowledge several very fruitful discussions with Professor N. Kopell of the Mathematics Department at Northeastern University, Professor E. Ippen of Bell Laboratory and Massachusetts Institute of Technology, and Professor M. H. Cohen of University of Chicago.

[1]R. G. Ulbrich and C. Weisbuch, Phys. Rev. Lett. 38, 865 (1977); G. Winterling, E. S. Koteles, and M. Cardona, Phys. Rev. Lett. 39, 1286 (1977).

[2]R. G. Brewer, *Phys. Today* 30, No. 6, 50 (1977), and references therein.

[3]M. Frankel and J. L. Birman, Opt. Commun. 13, 303 (1975), and Phys. Rev. A 15, 2000 (1977).

[4]L. Brillouin, *Wave Propagation and Group Velocity* (Academic, New York, 1962).

[5]J. D. Jackson, *Classical Electrodynamics* (Wiley, New York, 1962), Chap. 7.

[6]L. A. Segel, *Mathematics Applied to Continuum Mechanics* (Macmillan, New York, 1977).

[7]H. Jeffreys and B. S. Jeffreys, *Methods of Mathematical Physics* (Cambridge Univ. Press, New York, 1972), 3rd ed. Some of the numerical values derived here are in error.

[8]C. Chester, B. Friedman, and F. Ursell, Proc. Cambridge Philos. Soc. 53, 599 (1957).

[9]Refs. 6–8 expand Eq. (1) as an integral over wave number q. In order to make comparison with experiment it is convenient [especially for Eq. (10)] to work in the frequency regime. The only difference is that $x^{-1/3}$ in Eq. (4) will be replaced by $(v_g^* t)^{-1/3}$, which is no difference at all for an asymptotic expansion.

[10]M. Abramowitz and I. A. Stegun, *Handbook of Mathematical Functions* (Dover, New York, 1965).

[11]R. Wiegel *et al.*, University of California Wave Research Project Technical Report No. HEL 19-1, 1968 (unpublished). The key illustration is reproduced in Fig. 9.5 of Ref. 4.

[12]J. J. Hopfield and D. G. Thomas, Phys. Rev. 132, 563 (1963).

[13]F. Evangelisti, A. Frova, and F. Patella, Phys. Rev. B 10, 4253 (1974).

[14]S. I. Pekar, Zh. Eksp. Teor. Fiz. 34, 1176 (1958) [Sov. Phys. JETP 34, 813 (1958)], and references therein. Notation is somewhat different from the present article: $q_L = n_+\omega/c$, etc.

[15]It is assumed that the c axis lies in the surface plane and that the electric field is perpendicular to it.

[16]D. L. Johnson and P. R. Rimbey, Phys. Rev. B 14, 2398 (1976).

[17]V. A. Kiselev, B. S. Razbirin, and I. N. Uraltsev, Phys. Status Solidi (b) 72, 161 (1975), and references therein.

[18]A. Loubereau, G. Wochner, and W. Kaiser, Phys. Rev. A 13, 2212 (1976); R. R. Alfano, Opt. Commun. 18, 193 (1976); E. P. Ippen, C. B. Shank, and R. L. Woerner, Chem. Phys. Lett. 46, 20 (1977).

[19]F. Patella, F. Evangelisti, and M. Capizzi, Solid State Commun. 20, 23 (1976).