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Linear Pulse Propagation in an Absorbing Medium

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The pulse velocity in the linear regime in samples of GaP:N with a laser tuned to the bound A-exciton line is measured with use of a picosecond time-of-flight technique. The pulse is seen to propagate through the material with little pulse-shape distortion, and with an envelope velocity given by the group velocity even when the group velocity exceeds 3×10^{10} cm/sec, equals $\pm\infty$, or becomes negative. The results verify the predictions of Garrett and McCumber.

Electromagnetic pulse propagation in a dispersive medium has received a great deal of attention, but the subject continues to be plagued by widely held misconceptions. It is well known that the group velocity

$$v_g = \frac{d\omega}{dk} = \frac{c}{n(\omega) + \omega(dn/d\omega)} \quad (1)$$

describes the propagation of an electromagnetic pulse in a linear dispersive but nonabsorbing medium. However, in regions of strong anomalous dispersion, the group velocity can exceed c, or even become negative. The common belief is that the meaning of group velocity breaks down, and the behavior of the pulse becomes much more complicated.[1] Indeed, the well-known work of Sommerfeld and Brillouin[2] shows that for a pulse that turns on abruptly at some given instant and then follows a sinusoidal modulation, the original pulse becomes distorted, and although there are precursors of the pulse that travel at c, the main part of the pulse arrives at a "signal velocity" slower than c. Also, the energy velocity, v_E, defined as the rate of energy flow divided by the stored energy density, has been shown to be less than c.[3] These ideas of energy velocity, group velocity, and signal velocity are, of course, strongly reinforced by our concepts of special relativity, and physicists may be tempted to analyze pulse propagation experiments in

terms of energy velocities and not group velocities.[4] Nevertheless, Garrett and McCumber[5] have shown that under certain easily satisfied approximations, there is an analytic solution that predicts that the pulse will propagate with a velocity equal to the group velocity even when $v_g > c$, $v_g = \pm\infty$, or $v_g < 0$. (A negative pulse velocity occurs when the peak of the pulse emerges from the sample at an instant before the peak of the pulse enters the sample.) The work reported here verifies for the first time their predictions in the region where v_g passes through $\pm\infty$ and becomes negative. It also clearly demonstrates that v_E is not the measured quantity in this type of pulse propagation experiment.

A pulse velocity that exceeds c or is negative does not necessarily violate special relativity or causality. As discussed by Garrett and McCumber[5] and Crisp,[6] the effect is due to a pulse reshaping, and the leading edge of the pulse is less attenuated than the trailing edge. *The insidious aspect of the pulse shaping is that apart from an overall attenuation, the shape and width of the pulse can remain intact after it emerges from the sample.*

The essential ingredients to Garrett and McCumber's analysis are summarized as follows: They assume that the pulse described by $F(k,\omega)$, the Fourier-Laplace transform of an initial pulse $f(z,t)$, can be described by Maxwell's equations

in the form

$$[(\omega/c)n(\omega) - k]F(k, \omega) = S(\omega), \qquad (2)$$

where $S(\omega)$ is the envelope of the source field describing the incident signal $(z = 0^+)$. Then for $z > 0$,

$$f(z, t) = \int_{-\infty}^{\infty} d\omega\, e^{-i\omega[t - i\omega z n(\omega)/c]} S(\omega). \qquad (3)$$

Instead of a step-function envelope used by Sommerfeld and Brillouin, they assumed (1) that $S(\omega)$ is a Gaussian Fourier-transform–limited pulse, (2) that $\Delta\nu_L \ll \Delta\nu_{abs}$, where $\Delta\nu_L$ and $\Delta\nu_{abs}$ are frequency spreads of the laser and absorption line, and (3) that the number of absorption lengths of the sample is much less than $(\Delta\nu_{abs}/\Delta\nu_L)^2$. Then a Taylor series expansion of $\omega n(\omega)$ is possible, and a straightforward integration yields the result that (i) the pulse propagates with the group velocity, and (ii) the emerging pulse is a Gaussian with an almost identical pulse width.

These dispersion effects have been previously studied in both amplifying and absorbing media using the 0.63-μm line in neon[7] and the 3.5-μm transition in xenon[8] by measuring the change in pulse repetition frequency, $c/2L$, of a mode-locked laser as a function of laser power. In the case of neon, the changes in v_g were less than 1 part in 1000, and in the case of xenon, v_g in the gain medium was shown to be 2.5 times slower than c.

By using a picosecond time-of-flight technique, we have mapped pulse velocity over the entire line and compared it to an *independent measurement of* v_g. In this work, the pulse velocity not only exceeds c, but is measured to go smoothly through $\pm\infty$, and, in some samples, to have a negative value as low as $-1 \times 10^{+8}$ cm/sec.

The experimental arrangement is shown in Fig. 1, and is very similar to the scheme used by Ulbrich and Fehrenbach.[9] The material used was epitaxially grown GaP:N, and the laser was tuned in the vicinity of the well isolated bound A-exci-
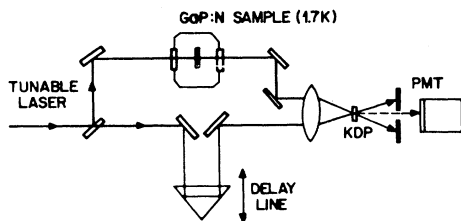

FIG. 1. A schematic of the experimental arrangement.

ton line at 534 nm. Data were taken on samples with nitrogen concentrations of $[N] = 1.5 \times 10^{17}$ cm^{-3}, 2.5×10^{18} cm^{-3}, and 3.8×10^{18} cm^{-3}. The thickness of the epilayer (between 9.5 and 76 μm) was adjusted by careful polishing so that the peak absorption never exceeded ~6 absorption lengths. The laser was a sync-pumped dye laser with a 3-plate birefringent filter. The frequency width of the laser was changed by use of additional uncoated intracavity etalons of 0.12, 0.5, or 2.0 mm thickness. The 0.5- and 2.0-mm-thick etalons produce laser pulses with $\Delta\tau_L = 22$ and 48 ps, respectively, and with $\Delta\nu_L\Delta\tau_L \simeq 0.35$ [$\Delta\nu_L$ = laser bandwidth, full width at half maximum (FWHM), and $\Delta\tau_L$ = autocorrelation pulse width, FWHM]. This $\Delta\nu_L\Delta\tau_L$ is between the Fourier-transform limit of a hyperbolic secant and exponential pulse.[10] The pulse delays measured are independent of peak intensities used between 100 and 3 W/cm^2. These intensities are 4 or more orders of magnitude below any saturation intensities in this time scale.[11] Finally, we moved the sample in and out of the laser beam to confirm that the off-resonance pulse in fact propagates with the known velocity c/n_0 where n_0 is the off-resonance index of refraction.
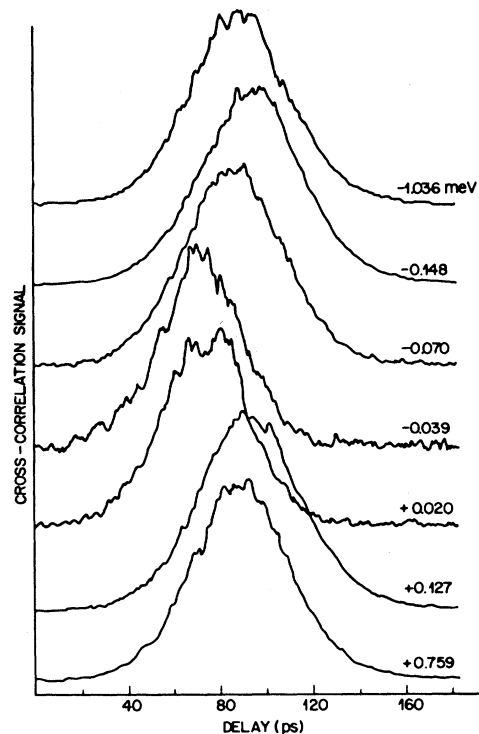

FIG. 2. A sample of the cross-correlation data as the laser is tuned through the exciton line of the $[N] = 1.5 \times 10^{17}$ cm^{-3} sample.

A sample of the cross-correlation scans is shown in Fig. 2. When $\Delta\nu_L$ is appreciably smaller than the width of the absorption line $\Delta\nu_{abs}$, no statistically significant pulse shaping occurs as the laser is tuned through the resonance line. However, when $\Delta\nu_L \simeq \Delta\nu_{abs}$, one can see noticeable pulse shaping and/or an overall decrease in the pulse advance. [See Fig. 3(b).] The center of each pulse relative to an arbitrary delay time is plotted as a function of laser frequency in
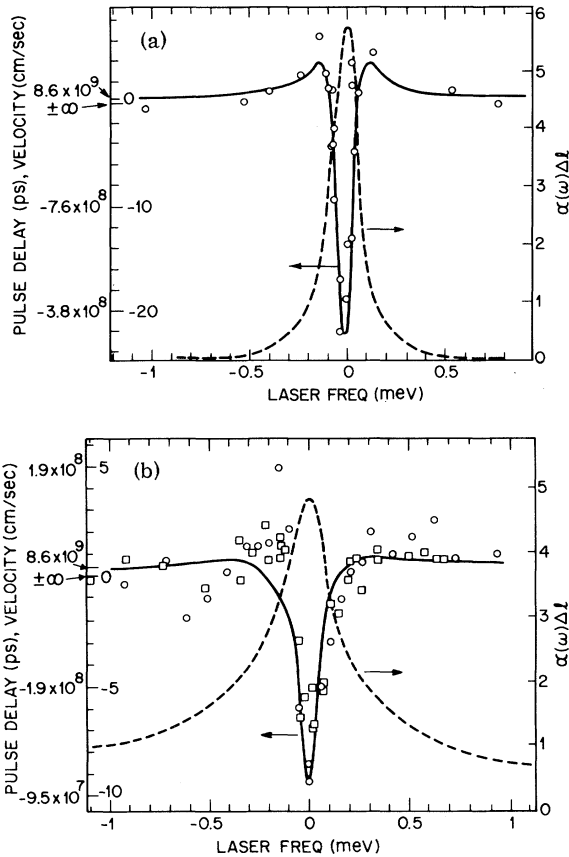

FIG. 3. The pulse delay as a function of laser frequency for (a) [N] = 1.5× 10^{17} cm^{-3}, Δl = 76 μm, and (b) [N] = 3.8× 10^{18} cm^{-3}, Δl = 9.5 μm. The pulse delay is plotted on a linear scale in picoseconds and also in units of velocity = Δl /delay. Zero delay (infinite velocity) corresponds to pulse whose peak emerges from the sample at the same instant the peak enters the sample. The solid lines are the delays expected if the pulse propagates with the group velocity. In (b), the solid line is a slightly smoothed curve of the computed values of v_g. The scatter in the data and in v_g and the disagreement between v_p and v_g at -0.2 meV may be due to small "etalon effects" present in this sample. Open square data and open circle data correspond to $\Delta \tau_L$ = 22 and 48 ps. Dashed curves are the measured absorption curves.

Figs. 3(a) and 3(b). The center of the pulse is found by taking an average of the midpoints at four predetermined pulse heights of a smoothed version of the pulses. The absolute pulse delay or advance values are determined by fixing the off-resonance pulse velocity to be $c/n_0 = 8.57$ ×10^9 cm/sec. Although the delay as a function of laser frequency is a smooth, well behaved function, the pulse velocity goes through some rather counter-intuitive singularities.

The solid lines in Figs. 3(a) and 3(b) are the pulse advances or delays that would occur if the pulse propagated with the group velocity. The group velocity is obtained by measuring the absorption coefficient $\alpha(\omega)$ with a transmission measurement of $I_t = I_0 e^{-\alpha(\omega)\Delta l}$. The measurement of $\alpha(\omega)$ extends over ~10 FWHM's of the absorption line. A least-squares spline fit (of polynomials piecewise continuous to the third derivative) of $\alpha(\omega)$ is made and the Kramers-Kronig relations are numerically applied to obtain the real part of the index of refraction, $n(\omega)$. Once $n(\omega)$ is known, $k_r = \omega n(\omega)/c$ and $\Delta t = \Delta l /v_g = \Delta l dk_r /d\omega$ can easily be obtained. Note that no adjustable parameters or models of the dielectric function $\epsilon(k, \omega)$ have been used to determine the group velocity. The differences in the group-velocity curves between Figs. 3(a) and 3(b) are the result of measured differences in $\alpha(\omega)$.

If one models the absorption line with some $\epsilon(k, \omega)$, e.g.,

$$\epsilon(k, \omega) = \epsilon_0 + \omega_p{}^2/(\omega_0{}^2 - \omega^2 - i\omega\Gamma), \qquad (4)$$

the coupling parameter $\omega_p{}^2$ and the damping constant Γ must produce the measured width of the absorption line. We have derived v_g from a best fit of the absorption curve to $\alpha(\omega) = (2\omega/c) \mathrm{Im}\sqrt{\epsilon}$, where ϵ is given by Eq. (4), and were able to obtain a rough fit to the data. Also, using Eq. (4) as a starting point, Loudon[3] has derived an expression for v_E:

$$\frac{1}{v_E} = \frac{1}{(c/n_0)}\left(1 + \frac{2\omega\kappa}{\eta\Gamma}\right), \qquad (5)$$

where η and κ are the real and imaginary parts to the index of refraction. When $\omega = \omega_0$, our fitting parameters give $v_E([N] = 10^{19}$ cm^{-3}) = +1.5 ×10^8 cm/sec, so that if the pulse propagated with the energy velocity, a pulse *delay* of 6.7 ps, rather than a pulse *advance*, would have been seen.

Finally, we discuss the relation of this work to polariton time-of-flight experiments.[9,12] In the case reported here, an effective $\omega_p{}^2$ and Γ give a maximum change in $\epsilon(\kappa, \omega)$ of roughly $0.04\epsilon_0$.

Thus, we are in the limit where the light is weakly coupled to the oscillators; i.e., when $\omega_p{}^2/\omega\Gamma \ll \epsilon_0$. In some exciton systems, a sufficiently pure, strain-free sample can produce couplings such that $\omega_p{}^2/\omega\Gamma \gtrsim \epsilon_0$. In these "polariton" cases (setting aside complications due to spatial dispersion), a large contribution from the rapidly varying part of $n(\omega)$ might lead one to believe that the Taylor series approximation of $\omega n(\omega)$ will need more than the the first three terms, so that the analysis of Garrett and McCumber will break down. However, one should not lightly dismiss their analysis as irrelevant to the polariton case. (i) Large changes in $n(\omega)$ can be compensated in the laboratory by decreasing $\Delta\nu_L$ and the sample thickness so that the Taylor series expansion can still be valid. We are now in the process of determining what the limits to a Garrett-McCumber type of analysis are in the polariton case. (ii) The results of these experiments clearly demonstrate that v_E does not describe the pulse propagation since the weak-coupling limit is merely a special case of the strong-coupling case. (iii) We have demonstrated that $\Delta\nu_L \ll \Delta\nu_{abs}$ is necessary for the pulse to propagate with the group velocity. We see deviations even when $\Delta\nu_L \simeq \frac{1}{2}\Delta\nu_{abs}$. Thus, in time-of-flight experiments where $\Delta\nu_L \simeq \Delta\nu_{abs}$, the results near the absorption peak will not have the simple interpretation given here, and there may be significant pulse shaping.[9,12] (iv) When $\omega_p{}^2/\omega\Gamma \gtrsim \epsilon_0$, saturation effects are easily seen. If the intensity of the laser is not carefully limited, self-induced transparency behavior or incoherent hole-burning effects may be seen.[13] Both of these effects can crudely mimic an energy-velocity behavior, and certainly obscure the linear pulse propagation behavior.[14]

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