Linear pulse propagation in an absorbing medium: Effect of film thickness

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(Received 30 August 1993)

We compute the peak velocity of a broad Gaussian pulse propagating in an absorbing thin film modeled with a one-oscillator dielectric function. The exact numerical calculation is done for a GaP:N film in the frequency region of the bound A exciton. For absorption lengths greater than the film thickness, the pulse velocity essentially coincides with the group velocity, and with the experimental results of S. Chu and S. Wong [Phys. Rev. Lett. **48**, 738 (1982)]. However, outside the region of strong absorption the pulse velocity depends sensitively upon the thickness of the slab. Our results give a better account of the Chu and Wong experiment than the calculation based on the group velocity. For film thickness with smaller than one absorption length the pulse velocity disagrees with the group velocity even in the strong absorption region, as a consequence of Fabry-Pérot reflections of the pulse inside the film.

Since the pioneering work of Sommerfeld and Brillouin[1] in light pulse propagation, a great deal of attention has been devoted to the subject of group velocity in the anomalous dispersion region of absorbing media. This is because the group velocity v_g can become infinite or even negative, thus challenging our conception of causality. In this regard, for light pulses with a sharp beginning in space, it has been shown by Sommerfeld and Brillouin[1] that (even in a material medium) the fastest part of the pulse propagates with the speed of light in vacuum, c. On the other hand, for Gaussian light pulses whose spectral width is much smaller than the width of the absorption line, it has been shown by Garrett and McCumber[2] that their peak velocity is equal to the group velocity, and that the pulse remains substantially Gaussian and unchanged in width (however, not in height) even if v_g is greater than the velocity of light, or negative. This fact was later interpreted by Crisp[3] as due to an asymmetric absorption of energy from the light pulse.

The first experimental verification of the assertions of Garrett and McCumber[2] for an absorbing solid was performed by Chu and Wong,[4] referred to hereafter as CW, in a GaP:N sample with the laser tuned to the bound A-exciton line. They measured the time delay of the peak of the light pulse using a cross-correlation technique. Near to the center ω_0 of the absorption line, the pulse velocity follows the curve $v_g(\omega)$. However, at the wings there are perceptible deviations from $v_g(\omega)$. As argued by Halevi and Fuchs,[5] this is due to interference arising from the internal reflections of the pulse at the faces of the slab. An account of this effect *for nonabsorbing dielectrics* has been reported by Halevi[6] showing an oscillatory behavior of the peak velocity with slab thickness.

For pulse propagation in an absorbing slab, Halevi and Valenzuela,[7] referred to hereafter as HV, have found a damped oscillatory behavior; in the limit of very large thickness, the peak velocity becomes equal to the group velocity.

A key parameter for an absorbing medium is the spectral width of the light pulse. This should be chosen very small compared to the width Γ of the absorption line, otherwise the spectrum of the pulse can be severely affected as it advances in the medium. This is because the frequency components with $|\omega-\omega_0|\lesssim\Gamma$ are much more heavily damped than the other components, also pointed out by Tanaka, Fujiwara, and Ikegami[8] for pulse propagation in an infinite medium. It was suggested by Katz and Alfano[9] that pulse compression or reshaping was occurring in Chu and Wong's experiment. This was explained by Chu and Wong[10] as due to the uneven damping of the pulse frequency components.

In the following we will present an exact numerical calculation for a Gaussian pulse propagating through a GaP:N film with parameters appropriate to CW's experiment. We obtain the theoretical pulse velocity by looking at the time that it takes for the peak of the pulse to traverse a given film thickness.

The experimental absorption line is reproduced by using a one-oscillator model for the dielectric function,

$$\varepsilon(\omega)=\varepsilon_0+\frac{\omega_p^2}{\omega_0^2-\omega^2-i\omega\Gamma} . \tag{1}$$

We determine the coupling parameter $\hbar\omega_p=2.89$ meV and the damping constant $\Gamma=0.16$ meV from this comparison. The vacuum wavelength is $\lambda_0=2\pi c/\omega_0=534$ nm, corresponding to the bound A-exciton line. The off-resonance pulse velocity is taken as $c/\sqrt{\varepsilon_0}=8.57\times10^9$ cm/sec as given by CW.

For an incident Gaussian light pulse centered at the "laser frequency" ω_l and having a width $\Delta\omega$ the corresponding transmitted electric field is given by

$$E_T(d^+,t)=\int_{-\infty}^{+\infty}d\omega\exp[-(\omega-\omega_l)^2/(\Delta\omega)^2]\times E_T(\omega)\exp[i\omega(d/c-t)] , \tag{2}$$

where E_T is the spectral amplitude of the transmitted electric field and d is the thickness of the film.

We assume that $\Delta\omega\ll\Gamma$. Our results are unchanged for different values considered for $\Delta\omega$, as long as this condition is fulfilled.

We studied the transit time of a pulse for a nitrogen

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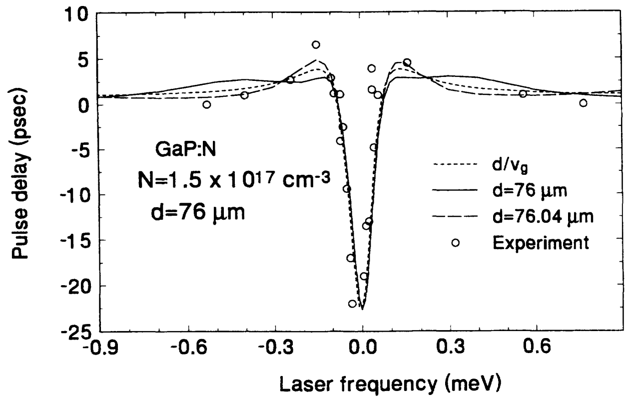

FIG. 1. Pulse delay as a function of frequency ($\omega-\omega_0$) for a GaP:N thin film of thickness $d=76$ μm. Open circles correspond to CW's data (Ref. 4). The short-dashed line is $d/v_g(\omega)$ and was obtained directly from the group velocity. The solid line corresponds to our exact calculation, and the long-dashed line is the same as the solid line but for $d=76.04$ μm. Notice that all three theoretical curves give good agreement near the center of the absorption line. Away from the central region, the line based on the group velocity yields poor agreement, while the exact calculation based on the thickness 76.04 μm (rather than the nominal 76 μm) gives best fit.

impurity density equal to 1.5×10^{17} cm^{-3} [Fig. 3(a) of CW's], with a thickness $d=76$ μm for which the absorption line is very well reproduced with the dielectric constant model given by Eq. (1). In Fig. 1, we compare CW's experimental results (open circles) with $d/v_g(\omega)$ (short-dashed line) and with our results for two slightly different thicknesses (solid line for $d=76$ μm, long-dashed line for 76.04 μm). The vertical axis is the pulse delay in psec while the horizontal axis is the frequency in meV measured from ω_0.

Before we analyze our results, we note the main features of CW's experimental data in comparison with the calculation based on the group velocity. Inside the region of strong absorption the experimental data follow closely the curve obtained from the group velocity. In the wings, on the other hand, there are notable discrepancies, as pointed out by Halevi and Fuchs.[5] In general, sufficiently far from the absorption region the experimental data disagree with the curve obtained from the group velocity.

Our curve for $d=76$ μm (solid line), follows the group-velocity curve in the absorption region; however, outside this region there are visible differences. The reason for this behavior is explained as follows: for strong absorption only one light pulse reaches the other side of the slab with an appreciable amplitude and this pulse propagates with the group velocity. For smaller dissipation, however, the pulse suffers multiple reflections at the slab faces, producing interference, and the outcoming pulse is the result of adding up several pulses, as explained by HV. As this effect is periodic in the slab thickness, there are to be expected different results for different thicknesses outside the region of strong absorption.

This oscillatory effect of the peak velocity for GaP:N is such that the period of oscillation is less than 0.1 μm for a laser frequency in the wings of the absorption line as will be shown later (indeed, it is approximately constant in the frequency region of the bound A exciton). The disagreement between CW's results and ours for $d=76$ μm can be diminished if we choose the slightly different thickness of $d=76.04$ μm as can be seen from the long-dashed line in Fig. 1. The very small change in thickness ($\sim0.01\%$) needed to improve the fit by 40%, indicates that, for pulse propagation in thin films at frequencies where absorption is relatively small, it is necessary to take into account the detailed behavior of the electric field in the medium. In other words, the thin film cannot be replaced by a semi-infinite medium.

Now we study in more detail the oscillatory effect of the peak velocity. According to Ref. 6, the peak velocity of the pulse oscillates with a period equal to $\lambda_2/2$, where λ_2 is the wavelength of the light in the slab. For the given parameters, at $\omega=\omega_0$, $\mathrm{Re}n_2\cong3.5$, then $\lambda_2\cong0.15$ μm. Since n_2 is approximately constant at about ω_0, then the period of oscillation in the vicinity of the absorption peak is ~0.075 μm. Now, at $\omega=\omega_0$, $\mathrm{Im}k=0.038$ μm^{-1} and for $d=76$ μm we have $d(\mathrm{Im}k)=2.9>1$, which explains why multiple reflections do not play a role for this frequency. On the other hand, if we choose $\omega_l=\omega_0-0.15$ meV, then $\mathrm{Im}k=8.37\times10^{-3}$ μm^{-1}, and $d(\mathrm{Im}k)=0.64<1$, thus multiple reflections of the pulse appreciably change the pulse velocity from the value of v_g.

The previous discussion is vividly illustrated in Fig. 2 for a pulse centered at $\omega_l=\omega_0-0.15$ meV. The x axis shows the slab thickness in μm; the solid line and the left y axis correspond to the peak velocity, and the dashed line and the right y axis correspond to the time delay. A maximum (minimum) of the pulse velocity (pulse delay) is observed for $d=76$ μm, while for 76 $\mu\mathrm{m}+\frac{1}{4}\lambda$ we have a minimum (maximum) of the pulse velocity delay. The total change of the pulse velocity in this range of thickness is ~12 μm/psec, which represents $\sim60\%$ of the group velocity for the chosen frequency. A similar percentage

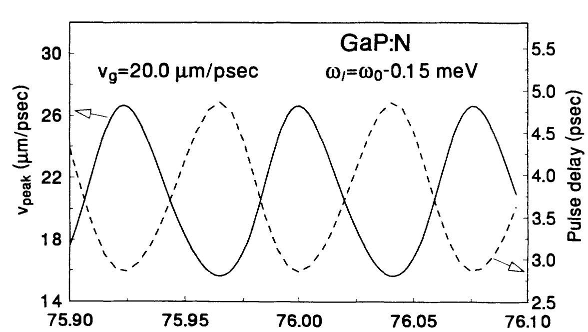

FIG. 2. Peak velocity and pulse delay as a function of thickness d of the film for a Gaussian light pulse centered at $\omega_l=\omega_0-0.15$ meV. The parameters of the GaP:N film are given in the text. Evidently the peak velocity and the time delay are very sensitive functions of the film thickness. Only for selected values of d does v_{peak} coincide with $v_g=200$ μm.

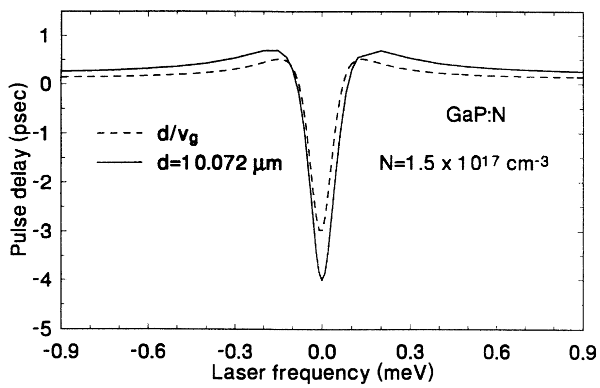

FIG. 3. Same as Fig. 1 but for a much smaller thickness, $d=10.07$ μm. The dashed line has been obtained from v_g. The solid line is gotten from our exact calculation. In this case the optical path is too short to damp out the wave, and the Fabry-Pérot oscillations in the film are important enough to cause disagreement between the exact calculation and that based on the group velocity—even at the very center of the absorption line.

is obtained for the pulse delay. Then for this central frequency, the pulse velocity depends strongly on thickness. For comparison, we performed the same calculation for $\omega_l=\omega_0$, obtaining a difference between the maximum and the minimum of the peak velocity of $\sim 0.5\%$ of v_g.

In order to improve the fit in the wings of the absorption line, we computed the peak velocity for a slightly different thickness ($d=76.04$ μm), where the pulse delay is a maximum for $\omega_l-\omega_0=-0.15$ meV. The result is shown as the long-dashed line in Fig. 1. This curve gives an overall better account of the experimental data.

The fact that near the center of the absorption line both results (long- and short-dashed lines) coincide has been foreseen by HV for thickness such that $\exp\{-2\,\mathrm{Im}[k(\omega)]d\} \ll 1$. In these cases, the peak velocity is very nearly v_g.

For smaller thicknesses such that the previous inequality does not hold, it is expected that the pulse velocity departs from v_g even for $|\omega_l-\omega_0| \leq \Gamma$. A qualitative view of this behavior was shown in Fig. 2 of the HV paper, and is illustrated in the next figure for a 10-μm-thick GaP:N slab whose parameters are the same as those of Fig. 1. The departure of the peak velocity from v_g over the range of frequencies considered is clearly seen in Fig. 3. In particular, for $\omega_l=\omega_0$, the pulse delay is larger than d/v_g by as much as $\sim 30\%$. From Figs. 1 and 3, and helped with HV's analysis, we can understand the behavior of the peak velocity in regions of absorption. CW's experiment corresponds to the case of large thickness such that multiple reflections are unimportant, and thus, $y_{\mathrm{peak}} \approx v_g$ near ω_0 (but not far from ω_0).

In conclusion, we have shown the importance of including the effect of film thickness for broad pulse propagation in an absorbing thin film. Under certain conditions, the peak velocity coincides with the group velocity in the strong absorption region. Outside of this region, the peak velocity is in general different than the group velocity and is given by the expressions computed by HV.

P.H. occupied a Cátedra Patrimonial while in Hermosillo, granted by CONACYT.

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