Pulse Propagation in an Absorbing Film

Recently Chu and Wong[1] (CW) measured the transit time of pulses of light traversing films of GaP:N in the vicinity of an exciton absorption line. They claimed that the pulses suffered little change in shape and width and propagated with the group velocity v_g even when $v_g > c$, $v_g = \pm\infty$, or $v_g < 0$. Such behavior was predicted by Garrett and McCumber.[2]

Figure 3 of CW shows that this claim is indeed true near the center of the absorption line. On the other hand, the agreement between the experimental points and the curve based on the calculated v_g is rather poor off the center of the absorption line. In this Comment we suggest an explanation for this descrepancy. The point is that Ref. 2 deals with an unbounded medium while in CW the absorbing medium is limited to epilayers of thickness $D \leqslant 76\ \mu\text{m}$. Then what is the condition for simulating unbounded propagation in a film of finite thickness?

The transmitted electric field may be written as the sum of partial waves transmitted after $n = 0, 1, 2, \ldots$ double internal reflections; for a pulse with input profile $S(\omega)$ this is

$$E(D,t) = \sum_{n=0}^{\infty} \int_{-\infty}^{\infty} d\omega \, [t(\omega) r^{2n}(\omega) t'(\omega)] \times S(\omega) \exp i[k(\omega) D (1+2n) - \omega t],$$

where $t(\omega)$ and $t'(\omega)$ are the transmission coefficients and $r(\omega)$ is the reflection coefficient for the vacuum-medium interface.[3] Assuming that $S(\omega)$ is centered at the laser frequency ω_L and that its width is much less than that of the absorption line, we expand

$$k(\omega) \simeq k(\omega_L) + (dk/d\omega_L)(\omega - \omega_L) \simeq k_r + \tfrac{1}{2} i \alpha + (\omega - \omega_L)/v_g,$$

where $k_r = k_r(\omega_L)$ and $\frac{1}{2}\alpha = \frac{1}{2}\alpha(\omega_L)$ are the real and imaginary parts of the wave vector $k(\omega_L)$, and $v_g = (dk_r/d\omega_L)^{-1}$. An additional crucial approximation needed is $\alpha D \gtrsim 2$. Then successive terms in the sum over n have the ratio $|r(\omega_L)|^2 \exp(-\alpha D) \ll 1$ and we can neglect the internal reflections in the film (the terms $n \geqslant 1$). Writing $T = |t(\omega_L) t'(\omega_L)|$ and

$$E_0(z,t) = \int d\omega \, S(\omega) \exp[i\omega(z/c - t)]$$

for the incident field, we get

$$|E(D,t)| \simeq T \exp(-\tfrac{1}{2}\alpha D) |E_0(0, t - D/v_g)|.$$

The amplitude of the field is then diminished by the transmission factor T, in addition to the usual exponential damping factor, but the transit time is still given by the group velocity.

If we go back to Fig. 3(a) of CW we note that a good experimental-theoretical agreement is obtained precisely for ω_L such that $\alpha D \gtrsim 2$, while the agreement is poor for $\alpha D < 2$. Allowance for the $n = 1$ term shows that the transit time t_0 of the peak is altered by

$$t_0 - D/v_g \simeq 2|r|^2 \exp(-\alpha D) \cos(2k_r D) D/v_g.$$

The sign of this difference depends on the phase $2k_r D$ and may cause an interesting variation in behavior from one sample to another.[3] As for the order of magnitude, $|t_0 - D/v_g| \sim 1$ psec for $\alpha D \sim 1$, in agreement with the deviations, from the theoretical curve, of the two uppermost circle data[4] in Fig. 3(a) of CW. The "counterintuitive singularities"[1] in the region $\alpha D \ll 1$ (not covered by our approximation) may have a similar origin.

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P. Halevi
Universidad Autonoma de Puebla
Puebla 72570, Mexico

R. Fuchs
Department of Physics and Ames
Laboratory–U.S. Department of Energy
Iowa State University
Ames, Iowa 50011

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[1] S. Chu and S. Wong, Phys. Rev. Lett. **48**, 738 (1982).

[2] C. G. B. Garrett and D. E. McCumber, Phys. Rev. A **1**, 305 (1970).

[3] In the actual experimental configuration the waves traversed a thick, undoped substrate before leaving the plate. Therefore, they suffered little reflection at the interface between the epilayer and the substrate. For the same reason the waves reflected back at the substrate-air interface were transmitted back into the epilayer *with a small change of amplitude.* Our derivation based on the vacuum-medium-vacuum configuration is still valid approximately if we allow for a phase change corresponding to an increased (epilayer + substrate) D. We thank S. Chu and D. W. Lynch for discussions of this point.

[4] We note that the greatest deviations occur at ω_L such that v_g has a minimum. Then it may be necessary to allow for a term proportional to $(\omega - \omega_L)^3$ in the expansion of $k(\omega)$, as studied by D. L. Johnson, Phys. Rev. Lett. **41**, 417 (1978).