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# Transient optical reflectivity from bounded nonlocal media

Govind P. Agrawal, Joseph L. Birman, Deva N. Pattanayak,* and Ashok Puri
*Physics Department, City College of City University of New York, New York, New York 10031*
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This paper considers the problem of reflection of a finite-duration optical pulse incident normally on a semi-infinite nonlocal medium. The light frequency is assumed to lie in the vicinity of an exciton-polariton resonance. The reflected pulse is found to have transients associated with its leading and trailing edges. It is shown that spatial dispersion enhances reflected transients when the laser frequency is at resonance with the exciton polariton. For the case of CdS and GaAs semiconductors, the transient intensities are about 10% of the incident intensity at a time 0.1 psec after the trailing edge of the reflected pulse and remain about 1% even after several picoseconds. We have obtained explicit expressions for the transient part of the reflected field and evaluated them numerically under certain simplifying assumptions; analytical results are presented in some limiting cases of interest. The theory predicts a crossover from exponential to inverse power-law decay rate of transient reflectivity; this occurs at a characteristic time of the order of 1 psec for CdS and GaAs crystals.

## I. INTRODUCTION

Electromagnetic wave propagation in a bounded nonlocal dielectric has attracted considerable attention in recent years.[1–12] The interest in nonlocal media arises from the fact that in some semiconductors, such as CdS and GaAs, the center-of-mass motion of excitons renders the dielectric response function $\epsilon(\vec{k},\omega)$ wave-vector dependent (spatial dispersion) near an exciton resonance. Various optical processes, such as reflection and transmission from a nonlocal interface and Raman and Brillouin scattering may thus provide valuable information about exciton parameters such as its mass and decay rate.

Transmission in a nonlocal semi-infinite medium has been well studied[1–8] in the steady state as well as transient regimes. A monochromatic plane-polarized wave, in general, excites two transverse waves and a longitudinal wave in the nonlocal medium.[4–8] For electromagnetic pulses of finite durations the transient effects give rise to precursors. Together with the well-known Sommerfeld and Brillouin precursors,[13,14] which are the only ones present in a local medium, spatial dispersion gives rise to a third precursor, namely the exciton precursor.[15] Johnson[16] has discussed transient oscillations in transmittivity of a plate.

It is important to note that the transient effects arise only when the light pulse has well-defined boundaries such that the optical intensity becomes strictly zero at the leading and the trailing pulse edges. The often used Gaussian pulse will not give rise to transients because it has no true beginning or end.

Reflection from a nonlocal interface has only been studied in the steady-state regime.[6–8] Transient reflectivity from a local frequency-dispersive dielectric was considered by Elert[17] more than 50 years ago. Generalization to nonlocal media was recently reported.[18] The purpose of this paper is to provide a detailed analysis of the effects of spatial dispersion on transient reflectivity. When an electromagnetic pulse of finite duration $T$ is incident on a nonlocal interface the reflected field consists of the steady-state signal (of duration $T$) and transients arising from both the leading and the trailing pulse edges. Experimentally it may be more convenient to look for trailing-edge transient reflectivity since steady-state reflectivity will then not interfere with measurements. For sufficient long pulses ($T >$ few psec), transients from the two edges will be essentially decoupled and can be considered independently. In this paper we consider the case of a square pulse and obtain expressions for steady-state and transient reflectivities.

Our results show that the transient reflectivity consists of a "local" part and a "nonlocal" part.

 

The former, although the only one present in a local medium, is at least an order of magnitude smaller than the latter. The measured transient reflectivity will thus almost completely arise from spatial dispersion: to the leading order it varies as $M^{-1/2}$ with the exciton mass $M$. Near an exciton resonance, transient reflectivity is resonantly enhanced. Its maximum magnitude in the beginning (just after the reflected laser pulse is cutoff) is about 10% of the incident intensity and decays exponentially. However, the time decay crosses over to an inverse power decay at about 1 psec in semiconductors of interest such as CdS and GaAs. The slow inverse power decay makes the transient reflected intensity persist for several picoseconds with magnitude 1% of the incident intensity. These effects should be measureable.

The plan of the paper is as follows. In Sec. II the model susceptibility for the semi-infinite nonlocal medium is introduced and an integral representation for the reflected field is obtained using the Fourier analysis in the time domain. Using the countour-integration method, the formal expressions for the steady-state and transient parts of reflectivity are obtained in Sec. III. Sections IV and V deal with the "nonlocal" and "local" parts of transient reflectivity, respectively. Detailed numerical results are presented for parameters appropriate to CdS and GaAs crystals. Whenever possible, the analytical expressions are obtained in certain limiting cases. The results are discussed in Sec. VI where various assumptions and approximations are also summarized. Necessary mathematical details are presented in the Appendix.

## II. INTEGRAL REPRESENTATION FOR THE REFLECTED FIELD

We consider a semi-infinite nonlocal medium occupying the half-space $z>L$, with $z<L$ being vacuum (see Fig. 1). A square pulse of the light frequency $\omega_0$ is incident normally at the interface $z=L$. Near an exciton resonance the coupling of an exciton state to a photon produces a quasiparticle exciton polariton. The origin of spatial dispersion or nonlocality is related to the center-of-mass motion associated with the exciton polariton. One particular model, a natural generalization of the classical Lorentz oscillator model, takes[1–3]

$$\epsilon(\vec{k},\omega)=\epsilon_0+4\pi\chi(\vec{k},\omega)$$
$$=\epsilon_0+\frac{4\pi\alpha_0\omega_t^2}{\omega_t^2-\omega^2-i\omega\Gamma+(\hbar\omega_t/M)k^2}\ , \tag{2.1}$$

where $\epsilon_0$ is the background dielectric constant, $\alpha_0$ is the oscillator strength, $M$ is the effective exciton mass, $\Gamma$ is a phenomenological damping rate, $\omega_t$ is the transverse exciton polariton frequency, and $c$ is the velocity of light. The space dependence of the susceptibility

$$\tilde{\chi}(\vec{r}-\vec{r}\,')=\int\chi(\vec{k},\omega)e^{i\vec{k}\cdot(\vec{r}-\vec{r}\,')}d^3k\ . \tag{2.2}$$

reflects the nonlocal nature of the optical dielectric response.

A number of authors[6,7] have used translationally invariant $\tilde{\chi}(\vec{r}-\vec{r}\,')$ for the wave propagation in a semi-infinite nonlocal medium. Zeyher *et al.*[5] pointed out that for a bounded medium reflection of the exciton polariton at the boundary makes the susceptibility translationally noninvariant and proposed that

$$\overline{\chi}(\vec{r},\vec{r}\,')=\tilde{\chi}(\vec{r}-\vec{r}\,')+\tilde{\chi}(\vec{\xi}-\vec{\xi}\,',z+z')\ , \tag{2.3}$$

should be used for the susceptibility. Here $\vec{\xi}$ is the transverse part of $\vec{r}$. Its use in Maxwell's equations determines the reflected and transmitted fields in a unique and self-consistent manner. Other forms of nonlocal susceptibility have been obtained by several authors. For example, Hyzhnyakov, Maradudin, and Mills[9] used an extended basis for a tight-binding exciton model and obtained a general expression for $\overline{\chi}(\vec{r},\vec{r}\,')$ which reduces to (2.3) in a limiting case. Also Ting, Frankel, and Birman[10] derived a nonlocal susceptibility for extended Wannier-type excitons. There have been numerous other derivations of the susceptibility for model bounded-nonlocal media; extensive references are given in the latest edition of

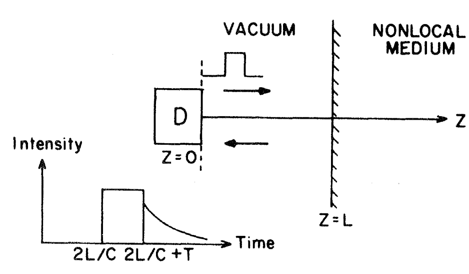


FIG. 1. Schematic illustration of the geometry and the notation used. At time $t=0$ a square pulse of duration $T$ is emitted from the plane $z=0$ where a detector is place to monitor the reflectivity. For $2L/c<t<2L/c+T$ the steady-state signal and for $\tau=t-2L/c-T\geq 0$ the transient signal is detected.

the well-known book by Agranovich and Ginzburg.[3] The general expressions for the reflected field, using (2.3) valid for arbitrary angle of incidence, were obtained by Frankel and Birman.[8] Here we consider the specific case of normal incidence. The amplitude reflection coefficient is given by[8]

$$\rho(\omega)=[1-\bar{n}(\omega)]/[1+\bar{n}(\omega)] , \qquad (2.4)$$

where $\bar{n}$ is the effective refractive index of the nonlocal medium,[19]

$$\bar{n}=(n_1 n_2+\epsilon_0)/(n_1+n_2) , \qquad (2.5)$$

and $n_1$ and $n_2$ are the solutions of the implicit relation

$$n_j=+[\epsilon(k_j,\omega)]^{1/2} , \quad k_j=n_j\omega/c . \qquad (2.6)$$

We assume that the normally incident laser pulse corresponds to a linearly polarized, monochromatic, plane-wave field (transverse variation of all optical fields is ignored). For a square pulse of unit intensity and duration $T$ at the frequency $\omega_0$, the electric field at the plane $z=0$ is given by

$$E_I(0,t)=\sin(\omega_0 t)[\Theta(t)-\Theta(t-T)] , \qquad (2.7)$$

where $\Theta(t)$ is the Heaviside step function. Owing to the linearity of the problem, one may work in the frequency domain and treat each frequency component by dispersion theory. The reflected field is obtained as a superposition of these reflected components and can be written as the frequency integral[12]

$$E_R(0,t)=\lim_{\eta\to 0}\mathrm{Re}\frac{1}{2\pi}\int_{-\infty}^{\infty}\frac{d\omega\,\rho(\omega)}{\omega_0-\omega-i\eta}\{1-\exp[i(\omega-\omega_0)T]\}e^{-i\omega(t-2L/c)} , \qquad (2.8)$$

where the time delay $2L/c$ corresponds to the round-trip time between $z=0$ and $z=L$.

## III. STEADY-STATE AND TRANSIENT REFLECTIVITY

The reflected field, Eq. (2.8), can be formally separated into steady-state and transient parts, after removing the usual harmonic time dependence, $\exp(-i\omega t)$. The steady-state part gives rise to a reflected pulse (of duration $T$) whose leading and trailing edges contain the rapidly time-varying transient contributions.

The frequency integral in Eq. (2.8) is evaluated using the method of contour integration in the complex $\omega$ plane. Using Eq. (2.4) in (2.8), the integrand is found to have the following singularities: (i) a simple pole at $\omega_0-i\eta$, (ii) four branch points $\omega_j$ $(j=1,4)$ in the lower half-plane, and (iii) two branch points $\omega_5$ and $\omega_6$ in the upper half plane. The explicit expressions for the location of these branch points are obtained using Eqs. (2.1) and (2.6) and are given by (see the Appendix)

$$\omega_{1,2}=-i\Gamma/2\pm[\omega_t^2(1+\beta^2/\epsilon_0)-\Gamma^2/4]^{1/2} , \qquad (3.1)$$

$$\omega_{3,4}=(1-\delta^2\epsilon_0)^{-1}\{-i(\Gamma/2+\beta\delta\omega_t)\pm[\omega_t^2-(\beta\delta\omega_t+\Gamma/2)^2]^{1/2}\} , \qquad (3.2)$$

$$\omega_{5,6}=(1-\delta^2\epsilon_0)^{-1}\{i(\beta\delta\omega_t-\Gamma/2)\pm[\omega_t^2-(\beta\delta\omega_t+\Gamma/2)^2]^{1/2}\} , \qquad (3.3)$$

where

$$\beta=(4\pi\alpha_0)^{1/2} , \quad \delta=(\hbar\omega_t/Mc^2)^{1/2} . \qquad (3.4)$$

Note that the dimensionless parameter $\delta$ is a measure of the extent of nonlocality of the medium. For semiconductors of interest such as CdS and GaAs, we estimate $\delta\sim 10^{-3}$ and $\beta\sim 10^{-1}$. It is interesting to observe that if the damping constant $\Gamma>2\beta\delta\omega_t$, all six branch points will lie in the lower half-plane. Presently we shall assume that this is not the case, thereby excluding very heavily damped exciton polaritons.

For $t<2L/c$ we close the contour in the upper half-plane which is chosen so as to exclude the branch-cut line joining $\omega_5$ and $\omega_6$. An evaluation of the integral (2.8) shows that the branch points $\omega_5$ and $\omega_6$ contribute in such a manner that $E_R(0,t<2L/c)=0$ as required by causality.

For $t>2L/c$ we close the contour in the lower half-plane. The four branch points are joined by a pair of branch-cut lines and the resulting contour is shown in Fig. 2. Evaluation of the integral (2.8) along this contour is somewhat lengthy, although straightforward. Algebraic details are presented in

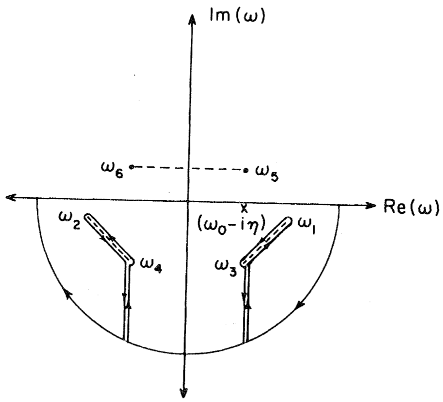


FIG. 2. Schematic illustration of the contour in the complex $\omega$ plane needed to evaluate the reflected field. A pole at $\omega_0 - i\eta$ and six branch-point singularities $\omega_j$, $j=1$ to 6 give rise to the steady-state and transient reflectivity, respectively.

the Appendix. Here we note that the reflected field is found to consist of three parts [Eq. (A15)]:

$$E_R(0,t)=\mathrm{Re}[\tilde{E}_s(t)+\tilde{E}_L(t)+\tilde{E}_{NL}(t)] . \quad (3.5)$$

The simple pole at $\omega_0 - i\eta$ contributes to the steady-state reflected field $\tilde{E}_s(t)$. The four branch points contribute to the transient reflected field which consists of a local part $\tilde{E}_L(t)$ and a nonlocal part $\tilde{E}_{NL}(t)$. Although $\tilde{E}_s$ and $\tilde{E}_L$ both are affected by spatial dispersion, $\tilde{E}_{NL}$ arises solely from it and vanishes as $\delta \to 0$ ($M \to \infty$). Further in the limit of $\delta \to 0$, $\tilde{E}_s$ and $\tilde{E}_L$ reduce to the previously obtained results.[17] We now consider the steady-state and transient parts of the reflected field separately.

### A. Steady-state reflectivity

Using Eq. (A16), $\tilde{E}_s(t)$ is given by

$$\tilde{E}_s(t)=i\rho(\omega_0)e^{-i\omega_0(t-2L/c)} , \quad (3.6)$$

for $2L/c < t < (2L/c+T)$ and zero otherwise. After an initial time delay $2L/c$, a reflected square pulse of duration $T$ arrives at the plane $z=0$. In Fig. 3 we have shown the reflectivity $R_0 = |\rho(\omega_0)|^2$ and the phase $\phi = \arg[\rho(\omega_0)]$ as a function of $\omega_0$ in the vicinity of exciton-polariton resonance for parameters appropriate to a CdS crystal ($\delta = 2.3\times 10^{-3}$). Reflectivity for the case of a local medium ($\delta=0$) is shown by a dashed line for comparison. We note that the main effect of spatial dispersion is to reduce the reflectivity peak height.

An analytic expression for $R_0$, correct to the first order in $\delta$, can be obtained for the case when the laser frequency $\omega_0$ is slightly off resonance, $|\omega_0-\omega_t|/\omega_t >> \delta^2$. With the use of Eq. (A4), the refractive indices $n_1$ and $n_2$ can be approximated by

$$n_1^2 \simeq \epsilon_0 + (\beta\omega_t/q\omega_0) , \quad n_2 \simeq q/\delta , \quad (3.7)$$

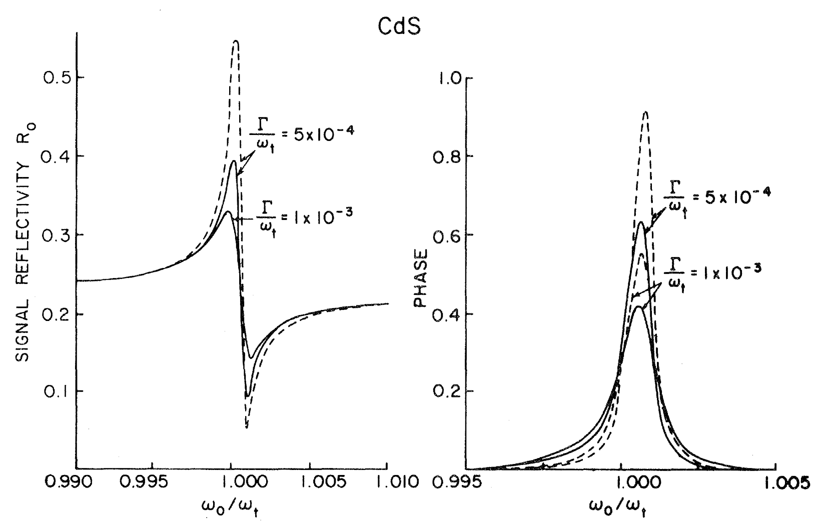


FIG. 3. Resonance enhancement of the steady-state reflectivity $R_0$ which persists for $2L/c \le t \le (2L/c+T)$. The parameters are chosen appropriate to a $CdS$ crystal with $\epsilon_0 = 8$, $\hbar\omega_t = 2.55$ eV, $M = 0.9m_e$ and $\beta^2 = 1.25\times 10^{-2}$ (LT splitting $\simeq 0.2$ meV). For comparison, the dashed line shows the reflectivity when spatial dispersion is neglected $M=\infty$). Phase is shown on the right.

where $q^2=(1-\omega_t^2/\omega_0^2)$ and we took $\Gamma=0$ for simplicity. With the use of Eq. (2.5) the effective refractive index

$$\bar{n}=(n_1+\epsilon_0\delta/q)/(1+n_1\delta/q)\simeq n_1+(\epsilon_0-n_1)\delta/q \; . \tag{3.8}$$

From Eq. (2.4), $R_0=|\rho(\omega_0)|^2$ is found to be given by

$$\frac{R_0}{R_0^{\rm L}}=\left[1+\delta\frac{2\sqrt{\epsilon_0}}{\sqrt{\epsilon_0}+1}\frac{\omega_0^2}{\omega_0^2-\omega_t^2}\right] , \tag{3.9}$$

where $R_0^{\rm L}=(n_1-1)^2/(n_1+1)^2$ is the reflectivity of a local medium. Equation (3.9) clearly shows how spatial dispersion affects reflectivity of a crystal in the vicinity of an excitonic resonance.

### B. Transient reflectivity

Transient reflectivity consists of a "local" part and a "nonlocal" part,

$$\tilde{E}_T(t)=\tilde{E}_{\rm L}(t)+\tilde{E}_{\rm NL}(t) \; ,$$

where from Eq. (A17)

$$\tilde{E}_j(t)=[E_j(t-2L/c)\Theta(t-2L/c) -e^{-i\omega_0 T}E_j(t-2L/c-T)\Theta(t-2L/c-T)] \; , \tag{3.10}$$

for $j=$L and NL and $E_{\rm L}(\tau)$ and $E_{\rm NL}(\tau)$ are given by Eqs. (A13) and (A14). The two terms in Eq. (3.10) correspond to the transients arising from the leading and trailing pulse edges, respectively. If the pulse duration $T$ is longer than the effective time during which transients significantly contribute, leading- and trailing-edge transients will not interfere and can be considered independently. Up to a constant phase factor, transients from both edges are identical in form and magnitude and are governed by $E_j(\tau)$. Experimentally it may be more convenient to look for trailing-edge transients which appear just after the reflected pulse is cut off at $t=2L/c+\Gamma$.

For arbitrary values of the crystal parameters, the evaluation of $E_{\rm L}(\tau)$ and $E_{\rm NL}(\tau)$ [Eqs. (A13) and (A14)] requires extensive numerical computation. Substantial simplification, however, occurs when we note the presence of three small dimensionless parameters ($<10^{-3}$), $\delta=(\hbar\omega_t/Mc^2)^{1/2}$, $\beta^2/2\epsilon_0=(\omega_l-\omega_t)/\omega_t$, and $\Gamma/\omega_t$. Here $\omega_l$ is the longitudinal exciton-polariton frequency and the LT-splitting value $\omega_l-\omega_t$ is $\sim 0.1$ meV. Neglecting products and higher powers of the three small parameters, we obtain somewhat simplified expressions for the transient fields given by Eqs. (A19) and (A20).

Using Eqs. (A3) and (A11) in Eq. (A19), the local part of the transient reflectivity is found to be given by

$$E_{\rm L}(\tau)=e^{-i\omega_t\tau}e^{-\Gamma\tau/2}\frac{p\omega_t}{2\pi}\int_0^1\frac{[\rho(n_1,n_2)-\rho(-n_1,n_2)]}{(\omega_t-\omega_0+pu\,|\,\omega_t\,|-i\Gamma/2)}\exp(-ip\omega_t\tau)du+\cdots \tag{3.11}$$

where the ellipses represents a similar expression obtained by $\omega_t\to-\omega_t$. Here dependence of $\rho$ on the two refractive indices $n_1$ and $n_2$ is explicitly shown, and $n_1$ and $n_2$ are to be evaluated at the frequency $\omega=\omega_t+pu\omega_t-i\Gamma/2$. As a reminder $p=2\pi\alpha_0/\epsilon_0=(\omega_l-\omega_t)/\omega_t$. It is easy to verify that Eq. (3.11) reduces to Elert's result[17] in the limit of infinite exciton mass ($\delta=0$). This follows by noting that when $\delta=0$, $\bar{n}=n_1$ and the term in the square brackets in Eq. (3.11) reduces to $4n_1/(1-n_1^2)$.

The nonlocal part is given by Eq. (A20) which together with (A3) and (A11) becomes

$$E_{\rm NL}(\tau)=e^{-i\omega_t\tau}e^{-\Gamma\tau/2}\frac{i\beta\delta\omega_t}{2\pi}\left[\int_0^1\frac{[\rho(n_1,n_2)-\rho(-n_1,n_2)]}{(\omega_t-\omega_0-i\beta\delta\,|\,\omega_t\,|\,u-i\Gamma/2)}\exp(-\beta\delta\,|\,\omega_t\,|\,\tau u)du + \int_1^\infty\frac{[\rho(n_1,n_2)-\rho(-n_2,n_1)]}{(\omega_t-\omega_0-i\beta\delta\,|\,\omega_t\,|\,u-i\Gamma/2)}\exp(-\beta\delta\,|\,\omega_t\,|\,\tau u)du+\cdots\right] \tag{3.12}$$

where again, the ellipsis represents two similar terms obtained by $\omega_t\to-\omega_t$. Here $n_1$ and $n_2$ are to be evaluated at the frequency

$$\omega=\omega_t-i\beta\delta\omega_t u-i\Gamma/2 \; .$$

Note that $E_{\rm NL}(\tau)$ is proportional to the spatial-dispersion parameter $\delta$ and vanishes for the case of a local medium in the limit of infinite exciton mass ($\delta=0$).

Equations (3.10) together with (3.11) and (3.12) give the total transient reflected field associated with a square pulse of duration $T$. In the following we focus our attention on transients arising from the trailing pulse edge and set $\tau=t-2L/c-T$. For $T>$ few psec, leading-edge transients would die out before the trailing edge of the pulse arrives and therefore

$$\tilde{E}_j(\tau)\simeq -e^{-i\omega_0 T}E_j(\tau), \tag{3.13}$$

where $\tau>0$ and $j=L$ and NL.

Since the transient effects are expected to be important only in the immediate vicinity of exciton-polariton resonance, we shall assume that the laser frequency $\omega_0\simeq\omega_t$. Under near-resonance conditions, the second term in Eq. (3.11) and the last two terms in Eq. (3.12) will not contribute significantly and henceforth will be neglected. It may be noted that these terms arise from the branch points lying in the fourth quadrant of the complex $\omega$ plane (see Fig. 2). In the next two sections we consider the nonlocal and local parts of transient reflectivity separately.

## IV. SPATIAL-DISPERSION-INDUCED TRANSIENT REFLECTIVITY

In this section we simplify the nonlocal part of the transient reflectivity, Eq. (3.12), and discuss pertinent features numerically and, whenever possible, analytically. For this purpose we need to evaluate $n_1$ and $n_2$ at the frequency $\omega=\omega_t-i\beta\delta\omega_t u-\Gamma/2$. Using Eqs. (A4) and (A5), to the leading order in $\delta$ we obtain

$$n_{2,1}=+[a\mp(a^2-\epsilon_0 b)^{1/2}]^{1/2}, \tag{4.1}$$

where

$$a\simeq(\epsilon_0/2-i\beta u/\delta), \quad b\simeq-\beta^2/(\epsilon_0\delta^2). \tag{4.2}$$

Substituting Eq. (4.2) in (4.1) and assuming $(\epsilon_0\delta/2\beta)<<1$, we obtain

$$\begin{aligned} n_1&=i(\beta/\delta)^{1/2}e^{i\theta/2}[1-(\delta\epsilon_0/4\beta)e^{-i\theta}],\\ n_2&=(\beta/\delta)^{1/2}e^{-i\theta/2}[1+(\delta\epsilon_0/4\beta)e^{i\theta}], \end{aligned} \tag{4.3}$$

where

$$\theta=\sin^{-1}u. \tag{4.4}$$

In the following we neglect the second term in Eq. (4.3). This sets an upper limit on the accuracy of our approximations. For parameters appropriate to crystals such as CdS and GaAs, we estimate $\delta\epsilon_0/4\beta\sim10^{-1}-10^{-2}$; our calculations estimate transient reflectivity within an error of 10%. Note that under such conditions

$$|n_1|=|n_2|=(\beta/\delta)^{1/2}=(4\pi\alpha_0 Mc^2/\hbar\omega_t)^{1/4}.$$

Using Eqs. (2.5) and (4.3) the effective refractive index is found to be given by

$$\begin{aligned} \bar{n}(n_1,n_2)&\simeq e^{i\pi/4}(\beta/2\delta)^{1/2}(1-u)^{-1/2},\\ \bar{n}(-n_1,n_2)&\simeq e^{-i\pi/4}(\beta/2\delta)^{1/2}(1+u)^{-1/2},\\ \bar{n}(-n_2,n_1)&=-\bar{n}(-n_1,n_2). \end{aligned} \tag{4.5}$$

Equations (2.4) and (4.5) are used to evaluate the integrand in Eq. (3.12). If we define $a_1=\bar{n}(n_1,n_2)$ and $a_2=\bar{n}(-n_1,n_2)$ we obtain

$$\rho(n_1,n_2)-\rho(-n_1,n_2)=(1-a_1)/(1+a_1)-(1-a_2)/(1+a_2)=2(a_2-a_1)/(1+a_1+a_2+a_1a_2)\simeq2/a_1-2/a_2 \tag{4.6}$$

where we assumed $\beta/\delta>>1$. Using (4.6) in (3.12), and using (3.13), $E_{\rm NL}(\tau)$ is given by

$$\mathrm{Re}[E_{\rm NL}(\tau)]=|R_1(\tau)|\sin(\omega_t\tau-\phi_1), \tag{4.7}$$

where $R_1(\tau)=|R_1(\tau)|\exp[i\phi_1(\tau)]$ is given by

$$R_1(\tau)=(2i\beta\delta^3/\pi^2)^{1/2}e^{-\Gamma\tau/2}\left[\int_0^1 du\frac{\omega_t\exp(-\beta\delta\omega_t\tau u)}{(\omega_t-\omega_0)-i(\beta\delta\omega_t u+\Gamma/2)}(\sqrt{1+u}+i\sqrt{1-u})+\int_1^\infty du\frac{\omega_t\exp(-\beta\delta\omega_t\tau u)}{(\omega_t-\omega_0)-i(\beta\delta\omega_t u+\Gamma/2)}(\sqrt{u+1}-\sqrt{u-1})\right]. \tag{4.8}$$

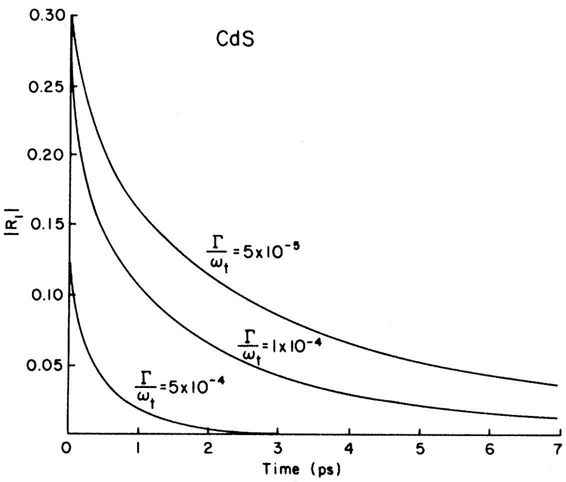


FIG. 4. Time decay of the nonlocal part of the reflectivity $|R_1|$ for several values of the exciton damping rate $\Gamma$ for a CdS crystal. The parameters are $\epsilon_0=8$, $\hbar\omega_t=2.55$ eV, $M=0.9m_e$, and $\beta^2=1.25\times10^{-2}$ (LT splitting = 0.2 meV). The spatial dispersion parameter $\delta=2.35\times10^{-3}$. When $\delta=0$ (infinite exciton mass). $|R_1|=0$. Note that significant reflectivity persists even after several psec.

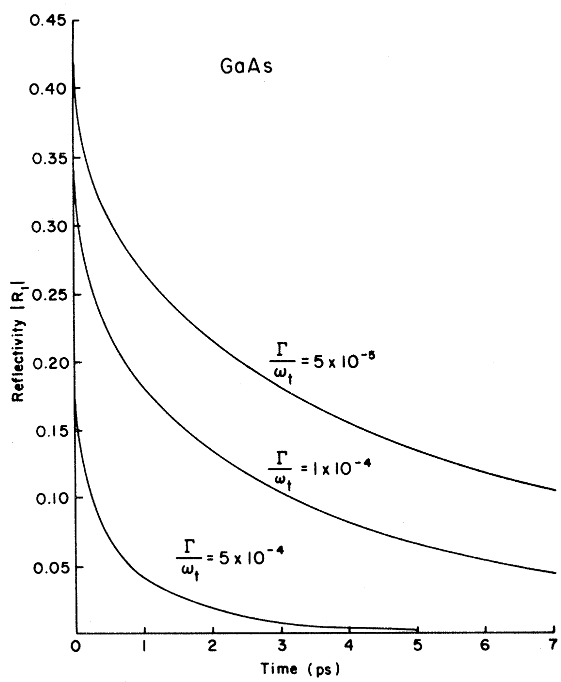


FIG. 5. Same as in Fig. 4 except that the parameters appropriate to a GaAs crystal were used: $\epsilon_0=12.55$, $\hbar\omega_t=1.515$ eV, and $\beta^2=1.3\times10^{-3}$ (LT splitting = 0.08 meV). The spatial dispersion parameter $\delta=2.22\times10^{-3}$. When $\delta=0$, $|R_1|=0$.

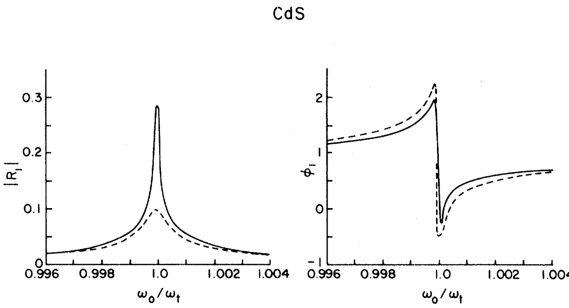


FIG. 6. Resonance enhancement of the nonlocal part of the transient reflectivity $|R_1|$ and its phase $\phi_1$ at fixed time 0.1 psec; full line, $\Gamma=5\times10^{-5}\omega_t$; dashed line, $\Gamma=5\times10^{-4}\omega_t$. The parameters appropriate to CdS are the same as those of Fig. 4.

We evaluated $R_1(\tau)$ numerically for parameters appropriate to CdS and GaAs crystals. The results are displayed in Figs. 4 and 5 where we show the time decay of $|R_1(\tau)|$, for several values of $\Gamma$, for the case of exact resonance $\omega_0=\omega_t$. Several features are noteworthy in the decay of transient reflectivity. At time $\tau=0$, when the trailing edge of the reflected pulse has just passed and steady-state reflectivity has dropped to zero, $|R_1(0)|^2$ is $\sim0.1$ indicating than 10% of the incident intensity is carried out by the transient reflected field. As $\tau$ increases $|R_1|$ begins to decrease exponentially but around $\tau\sim1$ psec a crossover from an exponential to a slow power-law decay takes place. The resulting tail in Figs. 4 and 5 shows that, if $\Gamma$ is sufficiently small, the on-resonance transient reflectivity is about 1% even if after several psec the reflected pulse is cut off. This feature should be observed experimentally.

Resonance enhancement of transient reflectivity is another remarkable feature and is illustrated in

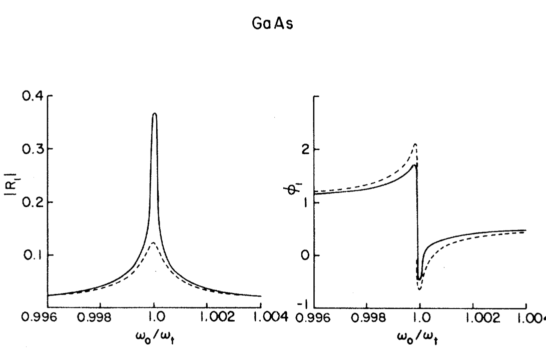


FIG. 7. Resonance enhancement of $|R_1|$ and its phase $\phi_1$ for two values of $\Gamma$ as in Fig. 6. The parame ters appropriate to GaAs are the same as those of Fig. 5.

Figs. 6 and 7 for CdS and GaAs semiconductors. At a fixed time $\tau=0.1$ psec, $|R_1|$ and the phase $\phi_1$ is shown as a function of the laser frequency $\omega_0$ in the vicinity of the exciton-resonance frequency $\omega_t$. Enhancement of $|R_1|^2$ by a factor of 5–10 is observed in a narrow frequency range. It is interesting to compare Fig. 6 or 7 with Fig. 3 and note a reversal in the form of the amplitude and the phase spectra.

Most of the features of the decay of $|R_1(\tau)|$ can be understood qualitatively by evaluating the integrals in Eq. (4.8) in a closed form for several limiting cases. The main contribution to $R_1(\tau)$ arises from the first integral in Eq. (4.8) which can be rewritten as follows

$$R_1(\tau)\simeq\frac{i}{\pi}\left[\frac{2i\delta}{\beta}\right]^{1/2}e^{-\Gamma\tau/2}\times\int_0^1\frac{\sqrt{1+u}+i\sqrt{1-u}}{u+(\Gamma/2+i\Delta\omega)\tau_c}\times\exp(-u\tau/\tau_c)du\ , \tag{4.9}$$

where $\Delta\omega=(\omega_t-\omega_0)$ and we have defined a characteristic time

$$\tau_c=(\beta\delta\omega_t)^{-1}\ . \tag{4.10}$$

It is useful to consider the limits $\tau<<\tau_c$ and $\tau>>\tau_c$ separately. In the former case we expand

$$\exp(-u\tau/\tau_c)\simeq(1-u\tau/\tau_c)$$

and the integration in Eq. (4.9) is straightforward. If we ignore the natural decay rate $\Gamma$, we obtain

$$R_1(\tau)\simeq C_1e^{-\tau/\tau_c}\ ,\quad \tau<<\tau_c \tag{4.11}$$

where $C_1$ is a constant ($\tau$ independent) whose exact form is not of interest here.

In the opposite limit $\tau>>\tau_c$ the major contribution to the integral in Eq. (4.9) comes from the region near $u=0$. To the lowest order we replace $(1+u)^{1/2}$ by 1 in the numerator and obtain

$$R_1(\tau)\simeq-\frac{2}{\pi}\left[\frac{\delta}{\beta}\right]^{1/2}e^{i\Delta\omega\tau}E_1(\Gamma\tau/2+i\Delta\omega\tau)\ ,\quad \tau>>\tau_c \tag{4.12}$$

where $E_1$ is the exponential integral.[20] At exact resonance, $\Delta\omega=0$, and $\Gamma\tau>>1$,

$$R_1(\tau)\simeq\frac{4}{\pi\Gamma}\left[\frac{\delta}{\beta}\right]^{1/2}\frac{e^{-\Gamma\tau/2}}{\tau}\ ,\quad \tau>>\tau_c\ . \tag{4.13}$$

Apart from the natural exponential decay $\exp(-\Gamma\tau/2)$, $R_1(\tau)$ begins to decrease exponentially and crosses over to a slow $\tau^{-1}$ decay around $\tau\sim\tau_c$. This feature is clearly seen in Figs. 4 and 5. Using Eq. (4.10) we estimate $\tau_c\sim1$ psec for CdS and GaAs crystals.

When the laser frequency is slightly off-resonance such that $\Delta\omega\tau_c>>1$, it is possible to obtain a closed form expression for $R_1(\tau)$ valid for all times. This is acheived by neglecting $u$ in the denominator of Eq. (4.9) and we obtain

$$R_1(\tau)=-\frac{1}{\pi}\left[\frac{2i\delta}{\beta}\right]^{1/2}\frac{e^{-\Gamma\tau/2}}{(\Delta\omega-i\Gamma/2)\tau_c}\left[\frac{\tau_c}{\tau}\right]^{3/2}\left\{e^{-\tau/\tau_c}\gamma\left[\frac{3}{2},-\frac{\tau}{\tau_c}\right]+e^{\tau/\tau_c}\left[\gamma\left[\frac{3}{2},\frac{2\tau}{\tau_c}\right]-\gamma\left[\frac{3}{2},\frac{\tau}{\tau_c}\right]\right]\right\}, \tag{4.14}$$

where $\gamma(a,x)=\int_0^x e^{-t}t^{a-1}dt$ is the incomplete gamma function.[20] It may easily be verified that for $\tau>>\tau_c$, $R_1(\tau)\sim\tau^{-1}$.

The nonlocal part of transient reflectivity, $R_1(\tau)$, vanishes in the limit of infinite mass, $\delta=0$. We now consider the local part $E_L(\tau)$ modified by spatial dispersion.

## V. LOCAL PART OF TRANSIENT REFLECTIVITY

The local part of the reflected field, $E_L(\tau)$ is given by Eq. (3.11). The integrand is to be evaluated at the frequency $\omega=\omega_t+pu\omega_t-i\Gamma/2$ and can be simplified following the procedure of Sec. IV. The two refractive indices are found to be given by

$$n_1=i(\beta/\delta p)^{1/2}e^{i\theta/2}\ ,\quad n_2=(\beta/\delta p)^{1/2}e^{-i\theta/2}\ , \tag{5.1}$$

where

$$u=-i(\beta\delta/p)\sin\theta\ ,\quad \beta=(4\pi\alpha_0)^{1/2}\ ,\quad p=2\pi\alpha_0/\epsilon_0=(\omega_l-\omega_t)/\omega_t\ . \tag{5.2}$$

Equations (5.1) and (5.2) are used to evaluate the integrand in Eq. (3.11) and we obtain

$$\mathrm{Re}[E_L(\tau)] = |R_2(\tau)| \sin(\omega_t \tau - \phi_2) , \tag{5.3}$$

where $R_2(\tau) = |R_2(\tau)| \exp[i\phi_2(\tau)]$ is given by

$$R_2(\tau) = \frac{\sqrt{2}}{\pi} p^3 e^{-\Gamma\tau/2} \int_0^1 du \frac{\omega_t \exp(-ip\omega_t \tau u)}{\omega_t - \omega_0 + p\omega_t u - i\Gamma/2} [(u + i\beta\delta/p)^{1/2} + (u - i\beta\delta/p)^{1/2}] . \tag{5.4}$$

Equation (5.4) shows how the local part $R_2(\tau)$ of transient reflectivity is affected by spatial dispersion. We first consider the case of exact resonance, $\omega_0 = \omega_t$. Figures 8 and 9 show the time decay of $|R_2(\tau)|$ and $|\phi_2(\tau)|$ for parameters appropriate to CdS and GaAs crystals, repectively. By a dashed line we have also shown the corresponding curves obtained for a local medium ($\delta = 0$). A comparison of the full and the dashed curves shows that the effect of spatial dispersion is to reduce $|R_2|$ in magnitude and damp out its oscil-

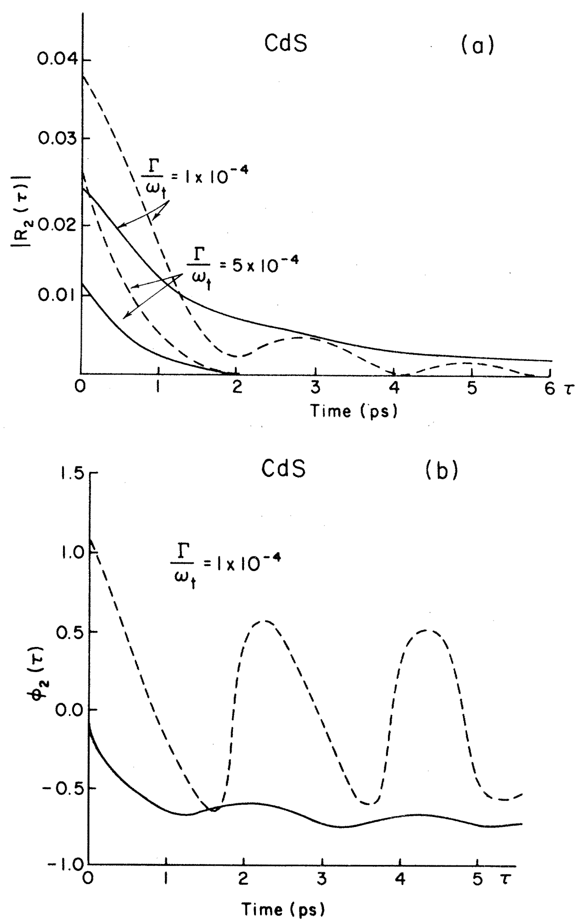


FIG. 8. Time decay of the local part of the transient reflectivity (a) the amplitude $|R_2|$ and (b) the phase $\phi_2$ for several values of the damping rate $\Gamma$ for a CdS crystal. The parameters used are the same as those of Fig. 4. For comparison dashed curves for the case $\delta = 0$ are also shown.

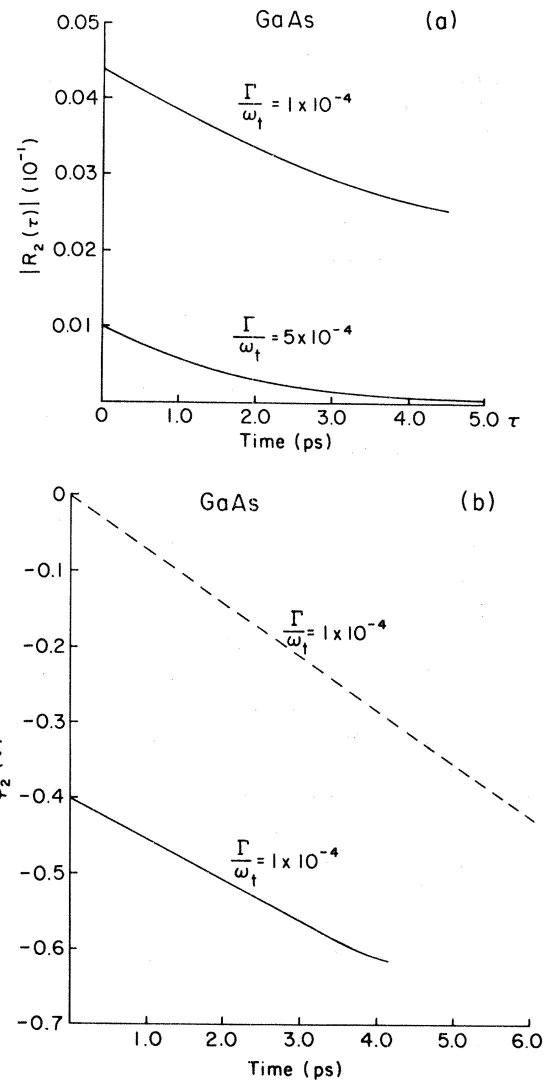


FIG. 9. The same as in Fig. 8 except the parameters appropriate to a GaAs crystal, the same as those of Fig. 5, were used. When $\delta = 0$, $|R_2|$ is too small to be shown on the present scale.

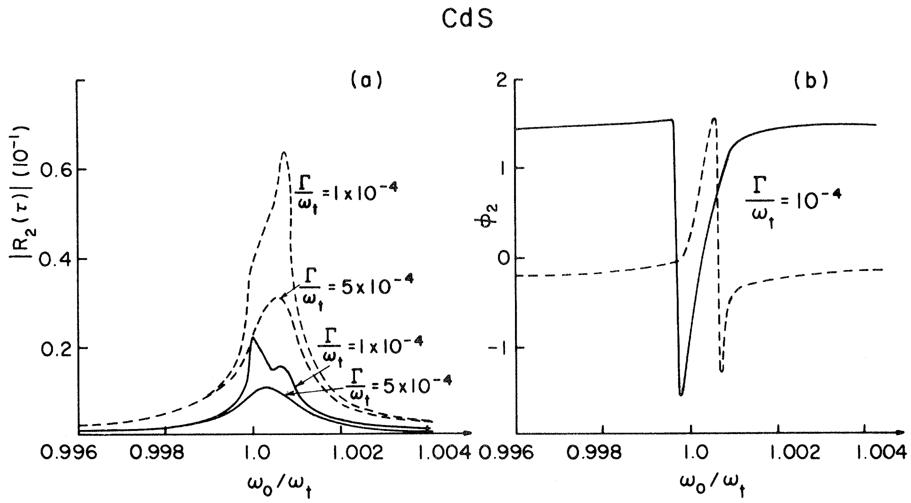


FIG. 10. Resonance enhancement of the local part of the transient reflectivity. (a) The amplitude $|R_2|$ and (b) the phase $\phi_2$ for several values of the damping rate $\Gamma$ for a CdS crystal. The parameters are the same as those of Fig. 4. Dashed line curves correspond to the case $\delta=0$ when spatial dispersion is neglected. Note the double-peak structure in $|R_2|$ for $\Gamma=10^{-4}\omega_e$. The two peaks correspond to the transverse and longitudinal exciton frequencies and are separated by the LT-splitting 2 meV ($\simeq 8\times10^{-4}\omega_t$) When damping rate $\Gamma$ is large, double-peak structure cannot be resolved.

latory variation with time. When we compare Figs. 8 and 9 with Figs. 4 and 5, respectively, we note that $|R_2|$ is smaller than $|R_1|$ by at least an order of magnitude. This observation is very significant: It implies that in crystals such as CdS or GaAs transient reflectivity almost completely arises from spatial dispersion and is much larger than that of a local medium.

Resonance enchancement of $R_2$ is shown in Figs. 10(a) and 11(a) for CdS and GaAs crystals, respectively. At a fixed time $\tau=0.1$ psec $|R_2|$ and the phase $\phi_2$ are, plotted as a function of the laser frequency $\omega_0$ in the vicinity of $\omega_t$ for two values of $\Gamma$. For comparison, the dashed curve shows the corresponding behavior when $\delta=0$. We note that the phase $\phi_2$ is affected by spatial dispersion in a qualitative manner. This is understood when we observe that the $\delta$-dependent term in Eq. (5.4) is purely imaginary.

An interesting feature of Fig. 10(a) is the double-peak spectrum of $|R_2|$ for $\Gamma=10^{-4}\omega_t$. The two peaks occur at $\omega_l$ and $\omega_t$, the longitudinal and transverse exciton-resonance frequencies. Their origin can be traced to the integrand in Eq. (5.4): when $\omega_0=\omega_t$, the region near $u=0$ contributes in a resonant manner; when $\omega_0=\omega_l=(1+p)\omega_t$, the region near $u=1$ contributes in a resonant manner. Spatial dispersion affects the height of two peaks in a different manner as is evident from Fig. 10(a). For a larger value of $\Gamma=5\times10^{-4}\omega_t$, both peaks broaden and cannot be resolved. In fact, this is the reason for not obtaining a double-peak spectrum for GaAs in Fig. 11(a). The LT splitting $\omega_l-\omega_t=0.08$ meV for GaAs, is smaller than that of CdS ($\simeq 0.2$ meV) and even for $\Gamma=10^{-4}\omega_t$, the two peaks are broader in comparison to their separation $\sim 5\times10^{-5}\omega_t$.

It is possible to obtain a closed-form expression for $R_2(\tau)$ in some limiting cases. With the definition of $\Delta\omega=(\omega_t-\omega_0)$, Eq. (5.4) can be rewritten as

$$R_2(\tau)=\frac{\sqrt{2}p^2}{\pi}e^{-\Gamma\tau/2}\int_0^1 du\frac{\exp(-iu\tau/\tau_L)}{u+(\Delta\omega-i\Gamma/2)\tau_L}[(u+i\tau_L/\tau_c)^{1/2}+(u-i\tau_L/\tau_c)^{1/2}]\ , \tag{5.5}$$

where

$$\tau_L^{-1}=p\omega_t=(\omega_l-\omega_t)\ , \tag{5.6}$$

and $\tau_c=(\beta\delta\omega_t)^{-1}$ was defined in Sec. IV.

Equation (5.5) indicates that the relative magnitudes of $\tau_L$ and $\tau_c$ govern the effect of spatial dispersion

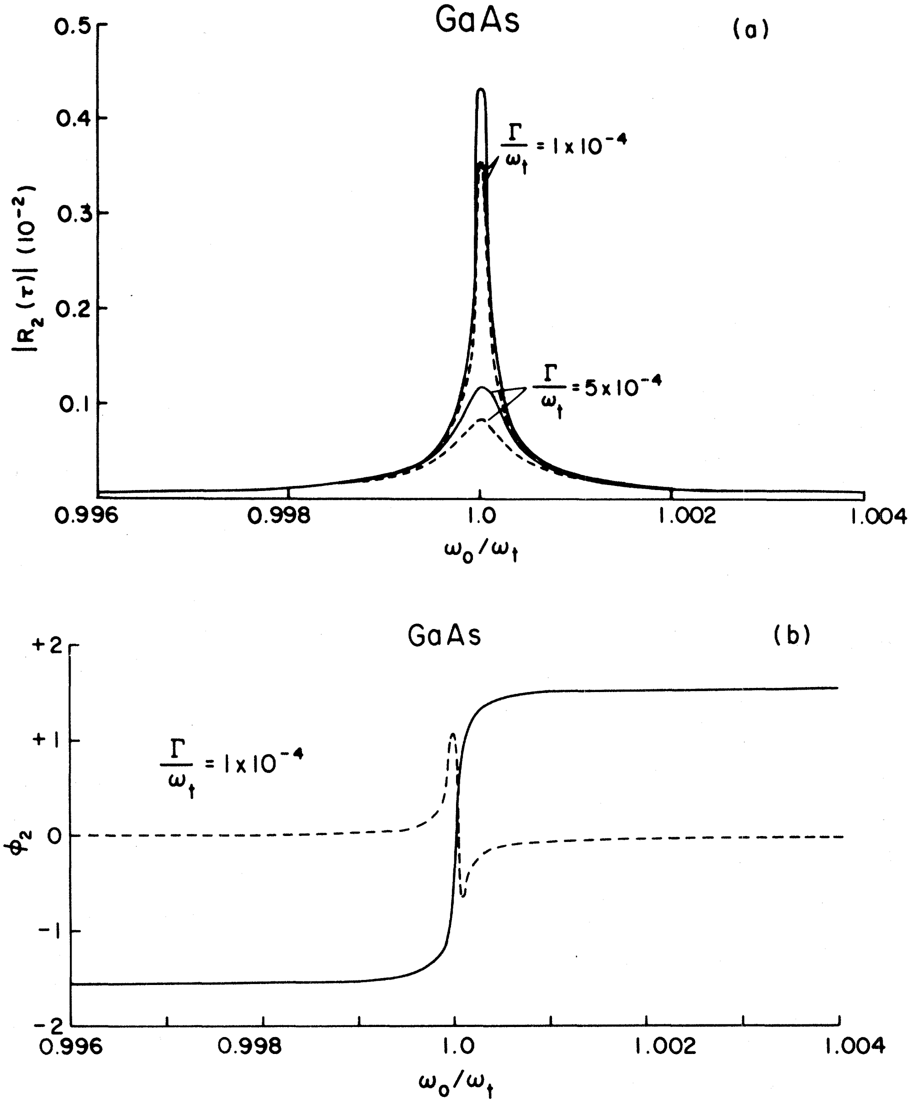


FIG. 11. The same as in Fig. 10 except that GaAs parameters, the same as in Fig. 9 are used. Note that the double-peak structure cannot be resolved for these values of Γ due to very small LT splitting $\simeq$0.08 meV.

on $R_2$. If $\tau_L << \tau_c$, $R_2$ remains essentially unaffected. For the case of CdS crystal shown in Fig. 10, $\tau_L \sim \tau_c$, while for the case of GaAs shown in Fig. 11, $\tau_L \sim 20\tau_c$. It is important to note that the LT splitting sets a time scale for the decay of the local part of transient reflectivity.

We evaluate the integral in Eq. (5.5) for two limiting cases, $\tau_L << \tau_c$ and $\tau_L >> \tau_c$. In the former case, at exact resonance $\Delta\omega = 0$ with $\Gamma << \tau_L^{-1} = \omega_l - \omega_t$, we obtain

$$R_2(\tau) \simeq \frac{\sqrt{2}p^2}{\pi} e^{-\Gamma\tau/2} \left[\frac{i\tau}{\tau_L}\right]^{-1/2} \gamma\left[\frac{1}{2}, \frac{i\tau}{\tau_L}\right], \quad \tau_L << \tau_c \tag{5.7}$$

where $\gamma(a,x)$ is the incomplete gamma function.[20] In the other limit $\tau_L >> \tau_c$, we obtain

$$R_2(\tau) \simeq -\frac{\sqrt{2}p^2}{\pi} \left[\frac{\tau_L}{\tau_c}\right]^{1/2} e^{i\Delta\omega\tau} \{ E_1[(1+\alpha)\tau/\tau_c] - E_1(-i\alpha\tau/\tau_c) \} , \tag{5.8}$$

where $\alpha=(\Delta\omega-i\Gamma/2)\tau_L$ and $E_1(Z)$ is the exponential-integral function.[20] It may be verified from Eqs. (5.7) and (5.8) that for large $\tau$, $|R_2(\tau)|$ crosses over to an inverse power-law decay as is also evident in Figs. 8 and 9.

## VI. DISCUSSION AND CONCLUSIONS

In this paper we have studied the problem of reflection of a finite-duration optical pulse, incident normally on the boundary of a nonlocal medium such as a CdS or GaAs crystal. The light frequency is assumed to lie in the vicinity of an exciton-polariton resonance frequency. Using a square pulse of duration $T$ with infinitely sharp edges, we have shown that the reflected pulse, also of duration $T$, has transients associated with its leading and trailing edges. We have obtained explicit expressions for the transient part of reflectivity, under certain simplifying assumptions, and have studied its dependence on various exciton parameters such as the mass $M$ and the decay rate $\Gamma$.

Transient reflectivity can be decomposed into a "local" part and a "nonlocal" part, the latter being absent when spatial dispersion is ignored. An important prediction of our work is that for some semiconductors such as CdS or GaAs, the nonlocal part contributes to transients much more significantly (an order of magnitude higher) than the local part. For instance, at a time 0.1 psec after the reflected pulse is cut off, the transient intensities are about 10% of the incident intensity and remain 1% even after several psec. Such reflectivities should be easily measurable.

A remarkable feature of our theory is that it predicts a crossover from exponential to slow inverse power-law decay rate of transient reflectivity. For CdS and GaAs crystals, the crossover time is $\sim 1$ psec after the trailing edge of the reflected pulse. Although the physical origin of the crossover is not so clear to us, its existence can be traced back to the structure of Eqs. (3.11) and (3.12) as a one-sided Fourier transform. Application of the Paley-Weiner theorem[21] to the integral indicates the need for a nonexponential decay of transient reflectivity.

Our results are obtained under certain simplifying assumptions. The finite length crystal is replaced by a semi-infinite nonlocal medium and a linearly polarized plane wave is assumed to be associated with the light pulse, thereby ignoring its transverse beam profile. Furthermore, advantage is taken of the smallness of three dimensionless parameters, namely, $\delta=(\hbar\omega_t/Mc^2)^{1/2}$, $p=(\omega_l-\omega_t)/\omega_t$, and $\Gamma/\omega_t$. For most semiconductors of interest each one of them is $\lesssim 10^{-3}$. The graphical results presented here are expected to be accurate within 10% of those obtained when a more accurate analysis is carried out. In the absence of significant experimental data, efforts required to improve upon them do not appear to be worthwhile. As a matter of fact, our simplified approach has allowed us to obtain most of the qualitative features in an analytical form.

Measurements of the predicted features of transient reflectivity can provide independently determined values of important exciton-polariton parameters such as the mass, the lifetime, the oscillator strength, and the resonance frequency. Recently group velocities of the exciton polaritons in GaAs and CuCl have been measured with use of transient spectroscopy methods.[22] This gives support to our belief that the predicted transient effects could be measured.

## ACKNOWLEDGMENTS

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## APPENDIX: CONTOUR-INTEGRATION METHOD TO EVALUATE THE REFLECTED FIELD [EQ. (2.8)]

In this appendix we outline the algebraic details to evaluate the integral in Eq. (2.8) in the complex $\omega$ plane for $t\geq 2L/c$. Care should be exercised in dealing with the integrand. It is helpful to break the integral in two parts arising from each term in the curly brackets: $\{-\exp[i(\omega-\omega_0)T]\}$. The first part contributes for $t>2L/c$ while the second part contributes only for $t>2L/c+T$. Equation (2.8) can therefore be written in the form,

$$E_R(0,t)=\mathrm{Re}[E(t-2L/c)\Theta(t-2L/c)-e^{-i\omega_0T}E(t-2L/c-T)\times\Theta(t-2L/c-T)]\,, \qquad \text{(A1)}$$

where

$$E(\tau)=\lim_{\eta\to 0}\int_{-\infty}^{\infty} f(n_1,n_2)d\omega \ , \tag{A2}$$

$$f(n_1,n_2)=\frac{1}{2\pi}\frac{\rho(\omega)}{\omega_0-\omega-i\eta}e^{-i\omega\tau} \ , \quad \tau>0 \ . \tag{A3}$$

For notational convenience, we have explicitly shown the dependence of the integrand $f(n_1,n_2)$ on the refractive indices $n_1$ and $n_2$ [see Eq. (2.4)] and the $\omega$ dependence is implicitly understood.

The choice of an appropriate contour depends on the singularities of $f(n_1,n_2)$. For this purpose we require an explicit form of $n_1$ and $n_2$. Using Eq. (2.1) in (2.6), we obtain

$$n_{2,1}=+[a\pm(a^2-\epsilon_0 b)^{1/2}]^{1/2} \ , \tag{A4}$$

$$a=(2\omega^2\delta^2)^{-1}[\epsilon_0\omega^2\delta^2+(\omega^2-\omega_t^2+i\omega\Gamma)] \ ,$$

$$b=(\omega^2\delta^2)^{-1}[(\omega^2-\omega_t^2+i\omega\Gamma)-\beta^2\omega_t^2/\epsilon_0] \ , \tag{A5}$$

where

$$\beta^2=4\pi\alpha_0 \ , \quad \delta^2=(\hbar\omega_t/Mc^2) \ . \tag{A6}$$

An examination of Eq. (A4) shows that the conditions $b=0$ and $a^2=\epsilon_0 b$ correspond to the branch-point singularities in $f(n_1,n_2)$. Using Eq. (A5), these two conditions yield 6 branch points (in the complex $\omega$ plane) $\omega_j$, $j=1-6$, whose location is given by Eqs. (3.1)–(3.3). The branch points $\omega_1$ and $\omega_2$ arise from the condition $b=0$. Around these points the integrand $f(n_1,n_2)$ is made single-valued by going to the Reimann sheet on which $n_1\to -n_1$ and $n_2$ remains unchanged. The branch points $\omega_3$, $\omega_4$, $\omega_5$, and $\omega_6$ arise from the condition $a^2=\epsilon_0 b$; on the corresponding Riemann sheet $n_1$ and $n_2$ are interchanged. The appropriate contour to evaluate Eq. (A2) is shown in Fig. 2 and contains only a simple pole at $\omega_0=\omega_0-i\eta$.

A straightforward application of Cauchy's theorem shows that

$$\oint f(n_1,n_2)dw=\int_{-\infty}^{\infty} f(n_1,n_2)dw+\int_{\omega_3}^{\omega_1}[f(n_1,n_2)-f(-n_1,n_2)]dw+\int_{\omega_3}^{\mathrm{Re}\omega_3-i\infty}[f(-n_2,n_1)-f(n_1,n_2)]dw$$
$$+\int_{\omega_4}^{\omega_2}[f(-n_1,n_2)-f(n_1,n_2)]+\int_{\omega_4}^{\mathrm{Re}\omega_4-i\infty}[f(n_1,n_2)-f(-n_2,n_1)]dw=2\pi iR \ , \tag{A7}$$

where $R$ represents the residue at the pole $\omega=(\omega_0-i\eta)$. Using Eqs. (A2) and (A7), we formally decompose $E(\tau)$ into the steady-state (pole contribution) and the transient (branch-point contribution) parts,

$$E(\tau)=E_s(\tau)+E_T(\tau) \ , \tag{A8}$$

where

$$E_s(\tau)=i\rho(\omega_0)\exp(-i\omega_0\tau) \ , \tag{A9}$$

and

$$E_T(\tau)=\int_{\omega_3}^{\omega_1} g(\omega)d\omega-\int_{\omega_4}^{\omega_2} g(\omega)d\omega-\int_{\omega_3}^{\mathrm{Re}\omega_3-i\infty} g'(\omega)d\omega+\int_{\omega_4}^{\mathrm{Re}\omega_4-i\infty} g'(\omega)d\omega \ , \tag{A10}$$

$$g(\omega)=[f(-n_1,n_2)-f(n_1,n_2)] \ , \quad g'(\omega)=[f(-n_2,n_1)-f(n_1,n_2)] \ . \tag{A11}$$

We have found it useful to further decompose the transient part $E_T(\tau)$ into a "local" part and a "nonlocal" part. This facilitates comparison with a local medium for which the nonlocal part, by definition, vanishes identically. Such a decomposition can be carried out by noting that for a local medium the branch-cut line (joining $\omega_1$ and $\omega_3$) in Fig. 2 is horizontal. This can be readily verified using Eqs. (3.1) and (3.2) with $\delta=0$. We then formally obtain

$$E_T(\tau)=E_{\mathrm{L}}(\tau)+E_{\mathrm{NL}}(\tau) \ , \tag{A12}$$

where

$$E_{\mathrm{L}}(\tau)=\int_{\mathrm{Re}\omega_3+i\mathrm{Im}\omega_1}^{\omega_1} g(\omega)d\omega-\int_{\mathrm{Re}\omega_4+i\mathrm{Im}\omega_2}^{\omega_2} g(\omega)d\omega \ , \tag{A13}$$

$$E_{\mathrm{NL}}(\tau)=\int_{\omega_3}^{\mathrm{Re}\omega_3+\mathrm{Im}\omega_1} g(\omega)d\omega-\int_{\omega_4}^{\mathrm{Re}\omega_4+i\mathrm{Im}\omega_2} g(\omega)d\omega-\int_{\omega_3}^{\mathrm{Re}\omega_3-i\infty} g'(\omega)d\omega+\int_{\omega_4}^{\mathrm{Re}\omega_4-i\infty} g'(\omega)d\omega \ . \tag{A14}$$

We now substitute $E = E_s + E_{\rm L} + E_{\rm NL}$ in Eq. (A1) and obtain

$$E_R(0,t) = {\rm Re}[\widetilde{E}_s(t) + \widetilde{E}_{\rm L}(t) + \widetilde{E}_{\rm NL}(t)] , \tag{A15}$$

where

$$\widetilde{E}_s(t) = i\rho(\omega_0)[\Theta(t-2L/c) - \Theta(t-2L/c-T)]e^{-i\omega_0(t-2L/c)} , \tag{A16}$$

$$\widetilde{E}_j(t) = [E_j(t-2L/c)\Theta(t-2L/c) - e^{-i\omega_0 T}E_j(t-2L/c-T)\Theta(t-2L/c-T)] , \tag{A17}$$

with $j = {\rm L}$ or NL.

The evaluation of Eqs. (A13) and (A14) can be somewhat simplified by noting that in most cases of practical interest the three parameters $\delta$, $\beta^2/2\epsilon_0$, and $\Gamma/\omega_t$ are much smaller than unity ($<10^{-3}$). We are therefore justified in neglecting their products and higher powers. The branch points $\omega_j$ given by Eqs. (3.1) and (3.2) then simplify and become

$$\omega_{1,2} \simeq -i\Gamma/2 \pm (1+p)\omega_t , \quad \omega_{3,4} \simeq -i(\Gamma/2 + \beta\delta\omega_t) \pm \omega_t , \tag{A18}$$

where we have defined $p = \beta^2/2\epsilon_0 = 2\pi\alpha_0/\epsilon_0$ and physically $p = (\omega_l - \omega_t)/\omega_t$, where $\omega_l - \omega_t$ is the so-called LT splitting. When Eq. (A18) is used in Eqs. (A13) and (A14), an appropriate change of variables permits to rewrite them in the following simplified form:

$$E_{\rm L} = p\omega_t \left[ \int_0^1 g(\omega_t + pu\omega_t - i\Gamma/2)du + \int_0^1 g(-\omega_t - pu\omega_t - i\Gamma/2)du \right] , \tag{A19}$$

$$\begin{aligned} E_{\rm NL}(\tau) = i\beta\delta\omega_t \Bigg[ & \int_0^1 g(\omega_t - i\beta\delta\omega_t u - i\Gamma/2)du - \int_0^1 g(-\omega_t - i\beta\delta\omega_t u - i\Gamma/2)du \\ & + \int_1^\infty g'(\omega_t - i\beta\delta\omega_t u - i\Gamma/2)du - \int_1^\infty g'(-\omega_t - i\beta\delta\omega_t u - i\Gamma/2)du \Bigg] . \end{aligned} \tag{A20}$$

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*Present address: Microelectronics Research and Development Center, Rockwell International, Anaheim, CA 92803.

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