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Resonance-Enhanced Transient Reflectivity via Exciton Polaritons

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The reflection of a light pulse from a spatially dispersive medium is investigated theoretically. For laser frequency at exciton resonance, spatial dispersion enhances the reflected transients associated with the light pulse. For the case of CdS and GaAs crystals, the transient intensities are about 10% of the incident intensity at a time 0.1 ps after the trailing edge of the reflected signal. The theory predicts a crossover from exponential to slow power-law decay rate of transient reflectivity; this occurs at a characteristic time $\zeta_c \sim 1$ ps after the trailing edge of the pulse for CdS and GaAs.

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This Letter reports results of a theoretical investigation of transient optical reflectivity from spatially dispersive media such as CdS and GaAs.[1] The laser frequency of an incident pulse is taken near one-photon resonance with the transverse exciton-polariton frequency ω_t. We show that enhanced (transient) reflectivity persists for several picoseconds after a laser pulse is cut off. In general both the leading and the trailing pulse edges give rise to transients. Experimentally it may be more convenient to look for trailing-edge transient reflectivity since steady-state reflectivity will then not interfere with measurements. For sufficiently long pulses ($T >$ a few picoseconds), transients from the two edges will be essentially decoupled and can be considered independently. Our results show that transient reflectivity consists of a "local" part and a "nonlocal" part. The former, although the only one present in a local media, 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 $M^{-3/4}$ with the exciton mass. Initially the time decay of reflectivity is exponential, crossing over to inverse-power decay, at about 1 ps in CdS or GaAs. Its maximum magnitude is about 10% of the incident intensity. These effects should be measurable.

The origin of spatial dispersion or nonlocality is due to coupling of an exciton state (with center-of-mass motion included) to a photon, producing an exciton polariton. This results in a wave-vector–dependent optical dielectric response. Consequently there is an "additional" transverse-propagating mode compared to the case of a local medium ($M = \infty$). In particular there is a (transverse) mode in the "pseudogap" frequency region, $\omega_t > \omega > \omega_l$, where ω_l is the longitudinal frequency. Steady-state reflectivity, transmittivity, and inelastic scattering in such media have been well studied[1]: Reflectivity is generally maximum in the pseudogap region although smaller than that of a local medium. Transient optical transmission was investigated theoretically by Birman and Frankel,[2] who showed that a new exciton precursor occurs; Johnson[3] discussed transient oscillations in transmittivity of a plate.

We consider transient reflectivity from a semi-infinite nonlocal medium occupying half-space $z > L$, with $z < L$ being vacuum. A detector is placed at $z = 0$. The nonlocal medium is taken to have a scalar isotropic dielectric susceptibility[4]

$$\chi(\vec{r}, \vec{r}\,') = \chi(\vec{r} - \vec{r}\,') - \chi(\vec{\zeta} - \vec{\zeta}\,', z + z'), \tag{1}$$

where $\vec{\zeta} = (x, y)$ is the transverse part of $\vec{r}$. At time $t = 0$ and at $z = 0$, a square optical pulse, with laser frequency ω_0 and duration T, is incident normally on the crystal surface. The electric field

$$\vec{E}(0,t)=\sin(\omega_0 t)[\theta(t)-\theta(t-T)]\hat{x}, \qquad (2)$$

where $\theta(t)$ is the Heaviside step function, corresponds to a polarized, monochromatic plane-wave field.

Because of the linearity of the problem, we may Fourier analyze (2) and treat each frequency component by dispersion theory. The reflected field is obtained as a superposition of these components and can be written as a complex-frequency integral,[5]

$$\vec{E}_r(0,t)=\hat{x}\lim_{\eta\to\infty}(1/2\pi)\int_{-\infty}^{\infty}d\omega[\rho(\omega)/(\omega-\omega_0+i\eta)]\exp[-i\omega(t-2L/c)]\{1-\exp[i(\omega-\omega_0)T]\}+\text{c.c.}, \qquad (3)$$

where $\rho(\omega)$ is the complex-amplitude reflection coefficient for susceptibility of Eq. (1) and is given by[4]

$$\rho(\omega)=[1-n(\omega)]/[1+n(\omega)], \qquad (4)$$

$$n(\omega)=(n_1 n_2+\epsilon_0)/(n_1+n_2). \qquad (5)$$

In Eq. (5), ϵ_0 is the background dielectric constant, and n_1 and n_2 are the refractive indices of the two propagating transverse modes in the nonlocal medium.[4,6]

The integral (3) is evaluated in the complex ω plane. For $t<2L/c$, we close the contour in the upper half-plane and find $\vec{E}_r=0$ as required by causality. For $t>2L/c$, the contour is closed in the lower half-plane. The integrand in (3) has the following singularities: (a) a simple pole at $\omega_0-i\eta$, (b) four branch points ω_j (joined by a pair of branch-cut lines) with $\omega_1{}^*=-\omega_2$ and $\omega_3{}^*=-\omega_4$. Details of the contour integration are given elsewhere.[7,8] Steady-state reflectivity arises from the pole contribution, while the branch points contribute to transient reflectivity. For a local medium ($M=\infty$) and far from exciton resonance, Elert[9] evaluated branch-point contributions analytically.

A simplification in the nonlocal case occurs by noting that a small dimensionless parameter exits: $\delta=(\hbar\omega_t/Mc^2)^{1/2}$, where c is the velocity of light, and $\delta\sim10^{-3}$ for CdS or GaAs. To the leading order in δ, neglecting off-resonance terms, the reflected field at time $t>2L/c+T$ can be written as[8]

$$\vec{E}_r(0,t)=-\hat{x}\sum_{j=1}^{2}|R_j(\zeta)|\sin(\omega_0 t-\varphi_j), \qquad (6)$$

where $R_j(\zeta)=|R_j(\zeta)|e^{i\varphi_j(\zeta)}$, $\zeta=t-2L/c-T$ is a suitably retarded time ($\zeta=0$ when the trailing edge of the reflected pulse arrives at the detector at $z=0$), and

$$R_1(\zeta)=\left(\frac{2i\delta^3}{\beta\pi^2}\right)^{1/2}e^{-\Gamma\zeta/2}\int_0^1 du\,\frac{\omega_t\exp(-\beta\delta\omega_t\zeta u)}{[\omega_t-\omega_0-i(\beta\delta u+\Gamma/2)]}[(1+u)^{1/2}+i(1-u)^{1/2}], \qquad (7)$$

$$R_2(\zeta)=\frac{\sqrt{2}}{\pi}p^3e^{-\Gamma\zeta/2}\int_0^1 du\,\frac{\omega_t\exp(-ip\omega_t\zeta u)}{[\omega_t(1+pu)-\omega_0-i\Gamma/2]}[(u+i\beta\delta/p)^{1/2}+\text{c.c.}]. \qquad (8)$$

Here $p=2\pi\alpha_0/\epsilon_0$, $\beta^2=4\pi\alpha_0$, and α_0 is the exciton oscillator strength. Further, Γ is an exciton damping constant. We note that as $M\to\infty$ ($\delta\to0$), $R_1(\zeta)\to0$, and $R_2(\zeta)$ reduces to Elert's result[9] in the limit of being far off resonance ($\omega_0\ll\omega_t$). A remarkable feature is that R_1 arises solely from spatial dispersion and is one order of magnitude larger than R_2. Although a finite exciton mass also introduces new features such as double peak in R_2 as a function of ω_0, in this Letter we neglect R_2 because major experimental features will be determined by R_1 alone.

We have numerically evaluated $R_1(\zeta)$ using parameters appropriate for CdS and GaAs. In Fig. 1, $|R_1(\zeta)|$ is plotted for CdS as a function of time for three values of Γ for which $\omega_0=\omega_t$ is assumed. Note appreciable reflectivity persists for a few picoseconds after the pulse cutoff. In Fig. 2, resonance enhancement of $|R_1|$ is shown at a fixed time $\zeta=0.1$ ps for two values of Γ. Enhancement by a factor of 5–10 is seen in a narrow frequency range. In Figs. 3 and 4, similar features are shown for GaAs parameters. In both materials the on-resonance transient reflectivity $|R_1(\zeta)|^2$ at 0.1 ps after the pulse cutoff is about 10% of the incident intensity and remains 1% even after several picoseconds.

An important feature of Figs. 1 and 3 is the crossover from a fast exponential decay to a slow power-law decay. An estimate of the crossover time is obtained from Eq. (7), which can be evaluated analytically for several limiting cases of interest.[8] Such an analysis reveals the existence

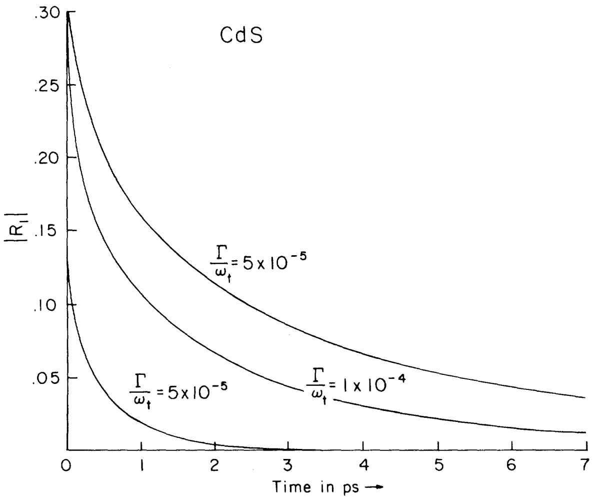

FIG. 1. Decay of $|R_1(\zeta)|$ with ζ for the on-resonance case ($\omega_0 = \omega_t$) for three values of Γ. At $\zeta = 0$, the trailing edge of the reflected pulse arrives at the detector. The parameters are appropriate to CdS: $\epsilon_0 = 8$, $4\pi\alpha_0 = 0.0125$, $\hbar\omega_t = 2.55$ eV, and $M = 0.9m_e$ (with m_e the electron mass).

of a characteristic time

$$\zeta_c = (\beta\delta\omega_t)^{-1} = [Mc^2/(4\pi\alpha_0\hbar\omega_t^3)]^{1/2}, \tag{9}$$

and one finds (setting $\Gamma = 0$ for simplicity) that

$$|R_1(\zeta)| \sim \begin{cases} C_1 \exp(-\zeta/\zeta_c) \text{ for } \zeta \ll \zeta_c \\ C_2/\zeta \text{ for } \zeta \gg \zeta_c, \end{cases} \tag{10}$$

where C_1 and C_2 are slowly varying functions of time. For materials of interest, such as CdS and GaAs, we estimate that $\zeta_c \sim 1$ ps, which should be experimentally observable. Hence to asymptotic accuracy, the decay of $|R_1(\zeta)|$ begins exponentially and shows a crossover to a ζ^{-1} power law around ζ_c.[10]

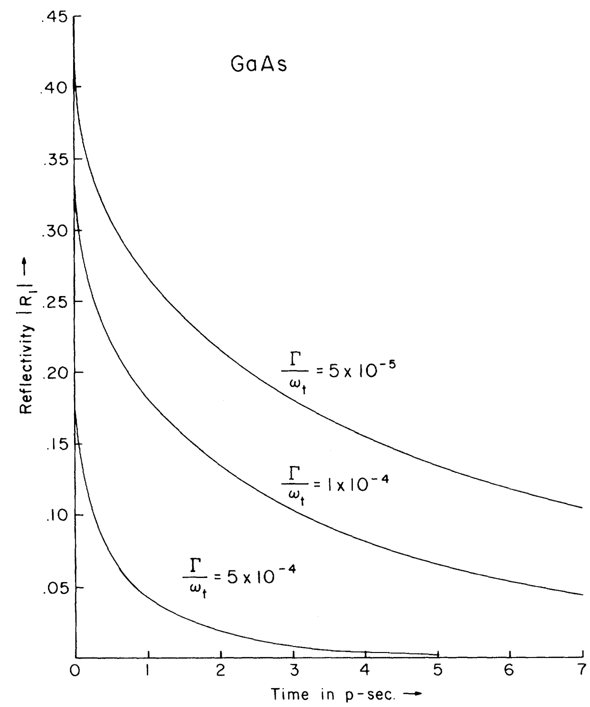

FIG. 3. Time decay of $|R_1(\zeta)|$ as in Fig. 1. The parameters correspond to GaAs: $\epsilon_0 = 12.55$, $4\pi\alpha_0 = 0.0013$, $\hbar\omega_t = 1.515$ eV, and $M = 0.6m_e$.

Measurement of the predicted features of transient reflectivity can provide independently determined values of important exciton-polariton features such as Γ, M, α_0, and ω_t. Recently group velocities of exciton polaritons in GaAs and CuCl have been measured with use of transient spectroscopy methods.[11] This gives sup-

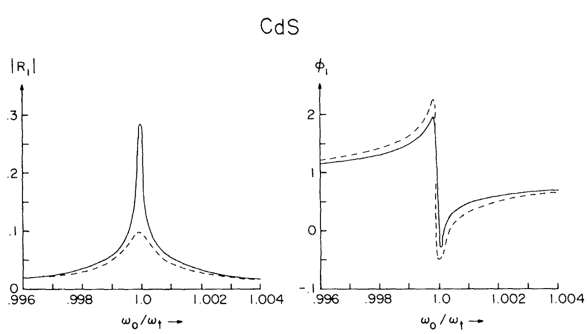

FIG. 2. Variation of $|R_1|$ with the laser frequency ω_0 at a fixed time $\zeta = 0.1$ ps; full line, $\Gamma = 5\times10^{-5}\omega_t$; dashed line, $\Gamma = 5\times10^{-4}\omega_t$. The phase φ_1 is shown on the right. CdS parameters are as in Fig. 1.

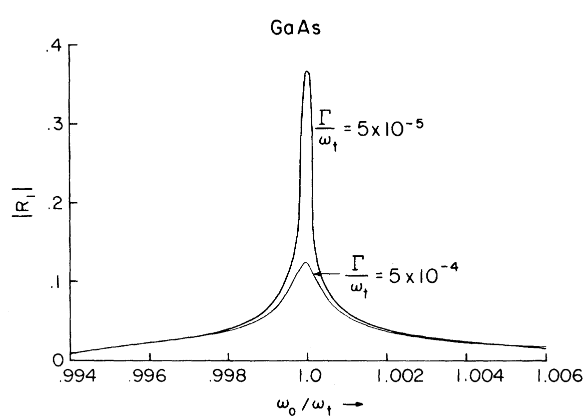

FIG. 4. Resonance enhancement of $|R_1|$ for GaAs at a fixed time $\zeta = 0.1$ ps for two values of Γ. The parameters are those of Fig. 3. The variation of phase φ_1 is similar to that of Fig. 2.

port to our belief that the predicted transient effects could be measured.

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Efficacy of Passive-Limiter Pumping of Neutral Particles

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Experiments have been performed to measure the neutral pressure buildup behind the limiter as a function of plasma density and gas species. The results indicate that a passive mechanical limiter effectively removes from the vacuum vessel up to 20% of the atoms injected during a discharge. The feasibility of mechanical limiters removing the fusion-reaction helium ash, thus negating a major need for magnetic divertors, is discussed.

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To sustain a controlled, steady-state D-T burn, one must be capable of maintaining a fixed α-particle density. For typical ignition-size plasmas and plasma parameters, the steady-state conditions may be achieved if approximately 10% or more of the α particles are not recycled; in other words, if they are pumped away.[1,2] One possible way to do this is by using a passive mechanical limiter, comparable to those promoted by Schivell.[3]

Experiments to investigate the effectiveness of a passive-limiter pumping scheme were performed on the Alcator-*A* tokamak. The vacuum vessel has a major radius of 54 cm and a minor radius of 12.5 cm. The total volume of the torus and the diagnostic port extension tubes is 451 liters; plasma volume is ≈ 117 liters.

A single molybdenum limiter is inserted from the horizontal port as shown in Fig. 1. The limiter is electrically floating and isolated from the vacuum vessel. The toroidal thickness of the limiter is 1.1 cm with a poloidal extension of 205°. The limiter inner radius (plasma radius) is 10.4 cm and the outer radius is 12.2 cm.

At the limiter flange there are three port extension tubes. During tokamak operation the ex-

