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Subpicosecond time-resolved reflection of ultrafast electrical pulses from GaAs in the presence of nonthermal photoexcited carriers

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The reflected waveform of a femtosecond electrical pulse incident on GaAs following hot-electron excitation by a subpicosecond optical pulse is calculated. Conditions under which the electrons initially relax mainly by emitting a cascade of longitudinal-optical (LO) phonons are assumed to hold. It is found that the electrical pulse can show amplification on reflection when the initial kinetic energy of the excited electrons equals an integral multiple of the LO-phonon energy within a sufficiently narrow energy spread.

Some years ago, Auston *et al.*[1] demonstrated that subpicosecond electromagnetic transients containing far-infrared frequencies can be generated in electro-optic materials during propagation of femtosecond optical pulses. A technique to use these electric field pulses for measuring far-infrared dielectric properties of materials was also developed.[2] In a recent experiment, Nuss, Auston, and Capasso[3] used such a technique to deduce time-dependent mobility of photoexcited hot electrons in GaAs with femtosecond time resolution for the first time. In that experiment, the carrier dynamics was studied by measuring the reflectivity of a femtosecond electrical pulse as a function of the delay time with respect to the initial carrier photoinjection. The carriers are excited by a femtosecond optical pulse. The electrical pulses are generated by optical rectification of femtosecond laser pulses[1,2] in a nonlinear medium ($LiTaO_3$) in contact with the GaAs surface. Both the incident and the reflected electric field waveforms at the GaAs surface are separately measured using the electro-optic effect in $LiTaO_3$. Such electrical pulses have been shown[2] to have a wide Gaussian-like spectral content with the frequency ranging from dc to a few THz and a peak at about 1 THz. The pulse duration is about 300 fsec.

In view of the importance of such measurements, we present a calculation of the time-dependent Fresnel reflection coefficient for a short electrical pulse incident on GaAs following hot electron excitation by a subpicosecond optical pulse. We compute the reflected electric field waveform for different conditions of photoexcitation and momentum relaxation rates due to quasielastic scattering mechanisms. We find the extremely interesting result that the incident electrical pulse can be amplified on reflection from the photoexcited GaAs surface when the initial photoelectron kinetic energies are restricted to be equal to an integral multiple of $\hbar\omega_{LO}$, the longitudinal-optical-phonon energy, within a sufficiently narrow energy spread ($=2\Delta < \hbar\omega_{LO} < 36$ meV in GaAs). Our results also bring out the role played by the momentum relaxation time due to quasielastic electron scattering in distorting the reflected electric field waveform.

To show this, we first calculate the time-dependent ac electrical conductivity of the photoexcited carriers with the dependence on the frequency of the incident electric field included. The calculation is performed within the framework of time-dependent Boltzmann transport equation for low-density photogenerated carriers ($n < 10^{17}$ cm^{-3}). We assume that a subpicosecond optical pulse excites electrons into the central Γ conduction valley predominantly from the heavy-mass valence band of a GaAs-like polar semiconductor, the light-mass valence band having a smaller density of states. The photon energy is taken to be small enough to avoid $\Gamma \rightarrow L$ intervalley transfer as a simplification. Under these conditions, the excited electrons initially relax to lower energies by emitting a series of longitudinal-optical (LO) phonons.[4] The carrier generation at the exciting photon energies of 1.6 eV and above typically occurs in a depth of about 0.2 to 0.4 μm in GaAs. In view of the long wavelengths contained in the incident electric fields ($\lambda \sim 300$ μm), the photocurrent carried by the excited carriers can be taken as a surface current. We may mention here that we focus attention only on the initial period of carrier relaxation of a few psecs and do not consider the regime of slow processes such as carrier recombinations, diffusion, etc., which occur on the scale of several tens of psecs or more. The surface current can be obtained from the volume photocurrent integrated over the depth of carrier generation. The probing electric field vector is taken here for simplicity to be in the plane of the GaAs surface (s polarization).

The generalized Fresnel reflection coefficient for a short electrical pulse can be calculated for weak electric fields by using the linearity of Maxwell's equations. The incident electrical pulse $E_i(t)$ can be Fourier decomposed as

$$E_i(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} E_i(\omega) \exp(-i\omega\bar{t}) d\omega , \qquad (1)$$

where $\bar{t} = t - t_0$, t_0 is the center of the pulse. The reflected field $E_r(t)$ is given by

$$E_r(t) = \frac{1}{\sqrt{2\pi}} \int_{-\infty}^{\infty} r(\omega,t) E_i(\omega) \exp(-i\omega\bar{t}) d\omega . \qquad (2)$$

(In what follows, it is understood that complex-conjugate quantities are added to obtain real fields and currents.) Here $r(\omega,t)$ is the Fresnel reflection coefficient at frequency ω for the s polarization obtained by solving Maxwell's equations subject to the usual boundary conditions expressing the continuity of the tangential components of the electric fields and of the magnetic fields in the two media at the interface. Writing

$$E_r(\omega) = r(\omega,t)E_i(\omega) , \tag{3}$$

it can be shown that the individual reflection coefficients are given in terms of the surface conductivity $\sigma_s(\omega,t)$ by[3]

$$r(\omega,t) = r_0 - (1+r_0)\frac{\sigma_s(\omega,t)}{\sigma_s(\omega,t)+\eta} , \tag{4}$$

where $r_0 = (n_1\cos\theta - n_2\cos\theta')/(n_1\cos\theta + n_2\cos\theta')$ is the usual reflection coefficient for s polarization in absence of photoexcitation. Here n_1, n_2 are the refractive indices of the media of incidence and transmission, respectively, both taken to be nonmagnetic and θ and θ' are angles of incidence and refraction, respectively. Also,

$$\eta = (\epsilon_0/\mu_0)^{1/2}(n_1\cos\theta + n_2\cos\theta')$$

in Eq. (4), ϵ_0 and μ_0, respectively, are the free-space permittivity and permeability.

If $E_i(t)$ and $\sigma_s(\omega,t)$ are known, $E_r(t)$ can be calculated for different values of the delay time t_0, measured from the onset of photoexcitation ($t=0$). As mentioned earlier, the surface conductivity $\sigma_s(\omega,t)$ can be obtained from the volume conductivity $\sigma(\omega,t)$ as

$$\sigma_s(\omega,t) = \int_0^\infty \sigma(\omega,t)e^{-x'/d}dx' , \tag{5}$$

where $d = \alpha^{-1}$ is the absorption depth, α is the absorption coefficient for the optical pulse, and

$$\sigma(\omega,t) = \int_0^t \Sigma(t,t')e^{i\omega(t-t')}dt' . \tag{6}$$

The generalized conductivity response function $\Sigma(t,t')$ is to be obtained by comparing the following expressions for the photocurrent $J(\omega,t)$ induced by the Fourier electric field $E(\omega,t)\sim E(\omega)\exp(-i\omega t)$:

$$J(\omega,t) = 2e\sum_{\mathbf{k}} f_{\mathbf{k}}(t)v(\mathbf{k}) , \tag{7}$$

and

$$J(\omega,t) = \int_0^t \Sigma(t,t')E(\omega,t')dt' , \tag{8}$$

where $E(\omega,t)$ is the total electric field in GaAs (i.e., a sum of the incident and the reflected fields) near the surface, and v is the particle velocity parallel to $E(\omega,t)$. The nonequilibrium independent-particle distribution function $f_{\mathbf{k}}(t)$ can be obtained within the framework of the time-dependent Boltzmann transport equation. Here we retain only the dominant contribution of the conduction electrons to $J(\omega,t)$, the excited heavy holes having much smaller mobility.

We approximate the incident electrical pulse by the following Gaussian form as an illustration:

$$E_i(t) = Ae^{-\bar{t}^2/2\tau^2}e^{-i\omega_0\bar{t}} , \tag{9}$$

where $\bar{t} = t - t_0$, t_0 is the center of the pulse, and A determines the strength of the field. From Eq. (1), we then have

$$E_i(\omega) = A\tau\exp[(\omega-\omega_0)^2\tau^2/2] . \tag{10}$$

We choose $\omega_0/2\pi$, the peak frequency, to be 1 THz and τ, the pulse width, to be 0.3 psec, giving a high cutoff (at 10%) of about 2 THz, similar to the experimental conditions.[2,3]

It is clear that once $\Sigma(t,t')$ is known, Eqs. (4)–(6) can be used to compute $E_r(t)$ using Eq. (2). An explicit expression for the nonequilibrium velocity-velocity correlation function $\Sigma(t,t')$ under the assumed conditions of photoexcitation and electron relaxation is readily available in literature, as obtained by us earlier.[5] Using that, we calculate the reflected electric field $E_r(t)$ [Eq. (2)] for $E_i(t)$ given by Eqs. (9) and (10) for different values of the excitation energies and of the quasielastic momentum relaxation rate.

That a variation of the excited electron energy has a rather dramatic effect in influencing the shape of the reflected pulse is anticipated for the following reasons. We have previously shown[5] for the low-density case that at low enough frequencies ($\omega\tau_m \ll 1, \tau_m$ = effective momentum relaxation time), σ is real and can attain absolute negative values in the subpicosecond and picosecond time domain, whenever the electron energy distribution at generation is peaked at $\varepsilon_x = n\hbar\omega_{\mathrm{LO}}$, n is the integer, with a peak width $2\Delta < \hbar\omega_{\mathrm{LO}}$. The conductivity subsequently approaches positive values determined by momentum relaxation time due to quasielastic scattering. The negative conductivity essentially arises[5] because the average electron velocity develops a direction opposite to that of the accelerating electric field when the electrons relax to energies $\approx \hbar\omega_{\mathrm{LO}}$. [The electrons accelerated by the field to energies beyond $\hbar\omega_{\mathrm{LO}}$ quickly lose energy by emitting an LO phonon but those moving against the field (i.e., with negative velocities) cannot]. This in principle can lead to very large $r(\omega,t)$ due to the singularity at $\sigma_s = -\eta$ [Eq. (6)] at low frequencies. For the more general response to electrical pulses considered here with complex $\sigma_s(\omega,t)$, the reflection coefficient $r(\omega,t)$ can still show a sharp enhancement whenever $\mathrm{Re}\sigma_s(\omega,t)$ nearly equals $-\eta$ for frequencies close to ω_0. Our computations of $E_r(t)$ indeed show that the reflected pulse is significantly distorted when the resonance condition $\varepsilon_x = n\hbar\omega_{\mathrm{LO}}$, n is the integer, is satisfied. In fact, $E_r(t)$ can exceed $E_i(t)$ implying amplification under suitable conditions of the time delay t_0 and the scattering time τ_1. Figure 1 provides an illustration of this interesting effect. A time delay of $t_0 = 2.25$ psec with respect to the onset of photoinjection at $t=0$ is chosen for this calculation. Normal incidence of the pulse is assumed for simplicity. The refractive index of the medium of incidence is set to be 6.4 while that of GaAs is assumed to be 3.8 in the frequency range contained in Eq. (10). The excited electron areal density n_s is taken to be 2.5×10^{12} cm^{-2}, generated with a laser pulse of duration of 0.2 psec. This corresponds to a volume density (N) at the surface of 1×10^{17} cm^{-3} if the absorption depth (d) is 0.25 μm. The electrons are assumed to initially have a Gaussian energy distribution with a peak centered at

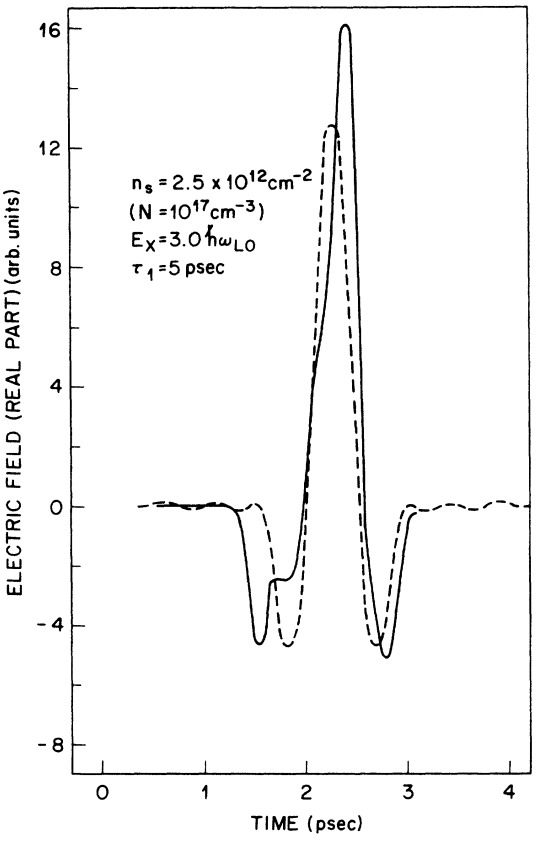

FIG. 1. The reflected electrical pulse waveform showing amplification when hot electrons are excited with energies ε_x at "resonance." τ_1 is the momentum relaxation time due to quasielastic scattering. The overall negative sign of the reflected pulse is removed for an easy comparison with the incident pulse shape given by Eq. (9) (dashed line).

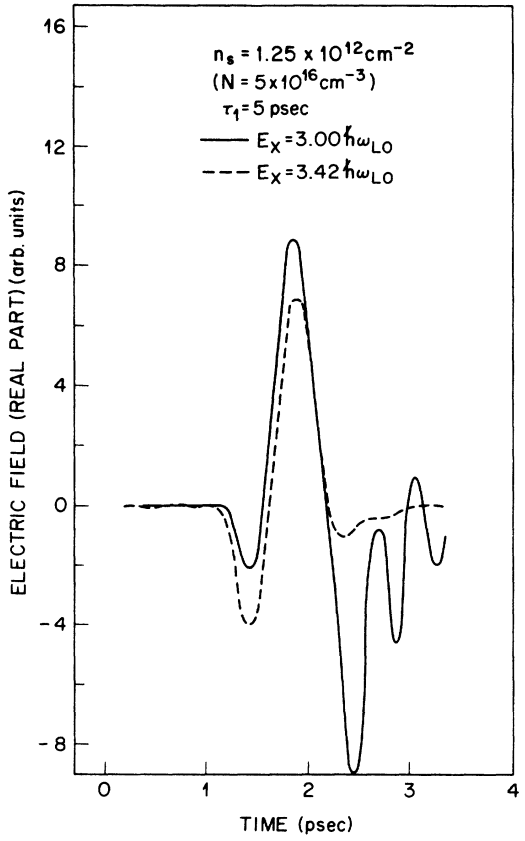

FIG. 2. The effect of changing the electron energy at generation from $3.42\hbar\omega_{LO}$ ("off-resonance") to $3.0\hbar\omega_{LO}$ ("on-resonance") in distorting the reflected pulse shape.

$\varepsilon_x = 3.0\hbar\omega_{LO}$ and a width 2Δ at e^{-1} of 10 meV. It can be shown that most of the excited electrons relax to energies below $\hbar\omega_{LO}$ in less than 1 psec so that τ_m is governed mainly by the quasielastic scattering mechanisms at the time of incidence of the electrical pulse. The momentum relaxation time τ_1 due to these scatterings is assumed for this illustration to be energy independent with a value of 5.0 psec. Figure 1 clearly demonstrates that by tuning the excitation energy to the resonance condition, it is possible to detect an enhancement in the reflected pulse.

The effect of changing ε_x from off-resonance to on-resonance condition is shown by another example in Fig. 2. A value of $n_s = 1.25 \times 10^{12}$ cm^{-3} ($N = 5 \times 10^{16}$ cm^{-3} if $d = 0.25$ μm) is used for this calculation. The delay time t_0 is set equal to 1.8 psec while τ_1 is 5.0 psec. The sharp change brought about in the distortion of the reflected pulse shape by shifting ε_x from $3.42\hbar\omega_{LO}$ to $3.00\hbar\omega_{LO}$ is quite remarkable.

The other parameter which may influence the shape of the reflected pulse is the momentum relaxation time, τ_m. To see this, we may refer to $\Sigma(t,t')$ of Ref. 5. It turns out that $\Sigma(t,t')$ contains factors such as $\exp[-(t-t')/\tau_m(\varepsilon)]$ where $\tau_m^{-1}(\varepsilon) = \tau_{LO}^{-1}(\varepsilon) + \tau_1^{-1}(\varepsilon)$ is the effective momentum relaxation rate given in terms of the characteristic LO-phonon emission rate τ_{LO}^{-1} and the quasielastic scattering rate $\tau_1^{-1}(\varepsilon)$, ε being the electron energy measured from the conduction-band edge. Typically, $\tau_{LO} \sim 150$ fsec and $\tau_1 \sim 1$–100 psec in undoped GaAs depending upon the background doping and the lattice temperature.[6] The initial carrier relaxation is dominated by $\tau_m \approx \tau_{LO}$ but as the electrons relax to energies less than $\hbar\omega_{LO}$, $\tau_m \approx \tau_1$. It is easy to see from Eq. (6) that the transient electrical conductivity becomes real and frequency independent only if $\omega\tau_m < 1$. This is basically a consequence of the fact that the transient photocurrent is in a "phase quadrature" with the impressed electric field.[2]

For the waveform given by Eqs. (8) and (9), the conductivity $\sigma_s(\omega,t)$ in general will be complex and frequency dependent, if the momentum relaxation time τ_m is much larger than $1/\omega_0 \simeq 0.2$ psec. The photocurrent then will not be in phase with the driving field. Also the reflected field pulse may have a waveform different in shape from that of the incident pulse due to the ω dependence of $\sigma_s(\omega,t)$. This indeed is seen in Fig. 3 when τ_1 is 5 psec. On the other hand, for a situation when τ_m has a much smaller value, it is expected that the reflected pulse will have a waveform not very different in shape from that of the incident pulse due to $\sigma_s(\omega,t)$ becoming nearly real and ω independent. Figure 3 very clearly shows this when the effects of $\tau_1 = 5$ and 0.25 psec are compared. The example with τ_1 of 0.25 psec corresponds to the conditions of

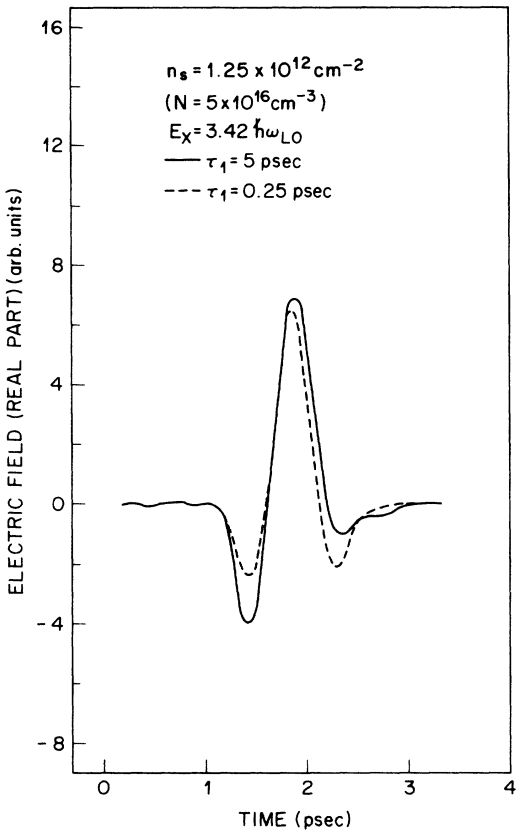

FIG. 3. The distortion in the reflected pulse waveform brought about by reducing the momentum relaxation rate due to quasielastic scatterings from 4×10^{12} sec^{-1} ($\tau_1 = 0.25$ psec) to 2×10^{11} sec^{-1} ($\tau_1 = 5$ psec).

the experiments of Nuss, Auston, and Capasso[3] where the transient electron mobility attains a small maximum value $\mu_0 (= e\tau_m/m^*) \simeq 4000$ cm^2/V sec with $\tau_m \simeq 0.2$ psec. The approximation of a frequency independent, real $\sigma_s(t)$ therefore is justifiable in this case. This confirms that the smaller the quasielastic scattering rate, the more the distortion of the reflected electrical pulse. It may be mentioned here that the time dependence of conductivity within the probe pulse duration ($\sim$300 fsec) is included in our calculations. The resulting small distortion of the pulse is present even when the effects of frequency dependence are insignificant.

Finally, we address the question of a practical test of the effects discussed in this paper. By suitably tuning the width t_p of the optical pulse used to generate hot carriers, 2Δ can be made sufficiently small ($\ll \hbar\omega_{LO} \simeq 36$ meV in GaAs). It was demonstrated earlier by us[5] that the negative σ effect persists even after increasing t_p to 0.5 psec. If the energy of these pulses can be varied so that the excited electron energy can be changed by at least 15–20 meV, the effects predicted in this paper may be tested. Femtosecond optical pulses will of course be required to generate and detect ultrashort electrical pulses. The large value of τ_1 needed to obtain the effect seen in Figs. 1 and 2 at a convenient value of t_0 can easily be arranged by performing the experiments on high mobility samples, if necessary at reduced temperatures. The $\Gamma \rightarrow L,X$ sidevalley transfer, not included in the analysis, can be avoided in the experimental conditions by using a III-V semiconductor with a sufficiently large intervalley transfer threshold in energy (e.g., InP, InGaAs). We may note that for values of the excitation volume densities beyond 10^{17} cm^{-3}, the model of carrier relaxation with a cascade emission of LO phonons may no longer be valid and the resonance effects described in this paper could be smeared out.

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