Damping Effects on the Polariton and Plasmariton Dispersion Curves in *n*-GaAs

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The effect on the temporal and spatial damped-dispersion curves of the polariton and plasmariton has been evaluated for different electron concentrations, lifetimes of the transverse optical (TO) phonon- and electron-collision times. These dispersion curves are important to optical spectroscopists who are investigating Raman and infrared processes.

INDEX HEADINGS: Raman effect; Infrared; Dispersion; Absorption.

The purpose of this paper is to calculate separately the temporal and spatial[1] damped-dispersion curves, ω vs q, of the polariton (a coupled TO phonon–photon) and plasmariton (a coupled TO phonon–dressed photon) in n-GaAs for different electron concentrations (N), optical-phonon lifetimes ($1/\gamma$), and electron-collision times ($1/\nu$).

Since the advent of laser Raman spectroscopy, it has been possible to measure experimentally the effect of the coupling among the various elementary excitations, in particular the coupled LO phonon–plasmon,[2] TO phonon–photon,[3] and the dressed photon–TO phonon[4] excitations. It is also possible to observe the coupled LO phonon–plasmon excitation by infrared absorption or reflection.[5] There appears to be no reported theoretical study of the effect of damping on the coupled temporal and spatial damped-dispersion curves except for the work of Tsu[6], who considered only spatial damping in SnTe. A damped excitation wave can be described by a complex frequency and/or momentum. The physical problem and conservation of momentum may define the complex nature of ω and $\bar{q}$. Two examples are: a temporal damped excitation, described by a real momentum $\hbar\bar{q}_1$ and complex frequency $\omega=\omega_1+i\omega_2$, and the waves involved, for example, in a Raman-scattering experiment[7] when the incident and scattered frequencies are outside the excitation-wave absorption region. A spatial damped excitation, described by a complex momentum $\hbar\bar{q}=\hbar(\bar{q}_1+i\bar{q}_2)$ or propagation constant $\bar{q}=\bar{q}_1+i\bar{q}_2$ and real frequency ω_1, takes part in infrared absorption or reflection experiments.[5]

The transverse plasmon[8,9] or dressed photon[6,10] is a quasiparticle that consists of a photon surrounded by an electron cloud. The electromagnetic field is strongly coupled to the electron, as witnessed by the modification of the photon's dispersion relation, which is given by

$$\omega^2=c^2q^2/\epsilon_\infty+\omega_p^2/\epsilon_\infty, \tag{1}$$

where c is the velocity of light, q is the magnitude of the propagation constant of the light, ϵ_∞ is the high-frequency dielectric constant of the medium, and ω_p is the plasma frequency for the electrons. Essentially, these transverse oscillations are electromagnetic waves modified by the fields arising from the collective electron response.[8] That is, the electromagnetic wave transfers energy to those electrons that move in a direction perpendicular to q. For $\omega_p \ll cq/\sqrt{\epsilon_\infty}$ the electromagnetic field is weakly coupled to the system; its frequency of oscillation is only slightly modified by the presence of the electrons. At $q=0$ the dressed photon's frequency is $\omega_p/\sqrt{\epsilon_\infty}$; below this frequency the photon is totally reflected.

We show that damping of TO phonons and electrons can have an important effect on the polariton and plasmariton dispersion curves. We have neglected the effect of a decaying polariton and plasmariton due to collision with another elementary excitation, in particular, with an acoustic phonon. In Sec. I the theoretical aspects of the total dielectric function of the coupled system are discussed; in Sec. II the dispersion curves for the undamped polariton and plasmariton are investigated, while in Secs. III and IV the temporal and spatial damped-dispersion curves are considered, respectively.

I. THEORY

The total dielectric function[6,11,12] of a system consisting of optical phonons and conduction-band electrons in GaAs, in the long-wavelength and SCF[13] approximation, is

$$\epsilon_{T,L}(\omega)=\epsilon_\infty-[(4\pi/i\omega)\sigma_{T,L}] \\ -(\epsilon_0-\epsilon_\infty)\omega_{\mathrm{TO}}^2/(\omega^2-\omega_{\mathrm{TO}}^2+i\gamma_{T,L}\omega), \tag{2}$$

where the subscripts T, L correspond to the transverse ($q_\perp$) and longitudinal ($q_\parallel$) modes, respectively. The first contribution to the total dielectric function is due to uv

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electronic resonances of the bound electrons, the second is due to the conduction-band electrons, and the last is due to the polar nature of the GaAs lattice. In Eq. (2), $\epsilon_0 = 13.3$ is the static dielectric constant, $\epsilon_\infty = 11.3$ is the high-frequency dielectric constant, $\omega_{TO} = 5.04 \times 10^{13}$ sec^{-1} is the transverse optical-phonon frequency in GaAs, $\gamma_{T,L}^{-1}$ is the lifetime of the transverse and longitudinal optical phonons, respectively, due to multiphonon processes, and $\sigma_{T,L}$ is the transverse and longitudinal conductivity, respectively.

The conditions on the dielectric function for the longitudinal modes and transverse modes obtained from Maxwell's equations[10] are

$$\epsilon_L(\omega) = 0 \tag{3}$$

and

$$\epsilon_T(\omega) = c^2 q^2 / \omega^2, \tag{4}$$

respectively. The longitudinal and transverse conductivity as given by Tsu[6] is

$$4\pi\sigma_L / i\omega \simeq (\omega_p^2/\omega^3)(\omega - i\nu) \tag{5}$$

and

$$4\pi\sigma_T / i\omega \simeq (\omega_p^2/\omega)(\omega - i\nu)/(\omega^2 + \nu^2), \tag{6}$$

respectively. In these equations, $1/\nu$ is the collision time of the electron with thermal phonons and impurities; $\omega_p^2 = 4\pi N e^2/m^*$, where N is the density of conduction electrons, $m^* = 0.07 m_e$ is their effective mass, and e is the electronic charge.

II. UNDAMPED POLARITON AND PLASMARITON

Figure 1 shows the dispersion relation ω_1 vs q_1 of the polariton and plasmariton, calculated from Eqs. (2) and (4) for various electron concentrations, taking the damping parameters γ_T and ν as zero. Curve 1 shows the well-known coupling[12] of the photon and TO phonon in GaAs, commonly called the polariton mode. The dash-dot curves are the uncoupled photon and TO phonon dispersion curves. These would be the stationary states of the hamiltonian if the modes were independent. However, the transverse field of the photon couples strongly with the TO phonon. The maximum coupling occurs at the intersection point. The transverse mode splits into two modes—a high-frequency mode (ω_+) and a low-frequency mode (ω_-). The stationary states of the low-frequency mode are photon-like at low q and phonon-like at large q. The stationary states of the upper mode are phonon-like at small q and photon-like at large q. However, in the vicinity of the intersection point, the stationary states are neither photon- nor phonon-like. A photon of frequency ω_0 and momentum $\hbar \bar{q}_0$ in free space belongs to the pure-photon curve; on entering a material medium, the excitation may be reflected or it may excite an internal excitation (on the polariton curve) at frequency ω_0 and momentum $\hbar \bar{q}'$. If the medium is perfect, on passing out of the medium the polariton will either be reflected or create an excitation (on the photon curve) with a frequency ω_0 and momentum $\hbar \bar{q}_0$.

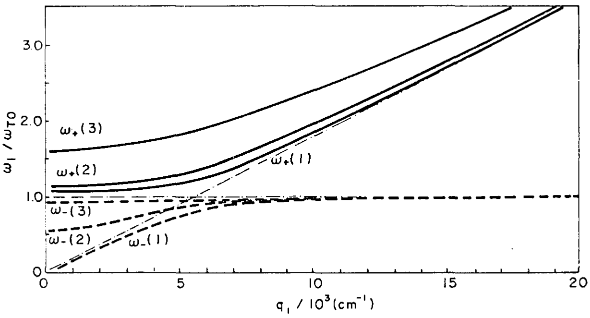

FIG. 1. Dispersion relation for GaAs for the polariton and the plasmariton. The constants used are $\epsilon_0 = 13.3$, $\epsilon_\infty = 11.3$, $\omega_{TO} = 5.04 \times 10^{12}$ sec^{-1}, and $\omega_{LO} = 5.47 \times 10^{13}$ sec^{-1}, where (1) $\omega_p/\omega_{TO} = 0$; (2) $\omega_p/\omega_{TO} = 2$ ($N = 2.2 \times 10^{17}$ cm^{-3}); (3) $\omega_p/\omega_{TO} = 5$ ($N = 1.38 \times 10^{18}$ cm^{-3}); and $\gamma_T = \nu = 0$.

Curves 2 and 3 of Fig. 1 show the coupling between the dressed photon and TO phonon—the plasmariton. The effect of free electrons on the coupling is readily noticed. The upper mode is phonon-like at low q and photon-like at large q, whereas the lower mode is phonon-like at large q and dressed-photon-like at low q. The coupling between the dressed photon and TO phonon decreases as ω_p increases, as shown in curve 2 (strong coupling) and curve 3 (weak coupling). This trend continues until the two modes uncouple at $\omega_p \gg \omega_{TO}$, which is partly shown in curve 3, where the lower mode is almost completely phonon-like and the upper mode is a dressed photon at small q and photon-like at large q. Essentially, in the weak-coupling regime, the electron field of the photon is completely screened out by the electrons, and no coupling occurs between it and TO phonons. This variation of coupling with concentration has been recently detected by small-angle Raman scattering.[4]

III. TEMPORAL DAMPED POLARITON AND PLASMARITON

The dispersion curves of a damped polariton and plasmariton have been evaluated assuming a temporal damped wave. The analysis consists of mapping from Eq. (2) the real q axis into the complex ω plane (q_1, $\omega_1 + i\omega_2$). A fifth-order complex polynomial in ω results and is solved by an iterative method.[14] Only two roots of ω for a given q turn out to be physically admissible ($\omega_1 \equiv$ positive). The real part of the complex frequency is the frequency of the coupled excitation mode, whereas the imaginary part governs the excitation's lifetime. In a Raman-scattering experiment, the real part of the frequency locates the Raman-scattered line center, and the imaginary part is a measure of the line's half-width at half-maximum.[7]

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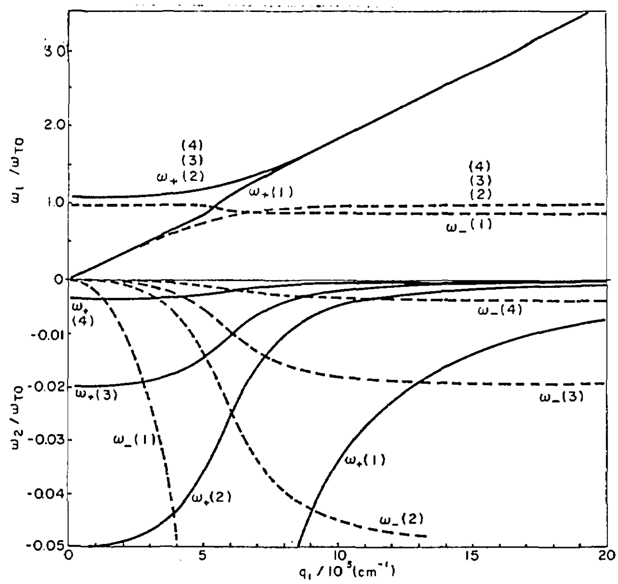

FIG. 2. Effect of the TO-phonon lifetime on the temporal damped-dispersion relation for GaAs for the polariton. The constants used are (1) $\gamma_T/\omega_{TO}=1$; (2) $\gamma_T/\omega_{TO}=10^{-1}$; (3) $\gamma_T/\omega_{TO}=4\times10^{-2}$; (4) $\gamma_T/\omega_{TO}=7\times10^{-3}$, where $\omega_p=\nu=0$.

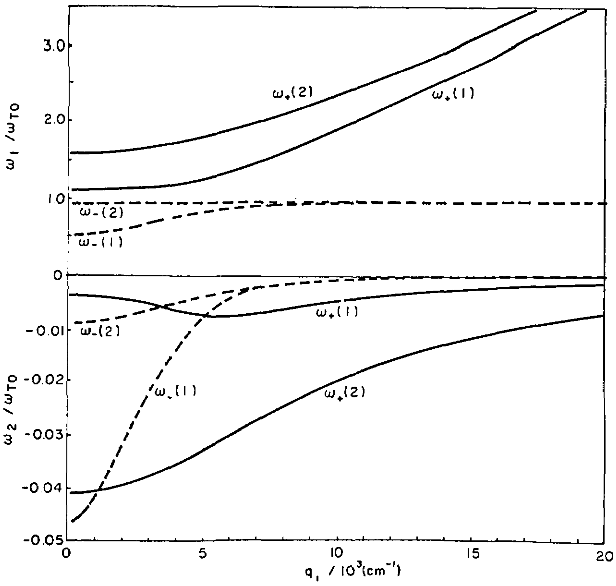

FIG. 4. Effect of the electron and phonon lifetime on the temporal damped-dispersion relation for GaAs for the plasmaritons (1) $\omega_p/\omega_{TO}=2$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T/\omega_{TO}=4\times10^{-2}$ and (2) $\omega_p/\omega_{TO}=5$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T/\omega_{TO}=4\times10^{-2}$.

Figure 2 shows the damped-polariton dispersion relation ω_1 and ω_2 vs q_1 for GaAs at different TO phonon lifetimes and infinite electron-collision times. Typical values[15] for γ_T/ω_{TO} at $q\simeq5\times10^3$ cm^{-1} in III–V semiconductors deduced from infrared-reflectivity measurements are 7×10^{-3} at 4 K and 4×10^{-2} at 300 K. There is essentially no effect on the shape of the real ω_1 vs q_1 dispersion curves due to typical TO-phonon lifetimes. On the other hand, the effect on the ω_2 vs q_1 dispersion curve is more pronounced. This variation in lifetime could easily be detected by measuring the line width by small- and large-angle Raman-scattering experiment. For a damping parameter γ/ω_{TO} of 4×10^{-2} for the ω_- mode, the line width at $q\simeq6\times10^3$ cm^{-1} is $\sim$3 cm^{-1} and at $q\simeq10^5$ cm^{-1} is $\sim$6 cm^{-1}.

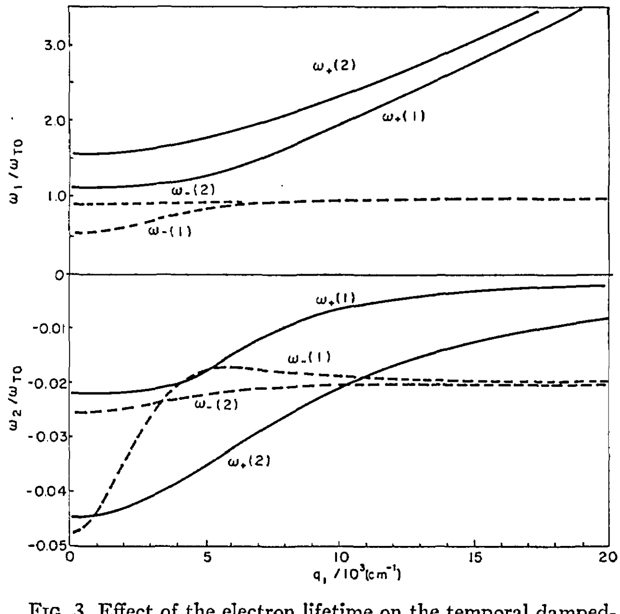

FIG. 3. Effect of the electron lifetime on the temporal damped-dispersion relation for GaAs for the plasmariton. The constants used are (1) $\omega_p/\omega_{TO}=2$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T=0$; (2) $\omega_p/\omega_{TO}=5$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T=0$.

Figure 3 shows the effect of a finite collision time of 2×10^{-13} sec (calculated from an assumed mobility of 5400 cm^2/V sec) on the dispersion curve for an electron concentration in the strong (curve 1) and weak (curve 2) coupling regimes. The TO-phonon lifetime is assumed infinite. The shape of the ω_1 vs q_1 part of the dispersion curve is not affected by the damping. The ω_- branch as shown by ω_2 vs q_1 is heavily damped at small q where the excitation mode is a dressed photon-like and slightly damped at large q where it is TO phonon-like. The upper branch of curve 1 is damped less than curve 2 at small q and large q because it is phonon-like at small q and photon-like at large q. The damping of upper branch of curve 2 decreases as q increases since it only slightly coupled to the TO phonon.

Figure 4 shows the temporal damped relation of the transverse modes including both electron and TO phonon damping. An electron-collision time of 2×10^{-13} sec ($\nu/\omega_{TO}=0.1$) and TO-phonon lifetime of 5×10^{-13} sec ($\gamma_T/\omega_{TO}=4\times10^{-2}$) were used for both the strong (curve 1) and weak (curve 2) coupling regimes. The shape of Reω_1 vs q_1 part of dispersion curves is not changed by the damping. The ω_2 vs q_1 dispersion curve,

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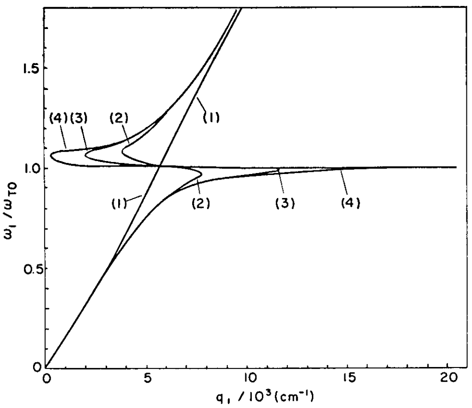

FIG. 5. Effect of the TO-phonon lifetime on the spatial damped-dispersion relation for GaAs for the polariton. The constants used are (1) $\gamma_T/\omega_{TO}=1$; (2) $\gamma_T/\omega_{TO}=10^{-1}$; (3) $\gamma_T/\omega_{TO}=4\times10^{-2}$; (4) $\gamma_T/\omega_{TO}=7\times10^{-3}$, where $\omega_p=\nu=0$.

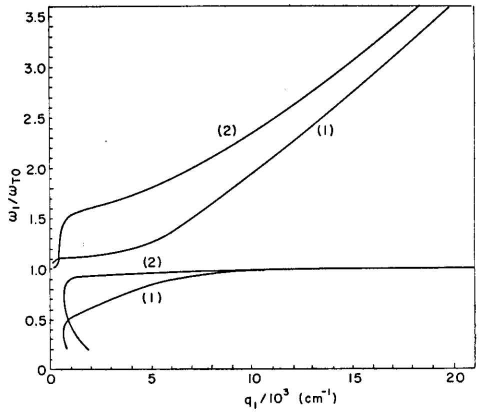

FIG. 6. Effect of the electron lifetime on the spatial damped-dispersion relation for GaAs for the plasmariton for (1) $\omega_p/\omega_{TO}=2$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T=0$ and (2) $\omega_p/\omega_{TO}=5$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T=0$.

which can be detected by Raman scattering by measuring the line width as a function of scattering angle, is essentially constant for $q_1>6\times10^3$ cm^{-1} of $\sim$6 cm^{-1}. Raman line-width measurements for small and large scattering angles have importance in stimulated polaritons and plasmariton processes because the stimulated Raman gain depends on the line width.

IV. SPATIAL DAMPED POLARITON AND PLASMARITON

The dispersion curves of the damped polariton and plasmariton have been evaluated assuming a spatial damped curve. The analysis consists of mapping from Eq. (2) the real ω_1 axis into the complex q plane.[1] Spatial damped-dispersion curves are important in the spatial absorption process and are relevant to infrared absorption and reflection work. The connection of the index-of-refraction and extinction coefficient to the propagation constant can be readily obtained, i.e., $n=(c/\omega_1)q_1$ and $K=(c/\omega_1)q_2$.

Figure 5 shows the dispersion relation ω_1 vs q_1 for GaAs at various TO-phonon lifetimes and infinite electron-collision times $1/\gamma$. The dispersion curves of Fig. 5 show that for the range of γ_T/ω_{TO} from unity to 7×10^{-3} there is no TO-phonon-like mode available with $q_1>2\times10^4$ cm^{-1}. A large-enough lattice anharmonicity could eliminate the dispersion due to the TO phonon, which is shown in curve 1 of Fig. 5.

Figure 6 shows the effect of a finite collision time of 2×10^{-13} sec ($\nu/\omega_{TO}=0.1$) on the dispersion curves (ω_1 vs q_1) for electron concentrations in the strong-(curve 1) and weak-(curve 2) coupling regimes. Here, the TO-phonon lifetime is assumed infinite. The effect of ν on the high- and low-frequency modes of the curves of Fig. 6 is most evident at small q.

Figure 7 shows the spatial damped-dispersion relation of the transverse modes, including both electron and TO-phonon damping. An electron-collision time of 2×10^{-13} sec ($\nu/\omega_{TO}=0.1$) and a TO-phonon lifetime of 5×10^{-13} sec ($\gamma_T/\omega_{TO}=4\times10^{-2}$) were used for both the strong-(curve 1) and weak-(curve 2) coupling regimes. The finite values of ν and γ_T affect primarily the transverse modes at small and large q, respectively. The dashed curve shown in Fig. 7 is a plot of ω_1/ω_{TO} vs q_2 for the parameters of the weak-coupling regime (curve). The q_2 governs the spatial absorption of the excitation mode at the frequency. Far-infrared transmittance and reflectance data obtained from measurements at an oblique angle of incidence[16,17] may show the presence of coupled modes. The finite lifetime of the excitations, as shown by the dispersion curves, would effect the absorptance and reflectance of the material.

In conclusion, we have shown that the finite lifetimes of optical phonons and/or electrons play important

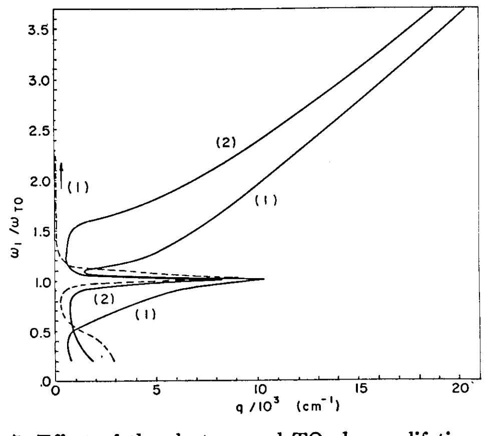

FIG. 7. Effect of the electron and TO-phonon lifetime on the spatial damped-dispersion relation for GaAs for the plasmaritons. (1) $\omega_p/\omega_{TO}=2$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T/\omega_{TO}=4\times10^{-2}$ and (2) $\omega_p/\omega_{TO}=5$; $\nu/\omega_{TO}=10^{-1}$; $\gamma_T/\omega_{TO}=4\times10^{-2}$. The solid curves are ω_1/ω_{TO} vs Req and the dash curve is ω_1/ω_{TO} vs q_2.

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roles in determining the dispersion curves of the coupling of these excitations with photons. These damped-dispersion curves have important consequences in Raman and infrared processes. The dispersion curves of other hybridizations should be affected when the finite lifetimes of the elementary excitations are included.

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