LIGHT SCATTERING FROM POLARITONS IN THE PRESENCE OF LATTICE DAMPING

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The difficulties encountered recently using the phenomenological approach to the polariton with damping are discussed.

Several authors [1,2] have recently encountered difficulties in describing damped polaritons in Raman scattering processes when they used a spatial damped dispersion curve derived from infrared reflectivity data. These difficulties include: a limiting momentum q_c that does not allow for 90° Raman scattering, and a turnabout at q_c which predicts a second polariton frequency at a given scattering angle that is not experimentally observed. The purpose of this paper is to calculate the dispersion curves of the polariton with damping. These dispersion curves can be used to determine the frequency of the mode excited in Raman and infrared processes. We show that when a temporally damped dispersion curve is used to describe the damped polariton in the Raman process, there is no limiting momentum q_c and hence no difficulties associated with it.

In the Raman process [3] when the incident and scattered frequencies lie outside the excitation absorption, the excitation wave may be described as a temporally damped wave. This requires that the momentum be a real quantity, $\hbar q_1$, and the frequency complex $\omega = \omega_1 + i\omega_2$. However, in far infrared absorption [1] or reflectivity [4,5] an excitation wave may be treated as spatially damped and is adequately described by a complex momentum $\hbar q = \hbar(q_1 + iq_2)$ and real frequency ω_1.

The use of a complex frequency $\omega = \omega_1 + i\omega_2$ is appropriate for describing polariton scattering because of the nature of the polariton in this process. In the region of small q, the predominant scattering mechanism is polar scattering in which most of the energy in the lattice mode is electromagnetic rather than mechanical. The velocity of energy propagation v_e in this region is that determined primarily by the electromagnetic wave and is $c/\epsilon_0^{1/2}$. In this region of propagating waves, the excitation wave may be described by Maxwell's equation, and the equation of motion of the lattice with retardation. The wave can then be characterized by either temporal damping (imaginary ω, i.e., ω_2 at constant q) or by spatial damping (imaginary q, i.e., q_2 at constant μ) where $q_2 = n\omega_2/c$. However, in the region of large q, where the predominant scattering mechanism is deformation potential scattering, most of the energy in the lattice mode is mechanical rather than electromagnetic. The electric interaction between the ions is then equivalent to an instantaneous Coulomb interaction with retardation neglected and the polariton in this region has a low velocity of propagation which is determined primarily by the mechanical nature of the wave. The appropriate equations which describe the polariton for large q are the mechanical equation of motion and the equation of electrostatics. The form of these equations is consistent with the concept of temporal damping and any description of the polariton in this region using spatial damping will be inadequate. In the following section, we show that a description of polariton dispersion using temporal damping is consistent with experimental observations in the Raman process while a spatial damping description of polariton leads to erroneous conclusions.

The phenomenological dielectric functions introduced by Born and Huang [5] of a system consisting of damped optical phonons in the long wavelength limit is written as

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$$\frac{c^2 q^2}{\omega^2} = \epsilon(\omega) = \epsilon_\infty - \frac{(\epsilon_0 - \epsilon_\infty)\omega_{TO}^2}{(\omega^2 - \omega_{TO}^2 + i\gamma_T \omega)} , \tag{1}$$

where ϵ_∞ is the high frequency dielectric constant, ϵ_0 is the static dielectric constant, ω_{TO} is the transverse optical phonon frequency, and γ_T is the damping constant of the transverse optical phonon. γ_T is assumed to be independent of ω and q.

The dispersion curves of a damped polariton in GaAs have been evaluated assuming a temporally damped wave. The analysis consists of mapping from eq. (1) the real q axis onto 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 [6]. 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 fixed q 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 [3]. The imaginary part is shown to be equal to the inverse of twice the lifetime through an analysis of the decay of polariton energy density.

The polariton lifetime may be calculated by considering the rate at which the polariton energy density decreases. Following Henry and Garrett [7], the energy density of the polariton, U, will damp out at a rate γ which is the inverse of the lifetime. That is,

$$\dot{U} = -\gamma U . \tag{2}$$

This damping is produced by the dissipative force of the oscillator and is given by ref. [7], eq. (C2):

$$\dot{U} = -N\mu\gamma_T \langle \dot{Q}^2 \rangle_{av} = -\frac{\mu\gamma_T \omega^2 \langle (Q')^2 \rangle_{av}}{(4\pi)^2 N e^2} = -\frac{\gamma_T \omega^2 (Q')^2 (n+\frac{1}{2})}{2\pi\omega_p^2} , \tag{3}$$

where ω_p^2 is the plasma frequency of the oscillator and is related to ω_{TO}^2 by $\omega_p^2 = \omega_{TO}^2(\epsilon_0 - \epsilon_\infty)$, μ is the reduced mass and e is the effective charge. Q' is related to the normal displacement coordinate Q by $Q' = 4\pi eNQ$ and satisfies the linear force equation for the lattice waves equation, (D1) of ref[7]:

$$\ddot{Q}' + \gamma_T \dot{Q}' + \omega_{TO}^2 Q' = \omega_p^2 E , \tag{4}$$

where E is the electric field. Combining eqs. (2) (3) and using $U = (\hbar\omega/V)(n + \frac{1}{2})$, we find

$$(Q')^2/\gamma = 2\pi\hbar\omega_p^2/\gamma_T \omega V . \tag{5}$$

Following Loudon [8] it can be shown that for the case of interest here namely $(\gamma_T \ll \omega_0, \omega_p/\epsilon_\infty)$ the polariton electric field is given by

$$|E|^2 = \{2\pi\hbar\omega/V\epsilon(\omega)\} |V_g|/|V_p| . \tag{6}$$

This is the appropriate expression for $|E'^2|$ because the polariton group velocity is equal to the polariton energy velocity for this case of interest. The polariton group velocity is obtained by applying the definition of group velocity to the dispersion relationship [eq. (1)]; we get

$$V_g = \frac{\partial\omega}{\partial q} = \frac{qc^2}{\omega} \left[\epsilon_\infty + \frac{\omega_p^2(\omega_{TO}^2 - \frac{1}{2}i\omega\gamma_T)}{(\omega_{TO}^2 - \omega^2 - i\omega\gamma_T)^2}\right]^{-1} . \tag{7}$$

The group velocity thus obtained is equal to the energy velocity obtained from eqs. (32) and (34) of ref. [9]. Using eqs. (6) and (7) and the definition of V_p, we get

$$|E|^2 = \left(\frac{2\pi\hbar\omega}{V}\right) \left| (\omega_{TO}^2 - \omega^2 - i\omega\gamma_T)^2 \times \{\epsilon_0(\omega_{TO}^2 - \omega^2 - i\omega\gamma_T)^2 + \omega_p^2(\omega_{TO}^2 - \frac{1}{2}\omega\gamma_T)\}^{-1} \right| . \tag{8}$$

With the results of eqs. (5) and (8) and using eq. (4), we finally arrive at the power attenuation constant:

$$\gamma = \gamma_T \left| \frac{\omega^2 \omega_p^2}{\epsilon_\infty(\omega_{TO}^2 - \omega^2 - i\omega\gamma_T)^2 + \omega_p^2(\omega_{TO}^2 - \frac{1}{2}i\omega\gamma_T)} \right| \tag{9}$$

This is the rate at which the total energy of the polariton decays, or equivalently it is the inverse of the polariton lifetime. Eq. (9) holds for small γ_T/ω_0 and is seen to be identical to the lifetime calculated in ref. [9], eq. (32), using energy velocity considerations with $\gamma_T/\omega_0 \to 0$. The appropriate ω to be used in eq. (9) is the frequency of the Raman scattered line center.

Fig. 1 shows the temporally damped-polariton dispersion relation ω_1 and ω_2 or $\gamma/2$ versus q_1 for GaAs at different TO photon lifetimes. Typical values of γ_T/ω_{TO} at $q \approx 5 \times 10^3$ cm^{-1} in III-V semiconductors are $\approx 7 \times 10^{-3}$ at 4°K and 4×10^{-2} at 300°K. Curves 1-4 (ω_{1-}) agree with the experimental work of Patel and Slusher [10] at small q and Mooradian and Wright [11] at large q. On the

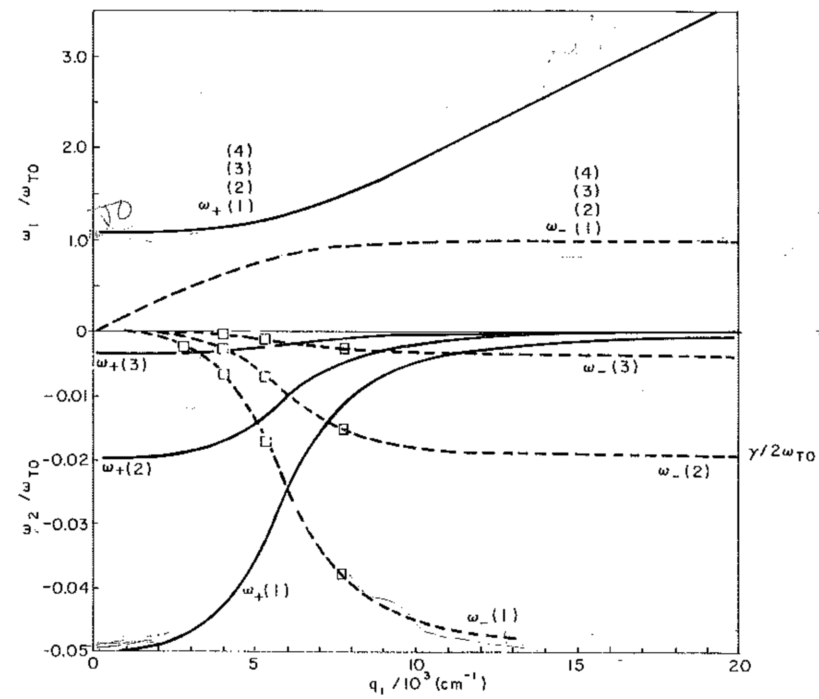

Fig. 1. Effect of the TO-phonon lifetime on the temporally damped-dispersion relation for GaAs for the polariton. The boxes in the ω_2 versus q curve represent $\gamma/2\omega_{TO}$, i.e., the lifetime calculated from energy consideration [eq. (9)] while ω_2, the imaginary part of the polariton frequency, is calculated from eq. (1) using an imaginary frequency and real q. The constants used are (1) $\gamma_T/\omega_{TO} = 10^{-1}$; (2) $\gamma_T/\omega_{TO} = 4 \times 10^{-2}$; (3) $\gamma_T/\omega_{TO} = 7 \times 10^{-3}$; (4) $\gamma_T/\omega_{TO} = 0$; $\epsilon_0 = 13.3$, $\epsilon_\infty = 11.3$, $\omega_{TO} = 5.04 \times 10^{13}$ sec^{-1}. The frequency difference between curves 1 and 3 for ω_-/ω_{TO} is $\approx 3 \times 10^{-5}$ at $q_1 = 10^3$ cm^{-1} and $\approx 10^{-3}$ at $q_1 = 10^4$ cm^{-1}.

other hand, the effect on the ω_2 versus q_1 dispersion curve is more pronounced. This plot of ω_2 versus q_1 or $\gamma/2$ versus q_1 clearly indicates the equivalence of ω_2 and $\gamma/2$. This variation in lifetime could be detected by measuring the line width by small- and large-angle fixed q Raman-scattering experiments. For a damping parameter γ_T/ω_{TO} of 4×10^{-2} for the ω_- mode, the linewidth at $q \approx 6 \times 10^3$ cm^{-1} is ≈ 3 cm^{-1} at $q \approx 10^5$ cm^{-1} is ≈ 6 cm^{-1}. Note, there are no shortcomings such as limiting momentum, turnarounds, or second frequency at a given scattering angle for the lower ω_- branch.

The spatially damped dispersion curve in GaAs has been evaluated by mapping from eq. (1) the real ω_1 axis into the complex q plane. Spatially damped curves are important in infrared absorption and reflection work. The connection between the index of refraction (n) and extinction coefficient (k) and the propagation constant q can be readily obtained, i.e., $n = (c/\omega_1)\, q_1$ and $k = (c/\omega_1)\, q_2$. Fig. 2 shows the dispersion relation ω_1 versus q_1 for GaAs at various TO-phonon

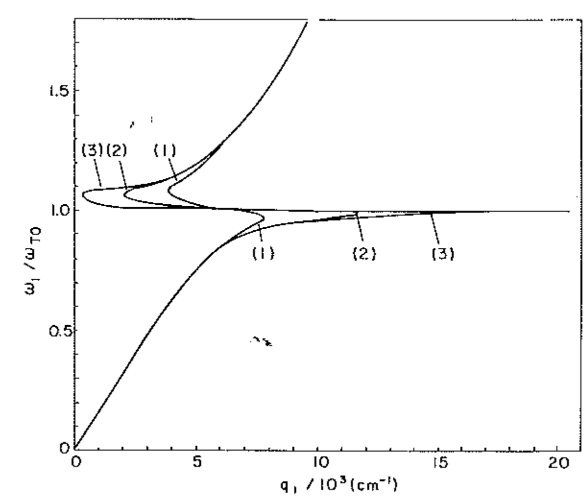

Fig. 2. Effect of the TO-phonon lifetime on the spatially damped-dispersion relation for GaAs for the polariton. The constants used are (1) $\gamma_T/\omega_{TO} = 10^{-1}$; (2) $\gamma_T/\omega_{TO} = 4 \times 10^{-2}$; (3) $\gamma_T/\omega_{TO} = 7 \times 10^{-3}$.

lifetimes. The dispersion curves of fig. 2 show various q_c for ranges of γ_T/ω_{TO} from 10^{-1} to 7×10^{-3}.

In conclusion, we have shown that the phenomenological approach of Born and Huang can be used to describe the coupled photon-transverse optical phonon with damping and the equivalent ω_2 to the inverse of twice the lifetime*.

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* A more complete theory will be published at a later date.

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