Dynamical conductivity of PbTe in the frequency range of LO-phonon–plasmon excitations

H. Burkhard
Forschungsinstitut der Deutschen Bundespost beim FTZ, P.O. Box 800, D-6100, Darmstadt, Federal Republic of Germany

G. Bauer
Experimentelle Physik IV, Universität Ulm, D-7900 Ulm-Donau, Federal Republic of Germany

A. Lopez-Otero
Institut für Physik, Experimentalphysik II, Universität Linz, A-4045, Linz-Auhof, Austria
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A strong resonantlike increase of the electron damping parameter was observed in the region of the coupled LO-phonon–plasmon excitations in n-PbTe at low temperatures. This behavior is interpreted as evidence of a generation of collective coupled plasmon-phonon excitations. The experimental results can be interpreted very well by a model calculation as reported recently by Mycielski *et al.*

The frequency dependence of the conductivity of semiconductors yields useful information on the carrier impurity as well as carrier-phonon interactions. Absorption of electromagnetic radiation is possible by individual carrier transitions as well as by generation of collective plasma oscillations (plasmons). In a recent paper, Mycielski[1] has shown that the interaction of electromagnetic radiation with plasmons in nonperfect lattices many become comparable or even higher than single-particle scattering. This is especially true for semiconductors with high-static dielectric constant ϵ_s. The volume-plasmon generation in the far-infrared region of the electromagnetic spectrum by a transverse electromagnetic wave has been reported by Mycielski *et al.*[2] for PbSe and $Pb_{1-x}Sn_xSe$, and by Gerlach *et al.*[3,4] for Bi. Bi does not exhibit infrared-active optical phonons, and therefore no influence due to the coupled plasmon-phonon modes occurs, in contrast to PbSe and $Pb_{1-x}Sn_xSe$. Experimentally, this excitation of collective modes was concluded from the observation of a peculiar frequency dependence of the dielectric function (and thus the dynamical conductivity). In PbSe and $Pb_{1-x}Sn_xSe$, e.g., an increase of the electron damping parameter was observed by about a factor of 2 slightly above the plasmon frequency.

The purpose of this paper is to demonstrate that the dissipative process considered here may lead just above the coupled LO-phonon–plasmon frequency to quite substantial changes in the damping parameter of the free carriers in PbTe, namely, by approximately two orders of magnitude.

In order to determine the frequency dependence of the electron damping parameter, we have performed reflectivity measurements in the range from 40 to 350 cm^{-1}. For carrier concentrations between 5×10^{16} and 5×10^{17} cm^{-3} the coupled LO-phonon–plasmon modes in n-PbTe are found between 130 and 230 cm^{-1}. We used epitaxial layers of PbTe-grown BaF_2 substrates[5] for this investigation which offers, owing to multiple reflections and interference effects within the PbTe film a considerable advantage as compared to data of halfspace reflectivity of bulk samples.

In Fig. 1 the reflectivity spectra of a PbTe-BaF_2 sandwich sample ($d=4.8$ μm, $n=8.10^{16}$ cm^{-3}) are shown at 5, 77, and 300 K. Due to the relatively thin PbTe layer, the observed reflectivity is not only determined by the optical constants of PbTe but also strongly influenced by those of BaF_2 which exhibits a reststrahlen region above $\omega_{TO}^{BaF_2} = 185.5$ cm^{-1}. As can already be seen in the reflectivity spectra, there is a remarkable difference between the spectra in the region near the coupled plasmon-phonon frequency ω_{LO}^* at $T=5$ and 300 K. Without damping, ω_L^+ is given approximately[6] by $\omega_{LO}^* = (\omega_p^{*2} + \omega_{LO}^2)^{1/2}$, where $\omega_p^{*2} = \omega_p^2/\epsilon_\infty = ne^2/m\epsilon_0\epsilon_\infty$, n is the carrier concentration, m is the plasma effective mass, $1/m = [\frac{1}{3}(1/m_l + 2/m_t)]$, m_l is the longitudinal, m_t is the transverse effective mass, and ϵ_∞ is the high-frequency dielectric constant. For a description of the optical properties of the PbTe film we use a simple model for the dielectric function

$$\epsilon = 1 + \chi_{\text{valence electrons}} + \chi_{\text{polar phonon}} + \chi_{\text{free carriers}}, \quad (1)$$

where $\chi_{\text{free carriers}} = i\sigma(\omega)/\epsilon_0\omega$, and $\sigma(\omega)$ is the dynamical conductivity. If a Drude expression for this last contribution is used, then

$$\chi_{\text{free carriers}} = -\omega_p^2/\omega(\omega + i\omega_\tau),$$

where $\omega_\tau \equiv 1/\tau$ denotes the carrier collision frequency (electron damping parameter). Using now

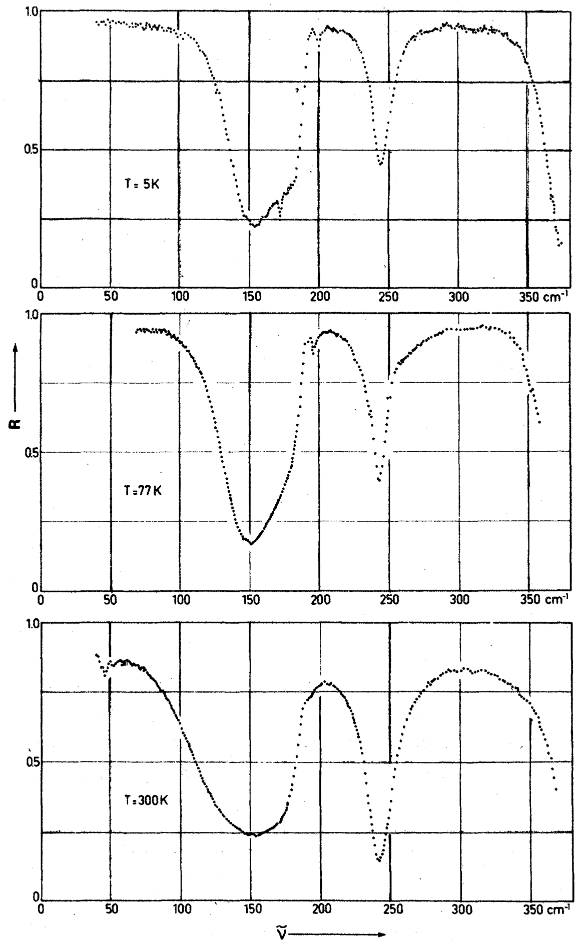

FIG. 1. Reflectivity vs frequency for PbTe-BaF_2 ($n = 8 \times 10^{16}$ cm^{-3}, $d^{PbTe} = 4.8$ μm) for three different temperatures. At frequencies below 150 cm^{-1}, the plasma edge of the PbTe film is visible, structures above 185 cm^{-1} ($\omega_{TO}^{BaF_2}$) are due to interference effects in the film and the reststrahlen region of the substrate.

for the phonon contribution the earlier determined values for ω_{TO}, $\Delta\epsilon$, and Γ,[5] we fit the reflectivity data using the full expression for R as given in Ref. 5. It turned out that for a reasonable fit of the experimentally observed data, a considerable frequency dependence of ω_τ must be assumed. The resulting $\omega_\tau(\omega)$ for the sample shown in Fig. 1 are demonstrated in Fig. 2. For all samples investigated, such a peak in $\omega_\tau(\omega)$ is observed, just above the corresponding values for ω_{LO}^* for the particular samples. In general, however, by performing this procedure one violates causality as a consequence of the linear response to the exciting electromagnetic wave. A frequency-dependent damping necessarily requires a frequency dependence of the resonance frequency of the system under consideration. According to Goetze and Wölfle,[7] the above inconsistency can be removed by considering the Kramers-Kronig transform $\xi(\omega)$ of $\omega_\tau(\omega)$.[7,8] Thus the free-carrier susceptibility contribution must be changed by including $\xi(\omega)$ given by

$$\xi(\omega) = \frac{2}{\pi}\int_0^\infty \frac{x f(x)}{x^2-\omega^2}dx\,;$$
$$f(x) = \begin{cases} -\omega_\tau(\infty)\,, & 0 \le x < \omega_{LO}^* \\ \omega_\tau(x) - \omega_\tau(\infty)\,, & \omega_{LO}^* < x < \infty \end{cases} \tag{2}$$

in the expression for $\chi_{free\ carriers}$

$$\chi_{free\ carriers} = -\omega_p^2/\omega[\omega + \xi(\omega) + i\omega_\tau(\omega)]\,, \tag{3}$$

which now leads to a dielectric function without the above outlined deficiency. Thus it is evident that $\omega_\tau(\omega)$ as presented in Fig. 2 has only a qualitative meaning because it is derived from the reflectivity spectra only in terms of a frequency dependence of the collision time, neglecting any shift of the resonance frequency.

For an interpretation of our results, we follow the calculations of Mycielski,[1] which yield absorption of electromagnetic radiation due to carrier-plasmon interaction in a polar semiconductor in the presence of lattice defects (vacancies).[9] In this case the interaction occurs via two perturbations, one connected with the electron potential energy in the presence of vacancies, the other in the field of the polarization charge density. Due to these perturbations, there is now a net power absorption due to creation of a plasmon excitation. From this power absorption the dynamical conductivity and thus $\omega_\tau(\omega)$ can be derived. According to Mycielski[1] it is given by

$$\omega_\tau(\omega) = (5^{3/2}\pi D/2^5 3^{3/2} w)\omega_{LO}^*(\hbar\omega_{LO}^*/E_F)^3\,[1-(\omega_{LO}^*/\omega)^2]^{1/2}\,(1-(1-\epsilon_\infty/\epsilon_s)\{1+\tfrac{9}{5}(\epsilon_\infty/\epsilon_s)\,[(\omega/\omega_{LO}^*)^2-1]^{-1}\}^{-1})^2\,, \tag{4}$$

when a degenerate plasma is considered. w is the number of equivalent valleys (4 in PbTe), E_F is the Fermi energy, ϵ_s is the static dielectric constant, $D = \sum_{l=1}^{s} Z_l^2(N_l/N_e)$, where s is the number of types of defects, $Z_l e$ and N_l are the charge and concentration of the l-type defect.[1]

In Mycielski's paper[1] the formalism leading to $\omega_\tau(\omega)$ as given by Eq. (4) was developed for the

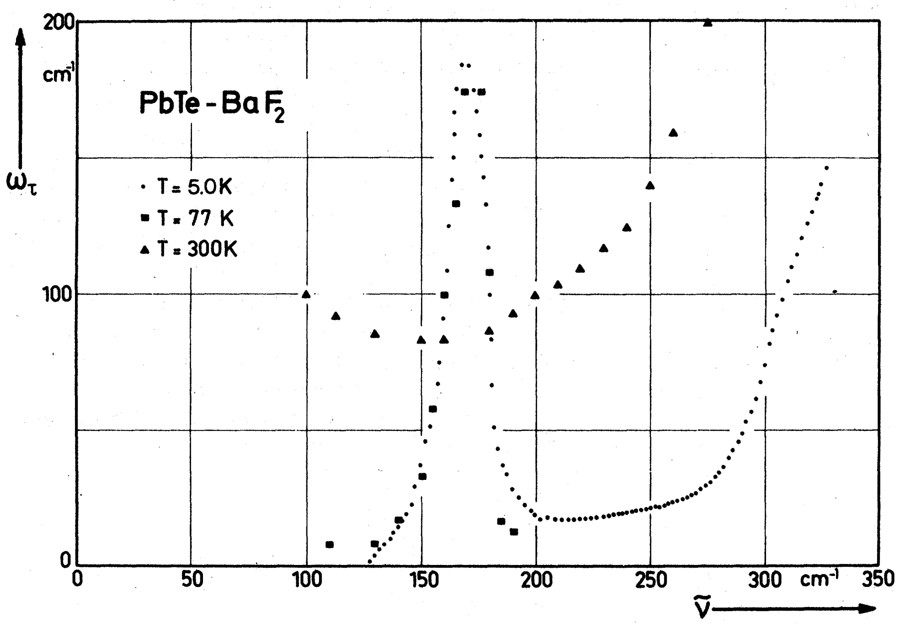

FIG. 2. Electron damping parameter ω_τ as a function of frequency for three temperatures as obtained from a fit to the reflectivity data shown in Fig. 1.

case of plasma frequency being much higher than the LO-phonon frequency to decouple the plasmon modes from the LO-phonon modes ($\omega_{LO}^* \simeq \omega_p^* > \omega_{LO}$), thereby using a simpler model for the plasma. In addition, Eq. (4) has been derived for an isotropic effective mass. Despite these limitations which are not fulfilled in our experiments with PbTe ($\omega_p^* \simeq \omega_{LO}$, four conduction-band minima located at the L point), we have applied Mycielski's theory for a description of $\omega_\tau(\omega)$ in our case. The main features, namely, the strong increase of ω_τ near ω_{LO}^* and its decrease towards higher frequencies,

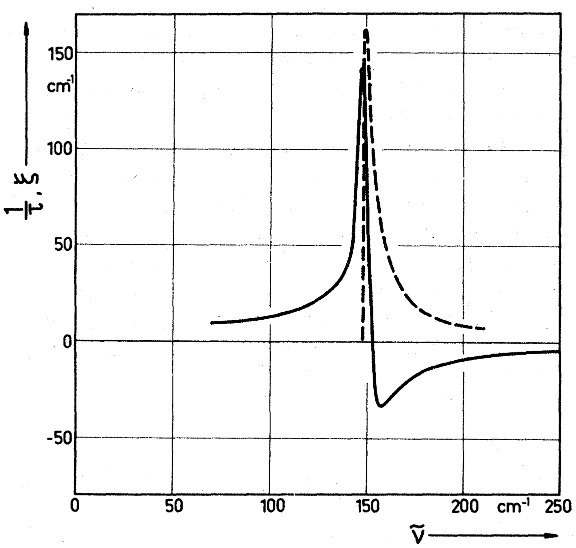

FIG. 3. Electron damping parameter [$\omega_\tau \equiv 1/\tau(\omega)$] calculated according to Eq. (4) as a function of frequency (---) for parameters of the PbTe-BaF$_2$ sample. ($T = 5$ K); (——): Kramers-Kronig transform $\xi(\omega)$ of $\omega_\tau(\omega)$, Eq. (2).

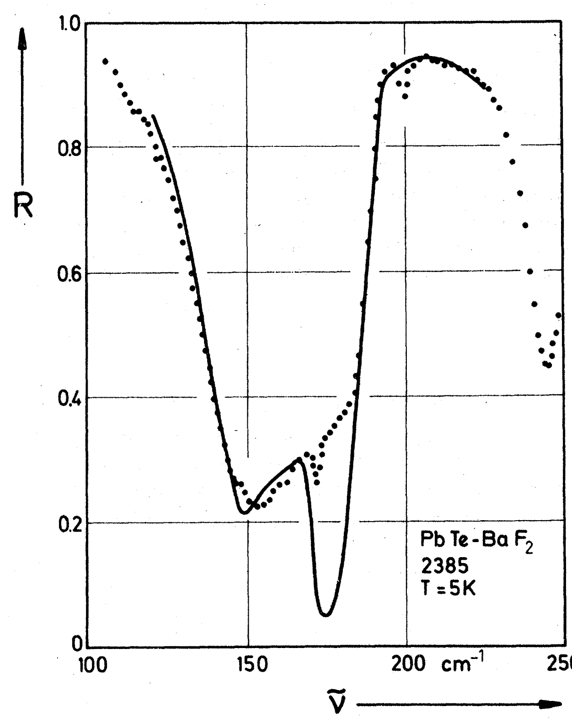

FIG. 4. Experimental (•) and calculated reflectivity vs frequency using for the free-carrier susceptibility Eq. (3) with data for $\xi(\omega)$ and $\omega_\tau(\omega)$ according to Fig. 3.

TABLE I. Optical-phonon parameters of PbTe and BaF_2.

$T=5$ K	ω_{TO} (cm^{-1})	$\epsilon_s-\epsilon_\infty$	ϵ_∞	Γ (phonon damping) (cm^{-1})
PbTe	17.5	1300	33	0.2
BaF_2	185.5	5.04	2.16	3.0

will also be present in a more refined theory, taking into account the effect of LO-phonon–plasmon coupling in the polarization charge density as well as the anisotropic band structure.[10]

The high static dielectric constant, which increases in PbTe with decreasing temperature, greatly suppresses the individual carrier-defect interactions. For ionized impurity scattering, ω_τ is proportional to ϵ_s^{-2}. Therefore PbTe ($\epsilon_s \simeq 1350$ for $T=5$ K) is a very suitable material to exhibit this interaction with a collective plasmon excitation not too much obscured by single-carrier excitations. At high temperatures the peak in $\omega_\tau(\omega)$ will disappear due to different screening ($\epsilon_s \simeq 385$) and smearing out. Therefore the temperature dependence of the damping process as shown in Fig. 2 is qualitatively correct.

In Fig. 3 $\omega_\tau(\omega)$ is calculated according to Eq. (4) for the parameters of the sample shown in Fig. 1. Its Kramers-Kronig transform $\xi(\omega)$ is also shown as a function of wave number. Using now these frequency-dependent expressions in Eq. (3) for the free-carrier susceptibility, and by considering the polar-phonon contribution, we calculated the frequency-dependent dielectric function for this material. From these data the reflectivity of the sandwich PbTe-BaF_2 was calculated using the values as tabulated in Table I. The results are shown together with the experimental data in Fig. 4. We have observed this peculiar behavior near ω_{LO}^* in the frequency dependence of the dielectric function PbTe at low temperatures also for samples with higher carrier concentrations ($\omega_p \gg \omega_{LO}$).

In a small frequency range around ω_{LO}^* our experimental data are very well represented by Mycielski's theory (where we have no adjustable parameter, see Fig. 4). For higher frequencies, where ω_τ due to the plasmon excitation process is already small (see Fig. 3), evidently other scattering mechanisms, caused by *single carrier* excitations due to interaction of the electromagnetic wave with defects or optical phonons[1,9] determine the frequency dependence of the free-carrier susceptibility (see Fig. 2). Thus the calculated reflectivity, based on Mycielski's expressions alone, deviates at higher frequencies from the measured values.

We have demonstrated that a plasmon-phonon defect process in a polar semiconductor may lead to a power dissipation process. The magnitude of it exceeds in a small frequency range even the contribution determined by individual carrier excitations.

ACKNOWLEDGMENT

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