A CRITICAL POINT ANALYSIS OF TWO PHONON STRUCTURE IN THE FAR INFRARED DIELECTRIC RESPONSE OF GaAs

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ABSTRACT

The technique of asymmetric Fourier transform spectroscopy has been used to determine the complex relative permittivities of undoped GaAs at frequencies near the transverse optic mode. It is demonstrated that the anharmonic damping function calculated from these results is sensitive to two phonon processes excited by the anharmonic decay of the $q \approx 0$ transverse optic phonon. Critical-point phonon frequencies are assigned from the structure revealed in damping function.

Key words: far infrared, dielectric function, anharmonic damping function, critical-point.

INTRODUCTION

Following the initial development (1,2) of asymmetric Fourier transform spectroscopy, also known as dispersive Fourier transform spectroscopy (DFTS), it has been established that suitable amplitude and phase spectra for solids, liquids, and gases can be determined by using variety of experimental techniques(3). In all these techniques the specimen is placed in one of the arms of a two beam Michelson interferometer rather than in front of the detector as for conventional power spectroscopy.

The infrared lattice absorption arising from multi-
phonon processes has been studied extensively in the crys-
tals of many III-V and II-VI compound semiconductors(4). In
case of binary semiconductors, in which the q=0 transverse
optic (TO) mode is infrared active and responsible for the
intense characteristic lattice absorption, there is an
additional possibility that multiphonon processes can be
excited indirectly via anharmonic decay of the q=0 TO
phonon. The detailed mechanism of the interaction of the
electromagnetic radiation with phonons in polar crystals has
been discussed by Klienman(5) and Cowley(6).

In the present work the technique of dispersive reflec-
tion spectroscopy has been used to determine the optical and
dielectric functions of GaAs in the far infrared in the
vicinity of the restrahl band, where the crystal is highly
absorbing. The measured values of dielectric response func-
tions are then used to calculate the anharmonic frequency
dependent damping functions of the q=0 TO mode. It appears
that no such measurements on GaAs have been reported before
using the technique of DFTS. The observed structure in the
spectrum of $\Gamma(oj,\nu)$ is usually dominated by the contribution
from two phonon decay processes at the critical points.
In many cases (7-9) excellent agreement has been obtained
between experimental and theoretical results. Here we
present an analysis of the features observed in the spectrum
of $\Gamma(oj,\nu)$ and compare them with the data published in the
literature.

EXPERIMENTAL METHODS

To study the far infrared optical properties of GaAs at
300K, dispersive reflection measurements have been made in
the spectral range of 200-350 cm^{-1} on an undoped GaAs sample
1mm thick and 30mm in diameter. These measurements were
performed at a resolution of 4cm^{-1} using the instrument
described by Russell and Bell(10). The average standard
deviation in the measured amplitude and phase spectra is of
the order of 0.5% over most of the frequency range.

Away from the reststrahlen band the measured phase is
dominated by systematic errors(3). At frequencies below the
transverse optic phonon frequency as well as above the lon-
gitudinal optic phonon frequency, the absolute value of
phase cannot be determined satisfactorily by reflection
DFTS, because it is very close to the phase of the reference
mirror.

RESULTS AND DISCUSSIONS

The values of amplitude and phase reflection spectra
measured were used to determine the optical constants of
GaAs at 300K. Refractive index $N(\nu)$ and extinction coeffi-
cient $K(\nu)$ are given in Figure 1. The real and imaginary
part of the dielectric functions ($\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ respec-
tively) obtained by using the data of Fig. 1 are shown in
Fig. 2.

FIG.1   FREQUENCY (CM-1)

FIG.2   FREQUENCY (CM-1)
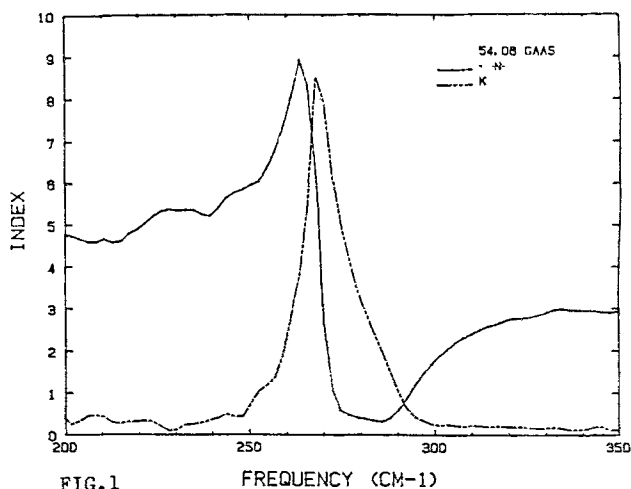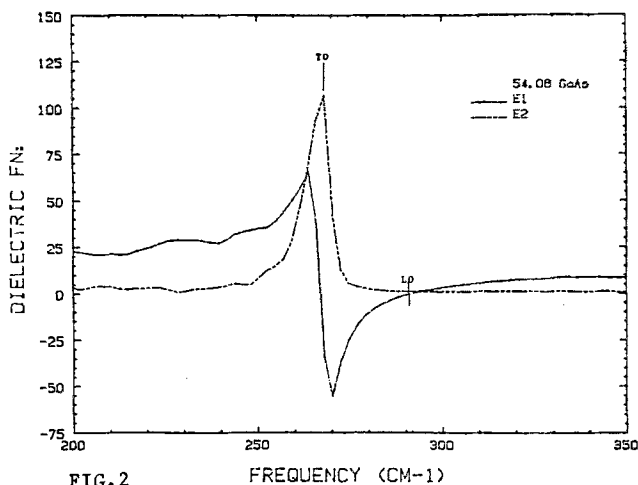
Fig. 1 (upper) and 2 (lower) presents respectively the
measured complex refractive index and real and imaginary
parts of dielectric function.

These quantities are defined by

$$\hat{\varepsilon}(\nu)=\varepsilon_1(\nu)+i\varepsilon_2(\nu)=[N(\nu)+iK(\nu)]^2 \qquad (1)$$

We obtained the frequencies of the transverse optic
mode ν_{TO} and the longitudinal optic mode ν_{LO} from the
positions of the maximum value of $\varepsilon_2(\nu)$ and zero
of $\varepsilon_1(\nu)$ the spectra shown in Fig. 2. These values are in
good agreement with those obtained by neutron scattering as
shown in Table I. We also verified the measured value
of ν_{TO} and ν_{LO} using Lyddayne-Sachs-Teller relation

$$\nu_{LO}/\nu_{TO}=\left(\varepsilon_0/\varepsilon_\infty\right)^{1/2} \qquad (1a)$$

A comparison can be seen in Table I below.

TABLE I

Comparison of long wavelength and optical frequencies ob-
tained by different methods.

Method	Room Temperature		
	ν_{TO} cm^{-1}	ν_{LO} cm^{-1}	LST ratio
Neutron Scattering(12)	267 ±2.6	285 ±6.6	1.139
Raman effect(14)	268.38±0.3	291.69±0.3	1.181
Transmission(15)	267.99±0.5	290.80±0.5	1.177
Reflectance (16)	267.50±0.5	291.44±0.5	1.187
DFTS (This work)	268.0	292.0	1.187

Although the characteristic resonant form of the dielectric
response of the q=0 TO mode in GaAs could be reproduced with
a model of a classical simple harmonic oscillator with a
frequency independent damping constant, this model fails to
account for the structure in the measured spectra because it
neglects the interaction between the normal modes which
accompany the decay of the TO phonon. If nonlinear contri-
bution to the dipole moment are neglected it follows from
the work of Cowley(6) that the complex frequency dependent
dielectric response can be written in the form

$$\hat{\varepsilon}(\nu) = \varepsilon(\infty) + \frac{\nu_{oj}^2 \left[\varepsilon(0) - \varepsilon(\infty)\right]^2}{\nu_{oj}^2 - \nu^2 + 2\nu_{oj}\left[\Delta(oj,\nu) - i\Gamma(oj,\nu)\right]} \qquad (2)$$

where ν_{oj} is the harmonic frequency of the TO phonon at wave
vector q=0, $\varepsilon(0)$ and $\varepsilon(\infty)$ are static and limiting high
frequency values of the dielectric constant, and $\Delta(oj,\nu)$
and $\Gamma(oj,\nu)$ are the real and imaginary parts of the irre-
ducible self energy of the TO phonons. It follows from Eq.
(2) that the frequency dependent damping function can be
written as

$$\Gamma(oj,\nu) = \frac{\nu_{oj}\left[\varepsilon(0) - \varepsilon(\infty)\right]\varepsilon_2(\nu)}{2\left\{\left[\varepsilon_1(\nu) - \varepsilon(\infty)\right]^2 + \varepsilon_2^2(\nu)\right\}} \qquad (3)$$

We have used Eq. (3) to calculate $\Gamma(oj,\nu)$ from the
measured dielectric functions shown in Fig. 2, using our
experimental value of ν_{TO} = 268cm^{-1} as an approximation
of ν_{oj} and taking the values of $\varepsilon(0)$ = 12.9 and $\varepsilon(\infty)$ = 10.9
from the data of Hass et. al. (11). The resulting spectrum
for $\Gamma(oj,\nu)$ is shown in Fig. 3.
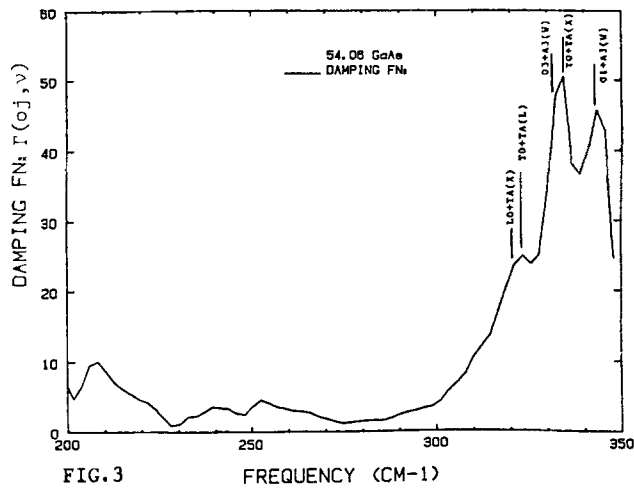
FIG.3   FREQUENCY (CM-1)

Figure 3: Measured anharmonic damping function for GaAs, assignments for the features marked are listed in Table II.

TABLE II

Observed two phonon summation bands in the anharmonic damping function for GaAs.

Observed cm^{-1}	Assignments	Calculated cm^{-1}
321	LO+TA(X)	320
323	TO+TA(L)	323
331	O3+A3(W)	329
334	TO+TA(X)	331
343	O1+A3(W)	342

It is apparent that the TO resonance is completely supressed, and that $\Gamma(oj,\nu)$ is more sensitive to weak structure in the measured spectra than are any of conventional optical (N,K,α) and dielectric functions (8), Here we have used the phonon frequencies of GaAs obtained from inelastic neutron scattering by Waugh and Dolling (12) to assign the different summation process in the measured spectrum of $\Gamma(oj,\nu)$. Considering the uncertainity of their phonon frequencies at the major symmetry points and elsewhere, all the features marked in Fig. 3 are in good agreement with the calculated frequencies, as shown in Table II. The observed features in the spectrum of $\Gamma(oj,\nu)$ have also been calculated for GaAs by Geick (13) using an approximate damping function which included nonlinear dipole moments in addition to anharmonic terms. The magnitude of damping is far below the measured values, which could be due to the approximation used in the calculations. As one can see that a number of weaker bands are present. In the absence of temperature dependent measurements we are unable to assign the difference phonon bands.

Despite the fact that many of the observed features in Fig. 3 are in a good agreement with the published data, measurements will be carried out at low temperatures and with higher resolution. Thus the technique of dispersive reflection spectroscopy gives an access to study the multiphonon processes in the reststrahel region, and should aid in establishing precise phonon frequencies at the major symmetry points.

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REFERENCES

1. Chamberlain, J.E., Gibbs, J.E., and Gebbie, H.A., Nature **198**, 874 (1963).
2. Bell, E.E., Infrared Physics **6**, 57 (1966).
3. Birch, J.I. and Parker, T.J. in Infrared and millimeter waves, Vol. 2, p. 137, Edited by K.J. Button (Academic Press, New York, 1979).

4. Spittzer, W.G., Semiconductor and semimetals, vol 3,
    Willardson, R.K. and Beer, A.C., eds. (Academic Press,
    New York, 1976).
5. Kleinman, D.A., Phys, Rev. 118, 118 (1960).
6. Cowley, R.A., Advances in Physics 12, 421 (1963).
7. Parker, T.J., Birch, J.R., and Mok, C.L., Solid State
    Commun. 36, 581 (1980).
8. Parker, T.J., Lowndes, R.P., and Mok, C.L., Infrared
    Physics 18, 565 (1978).
9. Memon, A. and Parker, T.J., International Journal of
    Millimeter Waves 2, 839 (1981).
10. Russell, E.E. and Bell, E.E., Infrared Physics 6, 75
    (1966).
11. Hass, M. and Hanves, B.W., J. Phys. Chem. Solid 23,
    1094 (1962).
12. Waugh, J.L.T. and Dolling, G., Phys. Rev. 132, 6, 2410
    (1963).
13. Geick, R., Phys. Rev. 138, 1495 (1965).
14. Mooradian, A. and Wright, D.G., Solid State Commun. 4,
    431 (1966).
15. Iwasa, S. Balslev, I., and Burstein, E. in Physics of
    Semiconductors Proc. 7th Int. Conf. (Dunod, Paris,
    1964) p. 1077.
16. Chandrashekhar, H.R. and Ramdas, A.K., Phys. Rev. B21,
    1511 (1980).