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

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Received April 2, 1984

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 $q=0$ 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 a 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 crystals 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 reflection spectroscopy has been used to determine the optical and dielectric functions of GaAs in the far infrared in the vicinity of the restrahlen band, where the crystal is highly absorbing. The measured values of dielectric response functions 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 $\Gamma(\omega,\nu)$ 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 $\Gamma(\omega,\nu)$ 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 restrahlen band the measured phase is dominated by systematic errors (3). At frequencies below the transverse optic phonon frequency as well as above the longitudinal 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 coefficient $K(\nu)$ are given in Figure 1. The real and imaginary part of the dielectric functions ($\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ respectively) obtained by using the data of Fig. 1 are shown in Fig. 2.

![Graphs showing refractive index and dielectric functions](image)

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
\[ \varepsilon(\nu) = \varepsilon_1(\nu) + i\varepsilon_2(\nu) = [N(\nu) + iK(\nu)]^2 \] (1)

We obtained the frequencies of the transverse optic mode \( \nu_{TO} \) and the longitudinal optic mode \( \nu_{LO} \) 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 \( \nu_{TO} \) and \( \nu_{LO} \) using Lyddayne-Sachs-Teller relation
\[ \frac{\nu_{LO}}{\nu_{TO}} = (\varepsilon_0/\varepsilon_\infty)^{1/2} \] (1a)

A comparison can be seen in Table I below.

<table>
<thead>
<tr>
<th>Method</th>
<th>( \nu_{TO} ) cm(^{-1} )</th>
<th>( \nu_{LO} ) cm(^{-1} )</th>
<th>LST ratio</th>
</tr>
</thead>
<tbody>
<tr>
<td>Neutron Scattering (12)</td>
<td>267 ± 2.6</td>
<td>285 ± 6.6</td>
<td>1.139</td>
</tr>
<tr>
<td>Kaman effect (14)</td>
<td>268.38 ± 0.3</td>
<td>291.69 ± 0.3</td>
<td>1.181</td>
</tr>
<tr>
<td>Transmission (15)</td>
<td>267.99 ± 0.5</td>
<td>290.80 ± 0.5</td>
<td>1.177</td>
</tr>
<tr>
<td>Reflectance (16)</td>
<td>267.50 ± 0.5</td>
<td>291.44 ± 0.5</td>
<td>1.187</td>
</tr>
<tr>
<td>DFTS (This work)</td>
<td>268.0</td>
<td>292.0</td>
<td>1.187</td>
</tr>
</tbody>
</table>
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 contributions 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

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

where \( \nu_{oj} \) 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 \( \Delta(\nu_{oj},\nu) \) and \( \Gamma(\nu_{oj},\nu) \) are the real and imaginary parts of the irreducible self energy of the TO phonons. It follows from Eq. (2) that the frequency dependent damping function can be written as

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

We have used Eq. (3) to calculate \( \Gamma(\nu_{oj},\nu) \) from the measured dielectric functions shown in Fig. 2, using our experimental value of \( \nu_{TO} = 268 \text{ cm}^{-1} \) as an approximation of \( \nu_{oj} \) 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 \( \Gamma(\nu_{oj},\nu) \) is shown in Fig. 3.
Figure 3: Measured anharmonic damping function for GaAs, assignments for the features marked are listed in Table II.

**TABLE II**

<table>
<thead>
<tr>
<th>Observed cm(^{-1})</th>
<th>Assignments</th>
<th>Calculated cm(^{-1})</th>
</tr>
</thead>
<tbody>
<tr>
<td>321</td>
<td>LO+TA(X)</td>
<td>320</td>
</tr>
<tr>
<td>323</td>
<td>TO+TA(L)</td>
<td>323</td>
</tr>
<tr>
<td>331</td>
<td>0(3)+A(3)(W)</td>
<td>329</td>
</tr>
<tr>
<td>334</td>
<td>TO+TA(X)</td>
<td>331</td>
</tr>
<tr>
<td>343</td>
<td>0(1)+A(3)(W)</td>
<td>342</td>
</tr>
</tbody>
</table>
It is apparent that the TO resonance is completely suppressed, and that $\Gamma(o_j, v)$ is more sensitive to weak structure in the measured spectra than are any of conventional optical ($N, K, \alpha$) 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 $\Gamma(o_j, v)$. Considering the uncertainty 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 $\Gamma(o_j, v)$ 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 multi-phonon processes in the reststrahlen region, and should aid in establishing precise phonon frequencies at the major symmetry points.

ACKNOWLEDGEMENTS

A. Memon gratefully acknowledges the Council for International Exchange of Scholars for their award of senior Fulbright postdoctoral scholarship, and wish to thank the Department of Physics University of Florida for their hospitality. We also thank Dr. T.J. Anderson for the loan of GaAs sample.

REFERENCES