phys. stat. sol. (b) 139, K81 (1987)
Subject classification: 63.20 and 78.30; S9.15
Department of Physics, University of Florida, Gainesville[1])

Phonon Combination Bands in the Far-Infrared Spectrum of $K_{0.5}Rb_{0.5}I$ Mixed Crystals

By
A. MEMON[2]) and D.B. TANNER

Introduction Critical point analysis of phonon combination bands in the far-infrared spectra have been widely used to establish precise values for alkali-halide phonon frequencies at major symmetry points in the Brillouin zone /1 to 4/. The high resolution available in far-infrared measurements enables more accurate determination of phonon frequencies than it is possible using neutron scattering alone. This note describes such measurements on mixed crystals of alkali halides.

The determination of the anharmonic self-energy is a difficult task, both theoretically and experimentally. The theory involves thermodynamic Green functions /5/ and requires substantial computer time to evaluate. The main experimental difficulties have been to measure precisely the frequency dependent complex dielectric response function in the far-infrared region. An important technique for measuring /6/ this quantity is dispersive Fourier transform spectroscopy (DFTS), wherein the sample is placed in one arm of a Michelson interferometer and affects the amplitude and phase of the frequency spectrum of the interferometer. At the present time, DFTS has reached the point that one can accurately measure the phase, and hence determine the complex dielectric function in the far-infrared region with high resolution. In this note we present an experimental determination of the frequency-dependent anharmonic self-energy function of the zone-center transverse-optic (TO) phonon in $K_{0.5}Rb_{0.5}I$ mixed crystals /7/. The features observed in this spectrum are satisfactorily explained using neutron scattering data /8, 9/.

Experimental Far-infrared DFTS studies of mixed alkali halide crystals, $K_{0.5}Rb_{0.5}I$, have been reported recently by the authors /7/. These measurements, performed on samples at room temperature, spanned 50 to 200 cm^{-1}

1) Gainesville, Florida 32611, USA.

2) Present address: Gulf Polytechnique, University of Bahrain, Bahrain.

at a resolution of 2 cm^{-1} and with an average standard deviation of the data of about 0.15 %. The complex dielectric function was determined from the amplitude reflectivity and phase shift.

Theory The theories of weakly anharmonic phonons in simple cubic ionic materials /5, 6/ lead to a lattice contribution to the complex dielectric function, $\varepsilon(\nu) = \varepsilon_1(\nu) + \varepsilon_2(\nu)$, at frequency ν of the form

$$\varepsilon(\nu) = \frac{(\nu(oj))^2 (f(\nu))^2}{(\nu(oj))^2 + 2\,\nu(oj)\,S(oj, \nu) - \nu^2} \quad , \tag{1}$$

where $\varepsilon(\nu)$ is the frequency dependent dielectric function, $\nu(oj)$ is the harmonic frequency of the transverse optic (TO) phonon at wave vector $\vec{q} = 0$, and $f(\nu)$ is the oscillator strength. The remaining quantity in (1), $S(oj, \nu)$ is the complex self-energy function of the $\vec{q} = 0$ TO mode. We write this as

$$S(oj, \nu) = \Delta(oj, \nu) - i\Gamma(oj, \nu),$$

where $\Delta(oj, \nu)$ and $\Gamma(oj, \nu)$ are, respectively, the real and the imaginary parts of the irreducible self-energy of the TO phonon.

The oscillator strength, $f(\nu)$, is not really determined experimentally. Here, we assume that $f(\nu)$ is frequency independent and thus equal to its zero-frequency value, $f(0) = \varepsilon(0) - \varepsilon(\infty)$, where $\varepsilon(0)$ is the static dielectric constant. Thus the oscillator strength may be estimated from measurements of the static and optical dielectric constants.

It then follows from (1) that the imaginary part of the self-energy function - the damping function - can be written as

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

Results We used our measured values /7/ of $\varepsilon_1(\nu)$ and $\varepsilon_2(\nu)$ to calculate $\Gamma(oj, \nu)$ according to (2). The TO frequency was taken as $\nu(oj) = 89$ cm^{-1}, a frequency between that of KI and RbI. The values of $\varepsilon(0)$ and $\varepsilon(\infty)$ for $K_{0.5}Rb_{0.5}I$ were taken to be 4.96 and 2.76, respectively, in accord with both our measurements and those of /10/. The resulting spectrum for $\Gamma(oj, \nu)$ is shown in Fig. 1.

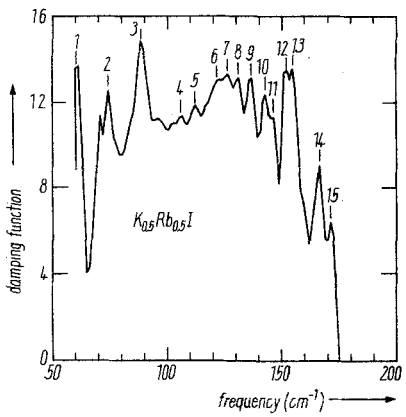

Fig. 1. Imaginary part of the phonon self-energy function for $K_{0.50}Rb_{0.50}I$ mixed crystals

The experimental uncertainty in this spectrum, an average of a few runs, is small. However, the absolute value of the damping function is very sensitive to the absolute value of the measured phase and thus should be viewed with lower confidence than the weak structure in $\Gamma(oj, \nu)$. The structure in Fig. 1 is very reproducible.

Discussion The dispersion relations in (1) contain six phonon branches. Because the TO branches in these dispersion curves are almost flat, suggesting a high phonon density of states, one can expect a number of critical points.

We now propose assignments for the observed peaks in Fig. 1, using two-phonon decay processes at the X and L symmetry points. These assignments are given in Table 1. The mode frequencies used in the critical point analysis are taken from /8/ and /9/. However, because the resolution of the neutron-scattering measurements is relatively low, we suggest some corrections to the frequencies in /8, 9/. After making these corrections, the agreement obtained between the expected and the measured frequencies is quite reasonable.

The largest changes that we find necessary occur at the X symmetry point. We have raised TA(X) by 4 cm^{-1} and reduced LA(X) by 3 cm^{-1}. These amounts are within the resolution of the neutron-scattering measurements. The revised frequencies of TA(X) = 30 cm^{-1} and LA(X) = 45 cm^{-1} enable satisfactory assignment of our measured features 1 through 7 and 9. In addition, $TO_1(X)$ and $TO_2(X)$ are reduced by 2 cm^{-1}, in order to obtain good agreement between the expected frequencies and observed features 4, 5, and 12.

Table 1 lists only two-phonon summation processes; some of the features might also be explained on the bases of difference processes, but in the absence of low temperature measurements one cannot assign them confidently.

Table 1

Assignment of observed features in two-phonon summation process (frequencies in cm^{-1})

feature	assignment	expected frequency	observed frequency
1	2TA(X)	60	61
2	TA(X) + LA(X)	75	74
3	2LA(X)	90	88
4	TO_2(X) + TA(X)	105	105
5	LO_2(X) + TA(X)	111	111
6	TO_2(X) + LA(X)	120	120
7	TO_1(X) + TA(X)	123	125
8	2LA(L)	129	131
9	TO_1(X) + LA(X)	138	136
10	TO_2(L) + LA(L)	143	143
11	TO_1(L) + TA(L)	145	147
12	$2TO_2$(X)	150	150
13	LO_2(X) + TO_2(X)	156	155
14	LO(L) + TA(L)	166	167
15	LO_2(X) + TO_1(X)	174	173

Conclusions This note shows that a critical point analysis can be performed for mixed alkali-halide crystals based on DFTS measurements. We conclude that the two-phonon process results predominantly from anharmonic decay of the $\vec{q} = 0$ TO phonon. Consequently, one can minimise the errors in the frequencies measured by neutron scattering, by performing high resolution measurements of phonon combination bands.

References

/1/ K.W. JOHNSON and E.E. BELL, Phys. Rev. 187, 1044 (1969).

/2/ J.I. BERG and E.E. BELL, Phys. Rev. B 4, 3572 (1971).

/3/ A. MEMON and T.J. PARKER, Internat. J. Infrared and Millimeter Waves 2, 849 (1981).

/4/ K. HISANO, F. PLACIDO, A.D. BRUCE, and G.D. HOLAH, J. Phys. C 5, 2511 (1972).

/5/ R.A. COWLEY, Adv. Phys. 12, 421 (1963).

/6/ K.F. PAI, A. RASTOGI, N.E. TORNBERG, T.J. PARKER, and R.P. LOWNDES, J. Opt. Soc. Amer. 67, 914 (1977).

/7/ A. MEMON and D.B. TANNER, Internat. J. Infrared and Millimeter Waves, to be published.

/8/ B. RENKER, N.M. BUTT, and N.E. MASSA, Phys. Rev. B 27, 1450 (1983).

/9/ S. ASLAM, M.M. ROHLANDSON, N.M. BEG, and Q.H. KHAN, phys. stat. sol. (b) 77, 693 (1976).

/10/ J.H. FERTEL and C.H. PERRY, Phys. Rev. 184, 874 (1969).

(Received October 1, 1986)