THE INFLUENCE OF TEMPERATURE ON PHONON PROCESSES IN $CdGa_2Se_4$

I.A. MAMEDOVA[1], A.A. MAKSIMOV[2], I.I. TARTAKOVSKII[2], F.M. SALEHLI[3], N.A. ABDULLAYEV[1,4]

[1]Institute of Physics of the Ministry of Science and Education of Azerbaijan, Baku, Azerbaijan
[2] Institute of Solid State Physics, Russian Academy of Sciences, Chernogolovka, Moscow region, Russia
[3] Department of Physics, Istanbul Technical University, Istanbul, Turkey.
[4]Baku State University, Baku, Azerbaijan
irada_mamedova@yahoo.com

The temperature dependences of phonon frequencies (104 cm^{-1}, 139 cm^{-1}, and 187 cm^{-1}) in $CdGa_2Se_4$ were studied. Raman scattering spectra in $CdGa_2Se_4$ crystals were measured with an accuracy of 0.1 cm^{-1} under excitation with a He-Ne laser with an emission wavelength of 632.817 nm. It was theoretically established that the change in phonon frequencies with temperature is described by exponential three-phonon interaction, and the experimental data well agree with calculations in the range of 5-175 K; above 180 K, linear dependence was observed.

Keywords: Raman scattering, phonon frequency, $CdGa_2Se_4$.
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1. INTRODUCTION

$CdGa_2Se_4$ belongs to the family of compounds $A^2B^3{}_2C^6{}_4$ (A–Zn, Cd; B–In, Ga; C-S, Se, Te), crystallizing in the space group $S^2{}_4$, are crystallochemical analogues of compounds crystallizing in the sphalerite and chalcopyrite structure. These compounds are characterized by optical anisotropy, birefringence, significant values of nonlinear susceptibility coefficients, high photosensitivity, and bright luminescence. The presence of these properties in combination with significant band gap values puts these compounds among promising materials for use in semiconducting nonlinear converters [1, 2]. In [3], a first-principles calculation of the phonon density of states and phonon dispersion at highly symmetric points and lines of the Brillouin zone of $CdGa_2Se_4$ was carried out, and in [4] the electronic spectra of $CdGa_2Se_4$ were studied theoretically and experimentally.

2. EXPERIMENTAL PROCEDURE

In the present work, we carried out temperature studies of Raman scattering spectra of $CdGa_2Se_4$ crystals at various temperatures from liquid helium to room temperature. Since the temperature intervals were small and the mode frequency shifts with temperature were small, it was necessary to increase the accuracy of mode frequency determination to 0.1 cm^{-1}. Raman scattering spectra in $CdGa_2Se_4$ crystals were studied under excitation by a He-Ne laser with an emission wavelength of 632.817 nm. To improve the accuracy of phonon frequency determination, light comparable to the Raman scattering intensity from a low-pressure Ne calibration lamp was sent into the optical path of the setup simultaneously with the recording of the Raman scattering spectra. Thus, the spectrum of the calibration lamp, consisting of known narrow lines and measured simultaneously with the Raman scattering spectrum under study, made it possible to significantly increase the accuracy of phonon frequency determination using appropriate mathematical processing [5, 6]. Single crystals $CdGa_2Se_4$ were obtained by the gas transport reactions method. Crystalline iodine was used as a transporter. The lattice parameters were determined a = 5.574Å, c = 10.756Å, c / a = 1.873 [3].

3. RESULTS AND DISCUSSION

The experimentally obtained dependences of the phonon frequencies of 104 cm^{-1}, 139 cm^{-1} and 187 cm^{-1} on temperature are shown in Figure 1 (dots).
As is known [7], the change in the frequencies of optical phonons with temperature is generally described by the relation:

$$\omega(T) = \omega_0 + C\left(1 + \frac{2}{e^x - 1}\right) + D\left[1 + \frac{3}{e^y - 1} + \frac{3}{(e^y - 1)^2}\right] \quad (1)$$

where, $x = \hbar\omega_0 / 2k_BT$, $y = \hbar\omega_0 / 3k_BT$. Here ω_0 is the phonon frequency at temperature T=0, the second term describes the three-phonon interaction, and the third term describes the four-phonon interaction. Since the four-phonon interaction makes a large contribution at high temperatures, for temperatures T<300 K one can restrict oneself to the expression [6, 7]:

$$\omega(T) = \omega_0 + C\left(1 + \frac{2}{e^x - 1}\right) \quad (2)$$

131, H.Javid ave, AZ-1073, Baku
Institute of Physics
E-mail: jophphysics@gmail.com

Figure 1 also shows the calculated curves (solid lines) according to relation (2) for phonons with frequencies of 104 cm^{-1} (a) (parameters ω_0=104.6 and C= - 0.16), 139 cm^{-1} (b) (parameters ω_0=140.4 and C= - 0.3), and 187 cm^{-1} (c) (parameters ω_0=189 and C= - 0.7) in $CdGa_2Se_4$.

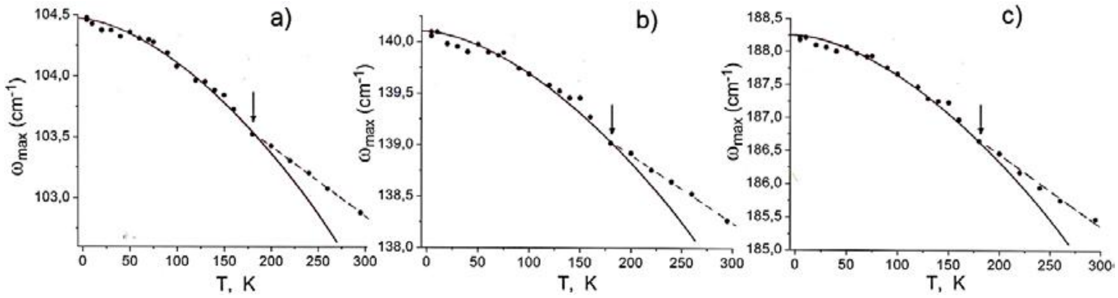

Fig. 1. Temperature dependence of phonon frequencies in $CdGa_2Se$ (dots-experiment, solid line-theory)

It is clearly seen that the calculated curves are in good agreement with the experimental data in the temperature range of 5-175 K; above 180 K, a linear decrease in phonon frequencies with increasing temperature is observed.

4. CONCLUSION

The temperature dependences of Raman scattering in $CdGa_2Se_4$ were studied over a wide temperature range of 5–300 K. It was found that the frequencies decrease with increasing temperature. Theoretical calculations, taking into account three-phonon processes, provide a good approximation of experimental data in the range of 5–175 K. Above 180 K, a linear decrease in frequencies with increasing temperature is observed.

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