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To cite this version:
D. Schmeltzer, R. Beserman. ANHARMONIC INTERACTION IN ZINC SELENIDE. Journal de Physique Colloques, 1981, 42 (C6), pp.C6-158-C6-160. <10.1051/jphyscol:1981648>. <jpa-00221584>

HAL Id: jpa-00221584
https://hal.archives-ouvertes.fr/jpa-00221584
Submitted on 1 Jan 1981

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ANHARMONIC INTERACTION IN ZINC SELENIDE

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Abstract.— Experimental evidence of anomalous temperature dependence of the double phonon Raman spectrum is brought and explained by a 4th order anharmonic coupling between two 2-phonon states.

We report experimental evidence of the fourth order anharmonic interaction in the continuum of ZnSe transverse acoustic-phonons, and its temperature dependence. The fourth order interaction produces a positive feedback which is the fourth order coupling parameter. The intensity of the response function increases with the increase of the feedback parameter.

The Raman spectrum (RS) intensity of the double-phonon band has been studied as a function of temperature. At room temperature this spectrum is mainly composed of the 2 TA peak from the X point of the Brilloin zone with some contribution from the <110> direction and from subtractive phonons.

Fig. 1 shows the $\Gamma_1+\Gamma_{12}$ representation of the ZnSe R.S, Excited by the 6471Å line, at 80K, 300K and 480K. As the temperature increases, the double phonon intensity increases with respect to the TO and LO modes. When the temperature varies from 80K to 660K, the variation of the Bose-Einstein factor ratio between TO and 2 TA should be 10 while experimentally we found a factor 2100. The width, shape and frequency of the double phonon peak do not vary appreciatively with temperature.

These results are the same when the 6764Å kryston laser line is used, and no anomalous behaviour is observed in the double phonon spectrum of ZnS.

These experimental facts show that at high enough temperature an instable double-phonon state exists which disappears at low temperature.

In order to analyze the behavior of the doublephonon in the acoustic range we assume a fourth order anharmonic interaction.

$$H = \sum_{k} \hbar \omega_{k} \left( b_{k}^{*} b_{k} + \frac{1}{2} \right) + \frac{\hbar^{4}}{4!} \int d^{3}x \phi^{4}(x)$$
The Raman Intensity is proportional to the time ordered green's function
\[ I^{(2)}(\omega) = -\left[ n(\omega) + 1 \right] 2 g_{ik} \omega^2 \frac{\hbar}{2kT} \text{Im} D_2^{\text{r}}(q=0, \omega) \]

In the mean field approximation
\[ \text{Im} D_2^{\text{r}}(q=0, \omega) \approx \frac{2 \text{Im} F(\omega)}{\left[ 1 - \frac{g_{ik}}{2} \text{Re} F(\omega) \right]^2 + \left( \frac{g_{ik}}{2} \text{Im} F(\omega) \right)^2} \]

\( \text{Im} F(\omega) \) is proportional to the double phonon density of states \( \rho(\omega) \). At high enough temperature \( T = \frac{\Delta}{2} \text{Re} F(\omega) \) gives the resonant condition.

For \( \frac{\hbar T}{2\omega_{ac}} \ll 1 \), the singularity of the density of states produces a large change in the second order Raman intensity
\[
\frac{I^{(2)}(T)}{I^{(2)}(T_0)} \propto 1 - \frac{T - T_0}{T} \sqrt{\frac{\text{Im} C(\epsilon)}{C(\epsilon)}} \quad \epsilon = \frac{\omega - 2\omega_{ac}}{2\omega_{ac}}
\]

When \( T > T_0 \) for high temperatures
\[
\frac{I^{(2)}(T)}{I^{(2)}(T_0)} \propto \frac{T}{T^* - T} \quad \text{with} \quad T^* = \frac{T_0}{\sqrt{2}} \frac{\sqrt{\epsilon}}{\sqrt{\sqrt{\epsilon} T_0}}
\]

We deduce
\[
F(T) = \left[ \frac{I^{(2)}(T)/T}{I^{(2)}(T_0)/T_0} \right]^{-1} \propto T^* - T
\]

In fig. 2 we have plotted \( F(T) \) as a function of temperature. The graph shows that for \( T \approx 600 \text{ K} \) the linear equation given for \( F(T) \) is valid and fits our experimental results well, with \( T^* = 600 \text{ K} \).
Fig. 1: ZnSe Raman Intensity at different temperatures, $\lambda =6471\text{Å}$.

Fig. 2: $F(T)$ plotted as a function of temperature; this equation has been normalized to 1 for $T=300\text{K}$, the experimental points are indicated by circles.