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Phonon dispersion properties of europium oxide (EuO)

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Abstract
The phonon dispersion properties of Europium Chalcogenides have been analysed by a lattice dynamical model which includes the effect of three-body interaction (TBI) in the framework of second neighbour rigid shell model (SNTRSM) and second neighbor rigid ion model (SNTRIM). The significance of these two approaches thus obtained, have been applied to study the phonon dispersion curves (PDCs) of Europium Oxide (EuO) along the principal symmetry directions. It is found from our prediction that SNTRSM explains well the phonon anomalies in the experimental PDCs of EuO as compared to SNTRIM and other models.

Keywords: phonon dispersion curves, rigid shell model, rigid ion model, DEBYE temperature

1. Introduction

The electronic structure of Europium Oxide (EuO) which is a family of Europium Chalcogenides (EuO, EuS, EuSe, EuTe) crystallize in f.c.c. NaCl structure and are also called as rare earth europium chalcogenides. Unlike other rare earth compounds europium chalcogenides generally, show non-mixed valance character. The study of lattice dynamical behavior of these chalcogenides is incomplete even today due to insufficient experimental data for phonon dispersion curves and a very few attention has been paid to it.

Although europium chalcogenides have a large application as magnetic semiconductors, yet no serious attention has paid for for. Only few information about the optical frequencies [3-5], elastic properties [2] and magnetic [29, 30] have been presented. Complete experimental data on phonon dispersion is not available for these compounds except for EuSe, for which limited information about phonon frequency has been reported by Silberstein *et al.* [1] Zeyher and Kress have applied a phenomenological model (OSM) [14] to discuss the complete phonon dispersion curves (PDCs) and combined density of states (CDS). Furthermore, Osaka *et al.* [11] have investigated the phonon frequencies only for EuSe using Breathing shell model (BSM). For better result Mischenko and Kikoin [19] have modified Zayher and Kress overlap shell model (OSM) to predict the phonon dispersions curves of EuS. These authors modified the dynamical matrix by incorporating the charge density deformation effects. But on the basis of overlap achievements, their results are for away from success because none has considered the many body interactions (the first important term is three body interactions) for these compounds. Due to the unfilled 4f shells, the radii of rare earth ion changes and therefore overlapping of the chalcogen ions, also changes. The Europium chalcogenides show deviations from the Cauchy relation $C_{12}=C_{44}$. The BSM used by Onsaka *et al.* [11] and Sakale *et al.* [25] of PDC only explains the acoustic branches well. Therefore it is evident that OSM and BSM fail to explain the optical branches of PDC of these crystals. It has been found that three body interactions explain well the optical branches and Cauchy discrepancy both simultaneously and successfully to almost all the ionic and semiconducting crystals [17]. This has motivated the present author to the basic need of two phenomenological lattice dynamical models. The aim of this paper is to test the applicability and utility of second neighbor three-body force rigid shell model (SNTRSM) and second neighbor three-body force rigid ion model (SNTRIM) for the satisfactory description of phonon dispersion relations and other phonon properties of these compounds.

2. Theory

The general formulation of present Lattice Dynamical model is given by:

1. Three-body force rigid shell model (SNTRSM)
2. Three-body force rigid ion model (SNTRIM)

The interaction system of present model thus consists of long range Coulomb and three body interactions (TBI) energies. The next term is the form of SR overlap repulsive energy extended to the next nearest neighbor ions in Europium Chalcogenides. As per the interaction system, the present model may positively be the successful attempt for the dynamical description of these materials.

The general formulation of SNTRSM can be derived from the crystal potential whose relevant expression per unit cell is given by

$TM = TM^{C} + TM^{R} + TM^{TBI}$............(1)

Where the first two terms represent, respectively, long range Coulomb and three body interactions (TBI) energies. The next term is the form of SR overlap repulsive energy extended to the next nearest neighbor ions. where first term Φ^{C} is

Coulomb interaction potential which is long-range in nature, second term Φ^R is short range overlap repulsion potential operative up to the second neighbors and third term Φ^{TBI} is three body interaction potential. The secular determinant D(q), is the (6x6) dynamical matrix which is given by:

$$D(q)=(R'+Z_mCZ_m)-(T+S_mCY_m)(S+K+Y_mCY_m)^{-1}(T^*+Y_mCZ_m) \text{-------} (2)$$

The Number of adjustable parameters has largely been reduced by considering the short range interaction to act only through the shells. This assumption leads to R=T=S. The expressions derived for elastic constants corresponding to SNTRSM have been obtained as:-

$$\frac{4r_0^4}{e^2}C_{11}=[-5.112Z_m^2+A_{12}+\frac{1}{2}(A_{11}+A_{22})+\frac{1}{2}(B_{11}+B_{22})+9.3204\xi'^2] \text{-----} \quad (3)$$

$$\frac{4r_0^4}{e^2}C_{12}=[0.226Z_m^2-B_{12}+\frac{1}{4}(A_{11}+A_{22})-\frac{5}{4}(B_{11}+B_{22})+9.3204\xi'^2] \text{------} \quad (4)$$

$$\frac{4r_0^4}{e^2}C_{44}=[2.556Z_m^2+B_{12}+\frac{1}{4}(A_{11}+A_{22})+\frac{3}{4}(B_{11}+B_{22})] \text{------} \quad (5)$$

In View of the equilibrium condition $[(d\,\Phi/dr)_0]$ We obtain.

$$B_{11}+B_{22}+B_{22}=-1.165Z_m^2 \text{------} \quad (6)$$

Where

$$Z_m^2=Z^2\left(1+\frac{12}{Z}f_0\right) \quad \text{and} \quad \xi^{12}=Zr_0f_0$$

The term f_o is a function dependent on the overlap integrals of the election wave-funtions and the subscript zero indicates the equilibrium value. By solving the secular equation along [q00] direction and subjection the short and long-range coupling coefficients to the long-wavelength limit $q\rightarrow 0$, two distinct optical vibration frequencies are obtained as:

$$(\mu\omega_L^2)_{q=0}=R_0'\frac{(Z'e)^2}{Vf_r}.\frac{8\pi}{3}\left(Z_m^2+6\xi'^2\right) \text{-----------} \quad (7)$$

$$(\mu\omega_r^2)_{q=0}=R_0'\frac{(Z'e)^2}{Vf_r}.\frac{4\pi}{3}Z_m^2 \text{---------} \quad (8)$$

Since, in these compunds, $\omega_L=\omega_r$ at Γ - point, therefore, Eqs(7) and (8) lead to the expression:

$$\frac{Z_m^2+6\xi'^2}{\xi'^2}=-\frac{f_L}{2f_r} \text{----------} \quad (9)$$

Where the abbreviations stand for

$$R_0'=R_0-e^2\left(\frac{d_1^2}{\alpha_1}+\frac{d_2^2}{\alpha_2}\right);Z'=Z_m+d_1-d_2$$

$$f_L=1+\left(\frac{\alpha_1+\alpha_2}{v}\right).\frac{8\pi}{3}\left(Z_m^2+6\xi'^2\right)$$

$$f_r=1-\left(\frac{\alpha_1+\alpha_2}{v}\right).\frac{4\pi}{3}Z_m^2$$

By solving the dynamical matrix along [0.5, 0.5,0.5] directions at L-Point modified expressions for $\omega_{Lo}(L)$, $\omega_{To}(L)$, $\omega_{LA}(L)$, and $\omega_{TA}(L)$, are as follows.

$$m_1\omega_{LA}^2(L)=R_0+\frac{e^2}{V}(2A_{11}+B_{11}-\frac{e^2d_1^2}{a_1}+\left(\frac{e^2}{V}\right)C_{1L}(Z_m+d_1)^2\left[1+\left(\frac{\alpha_1}{V}\right)C_{1L}\right]^{-1} \text{--------} \quad (10)$$

$$m_2\omega_{LO}^2(L)=R_0+\frac{e^2}{V}(2A_{22}+B_{22}-\frac{e^2d_1^2}{\alpha_1}+\left(\frac{e^2}{V}\right)C_{1L}(Z_m+d_2)^2\left[1+\left(\frac{\alpha_2}{V}\right)C_{1L}\right]^{-1} \text{--------} \quad (11)$$

$$m_2\omega_{TO}^2(L)=R_0+\frac{e^2}{2V}(2A_{22}+B_{22}-\frac{e^2d_2^2}{\alpha_2}+\left(\frac{e^2}{V}\right)C_{1T}(Z_m+d_2)^2\left[1+\left(\frac{\alpha_2}{V}\right)C_{1T}\right]^{-1} \text{----} \quad (12)$$

$$m_1\omega_{TA}^2(L)=R_0+\left(\frac{e^2}{2V}\right)(A_{11}+5B_{11}-\frac{e^2d_1^2}{\alpha_1}+\frac{e^2}{V}C_{1T}(Z_m+d_1)^2\left[1+\left(\frac{\alpha_1}{V}\right)C_{1T}\right]^{-1} \text{------} \quad (13)$$

Where

$$C_{1L}'=-\left[\left(C_{1xx}+2C_{1xy}\right)+\left(V_{1xx}+2V_{1xy}\right)Z_m^{-2}Zx_0f_0'\right]0.5,0.5,0.5$$

$$C_{1T}'=-\left[\left(C_{1xx}+C_{1xy}\right)+\left(V_{1xx}+2V_{1xy}\right)Z_m^{-2}Zr_0f_0'\right]0.5,0.5,0.5$$

Where $(C_{1xx}+C_{1xy})$ and $(V_{1xx}+V_{1xy})$ are Coulomb and three - body coupling coefficients evaluated at L-point. Polarizability is negligibly small and the negative ion polarizability of nitride ion is almost zero. Therefore, it has been considered to utilize the second neighbour three-body force rigid ion model (SNTRIM) for further calculations of phonon frequencies.

In an attempt to solve the expressions for SNTRIM, all the Eqs (1-6) will remain the same, only the difference is in the expressions from Eqs. (7-13), which can be written as follows.

$$\left(\mu\omega_L^2\right)_{q=0}=R_0+\frac{8\pi e^2}{3V}(Z_m^2+6\xi'^2) \text{-----------} \quad (14)$$

$$\left(\mu\omega_T^2\right)_{q=0}=R_0-\frac{4\pi e^2}{3V}(Z_m^2) \text{-----------} \quad (15)$$

Since, in these compounds, $\omega_L=\omega_T$ at $\Gamma-$ point, therefore, Eqs(14)and (15) lead to the expression:

$$Z_m^2=-4Zr_0f_0' \text{--------} \quad (16)$$

Again, by solving the dynamical matrix along [0, 0.5, 0.5] directions at L-point, the modified expressions for $\omega_{Lo}(L)$, $\omega_{To}(L)$, $\omega_{LA}(L)$, and $\omega_{TA}(L)$ are derived as follows

$$\omega_{LO}(L)=R_0+\frac{e^2}{v}(2A_{22}+B_{22})+\frac{e^2}{v}C_{1T}'Z_m^2 \text{-----} \quad (17)$$

$$m_2\omega_{TO}^2(L)=R_0+\frac{e^2}{v}(A_{22}+5B_{22})+\frac{e^2}{v}C_{1T}'Z_m^2 \text{---} \quad (18)$$

$$m_1\omega_{LA}^2(L) = R_0 + \frac{e^2}{v}(2A_{11}+B_{11}) + \frac{e^2}{v}C'_{1L}Z_m^2 \quad --- \quad (19)$$

$$m_1\omega_{TA}^2(L) = R_0 + \frac{e^2}{v}(A_{11}+5B_{11}) + \frac{e^2}{v}C'_{1T}Z_m^2 \quad --- \quad (20)$$

where R_0 and C'_{1T} have already been defined.

3. Computations and Results

The input data along with their relevant references and calculated model parameters from SMTRSM and SNTRIM for EuO are given in Table- 1. A comparative results on phonon dispersions curves from the two models have been shown in Figure 1. These results have also been compared with the observed data of Kress *et al.* [1] for visual comparison.
Table 1- Input data and model parameters for EuO

[$C_{ij}(in 10^{12} dyne\ \mathrm{cm}^{-2})$, ν(in THz), $r_0(in 10^{-8} cm)$ And $\alpha_i(in 10^{-24} cm^3)$]

Table 1

Input data			Model Parameters	
Properties	**Values**	**Parameters**	**SNTRSM**	**SNTRIM**
C_{11}	**19.2^a**	**$a\ Z^2$**	**0.7293**	**0.7238**
C_{12}	4.25^a	r f(r)	-0.0119	-0.0119
C_{44}	5.42^a	A	7.6342	7.564
ν (I)	13.05^b	B	□□.861	□0.8432
a^2	2.40	Y_2	-0.8448	….
a	2.07^c	Y_1	-2.0541	….
r_o	2.572^{**}	d_2	1.1721	….

a – [3] b – [1] c – [4]
* Value extrapolated from measured PDC
** Reasonable value taken from ionic radii.

4. Discussion and Conclusion

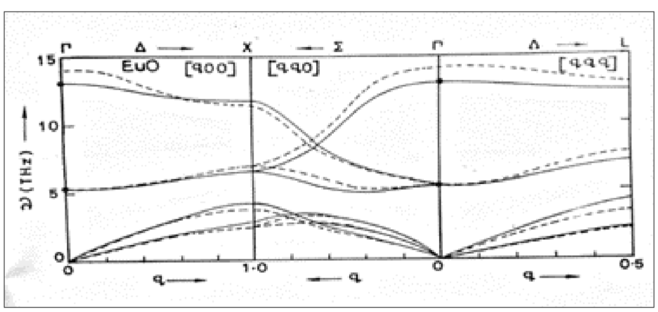

Fig 1: Phonon dispersion curves of EuO

From figure 1, it is clear that the results reported from SNTRSM for EuO are comparatively more close to the measured data on PDCs. These result are similar to the TMC but there are certain features in PDC of EuO which deserve special mention. The three body interactions have influenced both LO and TO branches much more than accoustic branches (LA and TA). Another striking feature of the present study is noteworthy from the excellent reproduction of optical and acoustic branches.
The model parameter of TRSM and TRIM have been used to calculate the phonon spectra for allowed 48 non-equivalent wave vector in first Brillouin zone. The frequency along with symmetry directions have been plotted against the wave vectors to obtain the phonon dispersion curves (PDCs) from both the models. These curves are compared with each other and with inelastic neutron scattering technique.
Since neutron scattering provide us only a very little data for symmetry direction, we have studied the specific heat and combined density of state for complete description of frequencies. For this purpose the specific heat has been computed at different temperature using Blackmann's technique [35] and corresponding Debye temperature have been plotted against absolute temperature (T).
It may be concluded that SNTRSM provides agreement which is certainly better than those fitted by experimental researchers and SNTRIM, are very much close to the experimental values. Although, qualitatively the agreement achieved from our present model SNTRSM is comparatively better than some of the model values. In addition, some other researchers[18-24] of the field has also tried their best to explain PDCs and other properties of europium chalcogenides but only with moderate success.
Furthermore, in order to increase the merit of this work, we have tested the adequacy of our model by calculating[33] two phonon Raman/IR spectra and variation of Debye temperatures shown in figure-2. Since no observed data on two phonon IR/Raman spectra are available, these Combined density of states peaks have been compared with the assignments calculated by using our present theoretical data shown in figure-3. In order to interpret them the critical point analysis have been used following the method prescribed by Burstein *et al.* [36]
It may be concluded that the inclusion of the effect of short range overlap repulsive interaction upto second neighbours in the framework of TRIM and TRSM is important in EuO. The present approach has revealed much better description of the crystal dynamics of the solid under consideration than those reported [1] by other models. Furthermore, the inherent shortcomings of the present models are most likely the same as the demerits of models RIM and RSM. It is expected that slight discrepancies still occurring between theory and experiment may be further improved by including the effect of free carrier screening (FCS), Van der Waals interactions (if data are available in future) and by including anharmoncity of vibrations in the present model (SNTRSM).

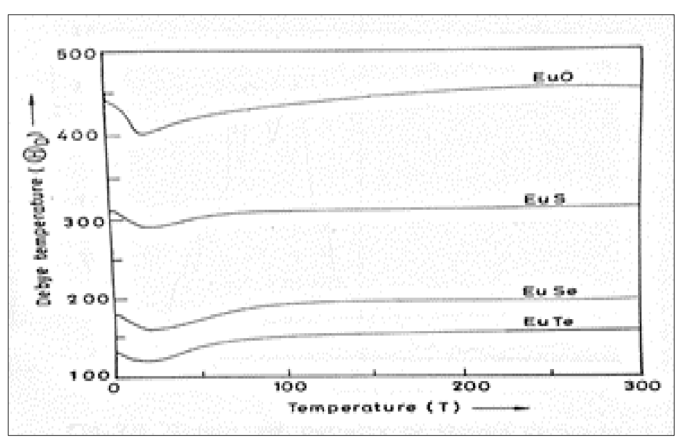

Fig 2: Debye temperature variation of EuO

(This is a combined picture of Debye temperature variation of EuO, EuS, EuSe, EuTe. But here consider only EuO)

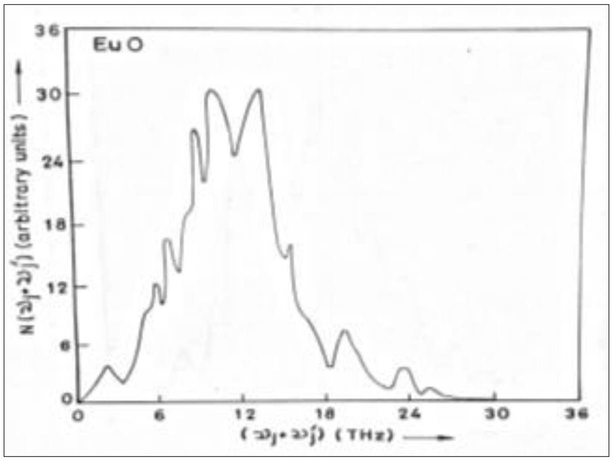

Fig 3: Combined density state curve of EuO

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