Phenomenological Description of the Acoustic Phonon Branch of K_2SeO_4

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Abstract

A phenomenological approach is used in investigating dispersion of the acoustic phonon branch of K_2SeO_4 in the extended zone scheme. The dispersion is expressed through coefficients of a simple thermodynamic potential. According to symmetry of the crystal (D_{2h} = *mmm*) the Lifshitz-type invariant exists in the potential. This invariant is responsible for the linear and dynamical interaction of the acoustic and optical branches. The soft optical branch lowering with decreasing temperature forces to lower the acoustic branch which becomes zero at some value of the wave vector.

Keywords Phenomenological approach; acoustic branch; K_2SeO_4

In the well-known paper by Iizumi et al. [1] the experimental dependence of acoustic branch of K_2SeO_4 on a dimensionless wave number is presented in an extended zone scheme (Fig. 1; see also review [2]). Many authors have developed a theory of this dispersion using different theoretical models (see e.g. [3-6]). We consider here a phenomenological approach to the problem, which was not used, as far as we know. This approach is based on the Landau theory of phase transitions, which in turn is based on symmetry considerations.

Our interest is only with the condition of stability loss of the initial (normal) phase. Thus we can present the thermodynamic potential density in the form:

$$\Phi=\alpha\eta^2+\alpha'\xi^2+\sigma(\eta'\xi-\xi'\eta)+\delta\eta'^2+\kappa\eta''^2+\delta'\xi'^2+\kappa'\xi''^2. \qquad (1)$$

Here only quadratic invariants in two variables η and ξ are taken into account. These variables are assumed to be dependent on the dimensionless coordinate xa^*. The primed (η', ξ') symbols indicate the derivatives of η and ξ with respect to xa^*. The variable η transforms according to one of the vector representations of the group D_{2h}, i.e. as the component P_z of the polarization vector P_i. The variable ξ transforms according to different one-dimensional representation of the group D_{2h}, as the component u_{xz} of the strain tensor u_{ij}. Then η is the amplitude of mode which belongs to the optical branch at q=0, while ξ belongs to the acoustic branch. Note that we consider the whole branch in

Fig. 1 as the acoustic branch.

The variables η and ξ interact via the Lifshitz-type (LT) invariant (it has the same form as the Lifshitz invariant for two-dimensional representations). The LT-invariant is a characteristic feature of potential (1). To seek the condition of stability loss of the initial phase we write η and ξ in the form:

$$\eta = P_z = \rho_1 \cos qa^*x, \quad \xi = 2u_{xz}/a^* = u'_z, \quad u_z = \rho_2 \sin qa^*x \qquad (2)$$

where u_z is the component of the displacement vector u_i, q is the dimensionless wave number.

Substituting Eq. (2) into Eq. (1) and integrating over xa^* we obtain the thermodynamic potential in the form:

$$\Phi = 1/2(\alpha+\delta q^2+\kappa q^4)\,\rho_1^2+\sigma\rho_1\rho_2+1/2\, q^2(\alpha'+\delta' q^2+\kappa' q^4)\,\rho_2^2. \qquad (3)$$

Minimizing Eq. (3) with respect to ρ_1, ρ_2 and q we arrive at the following expressions for the elastic coefficients A_{+-}^2 of the two branches as functions of q:

$$A_{+-}^2/Y^2 = 1/2\{\alpha(q)+\alpha'(q) \,{}^{+}_{-}\, [(\alpha(q)-\alpha'(q))^2+(2\sigma q^2)^2]^{1/2}\},$$

$$\alpha(q) = \alpha+\delta q^2+\kappa q^4, \quad \alpha'(q) = q^2(\alpha'+\delta' q^2+\kappa' q^4). \qquad (4)$$

The signs +- refer to the optical and acoustic branches, respectively. Y is a scale factor. The three-term polynomials $\alpha(q)$ and $\alpha'(q)$ correspond to the optical and acoustic branches without interaction.

For the purpose of presenting the dependence $A_-(q)$ in Fig. 2 at four temperatures as in Fig. 1, the values of coefficients have to be specified. The scale factor Y may be assigned arbitrary. Three coefficients α', δ' and κ' of the dependence $\alpha'(q)$ are determined from the requirement that the acoustic branch has the inclination at small q and a height of the maximum approximately the same as in Fig. 1 and also has the position of the second maximum at q=1. The frequency of acoustic branch becomes zero, according to Fig. 1, at the definite $q=q_0=0.69$ and $T=T_i=130$K. The requirement that this would be fulfilled also for the theoretical acoustic branch determines values of two coefficients; we choose α_0 and κ. Values of these coefficients must be taken with a high degree of accuracy, see below Eq. (5). As it is usually done in the Landau theory, the thermodynamic potential coefficients are assumed to have the simplest dependence on temperature T. Only one coefficient α at the square of the order parameter η depends on T linearly: $\alpha=\alpha_0+\alpha_T(T-T_i)/T_i$ (T_i is the temperature of the stability loss of the initial phase). The coefficient α_T is determined from experimental curves at $T- T_i$ =15, 45, 120K. The dispersion of the optical branch is of no interest for us. The point is that we could not identify a soft optical branch among different experimental branches for K_2SeO_4. Therefore we put for simplicity the coefficient δ=0. It remains only one coefficient σ, which we can vary in such a way as to obtain a better agreement between theoretical and experimental dispersions of the acoustic branch.

Thus, values of the coefficients are chosen as follows:

$$Y=100,\ \alpha'=3.5,\ \delta'=-2.2,\ \kappa'=0.4,\ \alpha_0=0.247,$$
$$\delta=0,\ \kappa=2.214,\ \alpha_T=0.15,\ \sigma=2. \qquad (6)$$

Figure 2 shows the dispersion of acoustic branch, which is computed according to Eqs. (4) and (5). There is agreement between theoretical and experimental dispersions as can be seen from comparison of Fig. 2 with Fig. 1. It should be emphasized that considerable change in the acoustic branch at T-T_i=45, 15, 0 K take place at comparatively small softening of the optical branch: A_+=54.7, 51.4, 49.7 at q=0.

To summarize we can say that the existence of the LT-invariant in the thermodynamic potential is the result of symmetry requirements. Due to the LT-invariant the soft optical and acoustic branches interact linearly and dynamically, so that the optical branch lowering with decreasing T pushes downwards the acoustic branch which becomes zero at $q=q_0$ and $T=T_i$.

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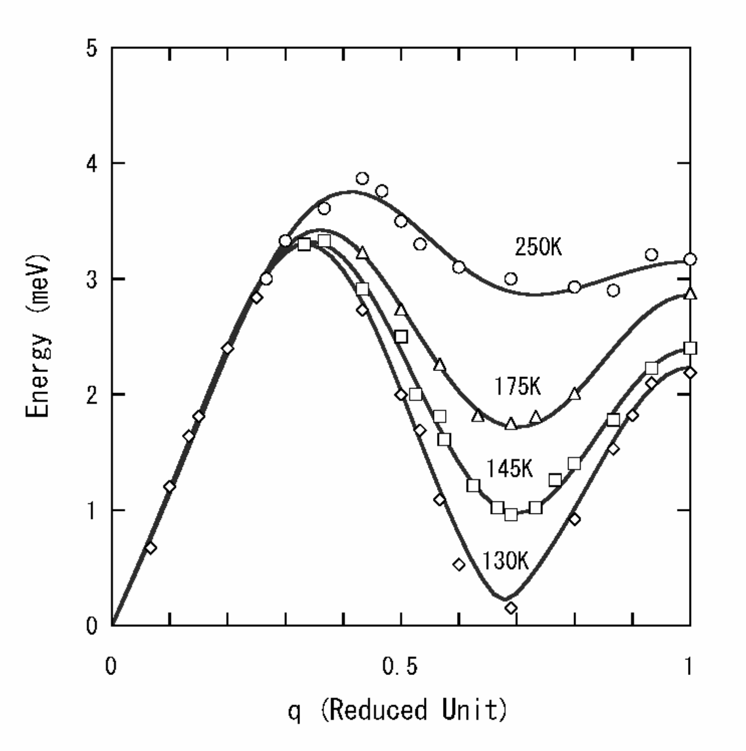

FIGURE 1. Phonon dispersion of the acoustic branch in K_2SeO_4 at different temperatures from Ref. [1].

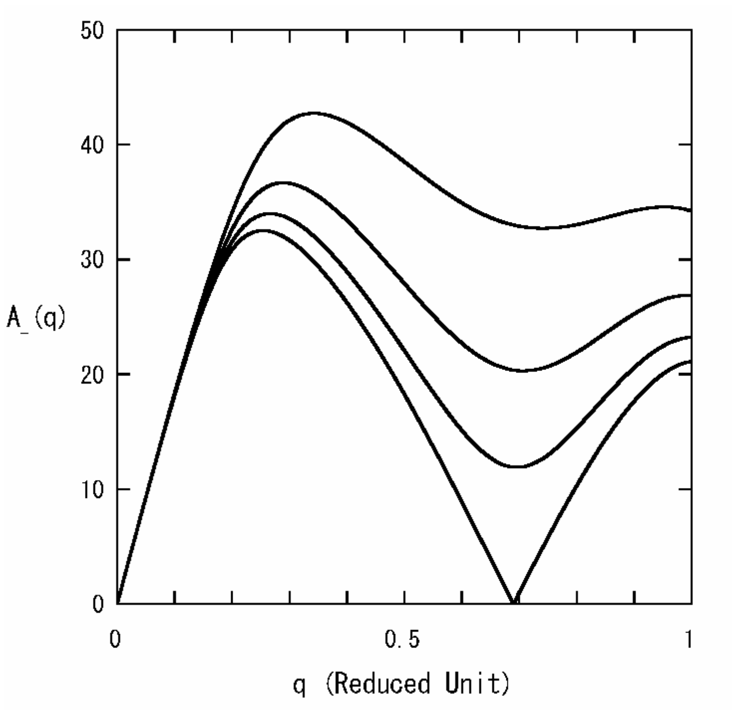

FIGURE 2. Dependence A_(q) plotted according to Eqs. (4) and (5).