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Elastic anomalies in strontium titanate

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Abstract. The anomalies of the second and third-order elastic constants have been considered for the phase transition of strontium titanate within the framework of Landau's theory. All the anomalies of the second-order elastic constants have been obtained in a single formula using Kronecker delta functions and relations among them have been established. The real parts of C_{11}^* and C_{44}^* decrease steeply across the transition temperature and thereafter flatly tend to their asymptotic values in the low temperature phase agreeing qualitatively with experimental observations. We have also derived expressions for the third-order elastic anomalies and discussed the temperature variation of the real part of C_{111}^*. We have derived expressions for the attenuation of the longitudinal and transverse waves along certain simple symmetry directions and have shown that there is nearly good agreement with experimental observations.

Keywords. Elastic anomalies; phase transition; strontium titanate

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1. Introduction

Strontium titanate has a cubic perovskite structure and undergoes an improper ferroelastic second order-phase transition near a temperature of 105 K. While at normal temperature, $SrTiO_3$ is paraelectric, it transforms on cooling to a tetragonal phase with a c/a ratio of 1·00056. The optical, dielectric, thermal and elastic properties associated with the phase transition have been investigated by many workers and these have been reviewed by various authors [1–4]. Experimental studies in $SrTiO_3$ as well as in similar crystals like $KMnF_3$, $LaAlO_3$, $PAlO_3$ and $NdAlO_3$ have demonstrated that spontaneous optic mode displacements and spontaneous strains appear during the phase transition at the low temperature phase [5–6].

Another aspect of the phase transition that has been extensively studied relates to second harmonic generation and the third-order elastic constants [7]. Peters and Arnold [8] studied the temperature variation of the third-order elastic constant C_{111} in the range 103–300 K using ultrasonic harmonic generation technique. Meeks and Arnold [9] used the second harmonic generation technique to measure combinations of several third-order elastic constants over a wide range of temperatures. The work of Bell and Rupprecht [10] showed that there were pronounced variations in the second-order elastic constants besides, large changes in ultrasonic attenuation occur near the phase transition. The attenuation of elastic waves and the changes in their velocity during phase transition have been investigated by Fossum and Fossheim [11] and also by Schwabl and Iro [12].

In this paper, we study systematically the anomalies near the phase transition of $SrTiO_3$ of the second and third-order elastic constants within the framework of the Landau theory. While the initial strains and the components of the order parameter in the low temperature phase can be obtained from the stability conditions, the fluctuations in these parameters from their equilibrium values have been derived by an appeal to the Landau-Khalatnikov equations in irreversible statistical mechanics. In §4 we derive the anomalies for all the second-order elastic constants in a single formula and discussed the temperature variation of the second-order elastic constants. In §5, we derive expressions for the anomalies in third-order elastic constants in terms of the coupling constants and discuss the temperature variation of C^*_{111} near the transition temperature. In §6, we make comments about the attenuation of the longitudinal and transverse modes along the principal axis and face diagonal of the crystal.

2. The equilibrium values of the order parameters and the strains

Strontium titanate undergoes a structural phase transition from cubic to tetragonal class at a temperature of 105 K. The transition is induced by the softening of the phonon mode at (π/a) $(1,1,1)$ corner of the Brillouin zone. Below transition temperature, there is coupling of the unit cell and the symmetry of the crystal changes from O_h to D^{18}_{4h}. The order parameter is the rotation of the alternate oxygen octahedra TiO_6 around cubic axes. The phase transition is an improper ferroelastic transition.

The free energy of the crystal is the sum of the elastic energy F_{el}, the Landau energy F_Q and the coupling energy F_c

$$F = F_{el} + F_Q + F_c \tag{1}$$

The elastic energy of the crystal in the cubic phase is the sum of the second-order (E_2) and third-order (E_3) deformation energies. Further,

$$E_2 = \frac{1}{2}\{C_{11}(\eta_1^2 + \eta_2^2 + \eta_3^2) + 2C_{12}(\eta_2\eta_3 + \eta_3\eta_1 + \eta_1\eta_2) + C_{44}(\eta_4^2 + \eta_5^2 + \eta_6^2)\} \tag{2}$$

where $\eta_1, \eta_2, \ldots, \eta_6$ are the six components of the strain tensor. The expression E_3 for the third-order deformation energy is well known and is not reproduced here for lack of space. It can be shown that the corrections brought about by third-order deformation energy to equilibrium values of order parameter and strain variables are smaller by a factor $Q_{30}^2 C_{ijk}(C_{ij})^{-2}$ in comparison with the contribution from second-order deformation energy. We shall therefore neglect them in future calculations. The order parameter is the octahedra rotation. The three components of the order parameter are denoted by Q_1, Q_2, Q_3 such that $Q^2 = Q_1^2 + Q_2^2 + Q_3^2$. The Landau free energy is given by Lemanov [1]

$$F_Q = \frac{1}{2}\alpha Q^2 + \frac{1}{4}\beta_1 Q^4 + \frac{1}{4}\beta_2(Q_2^2Q_3^2 + Q_3^2Q_1^2 + Q_1^2Q_2^2) \tag{3}$$

where $\alpha = \alpha'(T - T_c)$, α' being a constant. The coefficients β_1 and β_2 are constants, very nearly independent of temperature.

The term F_c due to coupling between the order parameter and the strains had been evaluated using symmetry consideration and is given by Lemanov [1]

$$F_c = d_1\{(Q^2 - 3Q_1^2)\eta_1 + (Q^2 - 3Q_2^2)\eta_2 + (Q^2 - 3Q_3^2)\eta_3\} + d_t\{(Q_2Q_3\eta_4 + Q_3Q_2\eta_5 + Q_1Q_2\eta_6\} \quad (4)$$

where d_1 and d_t are coupling constants.

The free energy is a function of the three components of the order parameter and the six strain components. For the stability of the crystal, the partial derivatives of F with respect to all these nine parameters should vanish.

It can be seen that the solution of these nine equations for the high temperature paraelectric phase corresponds to the case in which all the three components of the order parameter as well as the six strain components vanish.

The phase transition of $SrTiO_3$ to the tetragonal phase is ferroelastic and corresponds to the case in which the order parameter along the c-axis is nonzero while the other two components are zero. Hence the case $Q_1 = Q_2 = 0$ and $Q_3 \neq 0$ needs to be discussed. Solving the nine equations for the stability conditions, we find that the equilibrium values of the order parameter as well as the nonvanishing strain components are given by

$$Q_{30}^2 = (\alpha/a), \quad (5)$$

$$\eta_{10} = \eta_{20} = -d_1(C_{11} - C_{12})^{-1}Q_{30}^2, \quad (6)$$

$$\eta_{30} = 2d_1(C_{11} - C_{12})^{-1}Q_{30}^2. \quad (7)$$

3. The Landau–Khalatnikov equation and the fluctuations in the order parameter

The Landau–Khalatnikov equation expresses the fact that the time rate of change dQ/dt of the order parameter fluctuation towards equilibrium is proportional to the thermodynamic restoring force $\partial F/\partial Q$. From the Landau–Khalatnikov equation, the expressions for the fluctuations induced by an ultrasonic wave in order parameters from their equilibrium values can be obtained.

The Landau–Khalatnikov equation for the order parameter components can be written as

$$\dot{Q}_i = -\Gamma_i(\partial F/\partial Q_i) \quad (i = 1, 2, 3) \quad (8)$$

where $\Gamma_1, \Gamma_2, \Gamma_3$ are the three kinetic coefficients. The symmetry of the tetragonal phase ensures that $\Gamma_1 = \Gamma_2$. We shall consider the solution of the Landau–Khalatnikov equation for the ferroelastic phase and write

$$Q_i = Q_{i0} + q_i^* \quad (i = 1, 2, 3) \quad (9a)$$

$$\eta_i = \eta_{i0} + \eta_i^* \quad (i = 1 \text{ to } 6) \quad (9b)$$

Then q_i^* and η_i^* represent the time dependent fluctuations of these parameters from their equilibrium values.

By expanding $(\partial F/\partial Q_i)$ for $i = 1$ to 3 about the equilibrium values of the order parameter components and the strain variables and setting q_i^* proportional to $\exp(i\Omega t)$, we find that the solution of the Landau–Khalatnikov equations turn out to be

$$q_1^* = A_1 \eta_5^* \tag{10}$$

$$q_2^* = A_1 \eta_4^* \tag{11}$$

$$q_3^* = \sum_{i=1}^{6} \alpha_i \eta_i^* \tag{12}$$

where

$$A_1 = -(\Gamma_1/P_1) d_t Q_{30}, \tag{13a}$$

$$P_1 = i\Omega + \Gamma_1 (\partial^2 F/\partial Q_1^2)_0, \tag{13b}$$

$$\alpha_1 = \alpha_2 = -(2\Gamma_3 Q_{30} d_1)/P_3, \tag{13c}$$

$$P_3 = i\Omega + \Gamma_3 (\partial^2 F/\partial Q_3^2)_0, \tag{13d}$$

$$\alpha_3 = -2\alpha_1; \; \alpha_4 = \alpha_5 = \alpha_6 = 0. \tag{13e}$$

The above equations show that the fluctuations in the components of the order parameter about their equilibrium values are linearly related to the fluctuations in the strain variables.

4. Anomalies in the second-order elastic constants

If we substitute the equations (9a) to (13e) in the expression for the free energy, we find

$$F = F_0 + F_2 + F_3 + F_4 \tag{14}$$

where F_0, F_2, F_3 and F_4 denote the zero, second, third and fourth-order energies respectively. In view of the stability condition, F_1 vanishes. The zeroth order term F_0 gives the shift in the zero point energy at the transition temperature and its derivative with respect to temperature gives the specific heat anomaly at the transition temperature. By collecting all the terms which are quadratic in the strain variables, the expression for F_2 becomes

$$\begin{aligned} F_2 = \frac{1}{2}\sum_{ij} C_{ij}^* \eta_i^* \eta_j^* &= \frac{1}{2}\sum_{ij} C_{ij} \eta_i^* \eta_j^* \\ &+ \alpha/2 (q_1^{*2} + q_2^{*2} + q_3^{*2}) \\ &+ (\beta_1/4)\{2Q_{30}^2 (q_1^{*2} + q_2^{*2} + q_3^{*2}) + 4Q_{30}^2 q_3^{*2}\} \\ &+ (\beta_2/4) Q_{30}^2 (q_1^{*2} + q_2^{*2}) \\ &+ d_1 \{\eta_{10}(q_3^{*2} + q_2^{*2} - 2q_1^{*2}) + \eta_{20}(q_3^{*2} + q_1^{*2} - 2q_2^{*2}) \\ &+ \eta_{30}(q_1^{*2} + q_2^{*2} - 2q_3^{*2}) + 2Q_{30} q_3^* (\eta_1^* + \eta_2^* - 2\eta_3^*)\} \\ &+ d_t Q_{30} (q_2^* \eta_4^* + q_1^* \eta_5^*) \end{aligned} \tag{15}$$

where C_{ij}^* represents the modified second-order elastic (SOE) constants arising from the coupling of the order parameters with strains. By writing

$$C_{ij}^* = C_{ij} + \Delta C_{ij}^* \tag{16}$$

ΔC_{ij}^* represents the anomalies in the SOE constants. Using Kronecker delta functions, one can in fact write the anomalies for all the elastic constants in a single formula as

$$\begin{aligned}\frac{1}{2}\Delta C^*_{ij} = &\{A_1^2(\delta_{i5}\delta_{j5}+\delta_{i4}\delta_{j5})+\alpha_i\alpha_j\}\times\\ &\{\alpha/2+(\beta_1/2)Q_{30}^2+(\beta_2/4)Q_{30}^2\}+\\ &+Q_{30}^2\{(\beta_1-\beta_2/4)\}\alpha_i\alpha_j\\ &-3d_1\{A_1^2(\eta_{10}\delta_{i5}\delta_{j5}+\eta_{20}\delta_{i4}\delta_{j4})+\eta_{30}\alpha_i\alpha_j\}\\ &+d_1Q_{30}\{\alpha_i(\delta_{j1}+\delta_{j2}-2\delta_{j3})+\alpha_j(\delta_{i1}+\delta_{i2}-2\delta_{i3})\}\\ &+d_tQ_{30}A_1(\delta_{i4}\delta_{j4}+\delta_{i5}\delta_{j5}).\end{aligned} \tag{17}$$

The existence of anomalies in the SOE constants shows that the velocities of the elastic waves undergo a change during phase transition, and further the waves are attenuated as ΔC^*_{ij} is complex. One can notice that there exist relations among the elastic anomalies in the low temperature phase and all of them can be expressed in terms of two, namely ΔC^*_{11} and ΔC^*_{44}. The relations are

$$\Delta C^*_{11} = \Delta C^*_{22} = \Delta C^*_{12} \tag{18}$$

$$\Delta C^*_{23} = \Delta C^*_{13} = -2\Delta C^*_{11} \tag{19}$$

$$\Delta C^*_{33} = 4\Delta C^*_{11} \tag{20}$$

$$\Delta C^*_{44} = \Delta C^*_{55} \tag{21}$$

$$\Delta C^*_{11} = (8\Gamma_3^2 d_1^2\alpha^2\beta_1)/(P_3^2a^2) - (8d_1^2\Gamma_3\alpha)/(P_3a) \tag{22}$$

$$\Delta C^*_{44} = (2\Gamma_1^2 d_t^2\alpha^2)/(P_1^2a^2)\{(\beta_2/4)+(3K/4)\} - 2\Gamma_1^2 d_t^2\alpha/(P_1a) \tag{23}$$

where

$$K = 12d_1^2(C_{11}-C_{12})^{-1} \tag{24a}$$

and

$$a = -(\beta_1 - K). \tag{24b}$$

The expressions P_1 and P_3 given by (13b) and (13d) can be expressed in terms of the relaxation time τ. In terms of this parameter, the above expressions for ΔC^*_{11} and ΔC^*_{44} turn out to be

$$\Delta C^*_{11} = -(2d_1^2/\beta_1)(1+2i\Omega\tau)(1+i\Omega\tau)^{-2} \tag{25}$$

$$\Delta C^*_{44} = -\{2d_t^2/(\beta_2+3K)\}(1+2i\Omega\tau)(1+i\Omega\tau)^{-2} \tag{26}$$

where

$$\tau = \tau_0/|T_c - T|.$$

The numerical values of τ_0, (d_1^2/β_1), $d_t^2/(\beta_2+3K)$ for $SrTiO_3$ have been given by Lemanov [1] as $1{\cdot}6\times10^{-11}$ sK, $1{\cdot}6\times10^9\,N/m^2$ and $1{\cdot}9\times10^9\,N/m^2$ respectively. For numerical calculations, we shall choose experimental value of Ω as $1{\cdot}6\times10^9\,s^{-1}$ so that $\Omega\tau_0 = 10^{-2}$. We notice that both ΔC^*_{11} and ΔC^*_{44} are complex and temperature dependent. The real parts of the elastic anomalies are given by

$$R(\Delta C^*_{11}) = -(2d_1^2/\beta_1)(1+3\Omega^2\tau^2)(1+\Omega^2\tau^2)^{-2} \tag{27}$$

$$R(\Delta C^*_{44}) = -\{2d_t^2/(\beta_2+3K)\}(1+3\Omega^2\tau^2)(1+\Omega^2\tau^2)^{-2}. \tag{28}$$

From these expressions, it is possible to discuss the temperature variation of these anomalies numerically. At the transition temperature, $T = T_c$, $\tau \to \infty$ so that both $R(\Delta C_{11}^*)$ and $R(\Delta C_{44}^*)$ vanish. Next $\Omega\tau$ reaches the value unity when $(T_c - T) = \Omega\tau_0 = 10^{-2}$. For this value as well as for the point $\Omega\tau = 0$, we find that

$$R(\Delta C_{11}^*) = -2d_1^2/\beta_1 \tag{29}$$

and

$$R(\Delta C_{44}^*) = -2d_1^2/(\beta_2 + 3K). \tag{30}$$

Again the stationary values of these anomalies are reached for the temperature $T = T_c + \sqrt{3}10^{-2}$.

For this value

$$R(\Delta C_{11}^*) = -(9/8)(2d_1^2/\beta_1); \tag{31}$$

$$R(\Delta C_{44}^*) = -(9/8)\{2d_1^2/(\beta_2 + 3K)\} \tag{32}$$

With these numerical values, we notice that the elastic constants have uniform values in the paraelectric state, fall steeply within a range of the order 10^{-2} K to the values given in (31) and (32), and reach asymptotic values within a range of 10^{-1} K. The

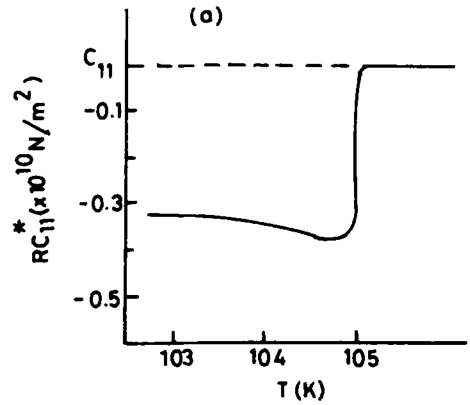

Figure 1a. Variation of the real part of C_{11}^* with respect to temperature close to the phase transition (theoretical).

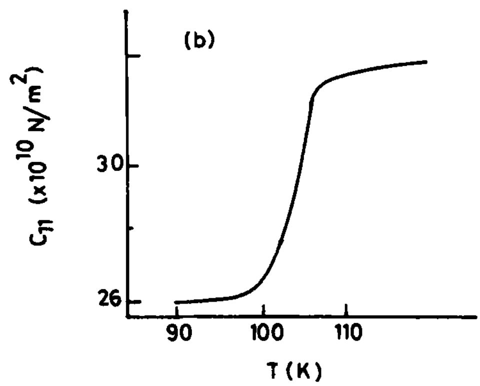

Figure 1b. Temperature variation of C_{11} near phase transition (experimental).

temperature variation of real parts of C^*_{11} and C^*_{44} has been plotted in figures (1a) and (2a) respectively.

Slonczewski and Thomas [5] have investigated the elastic anomalies of strontium titanate close to the transition temperature. They have derived the following relations among the elastic anomalies;

$$C_{11} = C^c_{11} - D;\ C_{33} = C^c_{11} - 4D \tag{33}$$

$$C_{12} = C^c_{12} - D;\ C_{13} = C^c_{12} + 2D \tag{34}$$

$$C_{44} = C^c_{44} - E \tag{35}$$

where

$$D = (4B_e^2 Q_s^2 / M\omega_3^2); \tag{36}$$

$$E = (B_t^2 Q_s^2 / M\omega_1^2). \tag{37}$$

Comparing the set of equations (33) to (35) with (18) to (21) we find that we have recovered all the relations for the elastic anomalies given by them. However, our formulae for the anomalies are different and go deeper than the results of Slonczewski

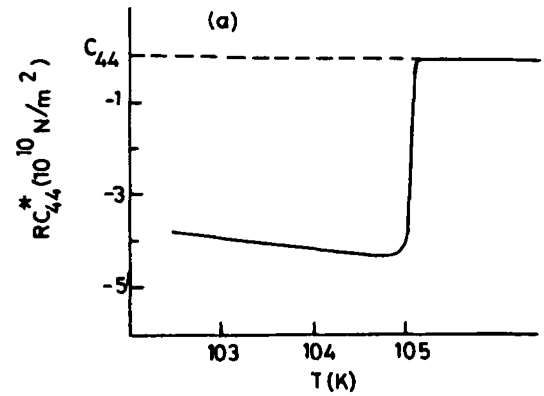

Figure 2a. Variation of the real part of C^*_{44} with respect to temperature close to the phase transition (theoretical).

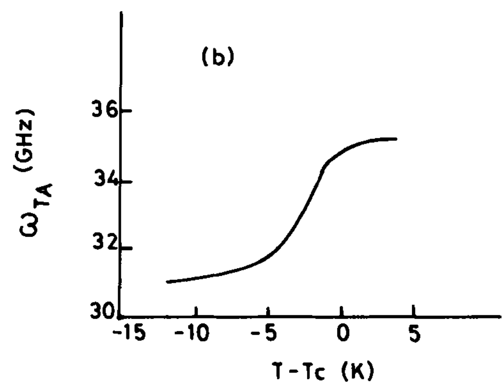

Figure 2b. Transverse acoustic mode ω_{TA} frequency as a function of $(T - T_c)$ (experimental).

and Thomas. If we substitute the expression for the soft mode frequencies from the formula

$$M\omega_1^2 = M\omega_2^2 = \left(\frac{\partial^2 F}{\partial Q_1^2}\right)_0 = \frac{\alpha}{2a}(\beta_2 + 3K) \tag{38}$$

$$M\omega_3^2 = \left(\frac{\partial^2 F}{\partial Q_3^2}\right)_0 = (2\alpha\beta_2/a) \tag{39}$$

in the formulae for D and E, we obtain

$$D = (2B_e^2/\beta_1) \text{ and } E = (2B_t^2)/(\beta_2 + 3K). \tag{40}$$

Comparing these with (27) and (28), we see that their formulae give the first factor in (27) and (28), but lack the factor depending on the relaxation time. Without this factor, it is not possible to explain the steep dip in the elastic constants occurring at the transition temperature.

The temperature variation of C_{11} near the transition temperature has been evaluated by Luthi and Moran [13] experimentally. The shape of our curve agrees qualitatively with their curve (1b). Similarly the shape of our curve agrees qualitatively with the experimental curve in figure (2b) for the variation of C_{44} near the transition temperature by the Brillouin scattering method given by Lyons and Fleury [14].

5. Anomalies in the third-order elastic (TOE) constants

The expression for the free energy contains cubic terms in the order parameters and strain variables. The anomalies in the TOE constants can be obtained by collecting the coefficients of all the third-order terms $\eta_i^* \eta_j^* \eta_k^* (i, j, k = 1 \text{ to } 6)$. Several of the anomalies become equal to each other and it can be verified that the following relations exist among the individual TOE constants

$$\Delta C_{111}^* = \Delta C_{222}^* = \Delta C_{112}^* = \Delta C_{122}^* = -\frac{1}{8}\Delta C_{333}^* \tag{41}$$

$$\Delta C_{123}^* = -2\Delta C_{111}^* \tag{42}$$

$$\Delta C_{144}^* = \Delta C_{255}^* \tag{43}$$

$$\Delta C_{344}^* = \Delta C_{355}^* \tag{44}$$

$$\Delta C_{155}^* = \Delta C_{244}^* \tag{45}$$

$$\Delta C_{456}^* = (d_t^3 \Gamma_1^2 \alpha)(P_1^2 a)^{-1}. \tag{46}$$

The real part of ΔC_{111}^* can be written as

$$R(\Delta C_{111}^*) = -\frac{6d_1^3(\beta_1 - K)(\Omega\tau)^2(3 - \Omega^2\tau^2)}{\beta_1^2(T_c - T)\alpha'(1 + \Omega^2\tau^2)^3} \tag{47}$$

It is seen that $R(\Delta C_{111}^*)$ is zero when $T = T_c$ and it is positive for all values of $\Omega\tau$ from infinity up to $\sqrt{3}$ where it becomes zero again. The graph for real part of C_{111}^* very close to the phase transition is given in figure 3a. It is seen that the real part of C_{111}^* decreases steeply close to the transition temperature and then increases within

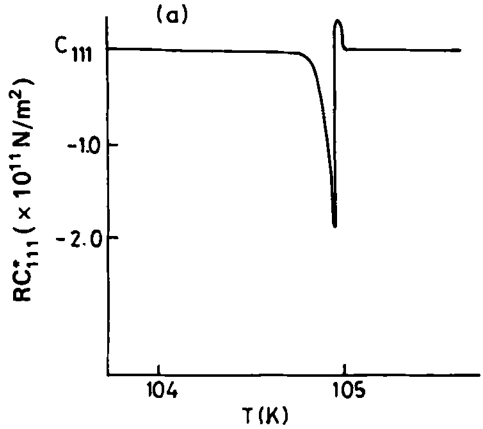

Figure 3a. Variation of the real part of C^*_{111} with respect to temperature close to the phase transition (theoretical).

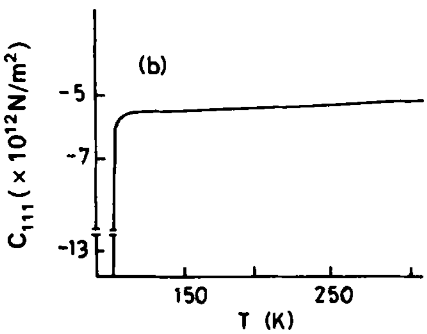

Figure 3b. Temperature variation of C_{111} as a function of temperature (experimental).

a range of the order 10^{-1}K to become zero again. Meeks and Arnold [9] have investigated experimentally (figure 3b) the temperature dependence of C_{111} at the transition temperature. They found that at the transition temperature C_{111} decreases steeply and our curve qualitatively agrees with their results. However, our theoretical results predict that the anomaly in $R(\Delta C^*_{111})$ will become zero again within a very small range of temperature.

6. Attenuation of elastic waves

Apart from the anomalies in elastic constants of various orders, which determine the sound wave velocities, another major effect of the phase transition is the attenuation of the sound waves. In this section, we give expressions for the attenuation of the elastic waves close to the transition temperature.

Let us write

$$C_{ij} = C'_{ij} + C''_{ij} \tag{48}$$

where C'_{ij} and C''_{ij} denote the real and imaginary parts of C^*_{ij}. Let us denote by α_1 the

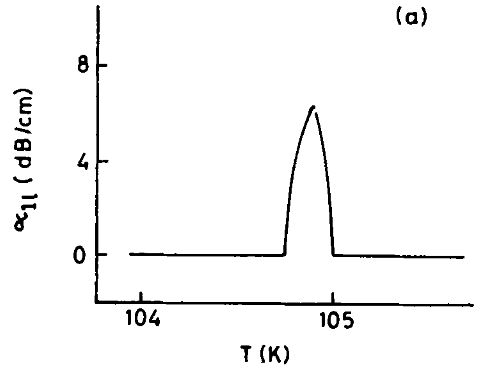

Figure 4a. Variation of attenuation of the longitudinal wave (α_{1l}) with respect to temperature close to the phase transition (theoretical).

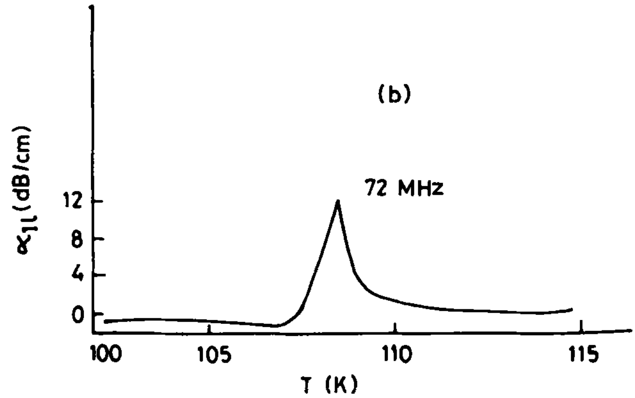

Figure 4b. Ultrasonic attenuation α_{1l} of longitudinal wave as a function of temperature at high biaxial pressure (experimental).

attenuation of the elastic waves. Then we have the following expressions for the attenuation of the longitudinal and transverse waves along the principal directions.

(1) (100) direction

$$\alpha_{1l} = (\Omega/2)(C''_{11}/C'_{11}) \tag{49a}$$

$$\alpha_{1t} = (\Omega/2)(C''_{44}/C'_{44}) \tag{49b}$$

(2) $\left(\frac{1}{\sqrt{2}}, \frac{1}{\sqrt{2}}, 0\right)$ direction

$$\alpha_{1l} = (\Omega/2)(C''_{11} + C''_{12} + 2C''_{44})/(C'_{11} + C'_{12} + 2C'_{44}) \tag{50a}$$

$$\alpha_{1t_1} = (\Omega/2)(C''_{11} - C''_{12})/(C'_{11} - C'_{12}) = \alpha_{1t_2} \tag{50b}$$

From the equation (49a), we find that the attenuation of the longitudinal wave along the [100] direction is given by

$$\alpha_{1l} = \frac{2\Omega d_1^3}{\beta C'_{11}} \frac{(\Omega\tau)^3}{(1 + \Omega^2\tau^2)^2} \tag{51}$$

The attenuation anomaly vanishes when $\Omega\tau \to \infty$ or at $T = T_c$. The attenuation steeply increases from zero to its maximum value and then again decreases to its asymptotic value of zero. Since C_{44}^* also has the same dependence on temperature, the variation with temperature of the transverse mode along [100] and the corresponding mode along [110] is also similar. The attenuation of longitudinal wave (72 MHz) as a function of temperature at high biaxial pressure of $SrTiO_3$ has been measured by Fossheim and Berre [15]. There is good agreement between the experimental curve (figure 4b) and our prediction (figure 4a).

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