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Temperature Dependence of Soft Mode Frequency Above Phase Transition*

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The temperature dependence of the soft phonon frequency, ν_r, near a second order phase transition, is calculated by means of diagram techniques. For ν_r small, compared to the four phonon vertex $g(T)$, one obtains $\nu_r^2 \sim \tau^2$, ($\tau = (T - T_c)/T_c$) and, for $\nu_r >> g(T)$, $\nu_r^2 \sim \tau^1$, which is the mean field result. The critical index, γ, thus changes from 1 to 2 when approaching transition. This is, qualitatively, in agreement with the value of γ obtained from inelastic neutron studies in $SrTiO_3$. It is too large, however, when compared with the value of γ resulting from EPR linewidth measurements.

Calcula-se, por técnicas diagramáticas, a dependência na temperatura, da freqüência ν_r dos fonons moles, perto da transição de fase de 2.ª ordem. Para ν_r pequeno, comparado com o vértice de quatro fonons, $g(T)$, obtém-se $\nu_r^2 \sim \tau^2$ ($\tau = (T - T_c)/T_c$) e, para $\nu_r >> g(T)$, $\nu_r^2 \sim \tau^1$, que é o resultado da teoria de campo médio. O índice crítico, γ, é então alterado de 1 para 2 quando se aproxima da transição. Qualitativamente, isso está de acôrdo com o valor de γ obtido em base aos estudos de difusão inelástica de neutrons em $SrTiO_3$. Esse valor, todavia, é muito grande quando comparado com aquele resultante das medidas de largura de linha EPR.

1. Introduction

Structural phase transitions in solids can be given in terms of unstable or "soft" optic phonons[1,2]. The temperature dependence of the soft phonon frequency, $\nu_r(T)$, well above the phase transition temperature, T_c, is described by the mean field result[3], $\nu_r^2 \sim \tau$. Closer to T_c, however, generally considerable deviations from the mean field behaviour are

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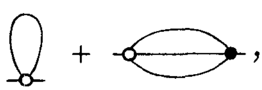
observed[4]. In $SrTiO_3$ e.g., where a soft mode, at the R corner of the Brillouin zone (B.Z.), drives the phase transition, v, deviates from linear behaviour for $T - T_c < 10°K$. For the critical coefficient, y, defined by $\nu_r^2 - \tau^\gamma$, Shapiro *et al.*[4] found, in the temperature interval $1.3°K < T - T_c < 10°K$, a value between 1.5 and 2.5. Using EPR[5] data, Schneider and Sto11[6] obtained $y = 1.29 \pm 0.10$. Such deviation has been attributed to the interaction of the soft phonon with low energy acoustic multiphonon fluctuations showing up as a central peak, in neutron scattering studies of $SrTiO_3$ (Ref. 7), around o = 0 and q = q, . Although such processes are certainly of importance in the following, only interactions between soft phonons will be considered. It will be shown that already in the Hartree approximation, when not treated in molecular field approximation, deviations from $y = 1$, close to T_c, do occur. The method applied is based on renormalization group techniques introduced in the study of critica1 phenomena by Wilson[8]. In the next Section, γ will be calculated in the Hartree approximation and, in the final one, modifications due to ladder diagrams will be considered.

2. The Hartree Approximation

The static wave-vector dependent susceptibility of the problem is given by

$$\chi_j(\mathbf{q}) = D_j^{\mathrm{R}}(\mathbf{q}, 0) = [(\omega_{\mathbf{q},j}^0)^2 - \textstyle\prod_j^{\mathrm{R}}(\mathbf{q}, 0)]^{-1}, \tag{1}$$

where $D^{\mathrm{R}}(q, o)$ is the retarded phonon propagator, $\omega_{\mathbf{q},j}^0$ the bare phonon frequency, $\prod_j^{\mathrm{R}}(\mathbf{q}, \omega)$ the retarded phonon self energy and, j, a polarization index. For a Hamiltonian with a quartic interaction term, the self energy is determined by the diagrams



(2)

where the first, or Hartree term, will now be considered. Due to the fact that the squared bare frequencies in a certain region around $\mathbf{q}_r$ are negative, whereas $[-\prod_j^{\mathrm{R}}(\mathbf{q}, 0)]$ is a positive increasing function of temperature, the denominator of (1) reaches zero at a certain T_c, indicating thus an instability of the lattice. Using the notation

$$\nu_r^2(T) + f_j(\mathbf{p}, T) = (\omega_{\mathbf{q},j}^0)^2 - \textstyle\prod_j^{\mathrm{R}}(\mathbf{q}, 0), \qquad \mathbf{q} = \mathbf{q}_{\mathrm{R}} - \mathbf{p}, \tag{3}$$

where $f_j(\mathbf{p}, T)$ is the dispersion of the soft phonon branch j referred to q_R, one can write

$$\prod_j^{H}(\mathbf{p}) = -\frac{1}{2}\frac{\hbar}{i} U_{j\mathbf{p},j'\mathbf{p}'} \frac{1}{(-i\hbar\beta)} \sum_{n'} D_{j'\mathbf{p}'}(\omega_{n'}), \tag{4}$$

where

$$U_{j\mathbf{p},j'\mathbf{p}'} \equiv U_{j\mathbf{q}_R+\mathbf{p},j'-(\mathbf{q}_R+\mathbf{p}),j'\mathbf{q}_R+\mathbf{p}',j'-(\mathbf{q}_R+\mathbf{p}')}$$

is the coupling function and, $D_{j\mathbf{p}}(\omega_n)$, the renormalized propagator in frequency representation; $\omega_n = 2\pi n/-i\hbar\beta$, $\beta = 1/k_B T$.

An excellent review of the field theory of phonon systems can be found in Ref. 9. It is understood that quantities occurring two times will be summed over. At $\mathbf{q}_R$, we can set up the following equation, which eliminates $(\omega^0_{\mathbf{q},j})^2$:

$$v_r^2(T) - v_r^2(T_c) = \frac{1}{2} U_{j0,j'\mathbf{p}'}\{\beta^{-1}\sum_n D_{j'\mathbf{p}'}(\omega_n) - \beta_c^{-1}\sum_n D_{j'p'}(\omega_n)\}, \tag{5}$$

where $v_r^2(T_c) = 0$. Separating, in (5), the most singular term, $\omega_n = 0$, from the rest, we can write

$$v_r^2(T) = \frac{1}{2} k_B \left\{ U_{j0,j'\mathbf{p}'}\left[\frac{T}{v_r^2(T) + f_{j'}(\mathbf{p}', T)} - \frac{T_c}{f_{j'}(\mathbf{p}', T_c)}\right] + U_{j0,j'\mathbf{p}'}\left[T\sum_{n\neq 0} D_{j'\mathbf{p}'}(\omega_n) - T_c \sum_{n\neq 0} D_{j'\mathbf{p}'}(\omega_n)\right]\right\}. \tag{6}$$

Evaluation of (6) requires some knowledge on the dispersion of the soft phonon branches around $\mathbf{q}_R$. Accordingly, an equation for $f_j(\mathbf{p}, T)$ has to be set up in addition to (5).

One easily obtains in the same approximation

$$f_j(\mathbf{p}, T) = (\omega^0_{\mathbf{p},j})^2 - (\omega^0_0)^2 - [v_r^2(T) - v_r^2(T_c)] + \frac{1}{2} U_{j\mathbf{p},j'\mathbf{p}'}\beta^{-1}\sum_n D_{j'\mathbf{p}'}(\omega_n) - \frac{1}{2} U_{j0,j'\mathbf{p}'}\beta_c^{-1}\sum_n D_{j'\mathbf{p}'}(\omega_n). \tag{7}$$

With

$$U_{j\mathbf{p},j'\mathbf{p}} = U_{j0,j'\mathbf{p}} + g(j\mathbf{p}, j'\mathbf{p}'), \tag{8}$$

where $g(j0, j'p') = 0$, we obtain from Eq. (7) for $T = T_c$,

$$f_j(p, T_c) = (\omega^0_{\mathbf{p},j})^2 - (\omega^0_0)^2 + \frac{1}{2} k_B T_c g(jp, j'p') \sum_{n'} D_{j'p'}(\omega_{n'}). \tag{9}$$

Eq. (9) defines a set of integral equations for the $f_j(\mathbf{p}, T_c)$. Since we are only interested in $f_j(\mathbf{p}, T_c)$ for small p, we obtain from (9), with $g(j\mathbf{p}, j'\mathbf{p}') \sim p^2 \bar{g}(j\mathbf{0}, j'\mathbf{p}')$, approximately,

$$f_j(\mathbf{p}, T_c) \sim \lambda_j(T)\, p^2. \tag{10}$$

Even if the bare frequency dispersion were of the form $(\omega_{p,j}^0)^2 - (\omega_0^0)^2 \sim \sim \lambda_j p^\alpha$, with a > 2, the critical behaviour will be determined by the smallest power term. Assuming that $\lambda_j(T) \sim \lambda_j(T_c) = \lambda_j$ and $U_{j\mathbf{p},j'\mathbf{p}'} \sim U_{j\mathbf{0},j'\mathbf{0}}$, we obtain, for (6),

$$v_r^2(T) \sim \frac{1}{4\pi^2} \frac{k_B U_{j0,j'0}}{\lambda_{j'}} \left\{ (T - T_c)\, q_c - \frac{\pi}{2} \frac{T}{\lambda_j^{1/2}} v_r(T) \right\} + a_{jj'}(T)(T - T_c), \tag{11}$$

where q, is a conveniently chosen cutoff and $a_{jj'}(T)$ represents the second term, of Eq. (6), with $a_{jj'}(T) > 0$.

It should be noted that, in the present model, the soft phonon momenta are not confined to the first B.Z. but that all soft phonons, belonging to the eight R-corners of the first B.Z., are translated by reciprocal lattice vectors into one R-corner. By this method, all kinds of processes between soft phonons, whether they are of the Umklapp-type or not, are taken into acount.

Disregarding polarization indices and using dimensionless quantities, we can write

$$\bar{v}_r(T) \equiv v_r(T)/\lambda^{1/2} q_c, \qquad g(T) \equiv \frac{1}{4\pi^2}(U k_B T/\lambda^2 q_c), \tag{12}$$

where λ corresponds to an average of the λ_j, then the relation (11) can be written in the form

$$\bar{v}_r^2(T) \sim g(T_c)\left[\tau - \frac{\pi}{2} \frac{T}{T_c} \bar{v}_r(T) \right] + a(T/T_c)\, \tau. \tag{11'}$$

From (11'), it follows that

$$\bar{v}_r^2 \sim \tau^2, \qquad \bar{v}_r << g(T_c), \tag{13a}$$

$$\bar{v}_r^2 \sim \tau, \qquad \bar{v}_r >> g(T_c), \tag{13b}$$

hold, where (13b) corresponds to the mean field. result.

The critical behavior of the static suceptibility is given by

$$\chi(q_r, 0) \sim 1/\tau^2, \qquad \bar{v}_r << g(T_c), \tag{14a}$$

$$\chi(q_r, 0) \sim 1/\tau, \qquad \bar{v}_r >> g(T_c). \tag{14b}$$

The critical coefficient γ would thus assume, in the critical region, the value 2 which is just in between the experimentally observed values[4].

3. Contribution of Chain Diagrams

From the invariance of the coupling function U, with respect to permutation of momenta, follows that higher order diagrams will involve the same $U_{jp,j'p'}$ as the Hartree term and the additional ones. Thus, however small $g(T)$ is, higher order diagrams will influence the critical behaviour the closer one comes to T_c because all diagrams, except the first order already considered, will diverge at T_c when involving only soft mode propagators. In particular, the second order self energy diverges as ln v, and, the third order, as $1/v_r$. The contribution of the chain diagrams can easily be summed up and one obtains

$$U \to \Gamma^{ch}(Q/q_c) \sim U\left\{1 + \frac{3}{2}\pi g(T)\frac{q_c}{Q}\left(\pi - 2\arctan\frac{2\bar{v}_r}{Q}\, g_c\right)\right\}^{-1}; \tag{15}$$

where only repeated scattering between modes of one branch has been considered and Q is the total momentum transfered by the chain. In the same way as before, one obtains now, instead of (11′),

$$\begin{aligned}\bar{v}_r^2(T) &\sim g(T_c)\left[\tau - \frac{\pi}{2}\,\frac{T}{T_c}\bar{v}_r(T)\right] \\ &-\frac{20}{27}g^2(T_c)\left\{\frac{T^2}{T_c^2}\ln[\bar{v}_R(T) + (10/3)\,g(T)]^{-1} - \ln[(10/3)\,g(T_c)]^{-1}\right\} + \\ &+ a(T/T_c)\tau.\end{aligned} \tag{16}$$

For $\bar{v}_r(T) << g(T_c)$, one obtains

$$\begin{aligned}\bar{v}_r^2(T) &\sim g(T_c)\left[\tau - \frac{\pi}{2}\,\frac{T}{T_c}\bar{v}_r(T)\right] \\ &- \frac{\pi^2}{6}g^2(T_c)\left[2\ln\frac{2}{3\pi^2 g(T_c)}\cdot\frac{T-T_c}{T} - \frac{1}{3\pi}\left(\frac{T}{T_c}\right)^2\frac{\bar{v}_r}{g(T)}\right. \\ &\left.- \frac{4}{9\pi^2}\,\frac{\bar{v}_r}{g^2(T)}\ln\frac{2}{3\pi^2 g(T)}\right]\end{aligned} \tag{17}$$

It follows, from (17), that the linear terms in $\bar{v}_r(T)$ have the same order of magnitude coefficient for Hartree and chain diagrams and the earlier result will be recovered. It is clear, however, that when the diagrams neglected are taken into account properly, the value of γ will change.

It has been shown in Ref. 10 that the dynamics of the soft phonon field is governed by an effective Lagrangian in which the effect of the acoustic phonons is incorporated in the new bare coupling functions. Furthermore, it has been shown that the effective Lagrangian defines a Heisenberg-type problem with a finite coupling constant. The critical coefficient γ of the latter problem assumes the value 1.375. In this, considerably more exact treatment one obtains thus a value of γ which compares well with EPR linewidth measiirements in $SrTiO_3$ (Ref. 5).

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