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Ultrasonic Attenuation at Structural Transitions above T_C

F. Schwabl

Institut für Festkörperforschung KFA, D-517 Jülich, Germany

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The anomalies of ultrasonic attenuation at structural transitions are studied theoretically by the mode-coupling theory. In situations like the 110 °K transition of $SrTiO_3$, where a central resonance couples to the order-parameter fluctuations, two regimes with different critical anomalies, are found. In addition, the attenuation depends on the dimensionality of the correlations The attenuation is anisotropic and depends on the polarization of the sound wave. Anomalies in the sound velocity are predicted. We also consider the sound attenuation at structural transitions other than in ABO_3 perovskites.

I. INTRODUCTION

Ultrasonic attenuation has proven to be very important for studying the dynamical behavior near second-order transitions.[1,2] Recently, the present author proposed a dynamical theory[3,4] for the structural transitions in ABO_3 perovskites from a cubic high-temperature phase to a tetragonal or trigonal low-temperature phase in which adjacent BO_6 octahedra are rotated in an alternating way. On the basis of this theory neutron scattering and the EPR linewidth were discussed.[3,4] The anomaly in the ultrasonic attenuation of longitudinal sound in the [100] direction was related to the critical behavior of the static susceptibility.[3] In this paper, this result is rederived by the mode-coupling theory; sound propagation in arbitrary directions and with an arbitrary polarization is considered. It will be shown that the coefficient of attenuation is anisotropic. Also the critical shift of the sound velocity depends on the direction and on the polarization. The results hold for a well-defined propagating soft mode and for an overdamped soft mode.

In the most general case the spatial correlations of the order parameter are three dimensional close to the critical temperature T_C and two dimensional further away from T_C. The crossover reflects itself in the ultrasonic attenuation. Not all systems will show this crossover but instead only one of these regimes may be exhibited.

The central resonance reflects itself not only in the neutron cross section but also in the ultrasonic attenuation. As has been shown in Papers I and II, far away from T_C (regime A) the frequency of the propagating part of the soft mode decreases and the strength of the central resonance rises as the square of the susceptibility. There is a second regime close to T_C, where the width of the central resonance shrinks like the inverse of the susceptibility, and the propagating part of the soft mode stays at a finite frequency. In this region the dynamic form factor is almost saturated by the central resonance and dynamical scaling holds. (The case of a completely overdamped soft mode is considered as well.) These two regimes are also characterized by different critical exponents of the sound attenuation. We shall see that sound attenuation offers a possibility to determine the width of the central resonance, which so far has not been possible by neutron scattering.

In Paper I a scaling argument was used to derive the anomaly of sound attenuation. Here mode-coupling theory[5-7] is employed. The damping of the ultrasonic wave results from the decay of the sound wave into two order-parameter fluctuations. The resulting attenuation coefficient is an average of the relaxation times of the order-parameter fluctuations of different $\vec{k}$. As near other critical points, not only the attenuation but also the velocity of sound shows an anomaly near T_C.

The main emphasis of this paper is on structural transitions such as the 105 °K transition in $SrTiO_3$. However, many of the results can easily be extended to other transitions, where the soft mode is an optic mode, as noted in Sec. V.

In Sec. II A the results of Paper I which will be needed in the following are summarized. In Sec. II B the general expression for the coefficient of sound attenuation is derived. The attenuation coefficient is evaluated by the mode-coupling theory in Secs. III and IV. In Sec. III longitudinal waves in the [100] direction are considered, in Sec. IV A, transverse waves in the [100] direction, and in Sec. IV B general directions are discussed. In Sec. V the application of the theory to other transitions is discussed briefly, and in Sec. VI the results are summarized.[8]

II. BASIC RELATIONS

A. Dynamical Theory

As in I the staggered rotation angles of the BO_6 octahedra are characterized by $\hat{\phi}^\alpha_{\vec{q}} = (N)^{-1/2} \times \sum_{\vec{l}} e^{-i(\vec{q}+\vec{q}_R)\vec{l}} \phi^\alpha_{\vec{l}}$, where $\phi^\alpha_{\vec{l}}$ is the angle of rotation of the octahedron at lattice site $\vec{l}$ around the Cartesian axis $\vec{n}^\alpha$ ($\alpha = 1, 2, 3$) and N is the number of unit cells. In terms of the lattice constant a, $\vec{q}_R$ is given by $\vec{q}_R = (\pi, \pi, \pi)a^{-1}$. In the low-temperature phase, $\langle \hat{\phi}^\alpha_0 \rangle$ is finite for at least one value of α and, therefore, constitutes the order parameter. (Note that the wave vector is measured relative to the R point in all quantities referring to the rotational degrees of freedom, but, of course, not for the acoustic phonons introduced below.)

For $T > T_C$ the dynamical susceptibility of $\hat{\phi}^\alpha_{\vec{q}}$ is given by[3,4]

$$\chi^{\alpha\alpha}(\vec{q}, \omega) = \chi^{\alpha\alpha}(\vec{q}) \frac{-[\omega^\alpha_0(\vec{q})]^2}{\omega^2 - [\omega^\alpha_0(\vec{q})]^2 + i\omega\Gamma^\alpha(\vec{q}, \omega)}, \tag{2.1}$$

where

$$\omega^\alpha_0(\vec{q}) = [\chi^{\alpha\alpha}(\vec{q}) I^\alpha(\vec{q})]^{-1/2}. \tag{2.2}$$

Here $\chi^{\alpha\alpha}(\vec{q})$ is the static susceptibility of $\hat{\phi}^\alpha_{\vec{q}}$ and $I^\alpha(\vec{q})$ is the static susceptibility of $P^\alpha_{\vec{q}}$. The damping function $\Gamma^\alpha(\vec{q}, \omega)$ is given by[3,4]

$$\Gamma^\alpha(\vec{q}, \omega) = \{i[b^\alpha(\vec{q})]^2/[\omega + i\gamma^\alpha(\vec{q})]\} + \sigma^\alpha(\vec{q}). \tag{2.3}$$

In (2.3) $b^\alpha(\vec{q})$ and $\gamma^\alpha(\vec{q})$ are uncritical, they remain finite for $q \to 0$ and $\epsilon \to 0$.

Under the condition $\omega^\alpha_s(\vec{q}) \gg \gamma^\alpha(\vec{q})$ and $\gamma^\alpha(\vec{q})\sigma^\alpha(\vec{q}) \ll [\omega^\alpha_s(\vec{q})]^2$, there is a central resonance of width

$$\Gamma^\alpha_c(\vec{q}) = \gamma^\alpha(\vec{q}) [\omega^\alpha_0(\vec{q})/\omega^\alpha_s(\vec{q})]^2. \tag{2.4}$$

Two situations can be realized in nature. (i) For $\sigma^\alpha(\vec{q}) \ll \omega^\alpha_s(\vec{q})$, the soft mode decomposes into the central resonance and a propagating part. The propagating part has the dispersion

$$\omega^\alpha_\pm(\vec{q}) = \pm\, \omega^\alpha_s(\vec{q}) - \tfrac{1}{2} i \Gamma^\alpha_s(\vec{q}), \tag{2.5}$$

with

$$\omega^\alpha_s(\vec{q}) = \{[\omega^\alpha_0(\vec{q})]^2 + [b^\alpha(\vec{q})]^2\}^{1/2} \tag{2.6}$$

and

$$\Gamma^\alpha_s(\vec{q}) = \left(\frac{b^\alpha(\vec{q})}{\omega^\alpha_s(\vec{q})}\right)^2 \gamma^\alpha(\vec{q}) + \sigma^\alpha(\vec{q}). \tag{2.7}$$

For $\Gamma^\alpha_s \ll \omega^\alpha_s$, the dynamic form factor

$$S^{\alpha\alpha}(\vec{q}, \omega) = \int_{-\infty}^{\infty} dt\, e^{i\omega t} \tfrac{1}{2} \langle \hat{\phi}^\alpha_{\vec{q}}(t) \hat{\phi}^\alpha_{-\vec{q}}(0) \rangle,$$

may be written in the form[4]

$$S^{\alpha\alpha}(\vec{q}, \omega) = \omega(1 - e^{-\omega/T})^{-1} \chi^{\alpha\alpha}(\vec{q}) \left[\left(\frac{b^\alpha(\vec{q})}{\omega^\alpha_s(\vec{q})}\right)^2 \frac{\Gamma^\alpha_c(\vec{q})}{\omega^2 + [\Gamma^\alpha_c(\vec{q})]^2} + \left(\frac{\omega^\alpha_0(\vec{q})}{\omega^\alpha_s(\vec{q})}\right)^2 \frac{[\omega^\alpha_s(\vec{q})]^2 \Gamma^\alpha_s(\vec{q}) - \Gamma^\alpha_c(\vec{q}) \{\omega^2 - [\omega^\alpha_s(\vec{q})]^2\} [b^\alpha(\vec{q})/\omega^\alpha_0(\vec{q})]^2}{[\omega^2 - (\omega^\alpha_s(\vec{q}))^2]^2 + [\omega \Gamma^\alpha_s(\vec{q})]^2} \right]. \tag{2.8}$$

(ii) For $\sigma^\alpha(\vec{q}) \gg \omega^\alpha_s(\vec{q})$, the soft mode is overdamped, and we get a superposition of two central components,[4]

$$S^{\alpha\alpha}(\vec{q}, \omega) = \omega(1 - e^{-\omega/T})^{-1} \chi^{\alpha\alpha}(\vec{q}) \times \left[\left(\frac{b^\alpha(\vec{q})}{\omega^\alpha_s(\vec{q})}\right)^2 \frac{\Gamma^\alpha_c(\vec{q})}{\omega^2 + [\Gamma^\alpha_c(\vec{q})]^2} + \left(\frac{\omega^\alpha_0(\vec{q})}{\omega^\alpha_s(\vec{q})}\right)^2 \frac{\Gamma^\alpha_0(\vec{q})}{\omega^2 + [\Gamma^\alpha_0(\vec{q})]^2} \right], \tag{2.8'}$$

with $\Gamma^\alpha_0(\vec{q}) = [\omega^\alpha_s(\vec{q})]^2/\sigma$. This is still under the conditions that $\gamma^\alpha\sigma^\alpha \ll (\omega^\alpha_s)^2$ and $\gamma^\alpha(\vec{q}) \ll \omega^\alpha_s(\vec{q})$.

Since neighboring octahedra share an oxygen atom, within the plane perpendicular to the axis of rotation ($\vec{n}^\alpha$), these octahedra will be strongly coupled, though only weakly coupled in a direction parallel to the rotation axis. This, of course, implies that the $\vec{q}$ dependence of $\chi^{\alpha\alpha}(\vec{q})$ will be anisotropic[9] [i.e., $\chi^{\alpha\alpha}(\vec{q})$ depends only weakly on q^α].

Because of these almost planar correlations there will be a crossover from three-dimensional to two-dimensional behavior.[10] Such effects may be described by a crossover temperature ϵ_Δ. For $\epsilon \ll \epsilon_\Delta$, the system will behave three dimensionally and for $\epsilon \gg \epsilon_\Delta$, it will behave two dimensionally $[\epsilon = (T - T_C)/T_C]$.[11] (The ϵ_Δ of this paper should not be identified with the much less general ϵ_Δ of Papers I and II, defined from a generalized Ornstein-Zernike susceptibility.) Needless to say, the existence of one or both of these regimes in a particular substance depends on the magnitude of the anisotropy. In some cases the two-dimensional regime may lie within the classical region. Static critical exponents in the d-dimensional region will be characterized by a suffix d $(\gamma_d, \nu_d \ldots)$.

In the three-dimensional regime the static susceptibility is given by

$$\chi^{\alpha\alpha}(\vec{q}) = \epsilon^{-\nu_3(2-\eta_3)}\, \tilde{\chi}(q_\perp/\kappa, q_\parallel/\kappa) \quad \text{for } \epsilon \ll \epsilon_\Delta , \tag{2.9a}$$

where κ is the inverse of the correlation length $\kappa = \kappa_0 \epsilon^{\nu_3}$. The components of $\vec{q}$ parallel (perpendicular) to $\vec{n}^\alpha$ are denoted by $q_\parallel$ $(q_\perp)$.

In the two-dimensional region the dependence on $q_\parallel$ will be neglected:

$$\chi^{\alpha\alpha}(\vec{q}) = \epsilon^{-\nu_2(2-\eta_2)}\, \hat{\chi}(q_\perp/\kappa) \quad \text{for } \epsilon \gg \epsilon_\Delta , \tag{2.9b}$$

with $\kappa = \kappa_0' \epsilon^{\nu_2}$.

Similar crossover effects are expected in an infinite array of plane Ising layers, with an exchange constant which is much weaker between spins in different layers than between spins in the same layer.[10] However the octahedron system is more complicated since it consists of three intersecting sets of layers corresponding to the three components ϕ^α and there are interactions between $\phi^{(1)}$, $\phi^{(2)}$, and $\phi^{(3)}$. Thus, this system corresponds to three intersecting and interacting sets of Ising layers. This is stressed because it may imply that the critical exponents are *different* from two-dimensional Ising exponents in the two-dimensional regime. They may be closer to usual three-dimensional static exponents.

At this point one might make comparison with the planar antiferromagnet K_2NiF_4, which has critical exponents[12] $\gamma = 1.0 \pm 0.1$, $\eta = 0.4 \pm 0.1$, and $\nu = 0.57 \pm 0.05$ compared to Ising values $\frac{7}{4}$, $\frac{1}{4}$, and 1. But this system is, of course, totally different since it is almost an isotropic Heisenberg system.

Before beginning our calculation $I^\alpha(\vec{q})$ has to be introduced, and ϵ_b characterizing the second crossover mentioned in the Introduction has to be defined. For small $\vec{q}$, $I^\alpha(\vec{q})$ may be taken to be[13]

$$I^\alpha(\vec{q}) \equiv \langle P^\alpha_{\vec{q}}, P^\alpha_{\vec{q}} \rangle \approx I[1 - \tfrac{1}{8}a^2(q^2 - q_\alpha^2)] ,$$

which reduces at the R point to $I^\alpha(0) = I = \frac{1}{2}M_0 a^2$. Hence $I^\alpha(0)$ is equal to one-half of the moment of inertia of the octahedron. Here M_0 is the oxygen mass and a is the lattice constant. For the study of critical anomalies $I^\alpha(\vec{q})$ could be replaced by I. We employ the standard bracket notation $\langle A, B\rangle$ for the susceptibility of two operators A and B. The quantity ϵ_b is defined through

$$\omega_0^\alpha(0, \epsilon = \epsilon_b) = b^\alpha(0, \epsilon = \epsilon_b) .$$

For $\epsilon \ll \epsilon_b$ (region B of Paper II), the width of the central peak is proportional to $1/\chi^{\alpha\alpha}(\vec{q})$, and the dynamic form factor is almost saturated by the central resonance, hence dynamical scaling[14,15] is valid. For $\epsilon \gg \epsilon_b$ (region A of Paper II), the width of the central resonance is γ^α and its strength is proportional to χ^2. (See the discussion in Papers I and II and Table I of Paper II.) Whether ϵ_Δ is smaller or larger than ϵ_b depends on the magnitude of b^α and the magnitude of the anisotropy. Both cases can be realized in nature and will be discussed.

B. Interaction of the Acoustic Phonons and the Soft Mode

As in the case of magnetostriction, where the exchange energy between spins is modulated by the presence of an acoustic phonon, one can here speak of a modulation of the coupling parameters of the octahedra. To second order in $\hat{\phi}^\alpha_{\vec{k}}$, the most general Hamiltonian for the interaction of the acoustic phonon with the soft mode has the structure[13,16]

$$\begin{aligned} H_{\mathrm{ph,s}} = (N)^{-1/2} \sum_{\vec{k},\vec{q}} \{ & A g^{11}_{11}(\vec{q},\vec{k}) \hat{\phi}^{(1)}_{\vec{k}} \hat{\phi}^{(1)}_{-\vec{k}-\vec{q}} \\ & + B[g^{22}_{11}(\vec{q},\vec{k}) \hat{\phi}^{(2)}_{\vec{k}} \hat{\phi}^{(2)}_{-\vec{k}-\vec{q}} \\ & + g^{33}_{11}(\vec{q},\vec{k}) \hat{\phi}^{(3)}_{\vec{k}} \hat{\phi}^{(3)}_{-\vec{k}-\vec{q}}] \} \tilde{E}_{11}(\vec{q}) \\ & + 2C g^{12}_{12}(\vec{q},\vec{k}) \hat{\phi}^{(1)}_{\vec{k}} \hat{\phi}^{(2)}_{-\vec{k}-\vec{q}} \tilde{E}_{12}(\vec{q}) \\ & + \text{cyclic permutations} . \end{aligned} \tag{2.10}$$

The terms which are obtained by cyclic permutation of the suffixes (1, 2, 3) must be added to Eq. (2.10). In (2.10), $\tilde{E}_{ij}(\vec{q}) = N^{-1/2}\sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{l}} E_{ij}(\vec{l})$ is the Fourier transform of the displacement field $E_{ij}(\vec{l})$ and A, B, and C are coupling coefficients. The functions $g^{\alpha\beta}_{ij}(\vec{q},\vec{k})$ appearing in Eq. (2.10) determine the wave-number dependence of the interaction. They would be identically equal to 1 for a contact interaction between the phonon and the soft modes. For more general interactions the cubic symmetry implies that A, B, and C can be defined such that all $g^{\alpha\beta}_{ij}(\vec{q},\vec{k})$ appearing in (2.10) have the property $g^{\alpha\beta}_{ij}(0, 0) = 1$. Hence, A, B, and C determine the coupling in the limit of small wave numbers. For the model of Feder and Pytte the wave number dependence of $g^{\alpha\beta}_{ij}(\vec{q},\vec{k})$ can be inferred from Ref. 13. An explicit expression for $g^{\alpha\beta}_{ij}(\vec{q},\vec{k})$ will not be needed since it will turn out that the anomalous part of the attenuation depends solely on $g_{ij}^{\alpha\beta}(0,0)=1$. Hence, for comparison with experiment and for a first reading of the paper the coefficients $g_{ij}^{\alpha\beta}(\vec{q},\vec{k})$ could everywhere be replaced by one.

Ultrasonic attenuation has been previously studied by Pytte[13] using a factorization approximation and molecular-field-theory (MFT) exponents. In view of the existence of the central resonance[17] and the appearance of nonclassical critical fluctuations the theory of ultrasonic attenuation has been reexamined by the use of scaling arguments and the dynamical equations in Paper I. It is convenient to introduce normal coordinates $Q^\lambda(\vec{q})$ for the acoustic phonons in terms of which the displacement field is given:

$$E_{ij}(\vec{1})=\tfrac{1}{2}i\,(NM)^{-1/2}\sum_{\vec{q},\lambda}e^{i\vec{q}\,\vec{1}}\,[q_i e_j^\lambda(\vec{q})+q_j e_i^\lambda(\vec{q})]Q^\lambda(\vec{q}). \quad (2.11)$$

Here λ is the polarization index, $\vec{q}$ the wave vector, $\vec{e}^\lambda(\vec{q})$ the polarization vector, and M the mass of the unit cell. Then the interaction Hamiltonian can be written in the form:

$$H_{\text{ph},s}=\sum_{\vec{q},\lambda,i}iq_i f_i^\lambda(\vec{q})Q^\lambda(\vec{q}), \quad (2.12)$$

from which it follows that the complex attenuation coefficient of the wave $(\vec{q},\lambda)$ is given by[18]

$$\alpha_{\vec{q}}^\lambda(\omega)+i\beta_{\vec{q}}^\lambda(\omega)=\sum_{i,j}\frac{q_i q_j}{2c_\lambda(\vec{q})}\int_0^\infty dt\,e^{i\omega t}\langle f_i^\lambda(\vec{q},t)[f_j^\lambda(\vec{q},0)]^\dagger\rangle. \quad (2.13)$$

The real part $\alpha_{\vec{q}}^\lambda(\omega)$ gives rise to sound attenuation while the imaginary part $\beta_{\vec{q}}^\lambda(\omega)$ produces a dispersion of the sound-wave frequency. For longitudinal waves in the [100] direction, f in (2.13) is given by

$$f_i^l[(q,0,0)]=\frac{\delta_{i,0}}{(NM)^{1/2}}\sum_{\vec{k}}[Ag_{11}^{11}(\vec{q},\vec{k})\,\hat{\phi}_{\vec{k}}^{(1)}\hat{\phi}_{-\vec{k}-\vec{q}}^{(1)}+B(g_{11}^{22}(\vec{q},\vec{k})\hat{\phi}_{\vec{k}}^{(2)}\hat{\phi}_{-\vec{k}-\vec{q}}^{(2)}+g_{11}^{33}(\vec{q},\vec{k})\hat{\phi}_{\vec{k}}^{(3)}\hat{\phi}_{-\vec{k}-\vec{q}}^{(3)})], \quad (2.14)$$

and for transverse waves in the [100] direction

$$f_i^t[(q,0,0)]=\frac{\delta_{i,0}}{(NM)^{1/2}}\sum_{\vec{k}}Cg_{12}^{12}(\vec{q},\vec{k})\hat{\phi}_{\vec{k}}^{(1)}\hat{\phi}_{-\vec{k}-\vec{q}}^{(2)}, \quad (2.15)$$

where $\delta_{i,0}$ is the Kronecker symbol.

The Hamiltonian of the octahedron system contains an interaction term[13] $\sum_{\vec{1},\vec{1}'}V_{\alpha\beta}(\vec{1}-\vec{1}')\phi_{\vec{1}}^\alpha\phi_{\vec{1}'}^\beta$, with $\alpha\neq\beta$. It shall be considered in this paper that $V_{\alpha\beta}(\vec{1}-\vec{1}')$ is small. In this weak coupling case we neglect correlation functions of the type $\langle\hat{\phi}_{\vec{k}_1}^{(1)}\hat{\phi}_{\vec{k}_2}^{(1)}\hat{\phi}_{\vec{k}_3}^{(1)}\hat{\phi}_{\vec{k}_4}^{(2)}\rangle\approx 0$. Hence, the only decay processes of a longitudinal sound wave propagating in the [100] direction are those into two $\hat{\phi}_{\vec{k}}^\alpha$ with the same α, while a transverse wave can decay only into two modes with different rotation axes (Fig. 1). In the evaluation of the damping of transverse sound also the statistical independence of $\hat{\phi}_{\vec{k}}^\alpha$ and $\hat{\phi}_{\vec{k}}^\beta$ for $\alpha\neq\beta$ is assumed.

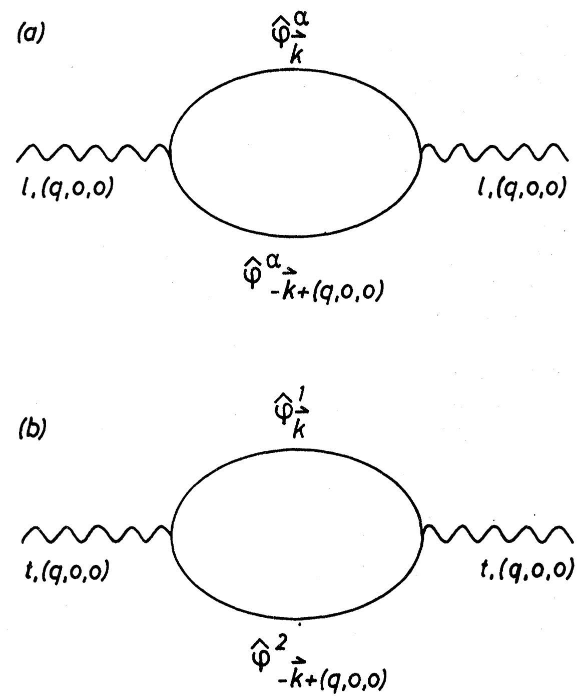

FIG. 1. Decay processes contributing to the damping of (a) longitudinal and (b) transverse acoustic mode in the [100] direction.

III. LONGITUDINAL SOUND IN THE [100] DIRECTION

A. Mode-Coupling Theory

For the attenuation coefficient $\alpha_{[100]}^l(\omega)$ of longitudinal waves in the [100] direction one finds from (2.13) and (2.14) $[\vec{q}=(q,0,0),\ \omega=c_l q]$

$$\alpha_{[100]}^l(\omega)=\{A^2\Lambda_{\vec{q}}^{(11)}(\omega)+B^2[\Lambda_{\vec{q}}^{(22)}(\omega)+\Lambda_{\vec{q}}^{(33)}(\omega)]\}/2c_l^3. \quad (3.1)$$

The four-point functions $\Lambda_{\vec{q}}^{(\beta\beta)}(\omega)$ are given in terms of the currents

$$J_{\vec{q}}^{\beta\beta}=\sum_{\vec{k}}g_{11}^{\beta\beta}(\vec{q},\vec{k})\,\hat{\phi}_{\vec{k}}^\beta\hat{\phi}_{-\vec{k}+\vec{q}}^\beta \quad (3.2)$$

by

$$\Lambda_{\vec{q}}^{(\alpha\beta)}(\omega)=(\omega^2/NM)\int_0^\infty dt\,e^{i\omega t}\langle J_{\vec{q}}^{\alpha\beta}(t)[J_{\vec{q}}^{\alpha\beta}(0)]^\dagger\rangle \quad (\alpha,\beta=1,2,3). \quad (3.3)$$

For $\vec{q}$ parallel to [100] only correlation functions of operators of the same kind are needed. [The currents containing different operators are defined after Eq. (4.1).]

The *mode-coupling theory*[6,7] is employed to evaluate $\Lambda_{\vec{q}}^{(\beta\beta)}(\omega)$, using arguments analogous to those in Kawasaki's theory for magnets.[6] To consider the decay of the acoustic mode into two soft modes—

one with wave number $\vec{k}$, the other with wave number $\vec{q}-\vec{k}$—the orthonormalized two-mode operators

$$\Phi^{\alpha\beta}_{\vec{k},\vec{q}-\vec{k}} \equiv [T\chi^{\alpha\alpha}(\vec{k})\chi^{\beta\beta}(\vec{k}-\vec{q})]^{-1/2}\,\hat{\phi}^{\alpha}_{\vec{k}}\,\hat{\phi}^{\beta}_{\vec{q}-\vec{k}} \; . \tag{3.4}$$

are introduced. The two-mode contribution to a general current $J^{\alpha\beta}_{\vec{q}}$ is determined by expanding this current in terms of these two-mode operators:

$$J^{\alpha\beta}_{\vec{q}} = \sum_{\vec{k},\sigma,\delta} \langle J^{\alpha\beta}_{\vec{q}},\, \Phi^{\sigma\delta}_{\vec{k},-\vec{k}+\vec{q}}\rangle\, \Phi^{\sigma\delta}_{\vec{k},-\vec{k}+\vec{q}} + \cdots \; . \tag{3.5}$$

The two-mode contribution to the time correlation function is then given by

$$\langle J^{\alpha\beta}_{\vec{q}}(t) J^{\alpha\beta}_{\vec{q}}(0)\rangle = \sum_{\vec{k},\sigma\delta} |\langle J^{\alpha\beta}_{\vec{q}},\; \Phi^{\sigma\delta}_{\vec{k},-\vec{k}+\vec{q}}\rangle|^2\,[T\chi^{\sigma\sigma}(\vec{k})\chi^{\delta\delta}(\vec{k}-\vec{q})]^{-1}$$

$$\times S^{\sigma\sigma}(\vec{k},\, t)\, S^{\delta\delta}(-\vec{k}+\vec{q},\, t) \; . \tag{3.6}$$

From now on the experimentally interesting case that the wave number q is much smaller than the inverse of the correlation length ($q \ll \kappa$) will be considered. Then the static expectation values appearing in Eq. (3.6) may be evaluated in the limit $q = 0$. The decay into two $\hat{\phi}^{\delta}_{\vec{k}}$ modes is determined by the matrix element

$$\lim_{q\to 0} \langle J^{\beta\beta}_{\vec{q}},\; \Phi^{\sigma\delta}_{\vec{k},\vec{q}-\vec{k}}\rangle = \delta^{\sigma\delta} D_{\beta\delta}\, [\chi^{\delta\delta}(\vec{k})]^{-1}\, \frac{\partial}{\partial T}\, \chi^{\delta\delta}(\vec{k}) \; , \tag{3.7}$$

where it has been noted that $J^{\beta\beta}_{0}$ scales like the energy. In Eq. (3.7) $D_{\beta\delta}$ are constants of proportionality. The diagonal terms $D_{\beta\beta}$, which are numerically most important, are assumed to be finite constants in the critical region. Thus, disregard here the possibility that $D_{\beta\beta}$ itself may contain a weak anomaly of the type ϵ^{α}, where α is the critical exponent of the specific heat, or more generally ϵ^{w} with a small exponent w.[6] Combining (3.3), (3.6), and (3.7) with $D^2 = \sum_\delta D^2_{\beta\delta}$ one obtains

$$\mathrm{A}^{(\beta\beta)}_0(\omega) = \frac{\omega^2}{\bar{\rho}}\, D^2 \int \frac{d^3k}{(2\pi)^3}$$

$$\times \left(\frac{\partial}{\partial T}\, \ln\chi(\vec{k})\right)^2 \mathrm{Re}\int_0^\infty dt\, e^{i\omega t} \left|\frac{S(\vec{k},\, t)}{\chi(\vec{k})}\right|^2 \; , \tag{3.8}$$

where $\bar{\rho}$ is the density and $S(\vec{k},\, t)$ is the Fourier transform of $S(\vec{k},\, \omega)$. Because of the cubic symmetry the index δ is omitted in the integrand, and $\mathrm{A}^{(\beta\beta)}_0(\omega)$ and D are independent of β. First the low-frequency limit will be considered.

B. Low-Frequency Regime $\omega \ll \Gamma_c(\kappa)$

For the case of a well-defined *propagating soft mode*, the leading contribution to the time integral in Eq. (3.8) is found by inserting (2.8):

$$\int_0^\infty dt\, |S(\vec{k},\, t)|^2 = \chi^2\left[\left(\frac{b}{\omega_s}\right)^4 \frac{1}{2\Gamma_C} + \frac{2b^2\omega_0^2\Gamma_s}{\omega_s^6} + \frac{1}{4}\left(\frac{\omega_0}{\omega_s}\right)^4 \left(\frac{2}{\Gamma_s} + \frac{\Gamma_s}{\omega_s^2}\right)\right] \; . \tag{3.9}$$

For brevity we omit the argument $\vec{k}$. We analyze (3.8) and (3.9) under the assumption that $\Gamma_s \approx \sigma \gg \gamma(b/\omega_s)^2$ and that σ is uncritical. The homogeneity of $\chi^{\alpha\alpha}(\vec{q})$ implies that $\partial/\partial T[\ln\chi^{\alpha\alpha}(\vec{q})]$ is of the form $\partial/\partial T[\ln\chi^{\alpha\alpha}(\vec{q})] = \epsilon^{-1}h(q_\perp/\kappa,\, q_\parallel/\kappa)$, where the function $h(q_\perp/\kappa,\; q_\parallel/\kappa)$ is readily found from (2.9a) or (2.9b).

The critical anomaly of the sound attenuation of longitudinal waves in [100] is characterized by an exponent ρ:

$$\alpha^{l}_{[100]}(\omega) \propto \mathrm{A}^{(\beta\beta)}_0(\omega) \propto \omega^2\epsilon^{-\rho} \; . \tag{3.10}$$

In the temperature regime $\epsilon \ll \epsilon_b$ (regime B of Paper II), one obtains from Eqs. (3.8)–(3.10) and (2.9a) in the three-dimensional region

$$\rho = \gamma_3 - 3\nu_3 + 2 \qquad \text{for} \quad \epsilon \ll \epsilon_\Delta,\ \epsilon_b \; , \tag{3.11a}$$

which, in agreement with Paper I, is equal to $\gamma_3 + \alpha_3$, if the static scaling law $3\nu_3 - 2 = -\alpha$ holds. For two-dimensional correlations one finds

$$\rho = \gamma_2 - 2\nu_2 + 2 = 2 - \nu_2\eta_2 \quad \text{for } \epsilon_\Delta \ll \epsilon \ll \epsilon_b \; . \tag{3.11b}$$

These power laws result from the first term in (3.9)(central-resonance term). For *Ising* exponents one finds $\rho \approx 1.25$ in the three-dimensional case and $\rho = 1.75$ in the two-dimensional case. However, for the reasons mentioned after Eq. (2.9b) the static critical exponents may be quite different from Ising exponents for $\epsilon > \epsilon_\Delta$. However, the critical exponents of ultrasonic attenuation for $\epsilon > \epsilon_\Delta$ are not very sensitive to the value of the static critical exponents. If the static exponents remain nearly three dimensional in the two-dimensional region, $\rho \approx 2$ for $\epsilon_\Delta \ll \epsilon \ll \epsilon_b$. The power law (3.11a) is in agreement with the value $\rho = 1.27 \pm 0.10$ found by Fossheim and Berre[19] in $SrTiO_3$ for $0.5 < T - T_C < 3\,^\circ$K. For $KMnF_3$, where ϵ_Δ is very small one expects a crossover from the three-dimensional law to the two-dimensional law further away.

In the limit $\epsilon \gg \epsilon_b$ (regime A of Paper II), the power law for three-dimensional correlations is[3]

$$\rho = 2\gamma_3 - 3\nu_3 + 2 \qquad \text{for} \quad \epsilon_b \ll \epsilon \ll \epsilon_\Delta \; , \tag{3.12a}$$

and for two-dimensional correlations it has the form

$$\rho = 2\gamma_2 - 2\nu_2 + 2 \qquad \text{for} \quad \epsilon_b,\ \epsilon_\Delta \ll \epsilon \; . \tag{3.12b}$$

The exponents for $\epsilon \gg \epsilon_b$ are larger than for $\epsilon \ll \epsilon_b$. With Ising exponents $\rho \approx 2.5$ for (3.12a) and $\rho = \frac{7}{2}$ for (3.12b). It depends on the magnitude of ϵ_b whether this regime is still in the *critical* region or not. For $\epsilon \gg \epsilon_b$ the third term in Eq. (3.9) makes a contribution to $\mathrm{A}^{(\beta\beta)}_0(\omega)$ of the order ϵ^0, which is numerically larger than the most singular terms (3.12a) and (3.12b). The fourth term in (3.9) gives a contribution proportional to $\approx \epsilon^{-\gamma_d}$ which is smaller as long as $(\omega_0^\alpha)^2\sigma\gamma^\alpha/4(b^\alpha)^4 \ll 1$. In the most general case, when both ϵ_Δ and ϵ_b lie well within the critical region there will be two crossovers. For ϵ_Δ

$\ll \epsilon_b$ the critical exponent ρ will change from (3.11a) to (3.11b) and to (3.12b) while for $\epsilon_b \ll \epsilon_\Delta$ the intermediate exponent is given by Eq. (3.12a).[20]

For the case of a central resonance plus an *overdamped soft mode* [Eq. (2.8′)] the same power laws (3.11a), (3.11b) and (3.12a), (3.12b) apply. The condition is again that the damping term σ^α is uncritical.

In Paper I a *scaling argument* was used to derive the power laws of sound attenuation: From (3.3) it can be seen that $A_{\vec{q}}^{(11)}(\omega)$ has the structure

$$A_{\vec{q}}^{(11)}(\omega) \propto \tau_r\, \epsilon^{-\alpha} \omega^2 ,$$

where τ_r is a typical relaxation time for $J_{\vec{q}}^{11}(t)$. $\{\tau_r \sim [\Gamma_c(0)]^{-1}$ for $\epsilon \ll \epsilon_b$, since then the central resonance dominates.$\}$ In the above relation for $A_{\vec{q}}^{(11)}(\omega)$ it has been noted that $\langle J_{\vec{q}}^{11}(0) J_{\vec{q}}^{11}(0)\rangle \sim \epsilon^{-\alpha}$ scales like the specific heat. A factorization of the four-point function $\langle J^{11}(t) J^{11}(0)\rangle$ entering (3.3) would violate this scaling property. Thus, if the time correlation function $\langle J^{11}(t) J^{11}(0)\rangle$ is factorized in order to determine τ_r and $A_{\vec{q}}^{(11)}$ for arbitrary ϵ, a scaling factor $\kappa^{-d-\alpha_d/\nu_d} f(\vec{k}/\kappa)[\chi^{11}(\vec{k})]^{-2}$ must be introduced to obtain the correct behavior for $t=0$. Then one finds

$$A_0^{(11)}(\omega) = (\omega^2/NM)\,\mathrm{Re} \int_0^\infty dt\, e^{i\omega t}$$
$$\times \sum_{\vec{k}} \kappa^{-d-\alpha_d/\nu_d} f(\vec{k}/\kappa)\,[\chi^{11}(\vec{k})]^{-2} |S^{11}(\vec{k}, t)|^2 . \quad (3.8')$$

If the static scaling law $d\nu_d = 2 - \alpha_d$ is valid, the power laws obtained from (3.8′), are the same as (3.11a), (3.11b), and (3.12a), (3.12b).

C. Range of Validity of the Power Laws

One may readily see from Eq. (3.8) that the divergence of the ultrasonic attenuation for $\epsilon \ll \epsilon_b$ does not continue right to the critical point. For arbitrary frequency ω the central resonance contributes

$$A_{\vec{q}}^{(\beta\beta)}(\omega) = \frac{\omega^2}{\bar{\rho}} D^2 \int \frac{d^3k}{(2\pi)^3}$$
$$\times \left(\frac{\partial}{\partial T} \ln\chi(\vec{k})\right)^2 \left(\frac{b(\vec{k})}{\omega_s(\vec{k})}\right)^2 \frac{2\Gamma_c(\vec{k})}{[2\Gamma_c(\vec{k})]^2 + \omega^2} . \quad (3.13)$$

Equation (3.13) combined with (2.9a) shows that for $\epsilon \ll \epsilon_b$, ϵ_Δ:

$$A_{\vec{q}}^{(\beta\beta)}(\omega) = \omega^2 \epsilon^{-2+3\nu_3-\gamma_3} f\left(\frac{\omega}{\Gamma_c(\kappa)}\right) , \quad (3.13')$$

where the dimensionless function $f(x)$ can be found from Eq. (3.13). Hence, the power law (3.10) applies only for $\omega \ll \Gamma_c(\kappa)$. The condition $q \ll \kappa$, which was used in the evaluation of the static correlation functions, is *less* restrictive than the condition $\omega \ll \Gamma_c(\kappa)$, and thus it is consistent to keep ω finite while letting $q \to 0$.

On approaching T_c the sound attenuation is first described by the power law (3.10). Since $\Gamma_c(\kappa) \sim \kappa^{2-\eta}$ decreases, the coefficient of attenuation reaches a maximum for $2\Gamma_c(\kappa) \approx \omega$. Hence, one could get an estimate for $\Gamma_c(\kappa)$ from ultrasonics and from that an estimate of γ, since b and ω_0 are known from neutron scattering. This *intrinsic* rounding effect at $\Gamma_c(\kappa) \approx \frac{1}{2}\omega$ should be observable in very pure samples.

D. Sound Velocity

One may readily show that the sound velocity c_l is shifted by Δc_l:

$$\frac{\Delta c_l}{c_l} = - \langle J_0^{11}(0) J_0^{11}(0)\rangle \frac{(A^2 + 2B^2)}{(2c_l MT)} \propto -\epsilon^{-\alpha} , \quad (3.14)$$

which is proportional to the negative of the specific heat since $J^{11}(0)$ scales like the energy.[3,21] In specific-heat measurements only the MFT-like discontinuity and no fluctuation contribution has been observed.[22] In the extension of the theory to $T < T_C$, the resonant interaction[13] must be taken into account and must be reexamined with nonclassical critical exponents. (See the note added in proof at the end of the paper.)

IV. GENERAL DIRECTIONS AND POLARIZATIONS

A. Transverse Sound in the [100] Direction

1. Sound Attenuation

From (2.15) it is seen that the coefficient of attenuation of a transverse wave in the [100] direction $\alpha^t_{[100]}(\omega)$, is given by

$$\alpha^t_{[100]}(\omega) = C^2 A_{\vec{q}}^{(12)}(\omega)/2c_t^3 , \quad (4.1)$$

where $A_{\vec{q}}^{(12)}(\omega)$ is found from Eq. (3.3) by inserting $J_{\vec{q}}^{(12)} = \sum_{\vec{k}} g_{12}^{12}(\vec{q}, \vec{k})\, \hat{\phi}_{\vec{k}}^{(1)} \hat{\phi}_{-\vec{k}-\vec{q}}^{(2)}$. In the weak coupling case, where the correlation between modes with different rotation axes can be neglected, the only important decay process is that into a $\hat{\phi}^{(1)}$ and a $\hat{\phi}^{(2)}$ mode [Fig. 1(b)].

$$J_{\vec{q}}^{12} = \langle J_{\vec{q}}^{12}, \Phi_{\vec{k},-\vec{k}+\vec{q}}^{12}\rangle\, \Phi_{\vec{k},-\vec{k}+\vec{q}}^{12} + \text{higher modes}. \quad (4.2)$$

Assuming the *statistical independence* of $\hat{\phi}_{\vec{k}}^{(1)}$ and $\hat{\phi}_{-\vec{k}}^{(2)}$ one gets

$$\lim_{q\to 0} \langle J_{\vec{q}}^{12}, \Phi_{\vec{k},-\vec{k}+\vec{q}}^{12}\rangle = [T g_{12}^{12}(0, \vec{k})\, \chi^{11}(\vec{k})\, \chi^{22}(\vec{k})]^{1/2}. \quad (4.3)$$

The statistical independence is not automatically implied by the smallness of the interaction $V_{\alpha\beta}(\vec{l} - \vec{l}')$, $\alpha \neq \beta$, but depends also on the fourth-order interaction terms of the type $\hat{\phi}_{\vec{k}_1}^{(1)} \hat{\phi}_{\vec{k}_2}^{(1)} \hat{\phi}_{\vec{k}_3}^{(2)} \hat{\phi}_{\vec{k}_4}^{(2)} \times U(\vec{k}_1, \vec{k}_2, \vec{k}_3, \vec{k}_4)$. Hence, the results for $\alpha^t_{[100]}(\omega)$, $A_0^{(12)}(\omega)$, and Δc_t (defined below) are less general than those of Sec. III. Combining Eqs. (3.3), (4.2), and (4.3) gives

$$A_0^{(12)}(\omega) = \frac{\omega^2}{\bar{\rho}} \operatorname{Re} \int_0^\infty dt\, e^{i\omega t} \int \frac{d^3\vec{k}}{(2\pi)^3} S^{11}(\vec{k}, t) S^{22}(\vec{k}, t) \,. \tag{4.4}$$

It is not surprising that one gets from the mode-coupling theory in this case just a factorization of (3.3). Inserting Eqs. (2.9) into (4.4) one finds for the contribution of the central resonance to $A_0^{(12)}$:

$$A_0^{(12)}(\omega) = \frac{\omega^2 T^2}{\bar{\rho}} \operatorname{Re} \int \frac{d^3k}{(2\pi)^3} \times \chi^{11}(\vec{k})\chi^{22}(\vec{k}) \frac{b^{(1)}(\vec{k}) b^{(2)}(\vec{k}) / [\omega_s^{(1)}(\vec{k})\omega_s^{(2)}(\vec{k})]}{\Gamma_c^{(1)}(\vec{k}) + \Gamma_c^{(2)}(\vec{k}) + i\omega} \,. \tag{4.5}$$

Defining the critical exponent of $A_0^{(12)}(\omega)$ by ρ'

$$\alpha^t_{[100]}(\omega) \propto A_0^{(12)}(\omega) \propto \omega^2 \epsilon^{-\rho'} \,, \tag{4.6}$$

one finds from (4.5) in the low-frequency regime $[\omega \ll \Gamma_c(\kappa)]$:

$$\rho' = 3(\gamma_d - \nu_d) \quad \text{for } \epsilon \ll \epsilon_b \,. \tag{4.7}$$

Since modes with different spatial anisotropy enter Eq. (4.5), there is no extra factor $\epsilon^{-\nu_2}$ for $\epsilon > \epsilon_\Delta$. For three- and two-dimensional Ising exponents one obtains $\rho' \approx 2$ and $\rho' = \frac{9}{4}$, respectively. [See the remarks after Eq. (3.11b).] In the temperature region $\epsilon \gg \epsilon_b$ one finds from Eq. (4.5)

$$\rho' = 4\gamma_d - 3\nu_d \quad \text{for } \epsilon \gg \epsilon_b \,. \tag{4.8}$$

The exponent ρ' Eqs. (4.7) and (4.8) is larger than ρ. Larger exponents for transverse waves in the [100] direction have already been found experimentally by Rehwald[23] in $SrTiO_3$. It has been noted in Paper I and in Ref. 24 that the interaction of the sound wave with two different rotational modes can be responsible for these effects.

2. Velocity of Sound

For the critical change in the velocity of sound one obtains

$$\frac{\Delta c_t}{c_t} = -\langle J_0^{12}(0) J_0^{12}(0) \rangle\, C^2/(2c_t MT) \,. \tag{4.9}$$

Using the statistical independence of modes 1 and 2 one gets

$$\frac{\Delta c_t}{c_t} \propto -\sum_{\vec{k}} \chi^{11}(\vec{k})\chi^{22}(\vec{k}) \,, \tag{4.9$'$}$$

which implies that

$$\frac{\Delta c_t}{c_t} \propto -\epsilon^{-\nu_d(1-2\eta_d)} \,. \tag{4.9$''$}$$

B. General Directions and Polarizations

From Eqs. (2.10) and (2.13) the attenuation of an acoustic wave with wave vector $\vec{q}$, polarization $\vec{e}$, and frequency ω is found to be

$$\alpha_{\vec{q}}^{\vec{e}}(\omega) = \tfrac{1}{2}[G_1(\vec{q}, \vec{e}) A_0^{(11)}(\omega) + G_2(\vec{q}, \vec{e}) A_0^{(12)}(\omega)] \,. \tag{4.10}$$

The coefficients $G_i(\vec{q}, \vec{e})$ are given for some longitudinal (l) and some transverse (t) waves in Table I (c_l and c_t depend, of course, on the direction). In Eq. (4.10) it has been noted that the system has cubic symmetry and that $A_{\vec{q}}^{(\alpha\beta)}(\omega)$ are independent of the direction of $\vec{q}$ in the limit $\vec{q} \to 0$. The only directional dependence comes through the coupling coefficients $G_i(\vec{q}, \vec{e})$. In the derivation of (4.10) one also recognizes that the singular part of the attenuation is determined by $g_{ij}^{\alpha\beta}(0, 0) = 1$.

Combining Eqs (3.10), (4.6), and (4.10) one gets

$$\alpha_{\vec{q}}^{\vec{e}}(\omega) = \omega^2 [G_1(\vec{q}, \vec{e}) r_1 \epsilon^{-\rho} + G_2(\vec{q}, \vec{e}) r_2 \epsilon^{-\rho'}] \,. \tag{4.11}$$

The coefficients r_i are uncritical. It would be interesting to check whether one can fit data on $SrTiO_3$ [19,23–25] with Eq. (4.11). The coefficients r_i are different in the various temperature regimes. The coefficients in the two-dimensional regime $r_i^{(2)}$ are related to the coefficients in the three-dimensional regime $r_i^{(3)}$ by $r_1^{(2)}/r_1^{(3)} \sim \epsilon_\Delta^{\rho_2 - \rho_3}$ and $r_2^{(2)}/r_2^{(3)} \sim \epsilon_\Delta^{\rho_2' - \rho_3'}$.

An equation similar to (4.11) can also be derived for the shift of the velocity of sound. For systems where (4.11) does not hold, this will be an indication that the weak coupling limit and (or) the statistical independence of $\phi^{(1)}$ and $\phi^{(2)}$ do not apply. In the strong coupling case, where the interaction between different modes is non-negligible, the factorization (4.3) is no longer valid. If the second-order coupling between different modes is large, J^{12} scales like the energy, and one expects the critical exponent ρ' to have the form $\rho' = \gamma_d - 3\nu_d + 2$. In a systematic theory one has to transform from $\phi_{\vec{q}}^\alpha$ to new orthogonal variables.

V. GENERALIZATION TO OTHER TRANSITIONS

The results of the Secs. II-IV can be generalized immediately to other transitions. As emphasized in Paper II the dynamical equations (2.1) and (2.2) hold generally for soft optic modes characterized by conjugate variables $\phi_{\vec{q}}$ and $P_{\vec{q}}$, where $\chi(\vec{q})$ is the susceptibility of the order parameter and $I(\vec{q})$ (the mass density) is the susceptibility of $P_{\vec{q}}$. Since the case of a central resonance has already been discussed, systems are considered where no slow

TABLE I. Coefficients $G_i(\vec{q}, \vec{e})$.

$\vec{q}$	$G_1(\vec{q}, \vec{e})$	$G_2(\vec{q}, \vec{e})$
l [100]	$(A^2 + 2B^2)/c_l^3$	0
l [110]	$[(A+B)^2/2 + B^2]/c_l^3$	C^2/c_l^3
l [111]	$(A+2B)^2/3c_l^3$	$4\, C^2/3\, c_l^3$
t [100]	0	C^2/c_t^3
t [110]		
($\vec{e} \parallel [1\bar{1}0]$)	$(A-B)^2/2\, c_t^3$	0

variable couples to the equations of motion, and hence, the first term in (2.5) is absent and there is no central resonance.

Interactions of the following type are considered:

$$H_{\text{int}} = \sum_{\vec{l},\vec{l}',\vec{l}''} C_\alpha(\vec{l},\vec{l}',\vec{l}'')\phi_{\vec{l}}\phi_{\vec{l}'}u^\alpha_{\vec{l}''}\,, \qquad (5.1)$$

where $u^\alpha_{\vec{l}}$ is the displacement field and $C_\alpha(\vec{l},\vec{l}',\vec{l}'')$ is a short-ranged interaction potential. If the damping coefficient σ is characterized by an exponent $\zeta(\sigma \propto \epsilon^{-\zeta})$, the critical exponent for ultrasonic attenuation found from Eq. (3.9) (with $b^\alpha = 0$) is

$$\rho = \gamma_d + 2 - d\nu_d + \zeta\,. \qquad (5.2)$$

If the dimensional scaling law $d\nu_d = 2 - \alpha$ is valid, ρ can be represented by $\rho = \gamma_d + \alpha + \zeta$. If dynamical scaling would hold for the soft mode, one would have $\zeta = -\frac{1}{2}\gamma_d$ and $\rho = \frac{1}{2}\gamma_d + \alpha$. However, we expect that the damping of optic soft modes arises mainly from interaction with uncritical acoustic modes so that $\zeta = 0$.

In the molecular-field-theory regime one obtains from Eq. (5.2) $\rho = 3 - \frac{1}{2}d$. This result should apply for the ferroelectric transition in $BaTiO_3$, at which a transverse optic mode softens at the Γ point. Since the correlations are one dimensional in $BaTiO_3$,[26] one expects $\rho = \frac{5}{2}$.

In cubic systems there will be several soft modes. The coupling of a sound wave to different modes can be treated as in Sec. IV.

VI. CONCLUDING REMARKS

The different temperature regimes apparent in the dynamic form factor reflect themselves in the coefficient of sound attenuation. For systems with a central resonance the critical exponents are summarized in Table II. In coordinate space the interaction of the sound wave with the fluctuations of the order parameter considered in this paper is of the form

$$H_{\text{int}} = \sum_{\vec{l},\vec{l}',\vec{l}''} C_{ij\alpha\beta}(\vec{l},\vec{l}',\vec{l}'')e_{ij}(\vec{l})\phi^\alpha_{\vec{l}'}\phi^\beta_{\vec{l}''}$$

with short-range coefficients $C_{ij\alpha\beta}(\vec{l},\vec{l}',\vec{l}'')$.

The terms with $\alpha = \beta$ make a contribution to the sound attenuation which is characterized by the exponent ρ. The terms with $\alpha \neq \beta$ lead to the critical exponent ρ'.

The regime $\epsilon > \epsilon_b$ will lie in the nonclassical critical region only for cases where b is very small. In $SrTiO_3$, for example, $\epsilon_b \approx 10^{-1}$, which is probably too large. If both ϵ_Δ and ϵ_b are small, there will be a dimensionality crossover at ϵ_Δ and a crossover due to the change in the characteristic temperature variation of the critical mode at ϵ_b. Depending on the material, ϵ_Δ may be smaller or larger than ϵ_b.

The width of the central resonance Γ_c, though not accessible to neutron scattering measurements up to now, should be measurable by an ultrasonic attenuation experiment. By comparing with the general formula (3.13), $\Gamma_c(\kappa)$ could be determined. Also, in a manner similar to EPR experiments, one could determine the ratio $(b^\alpha)^2/\gamma^\alpha(\omega_0^\alpha)^2$ from the amplitude of the attenuation coefficient in the region $\epsilon \ll \epsilon_b$ [see Eq. (3.9)]. Since b^α and ω_0^α can be found by neutron scattering one can estimate γ^α from both quantities.[20] The divergence of the ultrasonic attenuation is larger the lower the dimensionality of the correlations. On approaching T_C the critical exponent ρ becomes smaller at each of the crossover points.

At other structural transitions, ultrasonic propagation also gives valuable insight into the dynamics of the critical fluctuations.

Some of the approximations used in this paper are listed here. First, b^α, γ^α, and σ^α have been taken to be uncritical constants. The comparison of the results with experiments could test this assumption and could give insight into a possible temperature dependence of the parameters characterizing the central resonance. Second, the statistical independence of modes with different rotation axes was used, which need not be equally well satisfied in all substances. This would effect the results for transverse waves in the [100] direction and those results which contain the coefficient $A_0^{(12)}(\omega)$ but not the results for longitudinal waves in the [100] direction.

It is hoped, at least, that the experimental test of this theory could give some insight into how the theory has to be improved or modified if necessary.

Note added in proof. It should be mentioned that $I(\vec{q})$, the total intensity of light scattered with momentum transfer $\vec{q}$, also will show anomaly like the specific heat above T_C. Since light couples to $(\phi^\sigma_{\vec{l}})^2$, $I(\vec{q})$ contains a contribution $I_c(\vec{q}) \propto \sum_{\vec{l}} e^{-i\vec{q}\cdot\vec{l}} \times \langle(\phi^\sigma_{\vec{l}})^2(\phi^\sigma_{\vec{l}})^2(\phi^\sigma_0)^2\rangle$. The same scaling argument as in Sec. III D shows that $I_c(\vec{q}) \propto \epsilon^{-\alpha}$ for $\xi^{-1} > \frac{1}{2}q$. Such a weak singularity is indicated in experiments by Steigmeier, Auderset, and Harbeke (report of work prior to publication).

TABLE II. Critical exponents for ultrasonic attenuation.

ϵ	ρ	ρ'
$\epsilon < \epsilon_b$ (*B*)	$\gamma_d - d\nu_d + 2$	$-3\nu_d + 3\gamma_d$
$\epsilon > \epsilon_b$ (*A*)	$2\gamma_d - d\nu_d + 2$	$-3\nu_d + 4\gamma_d$

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Numerical Evidence for the Existence of a Phase Transition in a Two-Dimensional Exchange-Interaction Model

H. K. Charles, Jr. and R. I. Joseph
Department of Electrical Engineering, The Johns Hopkins University, Baltimore, Maryland 21218
(Received 27 April 1972)

High-temperature–low-field susceptibility series for the exchange-interaction model of ferromagnetism are analyzed by means of a reexpansion technique to obtain the ordering temperature. In particular, for all two-sublattice decomposible structures (e.g., linear chain, plain square, simple cubic, body-centered cubic) the formula $k_B T_C/J = (12/5)(z - 5/2)(4S+3)^{-1}$ reproduces our numerical results.

I. INTRODUCTION

Bogoliubov-type arguments have been used to rule out the possibility of a "phase transition" in two-dimensional lattices for a wide class of isotropic interaction Hamiltonians.[1] On the other hand, Stanley and Kaplan[2] have shown, using high-temperature series, that the numerical evidence for a phase transition in the two-dimensional (2-d) Heisenberg model (HSB) is just as convincing as for the three-dimensional (3-d) case. The major criticism of their result seems to have been that the available series are too short to yield reliable evidence for 2-d systems. A theoretical resolution to this dilemma has been proposed by Stanley and Kaplan[3] who point out that one must be clear in distinguish-