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Theory of Coupled Hydrodynamic Modes Applied Above Structural Phase Transitions

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Abstract. Starting from the fluctuating part of the lattice free energy the coupling between a soft shear mode and the heat diffusion mode is established. The soft mode response function is derived and compared with neutron scattering data for the two cases of a soft staggered (R-corner) mode ($SrTiO_3$, $LaAlO_3$) and a soft sound (zone centre) mode (Nb_3Sn). In an appendix the classical form of the fluctuation–dissipation theorem is derived in the framework of a classical many-body theory.

1. Introduction

In this paper we wish to demonstrate the fruitfulness of the hydrodynamic description of many-body systems by developing a purely classical many-body theory and applying it to the particular cases of the coupled modes discussed earlier [1].

Examples of couplings between hydrodynamic modes have been known for a long time. The most common and physically the most important example is the thermal expansion coupling between the isothermal sound mode $\omega = (1 - i\omega\tau_0/2)vq$ and the heat diffusion mode $\omega = -iDq^2$ which occurs in practically all hydrodynamic systems (see the review of Ref. [2]). This coupling gives rise to the transition from isothermal to adiabatic sound (velocity v_0) and to the occurrence of a central ($\omega = 0$) peak of width Dq^2 in inelastic light scattering, the socalled Landau–Placzek peak [2].

Another example is sound in a molecular liquid which couples to internal molecular degrees of freedom, as was first discussed by Mandel'shtam and Leontovich in 1937 [3]. This coupling is characterized by a relaxation time τ which governs the transition from ordinary (first) sound (velocity v_0) at low frequencies ($\omega\tau \ll 1$) to zero sound (velocity v_∞) at high frequencies ($\omega\tau \gg 1$). As was emphasized by Mountain [2] the effect of the coupling again is a central peak in inelastic light scattering.

The central peaks evidenced in inelastic neutron scattering near structural phase transitions in the perovskites $SrTiO_3$ [4] and $LaAlO_3$ [5] and in the A15-compound Nb_3Sn [6, 7] are of a similar nature. In fact, they have been interpreted as Mountain modes [3, 5, 6] in the sense that the relaxation time τ serves to distinguish between Cowley's first and zero sound in crystals [8]. In these cases the response function has been parametrized in the form

$$\rho\chi_i(\vec{q}, \omega) = \{\omega_i^2(\vec{q}) - \omega^2 - i\omega\Gamma(q, \omega)\}^{-1} \tag{1.1}$$

where [1, 7, 9]

$$\Gamma(q,\omega) = \Gamma_0 + \delta_0^2/(\gamma_0 - i\omega) \tag{1.2}$$

so that δ_0^2 describes the coupling between the modes $\omega = \omega_i(\vec{q}) - i\Gamma_0/2$ and $\omega = -i\gamma_0$ (here the index zero serves to distinguish γ_0 and δ_0^2 from the critical index γ and the particular δ^2 [1] occurring later). This parametrization gives rise to a three-pole structure of χ_i, as is explicitly shown in Appendix D.

The similarity with the examples mentioned before is brought out by the fact that the same parametrization also applies to the response function of the thermal expansion coupled sound where [2]

$$\begin{aligned} &\omega_i(\vec{q}) = vq, \quad \Gamma_0 = \tau_0 v^2 q^2 \\ &\delta_0^2 = (v_0^2 - v^2)\, q^2, \quad \gamma_0 = Dq^2 \end{aligned} \tag{1.3}$$

and of the molecular deformation coupled sound where

$$\begin{aligned} &\omega_i(\vec{q}) = v_0 q, \quad \Gamma_0 = 0 \\ &\delta_0^2 = (v_\infty^2 - v_0^2)\, q^2, \quad \gamma_0 = 1/\tau. \end{aligned} \tag{1.4}$$

While in these examples the coupling is not at all related to a phase transition it was the main idea of Ref. [1] that the strong temperature dependence of the soft mode frequency is the reason for the coupling of the soft mode to temperature fluctuations and hence for the central peak in perovskites. Without this idea it was indeed difficult to explain the value of the coupling constant δ_0^2 in the mentioned case of perovskites and A15's because of the difference with the examples of sound in fluids. In fact, all known cases of central peaks in perovskites and A15's refer to *shear* modes; in the perovskites they are in addition modes of the *staggered* displacement [1] (i.e. modes with wave vector near the R-corner $\vec{q}_R = (\pi/a)$ (1,1,1) instead of the centre of the Brillouin zone).

While in Ref. [1] only the case of the soft staggered mode was investigated we give in Section 3 below a general derivation of the response function χ_i valid also for the soft sound mode. Starting in Section 2 from an expression for the fluctuating part of the lattice free energy the mode coupling follows quite naturally. This will show convincingly that the soft frequency coupling near a phase transition is a feature of similar generality as the thermal expansion coupling.

In the case of A15-compounds a different phenomenological model, which also leads to the form (1.1), (1.2) of the response function, has recently been proposed by Kragler and Thomas [10]. In this work a coupling of the soft sound mode with the d-electrons is introduced with the result that $\omega_i(\vec{q}) = vq$, $\Gamma_0 = 0$, $\delta_0^2 = (v_0^2 - v^2)\, q^2$ where v_0 and v are, respectively, the unrenormalized and renormalized sound velocities and γ_0 describes the electron dissipation. Unfortunately this model yields no information about δ_0^2 and the estimate of γ_0 turns out to be much too large.

Experimentally, information about γ_0 is limited to order of magnitude bounds [4, 5]. The reason is that the central peaks have not been resolved in the neutron scattering experiments done so far [4–7]. The only existing estimate of the central peak width in $SrTiO_3$ is due to the electron paramagnetic resonance (EPR) measurements of Müller et al. [11], the theory of which is discussed in Sections 3 and 4. As already mentioned in Ref. [1] the wave-number average over γ_0 which in our theory is implied in this estimate, sets a limit to the validity of the hydrodynamic form of

γ_0. In terms of the wave-number cutoff q_m already introduced in Ref. [1] this means that the average $\bar{\gamma}_0 = D\bar{q}^2$ defined in Section 3 is meaningful only for $q_m \ll \pi/a$.

While it is evident from the above that little can be said at this moment about γ_0, the tests of δ_0^2 turn out to be quite explicit, with respect to both the wave-number and the temperature dependence. For the soft R-corner (staggered) mode the frequency behaves as $\omega_i^2(\vec{q}) \propto q^0 \epsilon^\nu$ and leads to [1]

$$\delta_0^2 \propto q^0 \epsilon^{2\nu-2} \tag{1.5}$$

where $\epsilon = (T - T_c)/T_c$. For the soft zone center (sound) mode, on the other hand, $\omega_i^2(\vec{q}) \propto q^2 \epsilon^\nu$ and

$$\delta_0^2 \propto q^2 \epsilon^{2\nu-2}. \tag{1.6}$$

Both these results are derived simultaneously in Section 4. The difference between the two cases is that only in the first case the response function has the Ornstein–Zernike form [9] so that $\omega_i(0) = \omega_0$ defines a correlation length.

On the other hand, neutron scattering at $SrTiO_3$ [4] and $LaAlO_3$ [5] indicates that $\delta_0^2 \propto q^0 \epsilon^0$. This suggests a discrepancy with (1.5) which, however, was shown in Ref. [1] not to exist within present accuracy. Similarly, while the experiments with Nb_3Sn [6] indicate that $\delta_0^2 \propto q^2 \epsilon^0$, there is at present again no discrepancy with (1.6). All problems related with experimental results are discussed in Section 5.

As shown in the general derivation of the response function χ_i in Section 3 the coupling parameter δ_0^2 is proportional to the (integrated) order parameter correlation function $S_i(q, \omega)$ which, in turn, is related to $\chi_i(\vec{q}, \omega)$ through the fluctuation–dissipation theorem. The classical form of this theorem needed here is derived in Appendix A which, more generally, sketches a framework of classical many–body theory. The idea is to cast the hydrodynamic equations of motion into a canonical form by extending the fluctuating part of the internal energy into a Hamiltonian. This formalism is in many respects complementary to the way Zwanzig [12] and Kawasaki [13] introduce fluctuation dynamics but is developed with the same goal. In turn our theory has many parallels with the works of Kadanoff and Swift [14] and of Halperin, Hohenberg and Ma [15].

Finally, it is worth mentioning that in limiting this analysis to the cubic symmetry above the phase transition we voluntarily eliminate unessential complications. Thus, the mode frequencies in the vicinity of both the zone centre and the R-corner have a particularly simple parametrization [16]. In Appendix B the related eigenvalue problem as function of the direction of $\vec{q}$ is solved algebraically and the connection with the group theoretical notation [17] established. In particular the form expressing $\omega_i(\vec{q})$ in terms of the anisotropy parameter Δ [9] is justified for $SrTiO_3$. With the explicit form of $\omega_i(\vec{q})$ the various integrals over the Brillouin zone, appropriately restricted by the cutoff q_m, then are easily evaluated in Appendix C.

2. The Hydrodynamic Equations

In the terminology of Ref. [2] hydrodynamics in a crystal is described by the equations of motion of the elastic continuum and of the fluid of thermal phonons. For the latter a flow or drift may safely be neglected here since effects of second sound or Poiseuille flow [2] are of no importance.

The elastic equation of motion may be written in the form

$$\rho\ddot{u}_i = -\rho\Gamma\dot{u}_i + \sum_{jkl} \eta_{ij,kl}\,\nabla_j\,\nabla_k\,\dot{u}_l - \delta F_L/\delta u_i. \tag{2.1}$$

Here u_i is a component of the local displacement vector $\vec{u}(\vec{r},t)$ of the soft mode with damping Γ ($\Gamma = 0$ for a sound mode) and $\eta_{ij,kl}$ are the components of the viscosity or staggered viscosity tensor for a zone center or an R-corner soft mode, respectively. F_L is the fluctuating part of the lattice free energy and ρ the mass density. The relation between density fluctuation and displacement,

$$\nabla\cdot\vec{u} = \delta(\Delta V)/\Delta V = -\delta\rho/\rho \tag{2.2}$$

where ΔV is a macroscopic volume element, shows that $\delta\rho = 0$ for a shear mode.

Without drift the only equation of motion of the thermal excitations is the entropy balance equation [2].

$$(\rho s)^{\cdot} + \nabla.\vec{I}_s = \sigma \tag{2.3}$$

where s is the entropy per unit mass and

$$\vec{I}_s = -\frac{\lambda^*}{T}\nabla(\delta T) \tag{2.4}$$

the entropy current which in the absence of a phonon dirft is entirely due to an effective heat conductivity λ^* (which for simplicity is assumed to be a scalar) and to the local temperature fluctuation $\delta T(\vec{r},t)$. The entropy production density σ can be shown [2] to be quadratic in $\nabla\delta T$, in $\dot{\vec{u}}$ and in $\dot{\theta}$ where θ is the deformation tensor defined by

$$\theta_{ij} = \tfrac{1}{2}(\nabla_i u_j + \nabla_j u_i). \tag{2.5}$$

Since terms proportional to $(\delta T)^2$, $\dot{\vec{u}}^2$ and $\dot{\vec{u}}\delta T$ and spatial derivatives thereof will be neglected we may drop σ and, according to (2.2), also $(\delta\dot{\rho})s$ in equation (2.3).

While in general the lattice does not contribute to the entropy [2] this is not so in the presence of a soft mode. Indeed, the soft mode parameters which occur in F_L strongly depend on temperature and hence

$$\rho s_L = -\delta F_L/\delta T \tag{2.6}$$

is an entropy density carried by the lattice.

Thus F_L is a functional of the local displacement $\vec{u}(\vec{r},t)$ and the local temperature fluctuation $\delta T(\vec{r},t)$ and has the form

$$F_L[\vec{u},\delta T] = \int_V d^3r \left\{ \frac{\rho}{2}\,\omega_0^2(T+\delta T)\,\vec{u}^2 + \tfrac{1}{2}\sum_{ijkl} C_{ij,kl}(T+\delta T)\,\theta_{ij}\,\theta_{kl} - \rho\vec{u}\cdot\vec{f} \right\}. \tag{2.7}$$

Here ω_0 is the soft mode frequency ($\omega_0 = 0$ for a sound mode). $C_{ij,nl}$ are the isothermal elastic or staggered elastic constants for a zone centre or an R-corner soft mode, respectively, and $\vec{f}(\vec{r},t)$ is a local external force per unit mass.

Performing a partial integration with the second term of (2.7), making use of (2.5), the equation of motion (2.1) takes the form

$$\left\{\omega_0^2+\frac{\partial\omega_0^2}{\partial T}\delta T-\frac{1}{\rho}C\nabla\otimes\nabla-\frac{1}{\rho}\frac{\partial C}{\partial T}[(\nabla\delta T)+\delta T\nabla]\otimes\nabla\right.$$

$$\left.+\frac{\partial^2}{\partial t^2}+\Gamma\frac{\partial}{\partial t}-\frac{1}{\rho}\eta\nabla\otimes\nabla\frac{\partial}{\partial t}\right\}\vec{u}=\vec{f} \tag{2.8}$$

valid up to bilinear terms in $\vec{u}$ and δT. Here $C\nabla\otimes\nabla$ is the symmetric matrix with elements

$$(C\nabla\otimes\nabla)_{il}=\sum_{jk}C_{ij,kl}\nabla_j\nabla_k \tag{2.9}$$

and $\eta\nabla\otimes\nabla$ is defined in the same way.

The lattice entropy per unit mass is found from (2.6), (2.7),

$$s_L=-\frac{1}{2}\frac{\partial\omega_0^2}{\partial T}\vec{u}^2-\frac{1}{2\rho}\sum_{ijkl}\frac{\partial C_{ij,kl}}{\partial T}\theta_{ij}\theta_{kl} \tag{2.10}$$

where the dependence on δT is of higher order and hence neglected. Thus the effective entropy fluctuation due to fluctuations $\vec{u}$ and δT is

$$\delta s=(c_V^*/T)\,\delta T+s_L \tag{2.11}$$

where c_V^* is an effective specific heat (per unit mass). Inserting equations (2.4), (2.10), (2.11) into (2.3) we find

$$\left(\frac{\partial}{\partial t}-D\nabla^2\right)\delta T=\frac{T}{2c_V^*}\frac{\partial\omega_0^2}{\partial T}\frac{\partial\vec{u}^2}{\partial t}+\frac{T}{2\rho c_V^*}\sum_{ijkl}\frac{\partial C_{ij,kl}}{\partial T}\frac{\partial}{\partial t}(\theta_{ij}\theta_{kl}) \tag{2.12}$$

where

$$D=\lambda^*/(\rho c_V^*) \tag{2.13}$$

is an effective heat diffusion constant. In (2.12) the entropy production σ as well as the term $(\delta\dot{\rho})s$ could be neglected because we have retained only linear terms in δT and in $\partial/\partial t$ while, as mentioned before, σ is quadratic in $\nabla\delta T$, in $\dot{\vec{u}}$ and in $\dot{\theta}$ and $(\delta\dot{\rho})s$ is proportional to $\nabla\cdot\dot{\vec{u}}\delta T$.

Equations (2.8) and (2.12) reduce to equations (3) and (4) of Ref. [1] if the temperature dependence of the staggered elastic constants and the viscosities are neglected. In introducing an effective heat conductivity λ^* and an effective specific heat c_V^* we want to take into account the facts i) that in general the transport coefficients depend on frequency [12] (which for the neutron scattering data in question is rather high) and ii) that not all the thermal degrees of freedom participate in the soft frequency coupling.

3. The Soft Mode Response Function

We define the soft mode response function by

$$\tilde{\chi}_{ij}(\vec{r},t;\vec{r}',t') = \frac{1}{\rho}\frac{\delta u_i(\vec{r},t)}{\delta f_j(\vec{r}',t')}. \tag{3.1}$$

Introducing in addition the temperature response function

$$\varphi_j(\vec{r},t;\vec{r}',t') = \frac{1}{\rho}\frac{\delta(\delta T(\vec{r},t))}{\delta f_j(\vec{r}',t')} \tag{3.2}$$

functional derivation of equations (2.8) and (2.12) with respect to the external force $\vec{f}$ yields, respectively,

$$\begin{aligned}
&\sum_k \Bigg\{ \omega_0^2 + \frac{\partial\omega_0^2}{\partial T}\delta T - \frac{1}{\rho} C\nabla\otimes\nabla - \frac{1}{\rho}\frac{\partial C}{\partial T}[(\nabla\delta T) + \delta T\nabla]\otimes\nabla \\
&\quad + \frac{\partial^2}{\partial t^2} + \Gamma\frac{\partial}{\partial t} - \frac{1}{\rho}\eta\nabla\otimes\nabla\frac{\partial}{\partial t}\Bigg\}_{ik}\tilde{\chi}_{kj} + \frac{\partial\omega_0^2}{\partial T}u_i\varphi_j \\
&\quad - \frac{1}{\rho}\sum_{mnl}\frac{\partial C_{im,nl}}{\partial T}[(\nabla_m\varphi_j)(\nabla_n u_l) + \varphi_j\nabla_m\nabla_n u_l] \\
&\quad = \frac{1}{\rho}\delta_{ij}\delta(\vec{r}-\vec{r}')\,\delta(t-t')
\end{aligned} \tag{3.3}$$

and

$$\left(\frac{\partial}{\partial t} - D\nabla^2\right)\varphi_j = \frac{T}{c_V^*}\frac{\partial\omega_0^2}{\partial T}\sum_k\frac{\partial}{\partial t}(u_k\tilde{\chi}_{kj}) + \frac{T}{\rho c_V^*}\sum_{hrsk}\frac{\partial C_{hr,sk}}{\partial T}\frac{\partial}{\partial t}[(\nabla_r u_h)(\nabla_s\tilde{\chi}_{kj})] \tag{3.4}$$

where we have used (2.5).

In order to eliminate φ_j we multiply equation (3.3) by $\partial' = -\partial/\partial t' - D\nabla'^2$, noting that this operator has the same effect as $\partial = \partial/\partial t - D\nabla^2$ when applied to functions which depend only on the coordinate differences $\vec{r}-\vec{r}'$ and $t-t'$. Substituting $\partial\varphi_j$ from (3.4) equation (3.5) can be written as

$$\sum_k A_{ik}\tilde{\chi}_{kj} + B_i(\partial' - \partial)\varphi_j = \frac{1}{\rho}\delta_{ij}\,\partial\delta(\vec{r}-\vec{r}')\,\delta(t-t') \tag{3.5}$$

where

$$\begin{aligned}
A_{ik} = \partial'\Bigg\{ &\omega_0^2 + \frac{\partial\omega_0^2}{\partial T}\delta T - \frac{1}{\rho}C\nabla\otimes\nabla - \frac{1}{\rho}\frac{\partial C}{\partial T}[(\nabla\delta T) + \delta T\nabla]\otimes\nabla + \frac{\partial^2}{\partial t^2} \\
&+ \Gamma\frac{\partial}{\partial t} - \frac{1}{\rho}\eta\nabla\otimes\nabla\frac{\partial}{\partial t}\Bigg\}_{ik} + \frac{T}{c_V^*}\left(\frac{\partial\omega_0^2}{\partial T}\right)^2 u_i\left(\dot{u}_k + u_k\frac{\partial}{\partial t}\right) + \frac{T}{\rho c_V^*}\frac{\partial\omega_0^2}{\partial T}
\end{aligned}$$

$$\times \sum_{mnl} \frac{\partial C_{ln,mk}}{\partial T} u_l \left[(\nabla_n \dot{u}_l) + (\nabla_n u_l) \frac{\partial}{\partial t} \right] \nabla_m - \frac{T}{\rho c_V^*} \frac{\partial \omega_0^2}{\partial T} \sum_{mnl} \frac{\partial C_{im,nl}}{\partial T}$$

$$\times \left\{ (\nabla_m \nabla_n u_l) \left(\dot{u}_k + u_k \frac{\partial}{\partial t} \right) + (\nabla_n u_l) \left[(\nabla_m \dot{u}_k) + (\nabla_m u_k) \frac{\partial}{\partial t} + \dot{u}_k \nabla_m + u_k \nabla_m \frac{\partial}{\partial t} \right] \right\}$$

$$- \frac{T}{\rho^2 c_V^*} \sum_{mnl} \sum_{hrs} \frac{\partial C_{im,nl}}{\partial T} \frac{\partial C_{hr,sk}}{\partial T} \left\{ (\nabla_m \nabla_n u_l) \left[(\nabla_r \dot{u}_h) + (\nabla_r u_h) \frac{\partial}{\partial t} \right] \right.$$

$$\left. + (\nabla_n u_l) \left[(\nabla_m \nabla_r \dot{u}_h) + (\nabla_m \nabla_r u_h) \frac{\partial}{\partial t} + (\nabla_r \dot{u}_h) \nabla_m + (\nabla_r u_h) \nabla_m \frac{\partial}{\partial t} \right] \right\} \nabla_s \qquad (3.6)$$

and

$$B_i = \frac{\partial \omega_0^2}{\partial T} u_i - \frac{1}{\rho} \sum_{mnl} \frac{\partial C_{im,nl}}{\partial T} [(\nabla_n u_l) \nabla_m + (\nabla_m \nabla_n u_l)].$$

Since the external force $\vec{f}$ was introduced only to generate the response functions (3.1) and (3.2) we now let $\vec{f}$ go to zero. Then $\vec{u}$ and δT become thermal fluctuations which essentially form a Gaussian distribution around zero with width proportional to the temperature T. All physically relevant information is then contained in the average $\langle \ \rangle_0$ over this distribution whose precise definition is given in Appendix A.

At this point we introduce a random phase type approximation by writing the averaged equation (3.5) as [1])

$$\sum_k \langle A_{ik} \rangle_0 \langle \tilde{\chi}_{kj} \rangle_0 + \langle B_i \rangle_0 (\partial' - \partial) \langle \varphi_j \rangle_0 = \frac{1}{\rho} \delta_{ij} \partial \delta(\vec{r} - \vec{r}') \, \delta(t - t'). \qquad (3.7)$$

From Appendix A it follows that

$$\langle \delta T(\vec{r}, t) \rangle_0 = 0, \ \langle \vec{u}(\vec{r}, t) \rangle_0 = 0 \text{ and } \langle \vec{u}_i(r, t) \, u_j(\vec{r}', t) \rangle_0 = 0 \text{ for } i \neq j \text{ and any } \vec{r}, \vec{r}'$$

and, by time reversal invariance, $\langle u_i(\vec{r}, t) \, \dot{u}_j(\vec{r}', t) \rangle_0 = 0$ for any $i, j, \vec{r}, \vec{r}'$. Invariance of averages under space and time translations (compare equations (A.30) of Appendix A) then implies that $(\partial' - \partial) \langle \varphi_j \rangle_0 = 0$ and that

$$\langle u_i(\vec{r}, t) \, u_j(\vec{r}, t) \rangle_0 = \phi_i \, \delta_{ij} \qquad (3.8)$$

with $\phi_i \geq 0$ and

$$\langle \nabla_r u_i(\vec{r}, t) \, \nabla_s u_j(\vec{r}, t) \rangle_0 = - \langle u_i(\vec{r}, t) \nabla_r \nabla_s u_j(\vec{r}, t) \rangle_0 = \psi_{ri} \, \delta_{rs} \, \delta_{ij} \qquad (3.9)$$

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[1]) Equation (3.7) is essentially a mean field approximation and hence may not be adequate in the critical region. However, since the goal of this paper is to understand the physics underlying soft mode-related central peaks we seek the simplest mechanism yielding the form (1.1), (1.2) of the order parameter response function. In an improved solution the basic 'interaction Hamiltonian' (2.7) would have to be treated in perturbation theory along the lines of Ref. [15], in which the critical behaviour is determined by recusion formulas. Presumably, such a treatment would also modify the ϵ-exponent in relations (1.5), (1.6). We plan to come back to this problem in the near future.

with $\psi_{rl} \geqslant 0$ are independent of $\vec{r}$, t and are the only correlation functions left in $\langle A_{ik}\rangle_0$. Hence we find from (3.6), after replacing ∂' by ∂,

$$\langle A_{ik}\rangle_0 = \left(\frac{\partial}{\partial t} - D\nabla^2\right)\left\{\omega_0^2 - \frac{1}{\rho}C\nabla\otimes\nabla + \frac{\partial^2}{\partial t^2} + \Gamma\frac{\partial}{\partial t} - \frac{1}{\rho}\eta\nabla\otimes\nabla\frac{\partial}{\partial t}\right\}_{ik}$$
$$+ \frac{T}{c_V^*}\left(\frac{\partial\omega_0^2}{\partial T}\right)^2\phi_i\,\delta_{ik}\frac{\partial}{\partial t} - \frac{T}{\rho^2 c_V^*}\sum_{mnrs}\frac{\partial C_{im,sr}}{\partial T}\frac{\partial C_{rs,nk}}{\partial T}\psi_{sr}\nabla_m\nabla_n\frac{\partial}{\partial t}. \tag{3.10}$$

Since $\langle\tilde{\chi}\rangle_0$ only depends on $\vec{r}-\vec{r}'$, $t-t'$ we may now go over to the Fourier representation

$$\chi_{ij}(\vec{q},\omega) = \int_V d^3r\int dt\langle\tilde{\chi}_{ij}(\vec{r},t)\rangle_0\exp[-i(\vec{q}\cdot\vec{r}-\omega t)]. \tag{3.11}$$

Multiplying the Fourier transformed equation (3.7) from the right by $(-i\omega + Dq^2)^{-1}\chi^{-1}(\vec{q},\omega)$ we find with (3.10)

$$\frac{1}{\rho}\chi_{ik}^{-1}(\vec{q},\omega) = (\omega_0^2 - \omega^2 - i\omega\Gamma)\,\delta_{ik} + R_{ik}(\vec{q})\,(1 - i\omega\tau_{ik}(\hat{q})) - i\omega\delta_{ik}^2(\vec{q})\,(-i\omega + Dq^2)^{-1}. \tag{3.12}$$

Here

$$R_{ik}(\vec{q}) = \frac{1}{\rho}\sum_{mn}C_{im,nk}\,q_m\,q_n \tag{3.13}$$

is the elastic matrix used in Ref. [1],

$$R_{ik}(\vec{q})\,\tau_{ik}(\hat{q}) = \frac{1}{\rho}\sum_{mn}\eta_{im,nk}\,q_m\,q_n \tag{3.14}$$

defines a relaxation time matrix τ and

$$\delta_{ik}^2(\vec{q}) = \frac{T}{c_V^*}\left(\frac{\partial\omega_0^2}{\partial T}\right)^2\phi_i\,\delta_{ik} + \frac{T}{\rho^2 c_V^*}\sum_{mnrs}\frac{\partial C_{im,sr}}{\partial T}\frac{\partial C_{rs,nk}}{\partial T}\psi_{sr}\,q_m\,q_n \tag{3.15}$$

are generalized coupling constants.

Equations (3.12) to (3.15) give the general response function valid for any crystal symmetry and any type of soft staggered or sound mode. We now assume *cubic symmetry* so that in the parametrization (B.3) of Appendix B

$$\frac{1}{\rho}C_{ij,kl} = (\lambda_1 - \lambda_2)\,\delta_{ij}\,\delta_{kl}\,\delta_{il} + \lambda_2\{\delta_{ik}\,\delta_{jl} + \delta_{il}\,\delta_{jk} - \delta_{ij}\,\delta_{kl}\} + \lambda_3\,\delta_{ij}\,\delta_{kl}(1 - \delta_{il}). \tag{3.16}$$

This reduces equation (3.13) to [15]

$$R_{ik}(\vec{q}) = [\lambda_2 q^2 + (\lambda_1 - \lambda_2)\,q_i^2]\,\delta_{ik} + \lambda_3\,q_i\,q_k(1 - \delta_{ik}). \tag{3.17}$$

Furthermore, for cubic symmetry ϕ_i in equation (3.8) becomes independent of i and ψ_{ri} in equation (3.9) has the form

$$\psi_{ri} = \psi_{\parallel}\,\delta_{ri} + \psi_{\perp}(1 - \delta_{ri}). \tag{3.18}$$

Then the coupling constants (3.15) may be written

$$\delta_{ik}^2(\vec{q}) = \delta^2\,\delta_{ik} + V_{ik}(\vec{q}) \tag{3.19}$$

where

$$\delta^2 = \frac{\phi T}{c_V^*}\left(\frac{\partial \omega_0^2}{\partial T}\right)^2 \tag{3.20}$$

is the coupling constant of Ref. [1],

$$\begin{aligned} V_{ik}(\vec{q}) = {} & \frac{T}{c_1^*}\{\psi_{\parallel}[\lambda_1'^2 + 2(\lambda_3' - \lambda_2')^2]\,q_i^2 + 2\psi_{\perp}\lambda_2'^2(q^2 - q_i^2)\}\,\delta_{ik} \\ & + \frac{T}{c_V^*}\{\psi_{\parallel}(\lambda_3' - \lambda_2')(2\lambda_1' + \lambda_3' - \lambda_2') + 2\psi_{\perp}\lambda_2'^2\}\,q_i\,q_k(1 - \delta_{ik}) \end{aligned} \tag{3.21}$$

and we have put $\lambda_i' = \partial\lambda_i/\partial T$.

In the case of the soft staggered mode of the perovskites $\omega_0 \neq 0$, $\Gamma \neq 0$ and $C_{ij,kl}$ are the staggered elastic constants. We may therefore neglect the temperature dependence of the $C_{ij,kl}$ as well as the staggered viscosities $\eta_{ij,kl}$, so that $\lambda_i' = 0$, $\tau_{ik} = 0$ and $V_{ik} = 0$. For $SrTiO_3$ a further simplification comes from the fact that to a good approximation $\lambda_3 = 0$ (see Appendix B) which makes the staggered elastic matrix (3.17) diagonal with eigenvalues $r_i(\vec{q})$ given by equation (B.14) of Appendix B. The response function (3.12) now assumes the form (1.1), (1.2) with

$$\begin{aligned} \chi_{ik}(\vec{q},\omega) &= \chi_i(\vec{q},\omega)\,\delta_{ik} \\ \omega_i^2(\vec{q}) &= \omega_0^2 + r_i(\vec{q}), \quad & \Gamma_0 &= \Gamma \\ \delta_0^2 &= \delta^2, & \gamma_0 &= Dq^2 \end{aligned} \tag{3.22}$$

where δ^2 is given by equation (3.20).

In the case of the soft sound mode of the A15 compounds $\omega_0 = 0$ and $\Gamma = 0$, so that $\delta^2 = 0$ and the elastic constants $C_{ij,kl}$ and viscosities $\eta_{ij,kl}$ become the leading terms of the soft mode. For Nb_3Sn, in particular $\lambda_3 \neq 0$ at T_c [18] so that diagonalization of the elastic matrix (3.17) is non-trivial (see Appendix B). We therefore restrict the discussion of the response function (3.12) to the soft T_1 mode [6, 18] with propagation direction $\hat{q}_0 \propto (1,1,0)$ and polarization direction $\hat{e}_0 \propto (1,-1,0)$ (see Table V in Appendix B). Projecting equation (3.12) onto $\hat{e}_0$, making use of (3.19) we recover again the form (1.1), (1.2) with

$$\begin{aligned} \chi_i^{-1}(\vec{q},\omega) &= (\hat{e}_0, \chi^{-1}(q\hat{q}_0,\omega)\,\hat{e}_0) \\ \omega_i^2(\vec{q}) &= (\hat{e}_0, R(q\hat{q}_0)\,\hat{e}_0) \equiv v_0^2 q^2 \\ \Gamma_0 &= (\hat{e}_0, R(q\hat{q}_0)\,\tau(\hat{q}_0)\,\hat{e}_0) \equiv \tau_0 v_0^2 q^2 \\ \delta_0^2 &= (\hat{e}_0, V(q\hat{q}_0)\,\hat{e}_0) \equiv \lambda^2 q^2 \end{aligned} \tag{3.23}$$

where from equation (3.17)

$$v_0^2 = \tfrac{1}{2}(\lambda_1 + \lambda_2 - \lambda_3) \tag{3.24}$$

and from equation (3.21)

$$\lambda^2 = 2\frac{\psi_\parallel T}{c_V^*}\left(\frac{\partial v_0^2}{\partial T}\right)^2. \tag{3.25}$$

From (3.8) and (3.9), (3.18) follows

$$\phi = \tfrac{1}{3}\langle \vec{u}^2 \rangle_0 \tag{3.26}$$

and

$$\psi_\parallel = \tfrac{1}{3}\langle (\nabla \cdot \vec{u})^2 \rangle_0. \tag{3.27}$$

Thus the existence of a central peak associated with the R-corner mode, equations (3.22), and with the T_1 mode, equations (3.23), crucially depends on the fluctuations (3.26) and (3.27), respectively. In particular, $\psi_\parallel \neq 0$ means, according to (3.27) that an appreciable amount of longitudinal thermal phonons must be present which evidently is always the case.

It is useful to express the fluctuations (3.26) and (3.27) in terms of the Fourier transformed correlation function or dynamical structure factor

$$S_{ij}(\vec{q}, \omega) = \int_V d^3 r \int dt \frac{\rho}{2\pi} \langle u_i(\vec{r}, t)\, u_j(0, 0) \rangle_0 \exp[-i(\vec{q} \cdot \vec{r} - \omega t)] \tag{3.28}$$

because S_{ij} is related to the response function (3.11) by the classical fluctuation–dissipation theorem

$$S_{ij}(\vec{q}, \omega) = \frac{\rho k_B T}{\pi \omega} \chi''_{ij}(\vec{q}, \omega) \tag{3.29}$$

derived in Appendix A (see equation (A.38)). Here χ''_{ij} is the dissipative part related to the response function by the dispersion relation (see equation (A.33))

$$\chi_{ij}(\vec{q}, z) = \int \frac{d\omega}{\pi} \frac{\chi''_{ij}(\vec{q}, \omega)}{\omega - z}; \quad \operatorname{Im} z > 0. \tag{3.30}$$

With (3.29) and (3.30) the dynamical structure factor associated to the form (1.1), (1.2) of the response function readily follows [4],

$$\frac{\pi}{k_B T} S_i(\vec{q}, \omega) = \left(\Gamma_0 + \frac{\delta_0^2 \gamma_0}{\gamma_0^2 + \omega^2}\right)\left\{\left(\omega_i^2(\vec{q}) - \omega^2 + \frac{\delta_0^2 \omega_0^2}{\gamma_0^2 + \omega^2}\right)^2 + \omega^2\left(\Gamma_0 + \frac{\delta_0^2 \gamma_0}{\gamma_0^2 + \omega^2}\right)^2\right\}^{-1}. \tag{3.31}$$

From (3.26) to (3.30) one also finds the fluctuations, namely with (3.22) and (1.1) [1]

$$\phi / k_B T = V^{-1} \sum_{\vec{q}} \tfrac{1}{3} \sum_i \chi_{ii}(\vec{q}, 0) = (\rho V)^{-1} \sum_{\vec{q}} \omega_i^{-2}(\vec{q}) \tag{3.32}$$

and with (3.12) and $\omega_0 = 0$, $\Gamma = 0$

$$\psi_{\parallel}/k_B T = V^{-1} \sum_{\vec{q}} \tfrac{1}{3} \sum_{ij} q_i q_j \chi_{ij}(\vec{q}, 0)$$

$$= (3\rho V)^{-1} \sum_{\vec{q}} (\vec{q}, R^{-1}(\vec{q})\, \vec{q}). \tag{3.33}$$

In certain paramagnetic impurity centres the local displacement generates an effective magnetic field via an effective coupling constant G. In such a case time-dependent fluctuations $\langle u_i(0,t)\, u_i(0,0)\rangle_0$ at the centre give a contribution ΔH to the EPR linewidth. Since the time imprecision of these fluctuations is $T \cong (\Delta H)^{-1}$ this contribution is roughly given by

$$(\Delta H)^2 \cong G^2 (2T)^{-1} \int_{-T}^{+T} dt\, \frac{\rho}{2\pi} \langle u_i(0,t)\, u_i(0,0)\rangle_0 \tag{3.34}$$

or making use of (3.28), by

$$(\Delta H)^2 \cong G^2 \int_0^{(\pi/2)\Delta H} \frac{d\omega}{\pi} \frac{1}{V} \sum_{\vec{q}} S_{ii}(\vec{q}, \omega). \tag{3.35}$$

If S_{ii} is given by the diagonal form (3.31) and if this expression is dominated by the central peak, i.e. close to T_c, the effect of the time averaging in equation (3.34) depends sensitively on the width of the central peak in S_i relative to the width $(\pi/2)\, \Delta H$ describing the time imprecision in (3.35). We assume here that the coupling constant G varies little with temperature. (For a detailed analysis of the EPR lineshape and crossover problem see Ref. [19].)

In the *fast motion regime* [19] the central peak averaged over $\vec{q}$ is broad compared to ΔH, or $\langle u_i(0,t)\, u_i(0,0)\rangle_0$ varies fast during the time imprecision $(\Delta H)^{-1}$. Hence we find from (3.35), (3.31) and (3.22)

$$\Delta H_f = \tfrac{1}{2} G^2 V^{-1} \sum_{\vec{q}} S_i(\vec{q}, 0)$$

$$= (G^2 k_B T/2\pi V) \sum_{\vec{q}} (\Gamma + \delta^2/Dq^2)\, \omega_i^{-4}(\vec{q}). \tag{3.36}$$

In the *slow motion regime* [19] the $\vec{q}$-averaged central peak is narrow compared to ΔH, or $\langle u_i(0,t)\, u_i(0,0)\rangle_0$ varies slowly during $(\Delta H)^{-1}$ so that we may use equations (3.26), (3.32) in (3.34),

$$(\Delta H_s)^2 = G^2 \frac{\rho}{2\pi} \langle u_i^2\rangle_0 = (G^2 k_B T/2\pi V) \sum_{\vec{q}} \omega_i^{-2}(\vec{q}). \tag{3.37}$$

For further discussion it is useful to normalize the $\vec{q}$-averaged central peak by defining the function

$$\sigma(\omega^2) = V^{-1} \sum_{\vec{q}} S_i(\vec{q}, \omega)/V^{-1} \sum_{\vec{q}} S_i(\vec{q}, 0). \tag{3.38}$$

The coupling constant G may be eliminated from (3.36) and (3.37) in the form

$$\int_0^\infty \frac{d\omega}{\pi}\,\sigma(\omega^2) = \frac{(\Delta H_s)^2}{2\Delta H_f} \tag{3.39}$$

and equation (3.35) may be written as

$$(\Delta H/\Delta H_s)^2 \cong \int_0^{(\pi/2)\Delta H} \frac{d\omega}{\pi}\,\sigma(\omega^2) \Big/ \int_0^\infty \frac{d\omega}{\pi}\,\sigma(\omega^2). \tag{3.40}$$

4. Critical Behaviour

Above T_c the isothermal susceptibility diverges with exponent γ [20]. For the parametrization (1.1), (1.2) this means that in the limit $q \to 0$

$$\chi_i^{-1}(\vec{q}, 0) = \rho\omega_i^2(\vec{q}) \propto \epsilon^\gamma. \tag{4.1}$$

Hence we have in the case (3.22) corresponding to the R-corner mode of $SrTiO_3$

$$\omega_0^2 = \lambda_2 \kappa_0^2 \epsilon^\gamma \tag{4.2}$$

and in the case (3.23) corresponding to the T_1 mode of Nb_3Sn

$$v_0^2 = \lambda_2 \beta_0 \epsilon^\gamma. \tag{4.3}$$

In the first case equations (3.22), (B.14) lead to the Ornstein–Zernike form [9]

$$\omega_i^2(\vec{q}) = \lambda_2\{\kappa^2 + q^2 - (1 - \Delta)\, q_i^2\} \tag{4.4}$$

where the inverse correlation length κ vanishes with exponent ν [20]

$$\kappa = \kappa_0 \epsilon^\nu. \tag{4.5}$$

In the limit $q \to 0$ equations (4.4), (4.5) lead to (4.2) with

$$\eta = 2 - \gamma/\nu = 0 \tag{4.6}$$

where η is the critical exponent describing the deviation from Ornstein–Zernike behaviour [20].

In the Ornstein–Zernike case we may define new variables

$$x = \kappa/q_m = (\epsilon/\epsilon_0)^\nu \tag{4.7}$$

where q_m is a cutoff wave number which comes from the fact that equations (3.26), (3.27) and (3.34) actually involve a spacial average of the correlation function $\langle u_i(\vec{r}, t)\, u_j(0,0)\rangle_0$ over microscopic distances. The *critical region* is then defined by

$$\gamma = 2\nu > 1, \quad x \ll 1, \quad \epsilon \ll \epsilon_0 < \epsilon_{mf} \tag{4.8}$$

whereas in the *mean-field region*

$$\gamma = 2\nu = 1, \quad x \gtrsim 1, \quad \epsilon \gtrsim \epsilon_0 \cong \epsilon_{mf}. \tag{4.9}$$

If a mean field region exists it is obvious from (4.9) that the changeover temperature from critical to mean-field behaviour determine by ϵ_{mf} should be close to ϵ_0 as defined by (4.7).

Since the critical behaviour occurs only in the limit $q \to 0$ of the soft modes (4.2) and (4.3) it is evident that the integration over the Brillouin zone smooths out the singularity in equations (3.32) and (3.33) so that ϕ and $\psi_\parallel$ are non-critical. Indeed with equations (C.6), (C.7) of Appendix C we find

$$\phi = (k_B T q_m / 2\pi^2 \rho \lambda_2 \sqrt{\Delta})\,(1 - x \tan^{-1}(1/x)) \tag{4.10}$$

and with (C.11)

$$\psi_\parallel = (k_B T q_m^3 / 18\pi^2 \rho \alpha \lambda_2)\vartheta(\epsilon) \tag{4.11}$$

where $\vartheta(\epsilon)$ as given by equation (B.27) is slowly varying and of order unity.

Parametrizing the specific heat c_V^* in terms of an effective number of thermal degrees of freedom $2f^*$ as

$$c_V^*(T) = \frac{f^* k_B}{\rho a^3} \frac{T}{T_c} \tag{4.12}$$

where a is the lattice constant and the last factor takes care of the leading temperature dependence near T_c (see Section 5), the coupling functions (3.20) and (3.25) may be written, respectively, as

$$\delta^2 = g(\epsilon)\, \epsilon^{4\nu - 2} \tag{4.13}$$

and

$$\lambda^2 = k(\epsilon)\, \epsilon^{2\gamma - 2}. \tag{4.14}$$

Here g and k are smooth functions of ϵ obtained with the help of (4.10) and (4.11), respectively,

$$g(\epsilon)/g(0) = (1 + \epsilon)\,(1 - x \tan^{-1}(1/x)) \tag{4.15}$$

with

$$g(0) = (2\nu^2 \lambda_2 \kappa_0 q_m (a\kappa_0)^3 / \pi^2 f^* \sqrt{\Delta}) \tag{4.16}$$

and

$$k(\epsilon) = (\gamma^2 \lambda_2 \beta_0^2 (a q_m)^3 / 9\pi^2 f^* \alpha)\,(1 + \epsilon)\,\vartheta(\epsilon). \tag{4.17}$$

By comparison with (3.32), equation (3.37) may be written, with (4.10), (4.15),

$$(\Delta H_s)^2 = (\Delta H_{0s})^2 g(\epsilon)/g(0) \tag{4.18}$$

where

$$(\Delta H_{0s})^2 = (G^2 k_B T_c q_m^s / 4\pi^3 \lambda_2 \sqrt{\Delta}) \tag{4.19}$$

and q_m^s is the cutoff for the slow motion regime. From equation (3.36) we obtain with (C.6), (C.8)

$$\Delta H_f = \Delta H_{0f} h(\epsilon) \epsilon^{-\nu} \tag{4.20}$$

where

$$h(\epsilon) = \frac{q_m^f}{\kappa_0}\left(1 + \frac{\delta^2}{\Gamma \bar{\gamma}(\epsilon)}\right)(1+\epsilon)\left(\tan^{-1}\frac{1}{x} - \frac{x}{1+x^2}\right) \tag{4.21}$$

and

$$\Delta H_{0f} = (G^2 k_B T_c \Gamma / 8\pi^3 \lambda_2^2 \sqrt{\Delta}\, q_m^f). \tag{4.22}$$

Here q_m^f is the cutoff for the fast motion regime and the average $\bar{\gamma}(\epsilon)$ is defined by

$$1/\bar{\gamma}(\epsilon) = (D\bar{q}^2(\epsilon))^{-1} = V^{-1} \sum_{\vec{q}} (Dq^2)^{-1} \omega_i^{-4}(\vec{q}) / V^{-1} \sum_{\vec{q}} \omega_i^{-4}(\vec{q}) \tag{4.23}$$

$\bar{q}(\epsilon)$ is explicitly calculated in equations (C.9) and (C.10). From (4.19) and (4.21) the coupling constant may be eliminated [1],

$$(\Delta H_{0s})^2 / \Delta H_{0f} = 2\lambda_2 q_m^f q_m^s / \Gamma. \tag{4.24}$$

The transition between the two regimes (4.18) and (4.20) is governed by temperature. At a fixed T the right hand side of (3.40) is a monotonically increasing function of ΔH, so that

$$\Delta H_s \gtrsim \Delta H \gtrsim \Delta H_f; \quad T = \text{const} \tag{4.25}$$

Since $\Delta H_s \propto \epsilon^0$ and $\Delta H_f \propto \epsilon^{-\nu}$ as $\epsilon \to 0$ it is evident from (4.24) that, sufficiently close to T_c, $\Delta H = \Delta H_s$, and according to (4.18), (4.7) [21]

$$(\Delta H_s)^2 = (\Delta H_{0s})^2 \{1 - C\epsilon^{\nu} + O(\epsilon^{2\nu})\} \tag{4.26}$$

with [1]

$$C = \frac{\pi}{2}(\epsilon_0^s)^{-\nu} = \frac{\pi \kappa_0}{2 q_m^s}. \tag{4.27}$$

For sufficiently small ϵ the dynamical structure factors S_i is dominated by the central peak part (D.11) of Appendix D for which equation (3.40) becomes

$$\left(\frac{\Delta H}{\Delta H_s}\right)^2 \cong V^{-1} \sum_{\vec{q}} \frac{\delta^2}{\Omega_i^2 \omega_i^2} \frac{2}{\pi} \tan^{-1}\left(\frac{(\pi/2)\,\Delta H}{\gamma_0'}\right) \Big/ V^{-1} \sum_{\vec{q}} \frac{\delta^2}{\Omega_i^2 \omega_i^2}$$

$$= \frac{2}{\pi} \tan^{-1}\left(\frac{\pi}{2} \Delta H / \langle \gamma_0' \rangle\right). \tag{4.28}$$

Here the second equality defines the ΔH-dependent average $\langle \gamma_0' \rangle$ over $\vec{q}$ of the central peak width γ_0'. This quantity defines a transition line-width

$$\Delta H_{tr} = \frac{2}{\pi} \langle \gamma_0' \rangle \cong \Delta H_s / \sqrt{2} \tag{4.29}$$

where the second equality determines the transition temperature to the fast motion regime as function of the coupling constant [11, 19], $\epsilon = \epsilon_{tr}(G)$.

5. Discussion of Experimental Results and Conclusion

i. $SrTiO_3$: Neutron scattering

In Table I are compiled the parameters for $SrTiO_3$ used in Ref. 1. $M = N_A \rho a^3$ is the molecular mass, N_A being Avogadro's number. The values of the parameters λ_2, λ_3 and Δ are discussed in Appendix B. $2f = 2\rho a^3 c_V(T_c)/k_B$ is the experimental number of thermal degrees of freedom. The temperature dependence of equation (4.12) is justified by the fact that near T_c, approximately, $c_V \propto T$ [17, 23].

Table I
Numerical values of the parameters characterizing the displacive phase transition of $SrTiO_3$, as used in Ref. [1]

$a = 3.904$ Å	[22]
$M = 183.5$ g/mole	
$\rho = 5.12$ g/cm^3	[22]
$\lambda_2 = 89$ (meV Å)^2	[17]
$\lambda_3 = 0$	[17]
$\Delta = 0.04$	[17]
$\Gamma = 0.88$ meV	[4]
$T_c = 105$ K	[4, 21]
$Mc_V(T_c) = 11.6$ cal/mole K	[17, 23]
$f = 5.84$	
$\lambda(T_c) = 0.167$ W/cm K	[24]
$\lambda(T_c)/\rho c_V(T_c) = 812$ meV Å^2	

The parameters of the fit of the neutron scattering data for $SrTiO_3$ [4] obtained in Figure 1 of Ref. [1] are collected in Table II. As has already been remarked in Ref. [1] it is not possible to fit the δ^2-data of Ref. [4] with the more realistic critical value $\nu = 0.65$ of Refs. [11] and [21]. The use of the value 0.92 obtained from the ω_0^2-data of Ref. [4] is justified only by the internal consistency with this reference achieved in this way.

This consistency is well born out by the values of ϵ_0 given in Table II which not only are in agreement with equations (4.8) and (4.9) but also with the changeover point $\ln \epsilon_{mf} \cong -1.2$ or $\epsilon_{mf} \cong 0.30$ of the ω_0^2-curve in Figure 1 of Ref. [1]. Also the values of the fraction of the Brillouin zone involved in the integral in (3.32) and measured by $q_m a/\pi$ is reasonable; in the critical region the neighbourhood of $q = 0$ is clearly enhanced.

The ratio f^*/f given in Table II may be interpreted as the fraction of thermal degrees of freedom not taking part in the freezing out of fluctuations. It therefore is expected to parallel the number $q_m a/\pi$ which is well born out in Table II. This interpretation of f^*/f suggests that the effective heat conduction is frozen out accordingly,

Table II
Numerical values of the parameters characterizing the critical and mean-field behaviour of $SrTiO_3$ above $T_c = 105$ K, as obtained from the neutron scattering data of Ref. [4] fitted in Fig. 1 of Ref. [1]

Parameter	Critical	Mean-field
ν	0.92	0.50
κ_0, Å^{-1}	0.676	0.405
$\ln g(0)$, (meV)^2	5.2	1.5
ϵ_0	0.080	0.30
$\ln \epsilon_0$	−2.53	−1.20
q_m, Å^{-1}	0.066	0.22
$q_m a/\pi$	0.082	0.27
f^*	0.34	1.8
f^*/f	0.058	0.31

so that $\lambda^*/\lambda \cong c_V^*/c_V = f^*/f$, and the value $D = \lambda/\rho c_V$ given in Table I is indeed realistic. Note that in this point we deviate from Ref. [1], where in addition the fraction $(f^*/f)M$ was related to the oxygen rotations. This fraction, however, has no obvious physical meaning (but rather $(f/f^*)M$).

With this new value of D we have to re-estimate the parameter $\gamma_0 = Dq^2$ of equation (3.22). A representative value of q is obtained with the help of the average $\bar{q}$ defined in (4.23), taken at $\epsilon_{res} \cong 0.013$ which corresponds to the temperature resolution of Ref. [4], see also [1]. From equation (C.9) we find $\bar{q}(\epsilon_{res}) = 0.021$ Å and

$$\gamma_{res} = D\bar{q}^2(\epsilon_{res}) = 0.12\,\text{meV} \tag{5.1}$$

which happens to be the same as the value obtained in Ref. [1] from the wave number resolution $q_{res} \cong 0.003$ Å^{-1} (note that here $Dq_{res}^2 = 0.0073$ meV). Since γ_{res} represents a lower bound of γ_0 the minimum $\delta^2 = 0.3 \pm 0.1$ (meV)^2 obtained in Ref. [4] is easily explained by the estimates of Ref. [1], so that there is indeed no discrepancy with (1.5).

With the above interpretation of the ratio f^*/f it is also possible that f^* goes to zero at T_c, which means that all the thermal degrees of freedom freeze out. In particular, $f^* \propto \epsilon^{4\gamma-2}$ would lead to a finite limit of δ^2 at T_c. On the other hand, as remarked in Ref. [1] the value of γ_{res} disagrees with the bound $\gamma_0 < 0.02$ meV given in Ref. [4].

Note that both values $\bar{q}(\epsilon_{res})$ and q_{res} are smaller than the critical q_m of Table II. And since $q_m a/\pi = 0.082 \ll 1$ the hydrodynamic form $\gamma_0 = Dq^2$ as well as the average $\bar{q}$ defined in (4.23) are well justified. We see, therefore, that, apart from (5.1) the consistency of our fit with the $SrTiO_3$-data of Ref. [4] is quite good. As to $LaAlO_3$ the data of Ref. [5] are unfortunately not sufficient to go beyond the comment made in Ref. [1].

ii. $SrTiO_3$: Electron paramagnetic resonance

The parameters of the fit of the EPR data for $SrTiO_3$ [21] obtained in Fig. 2 of Ref. [1] are collected in Table III. In the fast motion regime the impurity spin senses a time-averaged displacement fluctuation. This time average smears out the microscopically long range correlations in equation (3.34), so that the impurity centre appears strongly localized. Hence the $\vec{q}$-integration in equation (3.36) extends over the full Brillouin zone and $q_m^f = \pi/a$.

In the slow motion regime the impurity spin senses an instantaneous displacement fluctuation. Hence the microscopically long range correlations simulate a large ex-

Table III
Numerical values oft he parameters characterizing the critical behaviour of $SrTiO_3$ above $T_c = 105$ K, as obtained from the EPR data of Ref. [21] fitted in Fig. 2 of Ref. [1]. The slow and fast motion regimes are defined relative to the reciprocal EPR linewidth ΔH

Parameter	Slow motion	Fast motion
ν	0.65	0.65
κ_0, Å^{-1}	1.19	1.19
ΔH_0, 10^{-5} meV	9.0	0.25 $\times (1+\delta^2/\Gamma\bar{\gamma})^{-1}$
ΔH_0, Gauss	16	0.42 $\times (1+\delta^2/\Gamma\bar{\gamma})^{-1}$
ϵ_0	0.022	0.55
$\ln \epsilon_0$	−3.82	−0.60
q_m, Å^{-1}	0.10	0.805
$q_m a/\pi$	0.12	1

tention of the impurity centre which reaches over many unit cells and $q_m^s \ll \pi/a$. Since equations (3.32) and (3.37) contain the same $\vec{q}$-integral it is quite satisfying that the value of q_m^s in Table III is of the same magnitude as the q_m-values of Table II.

The value of κ_0 in Table III is obtained from $q_m^f = \pi/a$ and the fitted value of ϵ_0^f via equations (4.5) and (4.7). Using the same value of κ_0 and of ν in the slow motion regime the value of q_m^s is then determined from the fitted value of ϵ_0^s. With this ϵ_0^s equation (4.27) leads to $C = 18.6$ [1] in accord with Ref. [21]. As remarked in Ref. [1] the slow motion fit is quite insensitive to the value of ν; we also produced a fit with the value $\nu = 0.92$ of Table II which covers both the slow and fast motion data of Ref. [21].

Insertion of the values of Tables I and III into equation (4.24) yields

$$\Gamma(1 + \delta^2/\Gamma\bar{\gamma}) \cong \delta^2/\bar{\gamma} = 4400 \text{ meV} \tag{5.2}$$

(note that in Ref. [1] this value is too small by a factor of π). This is to be compared with the value obtained from equations (4.5), (4.13), (4.23) and (C.10) and Tables I, II, III (note that, according to the values of ϵ_0 of Table III, $x \ll 1$ holds practically for all data in Fig. 2 of Ref. [1])

$$\delta^2/\bar{\gamma} = (g(0)/D\kappa_0^2)\,\epsilon^{2\nu-2} = 0.16\,\epsilon^{-0.7} \text{ meV}. \tag{5.3}$$

The discrepancy with (5.2) comes from the fast motion fit where $\bar{\gamma}$ given by (4.23) is not justified since $q_m = \pi/a$ while $\gamma_0 = Dq^2$ is valid only for hydrodynamic values $q \ll \pi/a$. Therefore it was argued in Ref. [1] that (5.2) is preferable to (5.3) and may be used to estimate the average $\bar{\gamma}'$ of the central peak width γ_0' defined in equation (D.11) and approximately given in equations (D.8).

For $\omega_0^2 \ll \delta^2$ which according to Figure 1 of Ref. [1] holds for $\ln\epsilon \lesssim -2.2$ we deduce from (D.8) $\bar{\gamma}' \cong \omega_0^2(\delta^2/\bar{\gamma})^{-1}$. With equations (4.2), (5.2) and with λ_2 from Table I, κ_0 and ν from Table III we find, $\bar{\gamma}'$ being measured in Gauss [1],

$$\ln(T\bar{\gamma}') = 13.1 + 1.30\ln\epsilon; \quad \ln\epsilon < -2.2 \tag{5.4}$$

(note that in Ref. [1] the wrong factor π in (5.2) is just compensated by having used for κ_0 in (5.4) the critical value of Table I instead of the value of Table III). Equation (5.4) or $\bar{\gamma}'/\epsilon^{1.30} \cong 0.029$ meV $\cong 5000$ Gauss $\cong 4.4 \times 10^{10}$ sec^{-1}, valid for $\epsilon \ll 0.1$ is the main result here, giving the first estimate of a central peak width (see also Ref. [11]).

Equation (5.4) represents the dividing line between slow and fast motion regimes (the broken line in Figure 2 of Ref. [1]). This interpretation also follows from equation (4.29) if we identify the average $(2\sqrt{2}/\pi)\ \langle\gamma_0\rangle$ with the above $\bar{\gamma}'$. The transition temperature between the two regimes obtained in this way is the intersection between the slow motion line and equation (5.4), i.e. according to Figure 2 of Ref. [1], $\ln \epsilon_{tr} \cong -4.6$ or $\epsilon_{tr} \cong 0.010$.

iii. Nb_3Sn: Neutron scattering

The available parameters for Nb_3Sn are collected in Table IV. The elastic parameters λ_2, α, β_0 and the exponent γ are discussed in Appendix B. We note that the value of the exponent γ is almost the same as that for $SrTiO_3$ from Ref. [21]. The q-dependence of equation (1.6), $\delta_0 = \lambda q$ in equations (3.23), agrees with Figure 10 of Ref. [6]. This

Table IV
Numerical values of the parameters characterizing the Martensitic phase transition of Nb_3Sn

$a = 5.288$ Å	[25]
$M = 397.4$ g/mole	
$\rho = 4.46$ g/cm^3	
$\lambda_2\beta_0 = 3.34 \times 10^{10}$ cm^2/sec^2	[6, 18]
$\alpha = 5.88$	[18]
$\beta_0 = 1.05$	[18]
$T_c = 45$ K	[6, 18]
$Mc_V(T_c) = 25.6$ J/mole K	[26]
$f = 3.07$	
$\gamma = 1.35$	[6, 18]
$\lambda^2 = 0.417 \times 10^{10}$ cm^2/sec^2	[6]

Figure 10 also indicates that λ is at most weakly temperature dependent, which is consistent with the fact that from Figure 12 of Ref. [6] a unique value of λ^2 can be extracted. This Figure 12 also yields values for $\lambda_2\beta_0$ and γ which are consistent with those obtained from Ref. [18] in Appendix B.

From equations (4.14), (4.17) and with the values of Table IV we deduce

$$\frac{(f^*/f)\,\epsilon^{-2\gamma+2}}{(q_m a/\pi)^3\,(1+\epsilon)\,\vartheta(\epsilon)} = \frac{\pi\gamma^2\lambda_2\beta_0^2}{9\alpha f\lambda^2} = 0.30 \tag{5.5}$$

which is a very reasonable number. Since $(1+\epsilon)\,\vartheta(\epsilon)$ is of order one the most obvious conclusion from equation (5.5) is that, as discussed in the case of the neutron scattering data for $SrTiO_3$, $f^* \propto \epsilon^{2\gamma-2}$.

In conclusion we hope to have demonstrated the validity of our hydrodynamic understanding of soft mode related central peaks. The theory presented in this paper may be oversimplified in many respects and therefore fail to explain certain finer features such as the parameters γ_0, q_m and f^*. But the quality of the experimental results does not allow at present to go beyond the 10 to 30% accuracy of the fits discussed in this last Section.

APPENDIX A

Canonical Fluctuation Dynamics

In order to gain a systematic description of the dynamics of the fluctuations $\vec{u}$ and δT we here define canonical variables and construct a Hamiltonian which leads to the equations of motion (2.8) and (2.12).

It is important to realize, however, that dissipative terms cannot be described by a time-independent Hamiltonian because the corresponding terms in the equations of motion have the wrong signature under time reversal [2][2]). We therefore put $\Gamma = 0$, $\eta_{ij,kl} = 0$ and $D = 0$ in this appendix, it being understood that the corresponding terms can be generated by a linear response treatment based on the dynamics developed here.

The total free energy fluctuation is given by

$$F[\vec{u}, \delta T] = F_L - \int_V d^3r \frac{\rho c_V^*}{2T} (\delta T)^2 \tag{A.1}$$

where F_L is the lattice part (2.7), so that with (2.6)

$$-\frac{\delta F}{\delta(\delta T)} = \rho\left(s_L + \frac{c_V^*}{T}\delta T\right) = \rho\delta s. \tag{A.2}$$

Since the natural canonical coordinates turn out to be $\vec{u}$ and $\rho\delta s$ we perform a Legendre transformation to the internal energy fluctuation

$$U[\vec{u}, \rho\delta s] = F + \int_V d^3r\, \delta T \rho\delta s \tag{A.3}$$

so that, according to (A.2) and (2.10),

$$\frac{\delta U}{\delta(\rho\delta s)} = \delta T[\vec{u}, \rho\delta s] = \frac{T}{\rho c_V^*}\rho\delta s + \frac{T}{2c_V^*}\frac{\partial\omega_0^2}{\partial T}\vec{u}^2 + \frac{T}{2\rho c_V^*}\sum_{ijkl}\frac{\partial C_{ij,kl}}{\partial T}\theta_{ij}\theta_{kl}. \tag{A.4}$$

The dynamics of the system may now be obtained by associating canonical momenta $\vec{p}$ and π to $\vec{u}$ and $\rho\delta s$, respectively and by defining the Hamiltonian as

$$H[\vec{p}, \pi; \vec{u}, \rho\delta s; t] = U[\vec{u}, \rho\delta s] + \int_V d^3r \frac{\vec{p}^2}{2\rho}. \tag{A.5}$$

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[2]) A homogeneous equation of motion of type (2.8) is obtained with a *time-dependent* Hamiltonian of the form $H(t) = (1/2)(e^{-\Gamma t}p^2 + \omega_0^2 e^{+\Gamma t}q^2)$. I am grateful to W. Schlupp for this example.

Here the parametric time dependence comes from the external force $\vec{f}(\vec{r},t)$. The canonical equations of motion now follow with the help of (2.7) and (A.1) to (A.5),

$$\dot{\vec{u}} = \frac{\delta H}{\delta \vec{p}} = \frac{\vec{p}}{\rho}$$

$$\dot{\vec{p}} = -\frac{\delta H}{\delta \vec{u}} = -\rho\left\{\omega_0^2 + \frac{\partial \omega_0^2}{\partial T}\delta T - \frac{1}{\rho}C\nabla \otimes \nabla \right.$$

$$\left. -\frac{1}{\rho}\frac{\partial C}{\partial T}[(\nabla \delta T) + \delta T \nabla] \otimes \nabla + O(\vec{u}^2)\right\}\vec{u} + \rho\vec{f}$$

$$(\rho\delta s)^{\cdot} = \frac{\delta H}{\delta \pi} = 0$$

$$\dot{\pi} = -\frac{\delta H}{\delta(\rho\delta s)} = \frac{T}{\rho c_V^*}\rho\delta s + \frac{T}{2c_V^*}\frac{\partial \omega_0^2}{\partial T}\vec{u}^2 + \frac{T}{2\rho c_V^*}\sum_{ijkl}\frac{\partial C_{ij,kl}}{\partial T}\theta_{ij}\theta_{kl} \equiv \delta T[\vec{u}, \rho\delta s]. \quad \text{(A.6)}$$

These equations are the same as (2.8) and (2.12) with $\Gamma = 0$, $\eta_{ij,kl} = 0$ and $D = 0$.

In order to develop a many-body theory of classical observables we restrict the fields $\vec{u}(\vec{r},t)$, $\rho\delta s(\vec{r},t)$ and the associated momenta to the discrete set of points $\vec{R}$ spanning cells whose linear dimension, b, is a multiple of the lattice constant a (an *even* multiple for *staggered* fields). In renormalization group theory this corresponds to integrating out the irrelevant Fourier components $u_{i\vec{k}}$ etc. with $|\vec{k}| > \Lambda$ [27]. Writing the complete set of these discrete coordinates $\{u_i(\vec{R},t), b^3\rho\delta s(\vec{R},t)\}$ as vector $Q(t)$ and the canonically conjugate momenta $\{b^3 p_i(\vec{R},t), \pi(\vec{R},t)\}$ as vector $P(t)$ equations (A.6) are the special case $A = P,Q$ of the following general equation of motion:

$$\dot{A}(P,Q;t) = LA + \partial A/\partial t. \quad \text{(A.7)}$$

Here

$$L = \frac{\delta H}{\delta P}\cdot\frac{\delta}{\delta Q} - \frac{\delta H}{\delta Q}\cdot\frac{\delta}{\delta P} \quad \text{(A.8)}$$

is the Liouville operator where the dot designates a scalar product in the vector space of P and Q. In terms of Poisson brackets

$$[A,B] = -[B,A] = \frac{\delta A}{\delta Q}\cdot\frac{\delta B}{\delta P} - \frac{\delta B}{\delta Q}\cdot\frac{\delta A}{\delta P} \quad \text{(A.9)}$$

equation (A.7) may also be written as

$$LA = [A,H] \quad \text{(A.10)}$$

The formalism could be developed further into a Hilbert space formulation of classical mechanics [28]. This, however, is not our aim here.

Writing the set of external forces $\{b^3\rho f_i(\vec{R},t),0\}$ as vector $E(t)$ and making use of (2.7), (A.1) and (A.3) the Hamiltonian (A.5) may be written as

$$H(P,Q;t) = H_0(P,Q) - Q\cdot E(t) \tag{A.11}$$

where H_0 is independent of the external force and hence contains no parametric time dependence. Similarly we have from (A.8)

$$L(t) = L_0 + E(t)\cdot\delta/\delta P. \tag{A.12}$$

The classical time evolution depends on the initial values P, Q which we take at $t=0$. It is convenient to express this dependence by a double time argument [29],

$$\begin{aligned} &P(t,0) = P(P,Q;t); \quad P(0,0) = P \\ &Q(t,0) = Q(P,Q;t); \quad Q(0,0) = Q. \end{aligned} \tag{A.13}$$

Without parametric time dependence, $E=0$, the time evolution can be obtained in the form of a Taylor series, expressing the nth time derivative with the help of (A.7) as L_0^n,

$$A(t) = \sum_{n=0}^{\infty} \frac{(t-t_0)^n}{n!} L_0^n A(t_0) = \exp[L_0(t-t_0)]\, A(t_0). \tag{A.14}$$

Including parametric time dependence iteration of equation (A.7) yields successively more complicated expressions,

$$\ddot{A} = L^2 A + \dot{L}A + \partial^2 A/\partial t^2$$

$$\dddot{A} = L^3 A + 3L\dot{L}A + \ddot{L}A + \partial^3 A/\partial t^3$$

etc.

Restricting the class of functions A by the condition $\partial A/\partial t = 0$ the time evolution may be expressed as

$$A(t) = U(t,t_0)A(t_0) \tag{A.15}$$

where, according to (A.7), the operator U obeys the equation

$$\frac{\partial}{\partial t} U(t,t_0) = L(t)\, U(t,t_0). \tag{A.16}$$

Integration with $U(t,t) = 1$ yields

$$U(t,t_0) = T\exp\left\{\int_{t_0}^{t} dt'\, L(t')\right\} \tag{A.17}$$

where T is the chronological ordering operator and the group property

$$U(t,t')\,U(t',t_0) = U(t,t_0) \tag{A.18}$$

immediately follows.

Application of equations (A.15), (A.17), (A.18) and (A.12) to the response function (3.1) yields in the notation (A.13) and labelling vector components,

$$\begin{aligned}\frac{\delta Q_\alpha(t,0)}{\delta E_\beta(t')} &= \frac{\delta U_{\alpha\gamma}(t,0)}{\delta E_\beta(t')}\,Q_\gamma \\ &= \theta(t-t')\,\theta(t')\,U_{\alpha\gamma}(t,t')\frac{\delta}{\delta P_\beta}\,(U_{\gamma\delta}(t',0)\,Q_\delta) \\ &= \theta(t-t')\,\theta(t')\,U_{\alpha\gamma}(t,t')\,\delta Q_\gamma(t',0)/\delta P_\beta. \end{aligned} \tag{A.19}$$

This equation depends on three times but, due to the deterministic nature of the classical time evolution, only the time of preparation, $t=0$, and the time of perturbation, t', are relevant. Thus it is natural to define the classical response function $\tilde{\chi}(t)$ as the change in the initial values, induced by the perturbation,

$$\begin{aligned}\tilde{\chi}_{\alpha\beta}(t) &= \lim_{t'\to t-0} \delta Q_\alpha(t,0)/\delta E_\beta(t')\big|_{E=0} \\ &= \theta(t)\,\delta Q_\alpha(t,0)/\delta P_\beta \\ &= -\theta(t)\,[Q_\alpha(t,0), Q_\beta(0,0)] \end{aligned} \tag{A.20}$$

where we have made use of the relation

$$\delta A_\alpha/\delta P_\beta = [Q_\beta, A_\alpha] \tag{A.21}$$

which follows from (A.9).

In order to be physically meaningful the result (A.20) has to be freed from the particular initial values by taking a statistical average over the fluctuations P,Q. These fluctuations essentially form a Gaussian distribution around zero with a width depending on temperature. Hence, as in renormalization group theory [15, 27], the distribution is that of the canonical ensemble[3])

$$\mathscr{P}_0(P,Q) = \exp\{(\phi_0 - H_0(P,Q))/k_B\,T\}. \tag{A.22}$$

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[3]) In the case of friction type [30] or Ginzburg-Landau type [15] equations of motion the distribution function $G(u_0,u,t)$ at time t of the variables u with initial values u_0 (G is the fundamental solution, i.e. $G(u_0,u,0)=\delta(u-u_0)$, of the associated Fokker–Planck equation) can be constrcted with the device of fluctuating forces [30]. G has the property that (i) $G(u_0,u,\infty)=P_0(u)$, independent of u_0, and (ii) $\int du_0\,P_0(u_0)\,G(u_0,u,t) = P_0(u)$, independent of t. Thus the averaging over the initial values with the equilibrium distribution $P_0(u)$ leads to equilibrium values. Furthermore, $P_0(u)=\exp[(\phi_0 - H_0(u))/k_B\,T]$ is essentially a Gaussian distribution; for a velocity u (Brownian motion) $H_0(u)$ is the kinetic energy [30], for a thermodynamic variable u (diffusion) $-H_0(u)/k_B\,T$ is the entropy [31, 15]. The mentioned equations of motion [15, 30] are all of first order in the time derivative (rate equations). It is the virtue of the canonical formalism to cast the second order equations of motion of mechanics also into the form of rate equations. The difference is that the equations of motion mentioned before [15, 30] are essentially dissipative. It is therefore important to note that the parameters describing this dissipation (the friction coefficient f of Ref. [30] or the time scale Γ_0 of Ref. [15]) do not enter the equilibrium distribution function $P_0(u)$. In fact, the only relevant feature for the above properties (i) and (ii) to hold is that the equations of motion have the form of rate equations so that there exists an associated Fokker–Planck equation. This justifies both the use of averages (A.24) over initial values and the form (A.22) of the equilibrium distribution.

Here the normalization

$$\int_{-\infty}^{+\infty}\int dP\,dQ\,\mathscr{P}_0(P,Q) = 1 \tag{A.23}$$

determines the unperturbed 'free energy' $\phi_0(T)$ [27] (ϕ_0 of course has nothing to do with the thermodynamic free energy of the system) and the average is defined as

$$\langle A\rangle_0 = \int_{-\infty}^{+\infty}\int dP\,dQ(A\,\mathscr{P}_0). \tag{A.24}$$

A straightforward application of the definitions (A.8) and (A.9) leads to the identities

$$\begin{aligned} L_0(AB) &= (L_0 A)\,B + A(L_0 B) \\ L_0[A,B] &= [(L_0 A),B] + [A,(L_0 B)] \end{aligned} \tag{A.25}$$

which by iteration can be generalized into

$$\begin{aligned} e^{L_0 t}(AB) &= (e^{L_0 t}A)(e^{L_0 t}B) \\ e^{L_0 t}[A,B] &= [(e^{L_0 t}A),(e^{L_0 t}B)]. \end{aligned} \tag{A.26}$$

On the other hand it follows by partial integration that for any A bounded by a polynomial in P,Q

$$\int_{-\infty}^{+\infty}\int dP\,dQ\,L_0(A\,\mathscr{P}_0) = 0. \tag{A.27}$$

Since according to (A.10), (A.25) $L_0(H_0^n) = 0$ for $n = 0, 1, \ldots$ it follows by iteration of equation (A.25) with $B = H_0^n$ that for any power series f

$$L_0(Af(H_0)) = (L_0 A)f(H_0). \tag{A.28}$$

With this result iteration of (A.27) yields, in the notation of (A.24),

$$\langle e^{L_0 t}A\rangle_0 = \langle A\rangle_0. \tag{A.29}$$

In particular, application of (A.26) results in the time translation invariance [29]

$$\begin{aligned} \langle A(t)\,B(t')\rangle_0 &= \langle A(t+\tau)\,B(t'+\tau)\rangle_0 \\ \langle [A(t),B(t')]\rangle_0 &= \langle [A(t+\tau),B(t'+\tau)]\rangle_0 \end{aligned} \tag{A.30}$$

where the unperturbed time evolution (A.14) is understood.

Taking the average (A.24) the initial time arguments in equation (A.20) may obviously be dropped,

$$\langle \tilde{\chi}_{\alpha\beta}(t)\rangle_0 = -\theta(t)\langle [Q_\alpha(t),Q_\beta(0)]\rangle_0. \tag{A.31}$$

This response function has the correct causal behaviour, so that its Fourier transform

$$\chi(\omega) = \int dt\, e^{i\omega t} \langle \tilde{\chi}(t) \rangle_0 \tag{A.32}$$

satisfies the dispersion relation

$$\chi(z) = \int \frac{d\omega}{\pi} \frac{\chi''(\omega)}{\omega - z}; \quad \mathrm{Im}\, z > 0. \tag{A.33}$$

From (A.31), (A.33) one finds, with (A.30) and the antisymmetry (A.9),

$$2i\chi''_{\alpha\beta}(\omega) = -\langle [\hat{Q}_\alpha(\omega), Q_\beta(0)] \rangle_0 \tag{A.34}$$

where $\hat{Q}(\omega)$ is the Fourier transform of $Q(t)$. Making use of (A.21) equation (A.34) takes the form, after a partial integration and use of (A.22),

$$2i\chi''_{\alpha\beta}(\omega) = \frac{1}{k_B T} \left\langle \hat{Q}_\alpha(\omega) \frac{\delta H_0}{\delta P_\beta} \right\rangle_0. \tag{A.35}$$

Writing $\delta H_0 / \delta P = L_0 Q$ and shifting the Liouvillean to $\hat{Q}(\omega)$ with the help of (A.27), (A.28) we obtain

$$2i\chi''_{\alpha\beta}(\omega) = \frac{i\omega}{k_B T} \langle \hat{Q}_\alpha(\omega)\, Q_\beta(0) \rangle_0. \tag{A.36}$$

Defining the correlation function

$$\tilde{S}_{\alpha\beta}(t) = \frac{\rho}{2\pi} Q_\alpha(t, 0)\, Q_\beta(0, 0) \tag{A.37}$$

and using for it the analogous definition (A.32) we finally arrive at the classical fluctuation–dissipation theorem [29]

$$S(\omega) = \frac{\rho k_B T}{\pi\omega} \chi''(\omega) \tag{A.38}$$

in agreement with the limit $\hbar\,\omega \ll k_B T$ of the quantum mechanical case [32].

APPENDIX B

The Elastic or Staggered-Elastic Matrix for Cubic Symmetry

The matrix (3.17) may be written

$$R(\vec{q}) = \begin{pmatrix} \lambda_2 + (\lambda_1 - \lambda_2)\, x^2, \lambda_3\, xy, \lambda_3\, xz \\ \lambda_3\, yx, \lambda_2 + (\lambda_1 - \lambda_2)\, y^2, \lambda_3\, yz \\ \lambda_3\, zx, \lambda_3\, zy, \lambda_2 + (\lambda_1 - \lambda_2)\, z^2 \end{pmatrix} q^2 \tag{B.1}$$

where

$$\hat{q} = \vec{q}/q = (x, y, z) \tag{B.2}$$

and

$$\lambda_1 = c_{11},\ \lambda_2 = c_{44}$$
$$\lambda_3 = c_{12} + c_{44} \tag{B.3}$$

are the cubic elastic or staggered-elastic constants.

Introducing the unit polarization vector

$$\hat{e} = (u, v, w) \tag{B.4}$$

the eigenvalue equation

$$R(\vec{q})\,\hat{e}(\hat{q}) = r(\vec{q})\,\hat{e}(\hat{q}) \tag{B.5}$$

takes the form

$$\begin{aligned} s &= x^2 + \frac{\lambda_3}{\lambda_1 - \lambda_2}\frac{x}{u}(vy + wz) \\ &= y^2 + \frac{\lambda_3}{\lambda_1 - \lambda_2}\frac{y}{v}(wz + ux) \\ &= z^2 + \frac{\lambda_3}{\lambda_1 - \lambda_2}\frac{z}{w}(ux + vy) \end{aligned} \tag{B.6}$$

where the eigenvalue is written as

$$r = [\lambda_2 + (\lambda_1 - \lambda_2)\, s]\, q^2. \tag{B.7}$$

Defining the quantities

$$\mu = 1 - \frac{\lambda_3}{\lambda_1 - \lambda_2} \tag{B.8}$$

$$\zeta = \frac{\lambda_3}{\lambda_1 - \lambda_2}(\hat{e}\cdot\hat{q}) \tag{B.9}$$

and making use of (B.2) and (B.4) equations (B.6) can be solved for u, v and w with the result that

$$\hat{e}(\hat{q}) = \zeta\left(\frac{x}{s - \mu x^2}, \frac{y}{s - \mu y^2}, \frac{z}{s - \mu z^2}\right). \tag{B.10}$$

Insertion of (B.10) and (B.8) into (B.9) then leads to the cubic equation in s

$$s^2(s - 1) = \mu(2 - \mu)\, P(\hat{q})\, s - \mu^2(3 - 2\mu)\, Q(\hat{q}) \tag{B.11}$$

where

$$P(\hat{q}) = x^2 y^2 + y^2 z^2 + z^2 x^2 \leqslant 1/3$$
$$Q(\hat{q}) = x^2 y^2 z^2 \leqslant 1/27. \tag{B.12}$$

For given values λ_1, λ_2, λ_3 equations (B.8), (B.11) and (B.12) constitute an elementary determination of the eigenvalues (B.10) in terms of the wave normal (B.2). The three solutions of (B.11) can for example be determined graphically as intersections of the variable straight line on the right hand side with the fixed curve of third degree on the left hand side. In particular one finds for the symmetry directions one longitudinal and two transverse solutions as given in Table V.

Table V
Eigenvalue parameters of the elastic or staggered elastic matrix for the longitudinal and transverse modes along symmetry axes, for cubic symmetry. The labels are those of Refs. [17] and [6], respectively

Const. $\hat{q}$	r_l/q^2	Label	r_t/q^2	Label	Const. $\hat{e}_t$
(1, 0, 0)	λ_1	Δ_2	λ_2	Δ_5, T	(0, 1, 0) (0, 0, 1)
(1, 1, 0)	$\frac{1}{2}(\lambda_1 + \lambda_2 + \lambda_3)$	$\sum_1$	λ_2 $\frac{1}{2}(\lambda_1 + \lambda_2 - \lambda_3)$	$\sum_2$, T_2 $\sum_3$, T_1	(0, 0, 1) (1, −1, 0)
(1, 1, 1)	$\frac{1}{3}(\lambda_1 + 2\lambda_2 + 2\lambda_3)$	Λ_2	$\frac{1}{3}(\lambda_1 + 2\lambda_2 - \lambda_3)$	Λ_3	(1, −1, 0) (1,0, −1)

From Table 2 of Ref. [17] we deduce, for the high temperature phase of $SrTiO_3$, $\lambda_1 = 8 \pm 2$, $\lambda_2 = 205 \pm 27$, $\lambda_1 + \lambda_2 + \lambda_3 = 224 \pm 44$ and $\lambda_1 + 2\lambda_2 - \lambda_3 = 393 \pm 69$ in units of 10^8 cm^2/sec^2. The last two values combined with the first two yield, in the same units, $\lambda_3 = 11 \pm 52$ and 25 ± 88 which both are compatible with $\lambda_3 = 0$ or, according to (B.8), with $\mu = 1$. In this case the solutions of equation (B.11) are trivial, namely $s = x^2, y^2, z^2$, so that the eigenvalues (B.7) reduce to the expressions given in Ref. [1],

$$r_i(\vec{q}) = \lambda_2[1 - (1 - \Delta)\,\hat{q}_i^2]\,q^2 \tag{B.14}$$

where

$$\Delta = \lambda_1/\lambda_2 \tag{B.15}$$

is the anisotropy parameter of Schwabl [9] and has the value 0.039 ± 0.015 [17].

In equation (3.33) we need the inverse of the matrix $R(\vec{q})$. Introducing the abbreviations

$$a_1 = [\lambda_2 + (\lambda_1 - \lambda_2)\,x^2]\,q^2, \text{ etc.} \tag{B.16}$$

for the diagonal elements of R and

$$b_1 = \lambda_3\, yzq^2, \text{ etc.} \tag{B.17}$$

for the off-diagonal elements the corresponding elements of R^{-1} are

$$A_1 = (a_2 a_3 - b_1^2)/\|R\|, \text{ etc.} \tag{B.18}$$

and

$$B_1 = (b_2 b_3 - a_1 b_1)/\|R\|, \text{ etc.} \tag{B.19}$$

where

$$\|R\| = a_1 a_2 a_3 + 2b_1 b_2 b_3 - \sum_i a_i b_i^2 \tag{B.20}$$

is the determinant of R. With equations (B.16) to (B.20) the evaluation of

$$(\vec{q}, R^{-1}(\vec{q})\vec{q}) = \sum_i A_i q_i^2 + 2(B_1 q_2 q_3 + \text{cycl.}) \tag{B.21}$$

is straightforward and yields

$$\alpha\lambda_2(\vec{q}, R^{-1}(\vec{q})\vec{q}) = N(\hat{q})\Big/\left[N(\hat{q}) + \frac{\beta}{\alpha} + 4\,\frac{1-\beta}{\alpha}\,Q(\hat{q})\right]. \tag{B.22}$$

Here we have introduced two new parameters

$$\alpha = (\lambda_1 - \lambda_2 + \lambda_3)/(2\lambda_2) \tag{B.23}$$

and

$$\beta = v_0^2/\lambda_2 = (\lambda_1 + \lambda_2 - \lambda_3)/(2\lambda_2) \tag{B.24}$$

and the function

$$N(\hat{q}) = 1 - 4(1-\beta)\,P(\hat{q}) + 12(1-\beta)^2 Q(\hat{q}) \tag{B.25}$$

where $P(\hat{q})$, $Q(\hat{q})$ are defined in (B.12).

The parameters α and β are chosen such that α is to a good approximation independent of temperature while β becomes soft as

$$\beta = \beta_0\,\epsilon^{\gamma}. \tag{B.26}$$

With the identification of the modes in Table V we have extracted from Figure 1 of Ref. [18] $\alpha = 5.88 \pm 1.6$, valid for $T_c \leqslant T \lesssim 2\ T_c$, $\beta_0 = 1.05$, $\gamma = 1.35$ and $\lambda_2 = 3.17 \times 10^{10}$ cm^2/sec^2.

It is then obvious that the right hand side of (B.22) is never larger than one. And from (B.25) one finds that the numerator $N(\hat{q}) \geqslant \min(\frac{1}{9}, \beta)$. Hence the expression (B.22) is always between zero and one and the angular average

$$\vartheta(\epsilon) = \frac{1}{4\pi}\oint d\Omega_{\hat{q}}\,\alpha\lambda_2(\vec{q}, R^{-1}(\vec{q})\vec{q}) \tag{B.27}$$

is a slowly varying function of order one.

APPENDIX C

Evaluation of Wave number sums

In equations (3.32), (3.36) and (3.37) expressions of the form

$$I_n = (2\pi/\kappa)^3 V^{-1} \sum_{\vec{q}} (\omega_i^2(\vec{q})/\omega_0^2)^n \tag{C.1}$$

occur where integration is restricted by a cutoff. The simplest form of cutoff is obtained with the ellipsoid defined in (B.14),

$$r_z(\vec{q})/\lambda_2 = q_x^2 + q_y^2 + \Delta \cdot q_z^2 < q_m^2 \tag{C.2}$$

or in terms of the variable x defined in (4.7),

$$\nu^2 \equiv r_z(\vec{q})/\omega_0^2 = \xi^2 + \eta^2 + \zeta^2 < x^{-2} \tag{C.3}$$

where, according to (4.2),

$$\xi = q_x/\kappa, \quad \eta = q_y/\kappa, \quad \zeta = \sqrt{\Delta} q_z/\kappa. \tag{C.4}$$

With (4.4), (C.3) and (C.4) the integral (C.1) becomes

$$I_n(x) = (4\pi/\sqrt{\Delta}) \int_0^{1/x} (1+\nu^2)^n \nu^2 \, d\nu \tag{C.5}$$

or

$$V^{-1} \sum_{\vec{q}} \omega_i^{2n}(\vec{q}) = (\kappa/2\pi)^3 \omega_0^{2n} I_n(x). \tag{C.6}$$

The functions needed in Section 3 are

$$I_{-1}(x) = (4\pi/\sqrt{\Delta}) \left(\frac{1}{x} - \tan^{-1} \frac{1}{x} \right) \tag{C.7}$$

and

$$I_{-2}(x) = (2\pi/\sqrt{\Delta}) \left(\tan^{-1} \frac{1}{x} - \frac{x}{1+x^2} \right). \tag{C.8}$$

The average defined in (4.22) is also best evaluated in the coordinates (C.3), (C.4). Writing $q^2 = \kappa^2 \nu^2 [1 + ((1/\Delta) - 1) t^2]$ where $t = \zeta/\nu$ one finds

$$(\kappa/\bar{q})^2 = \frac{1}{2} \int_{-1}^{+1} \frac{dt}{1 + ((1/\Delta) - 1) t^2} \int_0^{1/x} (1+\nu^2)^{-2} \, d\nu \Big/ \int_0^{1/x} (1+\nu^2)^{-2} \nu^2 \, d\nu$$

$$= \frac{\tan^{-1} \sqrt{(1/\Delta) - 1}}{\sqrt{(1/\Delta) - 1}} \frac{\tan^{-1} \frac{1}{x} + \frac{x}{1+x^2}}{\tan^{-1} \frac{1}{x} - \frac{x}{1+x^2}} \tag{C.9}$$

or

$$\left(\frac{\bar{q}}{\kappa}\right)^2 \frac{\tan^{-1}\sqrt{(1/\Delta)-1}}{\sqrt{(1/\Delta)-1}} = \begin{cases} 1+O(x^2) \\ (1/3x^2)\,(1+O(x^{-2})). \end{cases} \tag{C.10}$$

Note that with other forms of the cutoff the integrals (C.7) to (C.9) become more complicated expressions which, however, vary little numerically. An exception is the integral in equation (3.33) which in terms of the angular integral (B.27) is simpler with a spherical cutoff $|\vec{q}|^2 < q_m^2$ instead of (B.2),

$$V^{-1} \sum_{\vec{q}} (\vec{q}, R^{-1}(\vec{q})\,\vec{q}) = (q_m^3/6\pi^2)\,\vartheta(\epsilon). \tag{C.11}$$

APPENDIX D

The Three-Pole Structure of the Response Function

Equations (1.1), (1.2) may be written as

$$\rho\chi_i = (\gamma_0 - i\omega)/D(\omega) \tag{D.1}$$

where the denominator is factorizable,

$$\begin{aligned} D(\omega) &= (\omega_i^2 - \omega^2 - i\omega\Gamma_0)\,(\gamma_0 - i\omega) - i\omega\delta_0^2 \\ &= (\omega_i'^2 - \omega^2 - i\omega\Gamma_0')\,(\gamma_0' - i\omega). \end{aligned} \tag{D.2}$$

The last equality leads to the following three conditions for the renormalized coefficients ω_i', Γ_0' and γ_0':

$$\begin{aligned} &\gamma_0' + \Gamma_0' = \gamma_0 + \Gamma_0 \\ &\gamma_0'\Gamma_0' + \omega_i'^2 = \gamma_0\Gamma_0 + \omega_i^2 + \delta_0^2 \\ &\gamma_0'\omega_i'^2 = \gamma_0\omega_i^2. \end{aligned} \tag{D.3}$$

Expressing the numerator of (D.1) in terms of the factors of $D(\omega)$,

$$\gamma_0 - i\omega = (a_i + i\omega b_i)\,(\gamma_0' - i\omega) + b_i(\omega_i'^2 - \omega^2 - i\omega\Gamma_0') \tag{D.4}$$

the real coefficients a_i and b_i are determined by the conditions

$$a_i + b_i(\Gamma_0' - \gamma_0') = 1, \qquad a_i\gamma_0' + b_i\omega_i'^2 = \gamma_0. \tag{D.5}$$

With these decompositions (D.2) and (D.4) the response function (D.1) splits into a central peak part and a soft mode part

$$\chi_i = \chi_i^{cp} + \chi_i^{sm} \tag{D.6}$$

where

$$\rho\chi_i^{cp} = b_i/(\gamma_0' - i\omega)$$
$$\rho\chi_i^{sm} = (a_i + i\omega b_i)/(\omega_i'^2 - \omega^2 - i\omega\Gamma_0') \tag{D.7}$$

explicitly exhibit the three-pole structure of χ_i.

The iterative solution of equations (D.3) used in Ref. [1] is

$$\omega_i'^2 = \Omega_i^2 + \gamma_0\Gamma_0\delta_0^2/\Omega_i^2 + O(\gamma_0^2)$$
$$\gamma_0' = \gamma_0\omega_i^2/\Omega_i^2 + O(\gamma_0^2)$$
$$\Gamma_0' = \Gamma_0 + \gamma_0\delta_0^2/\Omega_i^2 + O(\gamma_0^2) \tag{D.8}$$

where

$$\Omega_i^2(\vec{q}) = \omega_i^2(\vec{q}) + \delta_0^2. \tag{D.9}$$

Substituting into (D.5) we also find

$$a_i = 1 - \gamma_0\Gamma_0\delta_0^2/\Omega_i^4 + O(\gamma_0^2)$$
$$b_i = \gamma_0\delta_0^2/\Omega_i^4 + O(\gamma_0^2). \tag{D.10}$$

The fluctuation–dissipation theorem (3.29) then yields, to lowest order in γ_0,

$$\frac{\pi}{k_B T} S_i^{cp}(q,\omega) = \frac{\delta_0^2}{\Omega_i^2\omega_i^2}\frac{\gamma_0'}{\gamma_0' + \omega^2} \tag{D.11}$$

and

$$\frac{\pi}{k_B T} S_i^{sm}(q,\omega) = \frac{\Gamma_0}{(\Omega_i^2 - \omega^2)^2 + \omega^2\Gamma_0^2} \tag{D.12}$$

in accord with equations (6) and (7), respectively of Ref. [4] (except for the wrong factor γ'^2 in the numerator of equation (6) of Ref. [4]). The integrated intensities of the two parts are (equation (8) of Ref. [4])

$$I_i^{cp}(q) = \frac{2}{k_B T}\int_0^\infty S_i^{cp}(q,\omega)\,d\omega = \frac{\delta_0^2}{\Omega_i^2\omega_i^2} \tag{D.13}$$

and

$$I_i^{sm}(q) = \frac{2}{k_B T}\int_0^\infty S_i^{sm}(q,\omega)\,d\omega = \frac{1}{\Omega_i^2} \tag{D.14}$$

which lead to the sum rule (equation (5) of Ref. [4])

$$I_i^{cp} + I_i^{sm} = \frac{2}{k_B T}\int_0^\infty S_i(q,\omega)d\omega = \frac{1}{\omega_i^2}. \tag{D.15}$$

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