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# A phenomenological analysis of ultrasound near phase transitions

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**Abstract.** Phenomenological predictions for sound attenuation and velocity anomalies near critical points are presented. General symmetry properties of the linear elastic constants and the general properties of the low- and high-frequency behaviours of linear-response functions are used to develop phenomenological equations, where exponents, relaxation times and amplitudes are introduced as parameters to fit to the experimental data. Novel interpretation formulae, which combine the asymptotic high- and low-frequency limits of complex dynamic elastic constants, are constructed. It is shown that causality requirements give new relations between the amplitudes of ultrasonic attenuation and the velocity in the asymptotic limits of high and low frequency.

## 1. Introduction

Experimental methods used in investigations of phase transitions can be divided into two groups: (i) methods probing local properties and (ii) methods probing the macroscopic response. Examples from the first group are electron and nuclear magnetic resonance. Macroscopic properties are studied by dielectric constant and heat capacity measurements, and also by scattering experiments, where the external force is generated by, for example, a beam of light, x-rays, neutrons or acoustic phonons.

The latter case involves measurements of sound attenuation and velocity, since an applied sound wave may be considered as spatially homogeneous, i.e. $\xi^{-1} \gg q \simeq 0$ in the experimentally accessible region. Here, $\xi$ is the correlation length and $q$ is the wavevector ($\lambda \sim 50\ \mu$m is a typical ultrasound wavelength).

Nice illustrations of the statements made above with respect to local and macroscopic probes may be found in the papers by Halperin and Hohenberg (1967) and Leung *et al* (1979).

Ultrasound experiments near phase transitions in a number of systems (magnetic, liquid–gas, structural) have recently revealed *dynamic scaling within the asymptotic region*. This means that the dynamic elastic response function is found to behave according to the dynamic scaling hypothesis, with a macroscopic relaxation time obeying $\tau \sim \xi^z f(q\xi)$, where $z$ belongs to one of the dynamic universality classes. Some of the many examples to be found in the literature are given in a recent review by Fossheim

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and Fossum (1984). It is interesting to note that most examples of dynamical scaling of sound have been observed only during the past 3–4 years. These observations have lent increasing importance to the whole problem of the critical dynamics of sound.

The purpose of the present paper is to present a phenomenological description of these problems. The treatment will apply *general* symmetry arguments and simple analysis of linear response theory to construct theoretical models for sound near critical points. Thus, the treatment contains no theoretical technicalities. The general statements and results will be illustrated throughout the article on a system of cubic symmetry.

In § 2 of the present paper the connection is established between the complex elastic modulus and the sound velocity and attenuation. Section 3 contains symmetry considerations, concepts such as symmetrised and effective components of elastic constants are introduced. Various types of coupling between the order parameters and mechanical variables are considered in § 4. In § 5 simple equilibrium statistical mechanics is applied to derive scaling laws expressing the critical exponents of static elastic constants in terms of exponents of other thermodynamic quantities. It will be shown that only the symmetrised elastic constants can be expected to obey simple power law behaviour close to critical points, since the symmetrised components represent fundamental ways of distorting a given symmetry.

In § 6 the natural extension to include dynamics is presented. Linear response theory is applied to derive causal dynamic scaling functions. An exact result is found in the gaussian approximation, whereas a straightforward generalisation is made to other cases.

Results and expressions for sound velocity and attenuation are summarised and discussed in § 7, while an illustrative example is given in § 8.

Together with the present article we also publish an experimental paper on ultrasound experiments above the phase transition in $SrTiO_3$ (Fossum and Fossheim 1984, 1985, Fossum *et al* 1984) where the theory discussed below will be applied in the data analysis.

## 2. Ultrasound anomalies and mechanical response

The quantities most frequently measured in an ultrasonic experiment are (i) the sound velocity, $v$, and (ii) the sound attenuation, $\alpha$. A measurement of $v$ and $\alpha$ determines the full complex linear elastic modulus:

$$C = \mathrm{Re}C + \mathrm{i}\mathrm{Im}C. \tag{1}$$

$C$ may be defined through Hooke's law

$$\sigma = C\varepsilon \tag{2}$$

where $\sigma$ is the mechanical stress and $\varepsilon$ is the strain. For simplicity the tensor indices are dropped in equation (1). Anisotropy will be discussed in the following chapters. In general, the thermodynamic quantity $C$ contains information about all kinds of microscopic processes in the system which is studied. Near a phase transition $C$, as with any other susceptibility, will be split into two parts:

$$C = C_0 + \Delta C \tag{3}$$

where $\Delta C$ is the critical part and $C_0$ is the background part. $C_0$ contains *no* information

about the phase transition and is thus assumed to be a smooth function of the fields relevant to the critical behaviour (temperature, pressure, magnetic field etc).

Assuming $\mathrm{Im}\, C \ll \mathrm{Re}\, C$ and $\mathrm{Re}\,\Delta C \ll \mathrm{Re} C_0$, it is straightforward to derive the following expressions for the anomalous parts $\Delta v$ and $\Delta\alpha$ of the velocity and attenuation respectively:

$$\Delta v/v_0 = (1/2\rho v_0^2)\,\mathrm{Re}\Delta C \tag{4}$$
$$v_0\Delta\alpha/\omega = (1/2\rho v_0^2)\,\mathrm{Im}\Delta C$$

where $v_0^2 = \mathrm{Re}C_0/\rho$ is the background velocity squared, and $\rho$ is the density of the medium.

## 3. Linear elasticity: symmetry considerations

When crystal anisotropy is taken into account, Hooke's law, equation (2) is replaced by

$$\sigma_{ij} = C_{ijkl}\varepsilon_{kl} \tag{5}$$

or

$$\varepsilon_{ij} = S_{ijkl}\sigma_{kl} \tag{6}$$

where summation over repeated indices is understood. $\sigma_{ij} = \sigma_{ji}$ is the elastic stress tensor and $\varepsilon_{kl} = \varepsilon_{lk}$ is the linear elastic strain tensor.

The fourth-rank tensors $C_{ijkl}$ and $S_{ijkl}$ are the linear elastic constant and the linear compliance respectively.

Introductions to linear elasticity, including discussions of the validity of Hooke's law are, for example, given by Landau and Lifshitz (1959), Nye (1960) and Pollard (1977). Details will not be cited here.

Note, however, that the elastic constants measured in ultrasound experiments are the adiabatic ones. The reason is that the ultrasound frequency, $\omega$, is too high for the temperature variations accompanying the sound wave to be transported across a distance of one acoustic wavelength during times of the order of one period $(2\pi/\omega)$. One can then regard any part of the body as thermally insulated at all times, i.e. the wave motion is adiabatic. One case where a distinction between isothermal and adiabatic responses is important, is close to elastic phase transitions, i.e. structural transitions where the soft mode is an acoustic phonon (Rehwald 1973).

A fourth-rank tensor, such as $C$ or $S$, generally contains 81 independent components. This number is reduced drastically by symmetry. In the cubic case there are three independent components of the elasticity tensory, namely $C_{11}$, $C_{12}$ and $C_{44}$, where the short-hand Voigt matrix notation has been applied (see, for instance, Pollard (1977)). In isotropic systems there are only two independent components, since $C_{44} = \frac{1}{2}(C_{11} - C_{12})$, while in lower symmetries there are more than three independent components (Nye 1960).

The Helmholtz elastic free energy may be written to lowest order in the strain:

$$F = \tfrac{1}{2}C_{ijkl}\varepsilon_{ij}\varepsilon_{kl} = \tfrac{1}{2}C_{mn}\varepsilon_m\varepsilon_n = \tfrac{1}{2}C_r\varepsilon_r^2 \tag{7}$$

where summation over repeated indices is understood, and the $C$s are isothermal elastic constants.

The last equality in equation (7) may be obtained after diagonalising the elastic modulus matrix $[C_{mn}]$, or more easily by applying the group theory for the construction

of invariants. An important and useful group theoretical result in this context is the generalised Unsöld theorem, which states that (Tinkham (1964) p 81):

$$\sum_{k=1}^{l_r} |\varphi_k^{(r)}|^2$$

is invariant under all operations of the point group for which $\varphi_k^{(r)}$ are the basis functions of an irreducible representation $r$ of dimension $l_r$.

The elastic strain which is a second-rank tensor, transforms as the functions quadratic in the cartesian coordinates. These functions are usually listed in the character tables of the point groups, and a free-energy expansion on the diagonalised form $F = \frac{1}{2}C_r\varepsilon_r^2$ may thus easily be constructed. The strain components $\varepsilon_r$ (=some linear combination of various $\varepsilon_{ij}$), and elastic modulus components $C_r$ (=some linear combination of the various $C_{ijkl}$) obtained in this way are referred to as *symmetrised components*, since the various terms, $r$, correspond to the fundamental ways of distorting a given symmetry by external homogeneous strain. The $\varepsilon_r$ transforming under the identity representation are non-symmetry-breaking. If all the $C_r$ components are positive the crystal is stable against deformations. If, however, one of them, $C_{r_1}$ say, decreases to zero, which may happen close to *elastic* phase transitions, the crystal may distort into a new structure determined by the strain $\varepsilon_{r_1}$ (Cowley 1976). This property of the symmetrised energy expansion, then, determines the elastic stability of the crystal lattice.

As an example, we again consider the case of cubic symmetry. Using equation (7) the free energy in cubic symmetry may be written:

$$F = \tfrac{1}{2}C_{mn}\varepsilon_m\varepsilon_n = \tfrac{1}{2}C_{11}(\varepsilon_1^2 + \varepsilon_2^2 + \varepsilon_3^2) + C_{12}(\varepsilon_1\varepsilon_2 + \varepsilon_1\varepsilon_3 + \varepsilon_2\varepsilon_3) + \tfrac{1}{2}C_{44}(\varepsilon_4^2 + \varepsilon_5^2 + \varepsilon_6^2). \tag{8}$$

Applying the Unsöld theorem to the second-order functions listed in the character table of the cubic point group, $O_h$, (see, for example, Wooster (1973) p 326), one obtains:

$$F = \tfrac{1}{2}C_a\varepsilon_a^2 + \tfrac{1}{2}C_e(\varepsilon_{e_1}^2 + \varepsilon_{e_2}^2) + \tfrac{1}{2}C_t(\varepsilon_{t_1}^2 + \varepsilon_{t_2}^2 + \varepsilon_{t_3}^2) \tag{9}$$

where the connection between the quantities in equations (8) and (9) is given in table 1.

Generally in a crystal there is a possibility of three waves with different velocities for given directions of propagation. Only in certain directions of a crystal will these be one pure longitudinal and two pure transverse waves. One may define an *effective* elastic

**Table 1.** Symmetrised elastic constants and symmetrised strains in cubic symmetry. The strains are termed according to in which way the cubic structure is distorted in each case.

| Symmetrised elastic modulus | Symmetrised homogeneous strain | Term | Transforms under |
|---|---|---|---|
| $C_a = \frac{1}{3}(C_{11} + 2C_{12})$ | $\varepsilon_a = \varepsilon_1 + \varepsilon_2 + \varepsilon_3$ | Hydrostatic strain | $A_{1g}$ |
| $C_e = \frac{1}{2}(C_{11} - C_{12})$ | $\varepsilon_{e_1} = \varepsilon_1 - \varepsilon_2$ | Orthorhombic strain | $E_g$ |
| | $\varepsilon_{e_2} = 3^{-1/2}(3\varepsilon_3 - \varepsilon_a)$ | Tetragonal strain | |
| $C_t = C_{44}$ | $\varepsilon_{t_1} = \varepsilon_4$ | Trigonal strains | $T_{2g}$ |
| | $\varepsilon_{t_2} = \varepsilon_5$ | | |
| | $\varepsilon_{t_3} = \varepsilon_6$ | | |

constant for each of these waves:

$$v^2 = C_{\text{eff}}/\rho \tag{10}$$

where $C_{\text{eff}}$ is some linear combination of some, or possibly all, of the independent components $C_{ijkl}$.

Equivalently $C_{\text{eff}}$ may be expressed in terms of the symmetrised elastic constants $C_r$ as

$$C_{\text{eff}} = \sum_r{}' C_r \tag{11}$$

where $\Sigma_r'$ includes coefficients and also denotes that some of the $C_r$ may be missing. Equation (11) means that when a sound wave with a given direction of propagation and a given polarisation is applied the *effective* mode can generally be decomposed into some of the symmetrised modes.

In a cubic crystal, waves propagating along one of the major directions [100], [110] or [111] will be pure longitudinal or pure transverse. Some examples are given in table 2.

**Table 2.** Examples of effective elastic constants for some pure modes in cubic symmetry. The effective components are expressed in terms of the symmetrised components given in table 1.

| Direction of propagation $q$ | Direction cosines $n_1, n_2, n_3$ | Polarisation $u = [u_1 u_2 u_3]$ | $C_{\text{eff}} = \rho v^2 = \Sigma'_{mn} C_{mn} = \Sigma'_r C_r$ |
|---|---|---|---|
| [100] | 1, 0, 0 | Longitudinal ($u_1 \neq 0, u_2 = u_3 = 0$) | $C_{11} = C_a + \frac{4}{3}C_e$ |
| [100] | 1, 0, 0 | Transverse ($u_1 = 0, u_2$ or $u_3 \neq 0$) | $C_{44} = C_t$ |
| [110] | $1/\sqrt{2}, 1/\sqrt{2}, 0$ | Longitudinal ($u_1 = u_2 \neq 0, u_3 = 0$) | $\frac{1}{2}(C_{11} + C_{12} + 2C_{44}) = C_a + \frac{1}{3}C_e + C_t$ |
| [111] | $1/\sqrt{3}, 1/\sqrt{3}, 1/\sqrt{3}$ | Longitudinal ($u_1 = u_2 = u_3 \neq 0$) | $\frac{1}{3}(C_{11} + 2C_{12} + 4C_{44}) = C_a + \frac{4}{3}C_t$ |

## 4. Coupling between the order parameter and the mechanical variables

We want to specify the coupling between the strains, $\varepsilon_i$, and the ordering quantity $Q$, i.e. give the coupling term $H_c$ in the Hamiltonian. Depending on symmetry three main types of coupling may *dominate* (summation over repeated indices, and a space integration $\int d^d x$ is understood):

(i) Linear: $H_c \sim a_{ij}\varepsilon_i Q_j$
(ii) Quadratic: $H_c \sim b_{ijl}\varepsilon_i Q_j Q_l$
(iii) Quadratic: $H_c \sim d_{ijl}\varepsilon_i \varepsilon_j Q_l$.

Of these (i) and (iii) can only exist for certain symmetries, while (ii), which is the strictive type, will exist in all matter. It may therefore be said to be the most important one.

Case (iii) will not be discussed here, since it is rarely observed (see, for instance, Höchli and Scott (1971)).

A coefficient in the expansion of $H_c$ can exist only if the corresponding symmetric powers of $Q_j$ and $\varepsilon_i$ belong to the *same* irreducible representation. This is an extended version of the Unsöld theorem for construction of invariants, cited in § 3, i.e. one may replace the squares of the base functions, with products of *different* base functions belonging to the *same* irreducible representation (Rehwald 1973, Tinkham 1964).

As a simple example we derive the coupling Hamiltonian valid for cubic perovskites (see, for instance, Aharony (1978)).

In the perovskites the order parameter is an axial vector transforming according to the irreducible representation $T_{1g}$ of the cubic point group $O_h$. None of the symmetrised strain components (table 1) transform according to $T_{1g}$, i.e. no linear coupling term exists in this case. The squares of the order parameter components transform as an ordinary vector squared, i.e. in the same way as the symmetrised strains. Applying the extended Unsöld theorem one obtains

$$H_c = \int \mathrm{d}^d x \left\{ b_a \varepsilon_a (Q_1^2 + Q_2^2 + Q_3^2) + b_e [\varepsilon_{e_1} (Q_1^2 - Q_2^2) + 3^{-1/2} \varepsilon_{e_2} (3Q_3^2 - |\boldsymbol{Q}|^2)] \right.$$
$$\left. + b_t (\varepsilon_{t_1} Q_2 Q_3 + \varepsilon_{t_2} Q_1 Q_3 + \varepsilon_{t_3} Q_1 Q_2) \right\} \tag{12}$$

which may be transformed into a similar form as that of Aharony (1978) with $b_a = B_0$, $b_e = \frac{3}{2}B_1$, $b_t = B_2$. A more detailed treatment of $H_c$ in cubic symmetry is given by Henkel *et al* (1980). A listing of the symmetry properties of the order parameter in a large number of *structural* systems is given by Rehwald (1973) and Lüthi and Rehwald (1981).

## 5. Static electric constants near critical points

The static thermodynamic properties may in principle be determined from the singular part of the free energy:

$$F = -(kT/V) \ln Z \tag{13}$$

where $V$ is the volume and the partition function is

$$Z = \sum_{\{Q\}} \exp(-\beta H). \tag{14}$$

Here $\beta = 1/kT$, and $H$ is the order parameter dependent part of the Hamiltonian. For example, the following Landau–Ginzburg–Wilson (LGW) Hamiltonian $H$ is believed to be relevant to the critical behaviour in cubic perovskites (Aharony 1978):

$$H = \int \mathrm{d}^d x \left[ \tfrac{1}{2} r |\boldsymbol{Q}|^2 + \tfrac{1}{2} |\nabla \boldsymbol{Q}|^2 + u_0 |\boldsymbol{Q}|^4 + v_0 \sum_{\alpha=1}^{n} Q_\alpha^4 - \tfrac{1}{2} f \sum_{\alpha=1}^{n} \left( \frac{\partial Q_\alpha}{\partial X_\alpha} \right)^2 \right] + H_c. \tag{15}$$

Here the first three terms are the isotropic ones, while the terms with coefficients $v_0$ and $f$ represent cubic anisotropy and anisotropic dispersion respectively. $H_c$ takes coupling to strain degrees of freedom into account, and is given in equation (12). In equation (15) only $r = r_0(T - T_0)$ is assumed to depend strongly on temperature. The sum in equation (14) runs over all possible order parameter configurations. The singular part of the isothermal elastic constant is defined through equation (7):

$$\Delta C_{mn} = \partial^2 F / \partial \varepsilon_m \partial \varepsilon_n \tag{16}$$

where the Voigt notation is used. It is straightforward to combine the three equations (13), (14) and (16) into

$$\Delta C_{mn} = -\frac{\beta}{V}\left(\left\langle \frac{\partial H}{\partial \varepsilon_m}\frac{\partial H}{\partial \varepsilon_n}\right\rangle - \left\langle \frac{\partial H}{\partial \varepsilon_m}\right\rangle\left\langle \frac{\partial H}{\partial \varepsilon_n}\right\rangle\right) \tag{17}$$

where

$$\langle Y\rangle = \frac{1}{Z}\sum_{\{Q\}} Y \exp(-\beta H).$$

A term of the order of $\langle \partial^2 H/\partial\varepsilon_m \partial\varepsilon_n\rangle$ is omitted, since we do not consider the $\varepsilon^2 Q$-coupling here.

If the symmetric form of the strain-order parameter coupling Hamiltonian $H_c$ is known, the different components of the static elastic tensor may now be given in terms of order parameter correlation functions:

(i) Linear $H_c \sim a_{mi}\varepsilon_m Q_i$

$$\Delta C_{mn} \sim a_{mi}a_{nj}(\langle Q_i Q_j\rangle - \langle Q_i\rangle\langle Q_j\rangle)$$

(ii) Quadratic $H_c \sim b_{mij}\varepsilon_m Q_i Q_j$

$$\Delta C_{mn} \sim b_{mij}b_{nkl}(\langle Q_i Q_j Q_k Q_l\rangle - \langle Q_i Q_j\rangle\langle Q_k Q_l\rangle).$$

From this one expects $\Delta C_{mn}^{\text{static}} \sim t^{-\gamma}$ in the linear coupling case, where $t = (T - T_c)/T_c$ is the reduced temperature.

As a more explicit example we consider the cubic perovskites. $H_c$ is given in equation (12) and the symmetrised elastic constants in table 2. Equation (17) yields the following four-point correlation functions in agreement with Murata (1976):

$$\Delta C_a = -\frac{b_a^2}{kTV}\int \mathrm{d}^d\boldsymbol{r}\int \mathrm{d}^d\boldsymbol{r}'(\langle|\boldsymbol{Q}(\boldsymbol{r})|^2\,|\boldsymbol{Q}(\boldsymbol{r}')|^2\rangle - \langle|\boldsymbol{Q}(\boldsymbol{r})|^2\rangle\langle|\boldsymbol{Q}(\boldsymbol{r}')|^2\rangle) \tag{18}$$

$$\Delta C_e = -\frac{b_e^2}{kTV}\int \mathrm{d}^d\boldsymbol{r}\int \mathrm{d}^d\boldsymbol{r}'\langle(Q_1^2(\boldsymbol{r}) - Q_2^2(\boldsymbol{r}))(Q_1^2(\boldsymbol{r}') - Q_2^2(\boldsymbol{r}'))\rangle \tag{19}$$

$$\Delta C_t = -\frac{b_t^2}{kTV}\int \mathrm{d}^d\boldsymbol{r}\int \mathrm{d}^d\boldsymbol{r}'\langle(Q_1(\boldsymbol{r})Q_2(\boldsymbol{r}))(Q_1(\boldsymbol{r}')Q_2(\boldsymbol{r}'))\rangle \tag{20}$$

where $\langle Q_1^2(\boldsymbol{r}) - Q_2^2(\boldsymbol{r})\rangle = 0$ is used.

Note that in the isotropic approximation one must expect the same critical behaviour for $C_e$ and $C_t$ since $\langle(Q_1^2 - Q_2^2)(Q_1^2 - Q_2^2)\rangle = \langle(Q_1Q_2)(Q_1Q_2)\rangle$ as is seen by performing a rotation in the order parameter space (Murata 1976). Note also that the heat capacity is simply related to $\Delta C_a$

$$C \sim \frac{\partial^2 F}{\partial t^2} \sim \left\langle \frac{\partial H}{\partial t}\frac{\partial H}{\partial t}\right\rangle - \left\langle \frac{\partial H}{\partial t}\right\rangle\left\langle \frac{\partial H}{\partial t}\right\rangle \sim \Delta C_a$$

since the important temperature dependence of $H$ is given by $H \sim t|\boldsymbol{Q}|^2$. Therefore one expects $\Delta C_a \sim t^{-\alpha}$, which is generally valid for non-symmetry-breaking strains such as $\varepsilon_a$. The connections between elastic constants and the heat capacity are known as Pippard relations (see Hamano and Hirotsu 1979). These qualitative statements are in agreement with the results of Murata (1976), who calculated the exponents for $C_a$, $C_e$ and $C_t$. We

will not follow his development here. Instead we will derive exponents for the static elastic constants, starting from the scaling hypothesis (Fisher 1974):

$$F = t^{2-\alpha} Y_F(h/t^{\Delta}, \varepsilon_m/t^{\varphi_m}). \tag{21}$$

Here $h$ is the field conjugate to the order parameter, $\Delta = \beta + \gamma$, and the mechanical strain component $\varepsilon_m$ is included as a relevant parameter with corresponding crossover exponent $\varphi_m$. Note again that since we want to calculate elastic constants $\sim \partial^2 F/\partial \varepsilon_m^2$, we have chosen the strain $\varepsilon_m$ as the relevant field in equation (21). Usually one chooses the mechanical stress, which is the externally variable field in this case. Note also that the components of the elastic modulus we calculate in this way will be symmetrised ones, defined through equation (7), since they correspond to the fundamental ways of distorting the structure. Thus there will generally be one crossover exponent $\varphi_m$, associated with each symmetrised strain $\varepsilon_m$. In the following the subscript m on $\varphi$ will be dropped for simplicity.

Non-symmetry-breaking strains, for example, the hydrostatic strain $\varepsilon_a$ in the cubic case, or a strain along the $c$ axis in a tetragonal crystal, will not be relevant to the critical behaviour. This means that they will generally not transform the system into a new universality class. However, $T_c$ may be changed by such strains, since the $r|\boldsymbol{Q}|^2$ term in the Hamiltonian will be renormalised, as may be seen from equation (15) for non-zero $\varepsilon_a$. Denoting the non-symmetry-breaking strain $\varepsilon_a$ one can write:

$$F = (t + a'\varepsilon_a)^{2-\alpha} = t^{2-\alpha} f(\varepsilon_a/t). \tag{22}$$

Here $a'$ is some constant and $f(x) = (1 + a'x)^{2-\alpha}$. Thus

$$\Delta C \sim \partial^2 F/\partial \varepsilon_a^2 \sim t^{-\alpha} \tag{23}$$

for small perturbations $\varepsilon_a$.

*Symmetry-breaking* strains will generally change the universality class. Mechanical strains that couple linearly to the order parameter are equivalent to the field $h$ in the free-energy expression, equation (21). Hence, in the case of linear coupling ($H_c \sim \varepsilon Q$) one may write

$$F = t^{2-\alpha} Y_F(\varepsilon_m/t^{\Delta}) \tag{24}$$

and

$$\Delta C \sim \partial^2 F/\partial \varepsilon_m^2 \sim t^{2-\alpha-2\Delta} \sim t^{-\gamma} \tag{25}$$

where the scaling law $\alpha + 2\beta + \gamma = 2$ was used.

When the mechanical variables couple quadratically to the order parameter, $H_c \sim \varepsilon Q^2$, one has to consider the cases $T < T_c$ and $T > T_c$ separately. For $T < T_c$ the mean value of the order parameter $Q_0 = \langle Q \rangle \neq 0$, which means that $H_c \sim 2\varepsilon Q_0 \delta Q + \varepsilon \delta Q \delta Q$, where we drop the tensor indices for simplicity. There is now one 'field' $\sim \varepsilon Q_0$ that in effect couples linearly to the order parameter fluctuations, in addition to $\varepsilon$ which alone couples quadratically. Thus for $T < T_c$ one may write

$$F = t^{2-\alpha} Y_F(\varepsilon_m Q_0/t^{\Delta}, \varepsilon_m/t^{\varphi}) \tag{26}$$

which gives three contributions to the elastic constant

$$\Delta C \sim \partial^2 F/\partial \varepsilon_m^2 \sim t^{2\beta-\gamma} + t^{-\mu} + t^{2\beta-\varphi} \tag{27}$$

where the relation $Q_0 = \langle Q \rangle \sim t^{\beta}$ and the scaling law $\alpha + 2\beta + \gamma = 2$ have been used.

The exponent $\mu$ is given by $\mu = \alpha + 2(\varphi - 1)$. Amplitudes have been omitted in this discussion. Above $T_c$ linear coupling $\varepsilon_m Q_0$ is absent. Thus in this case

$$\Delta C \sim t^{-\mu} \tag{28}$$

$\varphi$ is in the cubic perovskite case believed to be equal to the crossover exponent away from an isotropic Heisenberg, or possibly cubic, behaviour to $XY$ or Ising behaviour.

As discussed above, the elastic constants may be expressed in terms of correlation functions. In the quadratic coupling case, these contained four order parameter components: $\Delta C \sim \langle QQQQ \rangle$, where subscripts have been dropped for simplicity. Writing $Q = Q_0 + \delta Q$ one may identify the three terms appearing in equation (37) as

$$Q_0 Q_0 \langle \delta Q \delta Q \rangle \sim t^{2\beta - \gamma}$$

$$\langle \delta Q \delta Q \delta Q \delta Q \rangle \sim t^{-\mu}$$

$$Q_0 \langle \delta Q \delta Q \delta Q \rangle \sim t^{2\beta - \varphi}.$$

Of these the term resulting from the three-point correlation function is usually neglected in the literature. The existence and importance of this term was stressed by Ferrell and Bhattacharjee (1981). Note that in mean-field theory, where fluctuations are neglected, all the exponents of the quadratic coupling case will be equal to zero. One will only observe a *step* at $T_c$ (Rehwald 1973). The divergence discussed above is due to fluctuations. Note also that although we have derived exponents for elastic constants, the same critical exponents will govern the symmetrised elastic compliances since we have considered the diagonalised version of the elastic free energy.

Apparently the present analysis gives a divergence of elastic constants close to critical points. This is, of course not physically correct since elastic constants become *smaller* near $T_c$ and in some cases, such as elastic phase transitions, go to zero. The quantities that diverge are the elastic compliances. However, as long as the anomalous part, $\Delta C = C - C_0$, is much smaller than the background part, $C_0$, it is correct to express $\Delta C$ and $\Delta S$ in terms of the same exponent. Here the total compliance $S = S_0 + \Delta S$ is the sum of a background part $S_0$ and an anomalous part $\Delta S$. Inserting into $C = 1/S$ one obtains $\Delta C \simeq -C_0^2 \Delta S$ when $\Delta C \ll C_0$. Since the elastic matrix is diagonal in the symmetrised version, the relation $C = 1/S$ is valid when $C$ and $S$ refer to corresponding symmetrised elastic constants and compliances.

Formally, it would therefore be slightly better to calculate the critical behaviour of the elastic compliances. The present calculation is a more direct approach, in which thermodynamics in a strict sense is sacrificed. In an ultrasound experiment, the *given* quantity, the external field, is the *stress*, and not the strain which was chosen for convenience in the present case. The correct potential to consider is then not the Helmholtz free energy $F$ as indicated for instance in equation (13), but rather the Gibbs free energy $G$. It is meaningless to take the derivatives of $G$ with respect to the strains. One must instead write the coupling Hamiltonian $H_c$ in equation (12) in terms of stress components, and the 'new' corresponding coupling constants. The second derivative of $G$ with respect to the symmetrised stresses yields symmetrised compliances which may then be inverted as described above to obtain the symmetrised elastic constants.

These formal considerations, which also apply to the treatment given in the following section where dynamics is discussed, do not invalidate the results as far as exponents of elastic constants are concerned. The results are summarised in table 3.

**Table 3.** Leading singular terms for *static symmetrised* elastic constants or compliances. Scaling law: $\mu = \alpha + 2(\varphi - 1)$. In static mean-field theory where fluctuations are neglected, only the linear case $H_c \sim \varepsilon Q$ will have an exponent different from zero (see also Rehwald (1973) and Aharony and Bruce (1974)).

| Coupling | $T < T_c$ | $T > T_c$ |
|---|---|---|
| Non-symmetry breaking | $a_1^- t^{-\alpha}$ | $a_1^+ t^{-\alpha}$ |
| Linear $\varepsilon Q$ | $a_2^- t^{-\gamma}$ | $a_2^+ t^{-\gamma}$ |
| Quadratic $\varepsilon Q^2$ | $a_3^- t^{-\mu} + a_4^- t^{2\beta-\gamma} + a_5^- t^{2\beta-\alpha}$ | $a_3^+ t^{-\mu}$ |

## 6. Dynamic elastic response near critical points

In this section we turn to the dynamics of the elastic constants near critical points. Parts of this discussion follow closely that given by Fossheim and Fossum (1984).

The response function, or susceptibility, which may be directly studied by ultrasonic methods is the complex elastic stiffness tensor defined through the (classical) fluctuation-dissipation theorem (Kubo 1966), as the following integral over the correlation of fluctuating internal stress $\delta\sigma_j(0, 0)$ and its time derivative $\delta\sigma_i(\boldsymbol{r}, t)$:

$$C_{ij}(\boldsymbol{q}, \omega) = \frac{1}{kT}\int \mathrm{d}^d\boldsymbol{r}\int_0^\infty \mathrm{d}t \exp(-\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{r})\exp(+\mathrm{i}\omega t)\langle\delta\dot{\sigma}_i(\boldsymbol{r}, t)\delta\sigma_j(0, 0)\rangle. \tag{29}$$

Here the spatial integration is over the volume of the system, $\boldsymbol{q}$ and $\omega$ are, respectively, the wave-vector and the frequency of the acoustic wave, $k$ is Boltzmann's constant, and $T$ is temperature. The fluctuation-dissipation theorem, equation (29), is derived assuming an external harmonic perturbation of the equilibrium system. In the present case this perturbation is the applied sound wave. The theorem states that the *linear* response of a given system is expressed in terms of the fluctuation properties in the system, in *thermal equilibrium*. The best way to proceed is by Fourier transformation of the spatial dependence of $\delta\sigma_i$. In addition, we choose to extract the statics by performing a partial integration over the time variable. After these changes equation (29) becomes:

$$C_{ij}(\boldsymbol{q}, \omega) = -\frac{V}{kT}\langle\delta\sigma_i(\boldsymbol{q}, 0)\delta\sigma_j(-\boldsymbol{q}, 0)\rangle$$
$$-\mathrm{i}\omega\frac{V}{kT}\int_0^\infty \mathrm{d}t \exp(\mathrm{i}\omega t)\langle\delta\sigma_i(\boldsymbol{q}, t)\delta\sigma_j(-\boldsymbol{q}, 0)\rangle. \tag{30}$$

Here the first term is purely static, and hence, as we shall see, contains all the terms discussed in the previous section on statics.

To proceed one must make some assumptions about the decay behaviour of the order parameter fluctuations $\delta Q_1$. The simplest case will, of course, be to assume purely relaxational behaviour $\delta Q_1(\boldsymbol{q}, t) = \delta Q_1(\boldsymbol{q}, 0)\exp(-t/\tau_q)$, corresponding to an overdamped phonon description in structural systems when $\tau_q$ is real (giving essentially mean-field-like results). To generalise the treatment, however, one may allow $\tau_q$ to be a time-dependent quantity. Further generalisation requires the use of scaling forms for the dynamical susceptibility.

Such a modification can be done approximately at the end of the calculation, since the results will be expressed in terms of the total relaxation time $\tau_q$. Even more sophisticated models, including the central mode in structural systems, may be approximated in the same way. We return briefly to this question below.

In the simplest case the relaxation time is expressed as $\tau_q = \xi^z f(q\xi)$ (Ferrell *et al* 1967, Halperin and Hohenberg 1967, 1977) where $\xi$ is the correlation length and $z$ is the dynamical exponent which is near two for a non-conserved order parameter and near four for a conserved one. We discuss only the quadratic coupling case ($\varepsilon Q^2$ coupling), since the results are easily generalised to other cases.

The fluctuation of internal stress is

$$\delta\sigma_i(\boldsymbol{r}, t) = b_{ijl}(Q_j(\boldsymbol{r}, t)Q_l(\boldsymbol{r}, t) - \langle Q_j(\boldsymbol{r}, t)Q_l(\boldsymbol{r}, t)\rangle)$$

or

$$\delta\sigma_i(\boldsymbol{q}, t) = \frac{Vb_{ijl}}{(2\pi)^d}\int \mathrm{d}^d\boldsymbol{k}(Q_j(\boldsymbol{k}, t)Q_l(-\boldsymbol{k}+\boldsymbol{q}, t) - \langle Q_j(\boldsymbol{k}, t)Q_l(-\boldsymbol{k}+\boldsymbol{q}, t)\rangle).$$

Writing $Q_j = Q_{0j} + \delta Q_j$, where $Q_{0j}$ is the average of $Q_j$, we obtain two terms in $\delta\sigma_i$:

$$\delta\sigma_i(\boldsymbol{q}, t) = 2b_{ijl}Q_{0j}\delta Q_l(\boldsymbol{q}, t) + \frac{V}{(2\pi)^d}b_{ijl}\int \mathrm{d}^d\boldsymbol{k}[\delta Q_j(\boldsymbol{k}, t)\delta Q_l(-\boldsymbol{k}+\boldsymbol{q}, t) - \langle\delta Q_j(\boldsymbol{k}, t)\delta Q_l(-\boldsymbol{k}+\boldsymbol{q}, t)\rangle]. \tag{31}$$

Inserting this in equation (30) we obtain three types of non-vanishing correlation functions, a two-point correlation which gives the so called Landau–Khalatnikov term (Landau and Khalatnikov 1951, hereafter LK), a four-point correlation which we may call *the critical scattering* term, and a three-point correlation which represents a mixing. The latter will not be discussed further, but the phenomenological extension of mean-field results to be given below, is valid for this term as well. See also table 3.

### 6.1. *The LK term*

The LK term is given by:

$$\Delta C_{ij}^{\mathrm{LK}}(\boldsymbol{q}, \omega) = -\frac{4V}{kT}b_{imn}b_{jpr}Q_{0m}Q_{0p}\left(\langle\delta Q_n(\boldsymbol{q}, 0)\delta Q_r(-\boldsymbol{q}, 0)\rangle + \mathrm{i}\omega\int_0^\infty \mathrm{d}t\exp(\mathrm{i}\omega t)\langle\delta Q_n(\boldsymbol{q}, t)\delta Q_r(-\boldsymbol{q}, 0)\rangle\right)$$
$$= 4b_{imn}b_{jpr}Q_{0m}Q_{0p}\chi_{nr}(\boldsymbol{q}, \omega) \tag{32}$$

Using the simple relaxation form for $\delta Q_n$ mentioned before would now lead to the well known mean-field results for the LK contributions:

$$\chi_{nr}(\boldsymbol{q}, \omega) = \chi_{nr}(\boldsymbol{q})/(1 - \mathrm{i}\omega\tau).$$

To obtain more general forms we have to use scaling arguments. We write the order parameter susceptibility as

$$\chi_{nr}(\boldsymbol{q}, \omega) \equiv \chi = \chi(\boldsymbol{q}, 0)f(\mathrm{i}\omega\tau)$$

where $f(i\omega\tau)$ is some scaling function. By the expansion of $f(i\omega\tau)$ we see that

$$\left.\begin{aligned}&\operatorname{Re}\chi \sim t^{-\gamma}\\&\operatorname{Im}\chi \sim t^{-\gamma}\,\omega\tau\end{aligned}\right\} \qquad \text{for } \omega\tau \ll 1.$$

Analogous to the static case where the susceptibility only diverges for $q = 0$, we must require that $\chi$ does not diverge at $T_c$ when $\omega \neq 0$, i.e. $\chi \sim t^0$ for $\omega\tau \gg 1$ (Landau and Lifshitz 1981). This leads to

$$\left.\begin{aligned}&\operatorname{Re}\chi \sim t^{-\gamma}(\omega\tau)^{-\gamma/\nu z}\\&\operatorname{Im}\chi \sim t^{-\gamma}\omega\tau(\omega\tau)^{-1+\gamma/\nu z}\end{aligned}\right\} \qquad \text{for } \omega\tau \gg 1.$$

Both limits may be combined in a scaling form, for instance as follows

$$\Delta C^{\mathrm{LK}}(0,\omega) = -A^{\mathrm{LK}}t^{2\beta-\gamma}(1-i\omega\tau)^{-\gamma/\nu z} \tag{33}$$

where this particular form is chosen to fulfil causality requirements. This function has no poles in the upper half-plane. The only instability occurs for $\omega \to 0$, $\xi \to \infty$. For a detailed discussion of linear response near phase transitions, we refer to Thomas (1976). In mean-field theory as described, for instance, by the gaussian approximation $\gamma = \nu z = 1$, i.e. equation (33) reproduces the classical Landau–Khalatnikov result.

### 6.2. *The critical part*

The critical scattering term contributes both above and below $T_c$. Combining equations (30) and (31) we have:

$$\begin{aligned}\Delta C_{ij}^{\mathrm{Crit}}(\boldsymbol{q},\omega) = {}& \frac{V^3}{(2\pi)^{2d}kT}\,b_{imn}b_{jpr}\int \mathrm{d}^d\boldsymbol{k}\,\mathrm{d}^d\boldsymbol{k}'\\ &\times\Big(\langle\delta Q_m(\boldsymbol{k},0)\delta Q_n(-\boldsymbol{k}+\boldsymbol{q},0)\delta Q_p(-\boldsymbol{k}',0)\delta Q_r(\boldsymbol{k}'-\boldsymbol{q},0)\rangle\\ &-\langle\delta Q_m(\boldsymbol{k},0)\delta Q_n(-\boldsymbol{k}+\boldsymbol{q},0)\rangle\langle\delta Q_p(-\boldsymbol{k}',0)\delta Q_r(\boldsymbol{k}'-\boldsymbol{q},0)\rangle\\ &+i\omega\int_0^\infty \mathrm{d}t\exp(+i\omega t)[\langle\delta Q_m(\boldsymbol{k},t)\delta Q_n(-\boldsymbol{k}+\boldsymbol{q},t)\delta Q_p(-\boldsymbol{k}',0)\\ &\times\delta Q_r(\boldsymbol{k}'-\boldsymbol{q},0)\rangle\\ &-\langle\delta Q_m(\boldsymbol{k},t)\delta Q_n(-\boldsymbol{k}+\boldsymbol{q},t)\rangle\langle\delta Q_p(-\boldsymbol{k}',t)\delta Q_r(\boldsymbol{k}'-\boldsymbol{q},t)\rangle]\Big).\end{aligned}$$

Performing the integration using $\delta Q(\boldsymbol{k},t) = \delta Q(\boldsymbol{k},0)\exp(-t/\tau_k)$, one obtains for $\boldsymbol{q} = 0$:

$$\begin{aligned}\Delta C_{ij}^{\mathrm{Crit}}(0,\omega) = {}& \frac{V^3}{kT(2\pi)^{2d}}\,b_{imn}b_{jpr}\int \mathrm{d}^d\boldsymbol{k}\,\mathrm{d}^d\boldsymbol{k}'\,\frac{1}{1-i\frac{1}{2}\omega\tau_k}\\ &\times[\langle\delta Q_m(\boldsymbol{k},0)\delta Q_n(-\boldsymbol{k},0)\delta Q_p(-\boldsymbol{k}',0)\delta Q_r(\boldsymbol{k}',0)\rangle\\ &-\langle\delta Q_m(\boldsymbol{k},0)\delta Q_n(-\boldsymbol{k},0)\rangle\langle\delta Q_p(-\boldsymbol{k}',0)\delta Q_r(\boldsymbol{k}',0)\rangle].\end{aligned} \tag{34}$$

We shall see below what approximate *general* forms this may take. Here we proceed to develop the 'mean-field' expressions. For this purpose we use a gaussian factorisation approximation for the four-point correlation function equivalent to neglecting inter-

actions between fluctuations. In this approximation the correlation function is a sum of three terms, one of which cancels, upon substitution in equation (34), with the product of two-point correlation functions, while the other two, because of symmetry, give exactly the same contributions. Equation (34) then becomes

$$\Delta C_{ij}^{\mathrm{Crit}}(0,\omega) = \frac{2V^3}{(2\pi)^{2d}kT} b_{imn}b_{jpr}\int \mathrm{d}^d\boldsymbol{k}\,\mathrm{d}^d\boldsymbol{k}'\,\frac{1}{1-\frac{1}{2}\mathrm{i}\omega\tau_k}\langle\delta Q_m(\boldsymbol{k},0)\delta Q_p(-\boldsymbol{k}',0)\rangle$$
$$\times\langle\delta Q_n(-\boldsymbol{k},0)\delta Q_r(\boldsymbol{k}',0)\rangle. \tag{35}$$

Using

$$\langle\delta Q_m(\boldsymbol{k},0)\delta Q_p(-k',0)\rangle = (kT/V)\chi_{mm}(\boldsymbol{k},0)\delta_{mp}\delta(\boldsymbol{k}-\boldsymbol{k}')$$

one obtains

$$\Delta C_{ij}^{\mathrm{crit}}(0,\omega) \simeq \frac{2kT}{(2\pi)^d} b_{imn}b_{jmn}\int \frac{\mathrm{d}^d\boldsymbol{k}}{1+\mathrm{i}\frac{1}{2}\omega\tau_k}\chi_{mm}(\boldsymbol{k},0)\chi_{nn}(-\boldsymbol{k},0). \tag{36}$$

Making use of the mean-field relations, in the case that $\tau_k$ is real we can write

$$\tau_k \sim \xi^2/[1+(k\xi)^2] \qquad \chi_{jj}(\boldsymbol{k},0) \sim \xi^2/[1+(k\xi)^2]$$

and equation (46) may be transformed into:

$$\Delta C_{ij}^{\mathrm{Crit}}(0,\omega) = [2kT/(2\pi)^d]\, b_{imn}b_{jmn}\xi^{4-d}\tilde{f}(\mathrm{i}\omega\xi^z) \tag{37}$$

Thus we have obtained a static ($\omega = 0$) exponent $\mu = 2 - d/2 = \alpha$ in the gaussian approximation (see, for instance, Ma 1976). The result $\mu = \alpha$ is not unexpected since the approximation made above does not distinguish between symmetry-breaking and non-symmetry-breaking modes. Performing an isotropic integration we find for $d = 3$

$$\Delta C^{\mathrm{Crit}}(0,\omega) = B^2(\xi/\mathrm{i}\omega\tau)[(1-\tfrac{1}{2}\mathrm{i}\omega\tau)^{1/2}-1] \tag{38}$$

where $B^2$ is a constant and $\tau \sim \xi^2$. A similar result was obtained by Levanyuk (1966) and Pytte (1971a, b), while Bhattacharjee (1982) calculated equation (38) from an $n^{-1}$-expansion to lowest order. For $\omega\tau \ll 1$ this gives

$$\mathrm{Re}\,\Delta C^{\mathrm{Crit}} \sim t^{-1/2} \qquad \mathrm{Im}\,\Delta C^{\mathrm{Crit}} \sim \omega t^{-3/2}.$$

For $\omega\tau \gg 1$

$$\mathrm{Re}\,\Delta C^{\mathrm{Crit}} \sim \omega^{1/2}t^0 \qquad \mathrm{Im}\,\Delta C^{\mathrm{Crit}} \sim \omega^{-1/2}t^0.$$

The total result which reproduces equation (38) in the asymptotic limits $\omega\tau \ll 1$ and $\omega\tau \gg 1$ is:

$$\Delta C^{\mathrm{Crit}} = \tfrac{1}{4}B^2t^{-1/2}\left(\frac{1}{1-\frac{1}{2}\mathrm{i}\omega\tau}\right)^{1/2}. \tag{39}$$

By a straightforward generalisation based on the same scaling arguments used for the two-point correlation function above, we adopt the following approximate dynamical scaling form:

$$\Delta C^{\mathrm{Crit}} \doteq -A^{\mathrm{Crit}}t^{-\mu}((1-\mathrm{i}\omega\tau)^{-\mu/\nu z}) \tag{40}$$

where $\mu = \alpha + 2(\varphi - 1)$ is the static exponent (see table 3). Note that the amplitude of $\tau$ in equation (40) is smaller than $\tau$ in equation (33) by a factor of about two. This is seen by comparing the mean-field results equations (32) and (39).

**Table 4.** The asymptotically correct, causal, scaling functions for dynamic *symmetrised* elastic constants. $\mu = \alpha + 2(\varphi - 1)$ and $\tau = \tau_0 t^{-\nu z}$ where $t$ is the reduced temperature. For non-symmetry-breaking strains, $\varphi = 1$. In the limit $\omega = 0$ this table reproduces table 3. In mean-field theory (gaussian approximation): $\gamma = \nu z = \varphi = 1$. $\alpha = 2 - d/2$. $\beta = \frac{1}{2}$. The relaxation time amplitude $\tau_0$ may be different above and below $T_c$, and it may be different for the different scattering terms.

| Coupling | $T < T_c$ | $T > T_c$ |
|---|---|---|
| Linear $\varepsilon Q$ | $-a_2^- t^{-\gamma}(1 - i\omega\tau)^{-\gamma/\nu z}$ | $-a_2^- t^{-\gamma}(1 - i\omega\tau)^{-\gamma/\nu z}$ |
| | $-a_3^- t^{-\mu}(1 - i\omega\tau)^{-\mu/\sigma z}$ | |
| Quadratic $\varepsilon Q^2$ | $-a_4^- t^{2\beta-\gamma}(1 - i\omega\tau)^{-\gamma/\nu z}$<br>$-a_5^- t^{2\beta-\varphi}(1 - i\omega\tau)^{-(\varphi-\beta)/z}$ | $-a_3^- t^{-\mu}(1 - i\omega\tau)^{-\mu/\nu z}$ |

The results of the above analysis, together with those of a similar treatment for $\varepsilon Q$-coupling to lowest order in the fluctuations, are given in table 4.

## 7. Summary and discussion of ultrasonic velocity and attenuation near critical points

We are now able to compute phenomenological expressions for the anomalous parts of the ultrasonic attenuation, $\Delta\alpha$, and the velocity, $\Delta v$. In § 2 we have expressed $\Delta\alpha$ and $\Delta v$ in terms of the changes in the elastic response function, $\Delta C(\omega)$, which we discussed in § 6. Here we will consider the quadratic coupling for $T > T_c$ in some detail. The treatment can easily be adapted to the other cases listed in table 4:

$$\Delta C^{\text{Crit}}(\omega) = -a_3^+ t^{-\mu}(1 - i\omega\tau)^{-\mu/\nu z} = -a_3^+ t^{-\mu} F(i\omega\tau)$$
$$= -a_3^+ t^{-\mu} H(\omega\tau) \exp[i(\mu/\nu z) \tan^{-1} \omega\tau)]. \tag{41}$$

When this is inserted into equation (9) we obtain

$$v_0 \Delta\alpha/\omega = A t^{-\mu} f_\mu(\omega\tau) \tag{42}$$

$$\Delta v/v_0 = A t^{-\mu} g_\mu(\omega\tau) \tag{43}$$

where $A$ is an unknown amplitude, $\mu = \alpha + 2(\varphi - 1)$ and

$$f_\mu(\omega\tau) = \sin[(\mu/\nu z) \tan^{-1} \omega\tau] H_\mu(\omega\tau)$$
$$g_\mu(\omega\tau) = \cos[(\mu/\nu z) \tan^{-1} \omega\tau] H_\mu(\omega\tau)$$
$$H_\mu(\omega\tau) = \{\cos(\tan^{-1} \omega\tau)\}^{-\mu/\nu z}.$$

The following points must be noted.

(i) In the asymptotic limits $\omega\tau \ll 1$ ($T \gg T_c$) and $\omega\tau \gg 1$ ($T \to T_c$), equations (42) and (43) satisfy the behaviours given in table 5 (Fossum and Fossheim 1984, Fossum *et al* 1984).

(ii) In the limit $\omega\tau \ll 1$ one obtains the well known result $\Delta\alpha = \alpha_0 \omega^2 t^{-\rho}$ where $\alpha_0 = (\mu/\nu z) A (\tau_0/v_0)$ and $\rho = \mu + \nu z$. Note also that it follows from equation (31) that

$$\Delta\alpha \sim t^{-\mu} \omega^2 \tau [1 - (F'''(0)/F'(0))(\omega\tau)^2 + \ldots]$$

**Table 5.** Critical parts of the sound velocity, $\Delta v$, and attenuation, $\Delta\alpha$, of symmetrised sound modes in the asymptotic limits $\omega\tau \ll 1$ ($T \gg T_c$) and $\omega\tau \gg 1$ ($T \to T_c$). The relaxation time satisfies $\tau = \tau_0 t^{-\nu z}$ in the critical asymptotic region. The exponents obey the scaling laws $\rho = \mu + \nu z = \alpha + 2(\varphi - 1) + \nu z$.

| | $\omega\tau \ll 1$ | $\omega\tau \gg 1$ |
|---|---|---|
| $v_0\Delta\alpha/\omega$ | $A(\mu/\nu z)\tau_0\omega t^{-\rho}$ | $A \sin(\pi\mu/2\nu z)(\omega\tau_0)^{-\mu/\nu z}$ |
| $\Delta v/v_0$ | $At^{-\mu}$ | $A \cos(\pi\mu/2\nu z)(\omega\tau_0)^{-\mu/\nu z}$ |

and

$$\Delta v \sim t - \mu[1 - (F''(0)/F(0))(\omega\tau)^2 + \ldots]$$

when $\omega\tau \ll 1$. Thus the lowest-order corrections to the $\omega\tau \ll 1$ asymptotic behaviour is proportional to $(\omega\tau)^2$ for both quantities. The amplitudes of the corrections are different, however, since

$$|F'''(0)/F'(0)| = |(\mu/\nu z - 1)(\mu/\nu z - 2)| > |F''(0)/F(0)| = |(\mu/\nu z)(\mu/\nu z - 1)|$$

giving a larger correction amplitude for the attenuation than for the velocity.

(iii) It is straightforward to derive a smooth function describing crossover from the $\omega\tau \ll 1$ regime to the $\omega\tau \gg 1$ regime. Simple *asymptotically correct* dynamic scaling forms were proposed by Golding (1975) who studied ultrasonic attenuation in the ferromagnet MnP, by Kawazaki (1977) and by Fossheim and Fossum (1984).

(iv) The present results equations (42) and (43) represent improvements compared with earlier published ones (Fossheim and Fossum 1984); they satisfy causality in a strict sense, since the *total* dynamic scaling function has no poles in the upper half-plane. This has the important implication that equations (42) and (43) fulfil the Kramers–Kronig relations automatically. It is a simple matter to perform the Kramers–Kronig integrals approximately in the asymptotic limits $\omega\tau \ll 1$ and $\omega\tau \gg 1$, to check that the amplitude relations of table 5 are the correct ones. For instance in the limit $\omega\tau \ll 1$ one obtains

$$\frac{\Delta v}{v_0}(\omega = 0) = \frac{1}{\pi} \mathrm{P} \int_{-\infty}^{\infty} \frac{v_0 \Delta\alpha(x)}{x^2} \, \mathrm{d}x \simeq \frac{2}{\pi} A \frac{\rho}{\mu} t^{-\mu} \simeq (\nu z/\mu) A t^{-\mu}$$

when $\nu z \geqslant \mu$. This reproduces the results in table 5, when $\omega\tau \ll 1$.

(v) The present results are based on phenomenological assumptions. Thus the relaxation time $\tau$ entering the expressions may represent critical cluster relaxation or any other anomalous relaxation time in the system, for instance the time associated with the soft-phonon dynamics in structural systems. In the latter case the simplest approximate modification of the results would be a simple insertion for $\tau$ with standard notation (see, for example, Shapiro *et al* (1972)):

$\tau = 2\Gamma/\omega_x^2$ purely relaxational (overdamped) mode

$\tau = \dfrac{2\Gamma}{\omega_x^2} - \mathrm{i}\dfrac{\omega}{\omega_x^2} = \tau_p$ propagating damped mode

$\tau = \dfrac{2\Gamma}{\omega_x^2} - \mathrm{i}\dfrac{\omega}{\omega_x^2} + \dfrac{\delta^2\omega_x^{-2}}{1 - \mathrm{i}\omega/\gamma}$ soft mode + central peak.

Thus the exponent $\nu z$, and also the pure static ones contained in $\mu$, may merely be *effective* exponents describing precritical or non-asymptotic exponential behaviour.

The results of the present analysis are summarised in table 6. A general sound mode can be decomposed into symmetrised modes with different exponents $\varphi$ (see equation (11)). Thus in a crystal a single power law behaviour like $t^{-\gamma}$ or $t^{-\mu}$ is not to be expected in the general case.

Note that the amplitude $A_i^{\pm}$ can be determined both from the velocity and the attenuation data. This is important since it represents a check on the internal (Kramers–Kronig) consistency of the results.

**Table 6.** Phenomenological dynamical scaling functions for the *symmetrised* components of sound attenuation, $\Delta\alpha$, and the sound velocity $\Delta v$. $\mu = \alpha + 2(\varphi - 1)$ where $\varphi = 1$ for non-symmetry-breaking sound modes. The $A_i$ are constants, while the $\tau_i^{\pm}$ are phenomenological relaxation times obeying $\tau_i^{\pm} = \tau_{0i}^{\pm} t^{-\nu z}$ in the critical asymptotic region, where the $\tau_{0i}^{\pm}$ are amplitudes. $t = (T - T_c)/T_c$ is the reduced temperature. The $f$ and $g$ are defined in equations (52) and (53). A general sound mode will be a linear combination of symmetrised modes. Outside the asymptotic region, correction terms must be included. Mean-field results (gaussian approximation) are $\varphi = \gamma = \nu z = 2\beta = 2\mu = 1$ for $d = 3$. Note that the so called mixing term with amplitude $A_5^{\pm}$ is possibly forbidden in *crystals* due to wave-vector conservation requirements.

| Coupling | Quantity calculated | $T < T_c$ | $T > T_c$ |
|---|---|---|---|
| Linear | $v_0\Delta\alpha/\omega$ | $A_2^{\pm} t^{-\gamma} f_\gamma(\omega\tau_2^{\pm})$ | $A_2^{\pm} t^{-\gamma} f_\gamma(\omega\tau_2^{\pm})$ |
| | $\Delta v/v_0$ | $A_2^{\pm} t^{-\gamma} g_\gamma(\omega\tau_2^{\pm})$ | $A_2^{\pm} t^{-\gamma} g_\gamma(\omega\tau_2^{\pm})$ |
| Quadratic | $v_0\Delta\alpha/\omega$ | $A_3^{\pm} t^{-\mu} f_\mu(\omega\tau_3^{\pm})$ $+A_4^{\pm} t^{2\beta-\gamma} f_\gamma(\omega\tau_4^{\pm})$ $+A_5^{\pm} t^{2\beta-\varphi} f_{\varphi-\beta}(\omega\tau_5^{\pm})$ | $A_3^{\pm} t^{-\mu} f_\mu(\omega\tau_3^{\pm})$ |
| | $\Delta v/v_0$ | $A_3^{\pm} t^{-\mu} g_\mu(\omega\tau_3^{\pm})$ $+A_4^{\pm} t^{2\beta-\gamma} g_\gamma(\omega\tau_4^{\pm})$ $+A_5^{\pm} t^{2\beta-\varphi} g_{\varphi-\beta}(\omega\tau_5^{\pm})$ | $A_3^{\pm} t^{-\mu} g_\mu(\omega\tau_3^{\pm})$ |

## 8. Example: explicit expressions for attenuation and velocity in cubic perovskites

In § 3 we gave the effective elastic constants measured experimentally in terms of the symmetrised components, $C_a$, $C_e$ and $C_t$. These correspond to the fundamental ways of distorting the cubic structure, and will therefore each have an associated crossover exponent $\varphi_a$, $\varphi_e$ and $\varphi_t$. In § 5 we argued that $\varphi_a = 1$ since this mode is non-symmetry breaking. We also argued that $\varphi_e \simeq \varphi_t = \varphi$. In the case of cubic perovskites $\varphi$ describes the crossover away from the isotropic Heisenberg or possibly cubic behaviour to $XY$ or Ising behaviour.

In the cubic high-temperature phase, the order parameter couples quadratically to the strain in the perovskites. Using tables 2 and 5 we obtain table 7. We neglect the contribution from the non-symmetry breaking term $C_a$, since this turns out to be negligible in experiments (Fossum and Fossheim 1984, 1985, Fossum *et al* 1984, Holt and Fossheim 1981). The results are in agreement with Murata (1976), Schwabl and Iro (1981) and Iro and Schwabl (1983).

**Table 7.** Theoretical predictions for sound attenuation and sound velocity in major directions of cubic systems. The $v_0$ are background velocities. The non-symmetry-breaking mode, $C_a$, is neglected. The exponents $\rho$ and $\mu$ are defined as $\rho = \mu + \nu z$ and $\mu = \alpha + 2(\varphi - 1)$. The scaling functions $f(\omega\tau) = g(\omega\tau) \to 1$ in the limit $\omega\tau \to 0$, while $f(\omega\tau) = (\nu z/\mu)\sin(\pi\mu/2\nu z)(\omega\tau)^{-\rho/\nu z}$ and $g(\omega\tau) = \cos(\pi\mu/2\nu z)(\omega\tau)^{-\mu/\nu z}$ when $\omega\tau \gg 1$. For comparison with table 6: $f(\omega\tau) = (\nu z/\mu)f_\mu(\omega\tau)/\omega\tau$, $g(\omega\tau) = g_\mu(\omega\tau)$.

| | |
|---|---|
| Attenuation | $\Delta\alpha_{L100} = \frac{1}{3}A_e \dfrac{\tau_0}{v_0^{L100}}\omega^2 t^{-\rho} f(\omega\tau)$ |
| | $\Delta\alpha_{T100} = A_t \dfrac{\tau_0}{v_0^{T100}}\omega^2 t^{-\rho} f(\omega\tau)$ |
| | $\Delta\alpha_{L110} = (\frac{1}{3}A_e + A_t)\dfrac{\tau_0}{v_0^{L110}}\omega^2 t^{-\rho} f(\omega\tau)$ |
| | $\Delta\alpha_{L111} = \frac{1}{3}A_t \dfrac{\tau_0}{v_0^{L111}}\omega^2 t^{-\rho} f(\omega\tau)$ |
| Velocity | $\Delta v_{L100} = \dfrac{\nu z}{\mu}\frac{1}{3}A_e v_0^{L100} t^{-\mu} g(\omega\tau)$ |
| | $\Delta v_{T100} = \dfrac{\nu z}{\mu} A_t v_0^{T100} t^{-\mu} g(\omega\tau)$ |
| | $\Delta v_{L110} = \dfrac{\nu z}{\mu}(\frac{1}{3}A_e + A_t) v_0^{L110} t^{-\mu} g(\omega\tau)$ |
| | $\Delta v_{L100} = \dfrac{\nu z}{\mu}\frac{1}{3}A_t v_0^{L111} t^{-\mu} g(\omega\tau)$ |

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