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Exponents for sound attenuation near critical points in solids*

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The expansion in $4-d=\epsilon$ is used to calculate exponents governing sound attenuation above a critical point. For $SrTiO_3$, exponents differing from previous predictions are predicted to govern the sound attenuation for *all* directions and polarizations. The predictions are also compared with recent measurements on $BaMnF_4$.

A number of authors[1,2] have discussed acoustic sound attenuation above a critical point in solids resulting from coupling to critical fluctuations. The fluctuations occur in an n-component field Φ_i, $i=1,2,\ldots,n$, describing the "soft" phonon normal modes at the phase transition. The most powerful method invoked thus far for deducing attenuation exponents involved the use of scaling arguments[2]; however, several incorrect assumptions were made.

The more general ϵ-expansion analysis indicates two or possibly three different exponents: ρ_1, governing the attenuation of sound which couples strictly to the scalar dot product $\vec{\Phi}\cdot\vec{\Phi}$ and ρ_2 or ρ_2', arising from coupling to any tensor $\Phi_i\Phi_j$. The exponent ρ_1, through Ward identities, can be expressed in terms of the usual static exponents (ν,η) and has approximately the value given before within various approximations for longitudinal sound. The exponents $\rho_2, \rho_2' \geq \rho_1$ it turns out *cannot be* expressed solely in terms of the static exponents ν and η but are also related to the so-called anisotropy crossover scaling exponents[3] φ. The interesting prospect raised here is the observability of the crossover exponents in sound-attenuation measurements. Estimates of the coupling constants in $SrTiO_3$ show that ρ_2 or ρ_2' should dominate for all directions and polarizations even far from T_c. This corrects the results of Ref. 2, where the analysis was oversimplified. Attenuation governed by only ρ_1 is, however, possible within a recently proposed Hamiltonian[4] for $BaMnF_4$ and the predictions are compared below with the measured exponents. From general arguments, the new exponents are also shown to appear in the generalized elastic constants near T_c.

For concreteness we shall discuss first the case appropriate to the cubic perovskites with $n=3$. Results below not valid for general n will be indicated. The Wilson functional, including coupling to strains e_{ij}, has the form[5]

$$\mathfrak{F}(\{Q\},\{\Phi\}) = \frac{1}{2}\sum_q\left(\sum_i(\omega_0^2+\lambda_0 q^2+\lambda_1 q_i^2)\Phi_i(q)\Phi_i(-q)+\sum_\mu c_0(\hat{q},\mu)q^2Q(\mu,q)Q(\mu,-q)\right)$$
$$+\sum_l\left[\frac{u_0}{4!}(\vec{\Phi}\cdot\vec{\Phi})^2+\frac{u_1}{4!}(\Phi_1^4+\Phi_2^4+\Phi_3^4)+Ae_{11}\Phi_1\Phi_1+Be_{11}(\Phi_2\Phi_2+\Phi_3\Phi_3)+Ce_{12}\Phi_1\Phi_2+\text{permutations}\right], \quad (1)$$

where we have assumed local coupling only and l denotes a sum over lattice sites. The $e_{ij}(l)$ are to be considered expressed in the acoustic normal coordinates $Q(\mu,q)$ in the usual way,[5] $c_0(\hat{q},\mu)$ is the generalized elastic constant in the absence of soft-mode coupling, μ denotes the polarization, and $\omega_0^2\propto T-T_c'$.

The dominant fixed point of (1), for $u_1\neq 0$ or $\lambda_1\neq 0$, has not been resolved[6,7] for $n=3$, even if strains are neglected. The competing fixed points are the isotropic Heisenberg fixed point (HFP) with u_1^*, $\lambda_1^*=0$, and the cubic fixed point (CFP), with u_1^* finite and $\lambda_1^*=0$.[6] The difference between the Heisenberg and cubic exponents is found almost negligible for $n=3$. However, the strain coupling in (1) creates a complicated directionally dependent Φ^4 interaction, which has not been fully studied. We shall assume for simplicity in the following that the Heisenberg or cubic fixed point is appropriate in spite of strains.[8]

The attenuation exponents are deduced from the time correlation functions

$$\Gamma_{ij;kl}(1-2)=\langle\Phi_i(1)\Phi_j(1)\Phi_k(2)\Phi_l(2)\rangle,$$

where the brackets represent the canonical ensemble for the Hamiltonian corresponding to (1). In the limit of small-sound wave number q and small-sound frequency ω ($q\xi\ll 1$ and $\omega\tau_{\text{crit}}\ll 1$ where ξ is the coherence length and τ_{crit} is the Φ relaxation time), the attenuation is proportional to some linear combination of $\Gamma_{ij;kl}(q=0,\ \omega=0)$. As shall become clear below, the $\Gamma_{ij;kl}\,(0,0)$ do *not* in general behave according to a single power law in $t = (T-T_c)/T_c$. Within the ϵ expansion this is indicated by the fact that if one assumes a single power law, then one does not in general obtain a well-behaved ϵ expansion for the exponent. However, one does obtain a well-behaved ϵ expansion and a good single exponent for the following:

$$\mathrm{Tr}\Gamma_{ii;jj}(0,0)\approx K_1 t^{-\rho_1};$$

$$\Gamma_{12;12}(0,0)\approx K_2 t^{-\rho_2};$$

and

$$\Gamma_{11;11}-\Gamma_{11;22}\approx 2K_2' t^{-\rho_2'}.$$

One can now obtain the other $\Gamma_{ij;kl}$ from symmetry; for example,

$$\Gamma_{11;11}=K_1 t^{-\rho_1}+12K_2' t^{-\rho_2'}/9$$

for $n=3$. At the HFP, a rotation in Φ space shows that $\rho_2=\rho_2'$. Note, however, that this does not imply $K_2=K_2'$ unless $u_1\equiv 0$, $\lambda_1\equiv 0$, since such coefficients depend on the approach to the fixed point.

We next discuss the exponent calculation. It has been argued that the dynamic critical behavior of Φ is not affected by coupling to propagating strain fields.[9] We therefore expect the soft-mode dynamic susceptibility to retain its decoupled form: $\chi(q,\omega)=q^{-2+\eta}f(\omega/q^{2+\sigma\eta}, q\xi)$. This form applies if the soft-mode response is completely overdamped. Neutron scattering measurements indicate this behavior is present[10] within 6 K of T_c in $SrTiO_3$. Second-order ϵ-expansion results have established that[11] in the above $\sigma=6\ln 4/3-1$, whereas conventional theories of dynamics assume $\sigma=-1$. We shall use the former value, although the distinction becomes significant only for $d=2$.

Through general scaling arguments, or even less general mode-mode coupling arguments,[2] one obtains $\rho_1=\nu[2\gamma_4(u_\infty)-(2-\sigma)\eta+6-d]$ where $\gamma_4(u_\infty)$ is the exponent for the static vertex function

$$\Gamma^{(4)}\delta_{ij}=\frac{1}{2}\sum_{1,2}\langle\vec{\Phi}(1)\cdot\vec{\Phi}(1)\Phi_i(2)\Phi_j(3)\rangle \sim t^{-\gamma_4(u_\infty)\nu},$$

in which the sum is taken over positions only and the times are set equal. The Ward identity $\Gamma^{(4)} = \partial\chi^{-1}(0,0)/\partial(T-T_c)$ allows us to equate $\gamma_4(u_\infty)$ with $\eta-2+1/\nu$, and therefore one can express ρ_1 in terms of usual static exponents:

$$\rho_1=2+(2+\sigma\eta)\nu-d\nu. \qquad (2)$$

Similar arguments lead to

$$\rho_2=\rho_1+2[\gamma_{4t}(u_\infty)-\gamma_4(u_\infty)]\nu,$$
$$\rho_2'=\rho_1+2[\gamma_{4t}'(u_\infty)-\gamma_4(u_\infty)]\nu, \qquad (3)$$

where $\gamma_{4t}(u_\infty)$ is defined as the exponent for $\langle\Phi_1(1)\times\Phi_2(1)\Phi_1(2)\Phi_2(3)\rangle$, and so forth. In contrast to γ_4, a Ward identity which relates γ_{4t} or γ_{4t}' to η and ν is not available. However, a straightforward differentiation of the free-energy scaling function[3] shows that the so-called crossover scaling exponent φ for the variable $\Phi_1\Phi_2$ is given by

$$\varphi=[\gamma_{4t}(u_\infty)-\gamma_4(u_\infty)]\nu+1$$

and that for the variable $\Phi_1^2-\Phi_2^2$ by

$$\varphi'=[\gamma_{4t}'(u_\infty)-\gamma_4(u_\infty)]\nu+1.$$

At the HFP, $\varphi=\varphi'$ was derived to $O(\epsilon^2)$ by Wilson.[12] At the CFP φ' was derived to $O(\epsilon^2)$ by Aharony.[13] Our results for γ_{4t} (HFP) and γ_{4t}' (HFP, CFP) below from Callan-Symanzik methods are consistent with their expressions. However, φ at the CFP has evidently not been previously derived, and thus the expression below for γ_{4t} at the CFP is a completely new result.

The method for calculating γ_{4t} follows that of Brézin *et al.*[14] The vertex $\Gamma^{(4)}$ is Brézin's $\Gamma^{(1,2)}$ and he finds for the HFP

$$\gamma_4(u_\infty)=-\frac{(n+2)\epsilon}{n+8}\left[1+\frac{6\epsilon(n+3)}{(n+8)^2}\right]+O(\epsilon^3)\ \text{(HFP)}.$$

In addition for the CFP, we obtain

$$\gamma_4(u_\infty)=-\frac{2(n-1)\epsilon}{3n}\times\left[1+\frac{\epsilon}{54n^2}(-212+160n-11n^2)\right]+O(\epsilon^3)\qquad \text{(CFP)}.$$

The result for γ_{4t} can be calculated in a similar way and we obtain

TABLE I. Sound attenuation exponents ρ_1, ρ_2, and ρ_2' from the ϵ expansion to $O(\epsilon^2)$ extrapolated to three dimensions.

	$n=1$ Ising	$n=2$ Heisenberg (planar)	$n=3$ Heisenberg	$n=2$ Cubic [a]	$n=3$ Cubic
ρ_1	1.39	1.36	1.34	1.39	1.33
ρ_2	...	1.73	1.86	1.39	1.84
ρ_2'	...	1.73	1.86	1.92	1.87

[a] Has less stability than the Heisenberg fixed point

TABLE II. Sound attenuation function g in Eq. (4) for various polarizations μ and propagation directions $\hat{q}$ for the $[\frac{1}{2}, \frac{1}{2}, \frac{1}{2}]$ R-point soft mode in the perovskites. The form of g' in Eq. (6) is also given by this table with the substitutions discussed after Eq. (6).

$\hat{q}$	μ	$g(\hat{q},\mu)$
(1, 0, 0)	longitudinal	$\frac{1}{9}K_1(A+2B)^2t^{-\rho_1}+\frac{4}{3}K_2'(A-B)^2t^{-\rho_2'}$
(1, 1, 0)	longitudinal	$\frac{1}{9}K_1(A+2B)^2t^{-\rho_1}+\frac{1}{3}K_2'(A-B)^2t^{-\rho_2'}+\frac{1}{4}K_2C^2t^{-\rho_2}$
(1, 1, 1)	longitudinal	$\frac{1}{9}K_1(A+2B)^2t^{-\rho_1}+\frac{1}{3}K_2C^2t^{-\rho_2}$
(1, 0, 0)	(0, 1, 0)	$K_2C^2t^{-\rho_2}$
(1, 1, 0)	$(\bar{1}, 1, 0)$	$K_2'(A-B)^2t^{-\rho_2'}$
(1, 1, 0)	(0, 0, 1)	$\frac{1}{4}K_2C^2t^{-\rho_2}$
(1, 1, 1)	transverse	$\frac{2}{3}K_2'(A-B)^2t^{-\rho_2'}+\frac{1}{12}K_2C^2t^{-\rho_2}$

$$\gamma_{4t}(u_\infty) = -\frac{2\epsilon}{n+8}\left[1-\frac{\epsilon(n^2-4n-36)}{2(n+8)^2}\right]+O(\epsilon^3)\ \text{(HFP)}$$

$$= -\frac{2\epsilon}{3n}\left[1+\frac{\epsilon}{54n^2}(-212+268n-65n^2)\right] + O(\epsilon^3)\qquad \text{(CFP)}.$$

At the CFP, γ_{4t}' is distinct from γ_{4t}:

$$\gamma_{4t}'(u_\infty) = -\frac{(n-2)\epsilon}{3n} \times\left[1+\frac{2\epsilon}{27n^2}(4n^2+40n-53)\right]+O(\epsilon^3)\ \text{(CFP)}.$$

Estimates for ρ_1, ρ_2, and ρ_2' from Eqs. (2) and (3), using results to $O(\epsilon^2)$, are given in Table I. We note that if one assumes $\gamma_{4t}(u_\infty)=0$, as done in Ref. 2, the difference $\rho_2-\rho_1$ would be somewhat larger.

The predicted behavior of sound attenuation in various orientations and polarizations for the perovskites is easily worked out now from symmetry arguments. The sound amplitude attenuation per length is given by

$$\alpha(\hat{q},\mu)=\frac{\omega^2}{4\bar{\rho}k_BTv^3(\hat{q},\mu)}g(\hat{q},\mu), \qquad (4)$$

where $\bar{\rho}$ is the unit-cell mass and $v(\hat{q},\mu)$ is the measured sound velocity. The functions $g(\hat{q},\mu)$ are given in Table II for various directions $\hat{q}$ and polarizations μ. The point we would like to make is that longitudinal sound even in high symmetry directions couples to the "transverse" exponents ρ_2 or ρ_2'. There is currently no good way to calculate K_1/K_2'. However, we note that K_2' is not negligible from Table II since attenuation of sound with $\hat{q}$ parallel to [110] and polarization $[\bar{1},1,0]$ has been observed[15] in $SrTiO_3$. From Ref. 5 we obtain $A/B=-1.6$ and $|C/B|=1.9$ for $SrTiO_3$. Since the ratio $(A+2B)^2/(A-B)^2$ is approximately 0.02, it seems likely that ρ_2' should completely dominate [1, 0, 0] longitudinal attenuation.

The experimental situation for $SrTiO_3$ is characterized by an extremely wide factor of 3 spread in quoted exponents,[15,16] with a mean of about 2. One trend is that exponents for sound involving ρ_2 are higher than those involving ρ_2' although both are comparable at the HFP and CFP. Since λ_1 scales to zero slowly,[6] a possible explanation is that one is not observing true exponents in the experimentally accessible region. The present finding that all exponents should correspond to ρ_2 or ρ_2' indicates that we are much farther from understanding $SrTiO_3$ than the results of Ref. 2 would indicate.

It may be that the problems associated with the perovskites are not as severe[4] in orthorhombic $BaMnF_4$. A free-energy functional consistent with experimental results obtained so far on $BaMnF_4$ has recently been proposed by Fritz[4]:

$$\mathfrak{F}=\mathfrak{F}_{\text{quad}}+\sum_l\left[\tfrac{1}{4}b_1(\Phi_1^4+\Phi_2^4)+\tfrac{1}{2}b_2\Phi_1^2\Phi_2^2 + \tfrac{1}{2}(\beta_1e_{11}+\beta_2e_{22}+\beta_3e_{33})(\Phi_1^2+\Phi_2^2) + \tfrac{1}{2}\beta_4e_{23}(\Phi_1^2-\Phi_2^2)\right]. \qquad (5)$$

For this functional, in contrast to (1), longitudinal attenuation along any axis depends only on ρ_1; transverse attenuation depends only on ρ_2'. The exponents measured by Fritz correspond to $\rho_1=2.2\pm0.3$ and $\rho_2'=3.9\pm0.1$. These values are much larger than those obtained from Table I for the $n=2$ fixed points of (5) for three dimensions. However, a comparison with the $\epsilon=2$, Heisenberg (planar) results $\rho_1=2.2$, $\rho_2'=3.5$, suggests two-dimensional fluctuations. This is consistent[4] with the planar nature of $BaMnF_4$.

The result for the generalized elastic constant follows from the observation that Q appears in (1) only in quadratic form and can be integrated out exactly. Since this is true in the presence of a field coupled to Q, the Q susceptibility or the generalized elastic constant must be expressible entirely in terms of Φ correlation functions. The correct expression is

$$\frac{1}{c(\hat{q},\mu)}=\frac{1}{c_0(\hat{q},\mu)}+\frac{1}{[c_0(\hat{q},\mu)]^2}g'(\hat{q},\mu). \qquad (6)$$

The $g'(\hat{q},\mu)$ is defined in terms of the $\Gamma_{ij;kl}$ at equal time. We define *singular* parts as follows: $\mathrm{Tr}\Gamma_{ii;jj}(q=0, t_1=t_2)=L_1t^{-\alpha_1}$; $\Gamma_{12;12}=L_2t^{-\alpha_2}$; $\Gamma_{11;11}-\Gamma_{11;22}=2L_2't^{-\alpha_2'}$ and obtain $\alpha_1=\alpha$, the specific-heat exponent; $\alpha_2=\alpha_1+\rho_2-\rho_1$; $\alpha_2'=\alpha_1+\rho_2'-\rho_1$. The form of $g'(\hat{q},\mu)$ is again given by Table II, provided one substitutes L's for K's and α's for ρ's. For positive α's Eq. (6) predicts a weak divergence at T_c in elastic constants which shows up initially as a "divergence" in the inverse velocity[17] $[v(\hat{q},\mu)]^{-1}\propto[c(\hat{q},\mu)]^{-1/2}$. Of course, α_2 and α_2' are also related to the crossover scaling exponents φ from Eq. (3) and the discussion following it.

Finally, we note that other second-order processes, not just ultrasonic attenuation will be affected by the α_2, α_2' exponents. For example, second-order Raman intensity measurements could be used to determine α_2. These could be quite important in view of the discrepancies in the observed behavior in $SrTiO_3$.

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[1]W. Rehwald, Phys. Kondens. Mater. 14, 21 (1971); E. Pytte, Phys. Rev. B 1, 924 (1970).

[2]F. Schwabl, Phys. Rev. B 7, 2038 (1973); Phys. Rev. Lett. 28, 500 (1972).

[3]For a review, see M. E. Fisher, Rev. Mod. Phys. 46, 597 (1974).

[4]I. J. Fritz, Phys. Rev. Lett. 35, 1511 (1975).

[5]J. Feder and E. Pytte, Phys. Rev. B 1, 4803 (1970).

[6]A. Aharony, Phys. Rev. B 8, 4270 (1973).

[7]R. A. Cowley and A. D. Bruce, J. Phys. C 6, L191 (1973); I. J. Ketley and D. J. Wallace, J. Phys. A 6, 1667 (1973).

[8]Studies of simpler models with longitudinal strain coupling suggest that a first-order transition results if $\alpha>0$ ($\alpha<0$ for $n=2$ and 3 in three dimensions). General cubic anisotropy may also drive the transition first order. However, we expect to "see" the unrenormalized fixed points if the transition is very weakly first order. See D. J. Bergman and B. I. Halperin (unpublished) for a good discussion.

[9]K. K. Murata, Phys. Rev. B (to be published).

[10]S. M. Shapiro, J. D. Axe, and G. Shirane, Phys. Rev. B 6, 4332 (1972).

[11]B. I. Halperin, P. C. Hohenberg, and S. Ma. Phys. Rev. Lett. 29, 1548 (1972).

[12]K. G. Wilson, Phys. Rev. Lett. 28, 548 (1972).

[13]A. Aharony, Phys. Lett. A 49, 221 (1974).

[14]E. Brézin, J. C. Le Guillou, and J. Zinn-Justin, Phys. Rev. D 8, 434 (1973).

[15]W. Rehwald, Sol. State Commun. 8, 607 (1970).

[16]K. Fossheim and B. Berre, Phys. Rev. B 5, 3292 (1972); J. M. Courdille and J. Dumas, Solid State Commun. 7, 1623 (1969).

[17]The "divergences" in the inverse velocities in $BaMnF_4$ are consistent with the difference in values in ρ_1 and ρ_2', cf. I. J. Fritz (private communication).