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Adiabatic and Isothermal Order-Parameter Susceptibilities and the Thermal Diffusion Mode

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Studies of the adiabatic order-parameter susceptibility near symmetry-breaking phase transitions show that the ratio $\chi_S(t)/\chi_S(-t)$ (in the limit $t\to 0^+$), and the proportion of the order-parameter spectral intensity associated with the thermal diffusion mode (in the limit $t\to 0^-$), are universal. The universal quantities are tabulated and discussed in the light of experiments on structural phase transitions, with particular emphasis on the dielectric properties of ferroelectrics and the scattering properties of ordered phases. New experiments are suggested.

This Letter is concerned with the adiabatic and isothermal order-parameter susceptibilities, χ_S and χ_T, near symmetry-breaking continuous phase transitions. The central result is simply expressed: The limiting $T\to T_c^-$ value of the ratio χ_S/χ_T is a constant unique to each universality class. This result is also easily derived: It follows immediately from results recently established by Aharony and Hohenberg.[1] Its consequences are, however, far reaching—and do not appear to have been previously appreciated. Firstly, it follows that the ratio of the amplitudes characterizing the $T\to T_c^\pm$ singularities of χ_S is itself universal. This result is particularly significant for the interpretation of experiments on ferroelectrics, since dielectric susceptibilities are almost invariably determined under adiabatic conditions. Secondly, it follows that, in the limit $T\to T_c^-$, the thermal diffusion mode carries a universal fraction of the intensity in the order-parameter spectral density. The implications for scattering measurements below classical tricritical points turn out to be particularly interesting.

According to fundamental arguments of thermodynamics the adiabatic and isothermal susceptibilities describing the response of any thermodynamic coordinate Q to its conjugate field E are related by[2,3]

$$\chi_S/\chi_T = C_Q/C_E = 1-(T/\chi_T C_E)(\partial Q/\partial T)_E^2, \qquad (1)$$

where C_Q and C_E denote, respectively, the constant-Q and constant-E specific heats. We apply Eq. (1) to those situations in which Q denotes the order parameter of a symmetry-breaking continuous transition, such that

$$Q(T)\equiv 0;\quad \chi_S\equiv\chi_T \quad (T>T_c). \qquad (2)$$

We assign to the quantities on the right-hand side of Eq. (1) their (most usual) $T\to T_c^-$ limiting forms[4]:

$$Q\approx B(-t)^\beta, \qquad (3a)$$

$$C_E\approx A'(-t)^{-\alpha'}+C_0, \qquad (3b)$$

$$\chi_T\approx\Gamma_T'(-t)^{-\gamma'}, \qquad (3c)$$

where $t\equiv(T-T_c)/T_c$. Within the scaling theory embodied in the renormalization-group approach to phase transitions, the Rushbrooke inequality, interrelating the exponents appearing in (3), holds as an equality[2]:

$$\alpha' + 2\beta + \gamma' = 2\,. \qquad (4)$$

Combining Eqs. (1), (3), and (4) one finds

$$\lim_{t\to 0^-} (\chi_S/\chi_T) = 1 - r_{\rm LP}^*, \qquad (5)$$

where

$$r_{\rm LP}^* = 0 \quad (\alpha' < 0), \qquad (6a)$$

$$r_{\rm LP}^* = \frac{B^2\beta^2}{\Gamma_T' A' T_c} = \frac{\Gamma_T}{\Gamma_T'}\frac{A}{A'}\frac{\beta^2}{R} \quad (\alpha' > 0)\,. \qquad (6b)$$

Here we have introduced the amplitudes A and Γ_T characterizing the $T \to T_c^+$ forms of the singularities (3b) and (3c), together with the quantity

$$R \equiv T_c \Gamma_T A / B^2\,. \qquad (7)$$

It has recently been established[1] that this quantity is universal—that is, its value is prescribed simply by the universality class to which the system concerned belongs. It is well known that the other quantities (Γ_T/Γ_T', A/A', and β) appearing in (6b) are also universal; the universality of $r_{\rm LP}^*$ follows immediately.

Before discussing specific universality classes we develop the two important general corollaries of this central result.

Firstly, we consider the limiting behavior of χ_S. Recalling Eq. (1) we find

$$\frac{\Gamma_S}{\Gamma_S'} \equiv \lim_{t\to 0^+} \frac{\chi_S(t)}{\chi_S(-t)} = \frac{\Gamma_T}{\Gamma_T'} \lim_{t\to 0^+} \frac{\chi_T(-t)}{\chi_S(-t)}, \qquad (8a)$$

so that the ratio

$$\Gamma_S/\Gamma_S' = (\Gamma_T/\Gamma_T')(1 - r_{\rm LP}^*)^{-1} \qquad (8b)$$

is itself universal.

Secondly, we consider the spectral density of the order-parameter fluctuations. It is well known[3,5] that a difference between χ_S and χ_T should be reflected in the existence of a thermal diffusion "central peak," in this spectral function. It follows from (5) that the ratio of the net (frequency-integrated) intensity of this central peak to the residual intensity in the spectral function is given (in the limit $T \to T_c^-$) by a renormalized "Landau-Placzek" constant

$$R_{\rm LP}^* \equiv r_{\rm LP}^*/(1 - r_{\rm LP}^*), \qquad (9)$$

which is, again, universal. We remark that the vanishing of $R_{\rm LP}^*$ in systems whose specific heat does not diverge [cf. Eq. (6a)] is a simple expression of a result that has been established by dynamic renormalization-group methods[6]: The coupling of the order parameter to the thermal diffusion mode is irrelevant near a continuous transition unless the specific heat is divergent.

Table I gives values for the various universal quantities appearing in Eqs. (5), (8b), and (9), for four universality classes in which the specific heat does exhibit a critical divergence. The $d = 3$ short-range-force Ising values (indexed a) are derived from the equations given above, with the aid of series-expansion results tabulated in Ref. 1. On the basis of a recent direct analysis of series expansions for χ_S and χ_T in the spin-$\frac{1}{2}$ Ising model, Betts[7] has suggested a limiting value for χ_S/χ_T which leads to rather different estimates (indexed b). The latter are presumably the more reliable.

The $d = 2$ short-range Ising results are exact. However, although the specific heat is divergent [logarithmically; Eq. (3b) must be modified accordingly] the adiabatic correction $r_{\rm LP}^*$ still vanishes in the limit $T \to T_c^-$.[8]

In the case of the $d = 3$ uniaxial ferroelectric the asymptotic forms given in Eq. (3) have to be modified to allow for the logarithmic corrections characteristic of borderline dimensionalities[4]; Eqs. (6b) and (7) remain appropriate. The values shown in Table I are exact—and again fol-

TABLE I. Values of the universal quantities discussed in the text, for different universality classes. For source references see the text.

Universality Class	$r_{\rm LP}^*$	Γ_T/Γ_T'	Γ_S/Γ_S'	$r_{\rm LP}^*$
Short-range Ising, $d = 3$	0.53[a]		10.90[a]	1.15[a]
		5.07		
	0.75[b]		20.28[b]	3.0[b]
Short-range Ising, $d = 2$	0	37.69....	37.69....	0
Uniaxial dipolar, $d = 3$	3/4	2	8	3
Classical tricritical	1	4	∞	∞

[a] Present work.

[b] Based on results of Betts, Ref. 7; see text.

low from previously established results.[1,9,10]

The final set of results are the simplest to derive and in some respects (see below) are the most interesting: They follow trivially from the *strictly* Landau theory of a tricritical point, appropriate in cases where the borderline dimensionality is *less* than three.

The results gathered in Table I have striking consequences for experiments probing either the susceptibilities or the scattering properties of ordered phases.

It is clear, firstly, that the ratio Γ_S/Γ_S' can be quite different from its isothermal counterpart. This difference may prove to be particularly important in understanding measurements of the dielectric susceptibilities in uniaxial ferroelectrics. The dielectric susceptibility is almost invariably measured at frequencies which guarantee adiabatic conditions. It has long been appreciated[11] that there must exist some "adiabatic correction" to the familiar "law of two" displayed by the ratio Γ_T/Γ_T'. However, it has not been recognized that, in the asymptotic regime, this correction establishes a universal "law of eight." A study of the literature on ferroelectrics shows that values of the observed ratio Γ/Γ' are, indeed, frequently larger than 2. Values of 4, 4.5, and 3 have been reported for $Pb_5Ge_3O_{11}$,[12] triglycine sulfate,[13] and colemanite.[14] At present, however, the interpretation of these results must be tentative since there are other effects—dispersion due to domain-wall motion[15] or piezoelectric coupling[13]—which can lead to quite spurious results in ordered-phase dielectric studies. Further measurements are therefore needed, on monodomain samples at frequencies low compared with those of piezoelectric resonances—and in the regime of the smallest accessible reduced temperatures.

Secondly, it is evident from Table I that a significant proportion of the $T \lesssim T_c$ order-parameter spectral intensity will typically reside in the thermal-diffusion central peak. The most striking case is the classical tricritical point, where the thermal diffusion mode is asymptotically dominant and both Γ_S/Γ_S' and $R_{\rm LP}{}^*$ are formally infinite. These results reflect the fact that, in this case, the adiabatic susceptibility diverges with a smaller exponent ($\frac{1}{2}$) than that characteristic of the isothermal susceptibility ($\gamma' = 1$). One immediate consequence is that, if the remaining portion of the spectral weight resides in a softening phonon, the square of its frequency will vanish, not with the familiar $T_c - T$ Curie law, but as $(T_c - T)^{1/2}$.

The most promising physical realization of this case is KH_2PO_4 whose borderline dimensionality for critical (and thus also tricritical) behavior is less than 3,[16] and which exhibits a tricritical point at a pressure of approximately 2 kbar.[17] Indeed, at atmospheric pressure (where the transition is slightly first order) a strong thermal diffusion mode *has* been observed in the light scattering spectrum immediately below the phase transition[18]—the only occasion, to the author's knowledge, that such a feature has been identified in a soft-mode spectral function. The dominance of the thermal diffusion mode also explains the very large values of Γ/Γ' observed in dielectric studies on KH_2PO_4: Results as high as 10 and 12 have been reported.[13,15] Further light-scattering and dielectric studies closer to the tricritical point should provide an explicit test of the novel predictions made here.

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[1]A. Aharony and P. C. Hohenberg, Phys. Rev. B 13, 3081 (1976); see also D. D. Betts, A. J. Guttman, and G. S. Goyce, J. Phys. C 4, 1994 (1971).

[2]See, e.g., H. Stanley, *Introduction of Phase Transitions and Critical Phenomena* (Clarendon, Oxford, 1971).

[3]H. Thomas, in *Anharmonic Lattices, Structural Transitions and Melting* (Noordhoff, Leiden, 1974), p. 231.

[4]It is convenient to omit the customary prefactor of $1/\alpha'$ in the relation (3b) defining the amplitude of the specific heat singularity. In the case of the uniaxial ferroelectric, the pure powers of $-t$ in Eqs. (3a)–(3c) are replaced by $(-t)^{1/2}|\ln(-t)|^{1/3}$, $|\ln(-t)|^{1/3}$, and $(-t)^{-1}|\ln(-t)|^{1/3}$, respectively.

[5]See, e.g., P. Heller, Int. J. Magn. 1, 53 (1970); R. A. Cowley and G. J. Coombs, J. Phys. C 6, 143 (1973).

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Remarks on the Differing Lifetimes of Charmed Mesons

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Differences in the D^+ and D^0 lifetimes may arise either from 6 dominance in the effective nonleptonic Hamiltonian, or from the dominance of a particular W^+-exchange diagram which behaves like a D^0-$\bar{K}^{0*}$ pole. If 6 dominance holds, then the F^+ will have a relatively short lifetime and a small semileptonic branching ratio, just like the D^0; but if W^+ exchange is dominant, the F^+ will have a long lifetime and large semileptonic branching ratio, like the D^+.

Two different experiments[1,2] report evidence that the charged D meson is much longer lived than its neutral companion. In one experiment,[1] the semileptonic branching ratio for D^+ is found to be at least four times the corresponding ratio for D^0; and in the other experiment,[2] direct measurements of the flight paths of charmed mesons in emulsions indicate that the D^+ lifetime is roughly five times the D^0 lifetime. This result is not consistent with the notion that charmed-meson decay is a process in which the charmed quark is the active partner, and the light quark merely a spectator. Here we examine ways in which this notion might be modified to accomodate the differing lifetimes, and discuss tests based on the F^+ lifetime.

In the standard model of quarks and their weak interactions,[3,4] the inclusive semileptonic decays of charmed mesons are all manifestations of the quark transition

$$c \to s + l^+ + \nu_l \tag{1}$$

and, apart from small corrections due to phase space and Cabibbo-suppressed processes, their rates are equal to one another:

$$\Gamma(D^+ \to l^+\nu_l x^0) \approx \Gamma(D^0 \to l^+\nu_l x^-) \approx \Gamma(F^+ \to l^+\nu_l x^0). \tag{2}$$

Differences in lifetimes must therefore arise from the nonleptonic decay modes that are engendered by the coupling of the standard hadronic current to the charged vector boson W^+:

$$H_{NL} = g[(\bar{s}c) + (\bar{d}u) + \dots]W^+ + \text{H.c.} \tag{3}$$

It is well known[3,5] that this interaction leads to an effective Hamiltonian involving the 6 and 15* representations of SU(3), and that short-distance effects of quantum chromodynamics (QCD) enhance the 6 component over the 15* by a factor of order 3–4. Could this enhancement be responsible for the relatively long lifetime of D^+?

To analyze this question, I note that 6 dominance requires the rate for the exclusive decay $D^+ \to K^-\pi^+$ to be much smaller than the rates for $D^0 \to K^-\pi^+$ and $D^0 \to \bar{K}^0\pi^0$.[5] Experimentally, all three decay modes turn out to have branching ratios in the neighborhood of 2%,[6] and so the absolute rate for the D^+ decay mode will be much smaller than the D^0 decay rates only if the lifetime of D^+ is much longer than the lifetime of D^0.[7] Thus the answer to my question appears, at first sight, to be positive. There are, however, some indications to the contrary from F^+ decay.

Because of the $u \leftrightarrow s$ symmetry properties of the 6 component of the effective Hamiltonian,[8] 6 dominance implies that the F^+ must have approximately the same total nonleptonic decay rate as the D^0. Therefore if the D^0 has a short life-