Scaling theory of nonlinear critical relaxation

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A scaling analysis of nonlinear critical slowing down on the basis of a Landau-type relaxation equation, shows that the critical exponents, $\Delta^{(l)}$ and $\Delta^{(nl)}$ of the linear and nonlinear relaxation times of the order parameter are related by $\Delta^{(nl)} = \Delta^{(l)} - \beta$. Generally if Q scales like $\Delta T^{\beta Q}$ one has $\Delta^{(l)}_Q - \Delta^{(nl)}_Q = \beta_Q$ but a different relaxation may occur in systems with oscillatory modes.

Consider a thermodynamic property $\Psi(t)$, which relaxes to zero with time t; for example, the order parameter of a system at temperature Tabove, but near its critical point, T_c . If $\Psi_0 = \Psi(0)$ is the initially prepared value, the relaxation time may be defined^{1,2} generally by

$$\tau(\Psi_{0}, T) = \int_{0}^{\infty} dt \, \Psi(t) / \Psi_{0} \,. \tag{1}$$

The limit $\Psi_0 \rightarrow 0$, of a vanishingly small initial disturbance, then defines the *linear relaxation time* $\tau^{(1)}(T)$; on approach to a critical point, $\tau^{(1)}$ will in general diverge as $\Delta T - \Delta^{(1)}$ where $\Delta T = T - T_c$. On the other hand, if one lets T approach T_c at fixed finite $\Psi_0 (\neq 0)$ one is studying the nonlinear relaxation time which, in principle, diverges with a distinct exponent $\Delta^{(n1)}$ as $\Delta T - 0$.

Previous discussions^{1,3} have lead one to expect the equality $\Delta^{(1)} \equiv \Delta^{(n1)}$, except for possible violations in nonergodic systems,¹ since the long-time relaxation always occurs in the linear region close to equilibrium. However, it has recently been discovered⁴ that the critical exponents of the linear and nonlinear relaxation may differ even in an ergodic system. In this note, as a step towards a better understanding of the phenomena of nonlinear critical slowing down, we examine the problem on the basis of scaling theory applied to a Landautype relaxation formulation. Our analysis leads to the exponent relation

$$\Delta^{(n1)} = \Delta^{(1)} - \beta , \qquad (2)$$

where β is the exponent characterizing the scaling of Ψ with ΔT . This relation is relevant,⁵ in particular, to the interpretation of the recent observation⁶ of critical slowing down near the order-disorder transition in the binary alloy Ni₃Mn. More generally for the relaxation of another quantity, such as the energy E, we find the corresponding exponent difference $\Delta_E^{(1)} - \Delta_E^{(n1)}$ to be $\beta_E = 1 - \alpha$ (where α is the standard specific-heat exponent), and so on. We also show that for systems in which the relaxation is governed by a non-Hermitian Liouville operator the relation between $\Delta^{(1)}$ and $\Delta^{(n1)}$ may differ from that in a purely dissipative system.

To develop the argument we postulate that the time evolution of the macroscopic order parameter ($\Psi = \langle \psi_{k=0} \rangle$) towards an equilibrium configuration of minimum free energy may be described by the dissipative equation

$$\frac{\partial \Psi}{\partial t} = -\Gamma \frac{\partial F}{\partial \Psi} , \qquad (3)$$

where $F(\Psi, T)$ is the appropriate free energy of the system. Near the critical point, the driving field h satisfies the scaling relation⁷

$$h = \frac{\partial F}{\partial \Psi} \approx \Psi^{\delta} H(\Delta T / \Psi^{1/\beta}), \qquad (4)$$

for $\Psi \ge 0$, where $\delta = 1 + \gamma/\beta$ is the critical isochore exponent, and γ is the susceptibility exponent.

Now in order to describe properly the analytic behavior of h for small Ψ and $T > T_c$, the scaling function H(x) must^{7b} vary as

$$H(x) \approx x^{\gamma} \left[H_{\infty} + H_{\infty}^{(2)} x^{-2\beta} + O(x^{-4\beta}) \right], \qquad (5)$$

as $x \to \infty$, where H_{∞} is directly related to the susceptibility amplitude for $T > T_c$. For finite x the requirement is simply that H(x) be analytic so that

$$H(x) = H_0 + H_1 x + O(x^2), \quad H_0 \neq 0.$$
 (6)

Following various microscopic calculations⁸ and, for example, Suzuki's Kadanoff-type cell analysis for time-dependent phenomena,⁹ it is appropriate to assume that Γ in (3) is a renormalized kinetic coefficient satisfying the scaling relation

$$\Gamma \approx \Delta T^{\sigma} G(\Delta T / \Psi^{1/\beta}) . \tag{7}$$

Note, indeed, that the important memory effects are neglected in (3) only seemingly. From Kawa-

13

5039

saki's work⁸ one can see that, at least near four dimensions, the memory terms lead, in the critical region, merely to a renormalization of the kinetic coefficient as incorporated here.

In order to describe properly the linear regime $(\Psi \rightarrow 0)$ and the relaxation on the critical isotherm the scaling function G(x) must have the following asymptotic properties

$$G(\infty) = \Gamma_0 \neq 0$$
 and $G(x) \approx A x^{-\sigma}$ (8)

as $x \rightarrow 0$. As $x \rightarrow \infty$ the correction terms are of the same form as in (5).

We may now rewrite the evolution equation (3) in terms of the scaled variables

$$m = \Psi / \Delta T^{\beta}, \quad t' = \Delta T^{\gamma + \sigma} t,$$
 (9)

and so obtain

$$\frac{\partial m}{\partial t'} \approx -m^{\delta} K(m^{-1/\beta}), \qquad (10)$$

where K(x) = G(x)H(x). If $m_x(t')$ is the solution of this equation satisfying $m_x(0) = x = \Psi_0 / \Delta T^\beta$, we may rewrite the relaxation time (1) in the scaled form

$$\tau(\Psi_0, T) \approx \Phi(\Psi_0 / \Delta T^{\beta}) / \Delta T^{\gamma + \sigma}, \qquad (11)$$

where the scaling function is

$$\Phi(x) = x^{-1} \int_0^\infty dt' \ m_x(t') \ . \tag{12}$$

Now the linear relaxation time is evidently given by

$$\tau^{(i)} \approx \Phi(0) \Delta T^{-\Delta^{(i)}}, \ \Delta^{(i)} = \gamma + \sigma, \tag{13}$$

provided $\Phi(0)$ is finite and nonzero; this will be shown below. Conversely the *non*linear relaxation time requires knowledge of $\Phi(x)$ for large x; we will demonstrate the behavior

$$\Phi(x) \approx \Phi_{\infty}/x$$
, as $x \to \infty$. (14)

From this we immediately deduce

$$\tau^{(nl)} \approx (\Phi_{\infty}/\Psi_0) \Delta T^{-\Delta^{(nl)}}, \quad \Delta^{(nl)} = \gamma + \sigma - \beta . \quad (15)$$

Comparison with (13) establishes the result (2) for the difference between $\Delta^{(n1)}$ and $\Delta^{(1)}$.

To study $\Phi(x)$ we integrate (10) to obtain

$$t' = V(m_{x}) - V(x),$$
(16)
$$V(y) = \int_{y}^{\infty} \frac{dm}{m^{\delta} K(m^{-1/\beta})} .$$

Since K(x) is positive, V(y) is a monotonic function and so has a monotonic inverse, $V^{-1}(u)$, in terms of which the solution of (10) is $m_x(t') = V^{-1}[t' + V(x)]$. It follows that the scaling function $\Phi(x)$ can be expressed as

$$\Phi(x) = x^{-1} \int_{V(x)}^{\infty} V^{-1}(u) \, du \,. \tag{17}$$

Using the properties (5), (6), and (8) of H(x) and G(x) we find from (16) the small-y behavior

$$V(y) \approx (\Gamma_0 H_{\infty})^{-1} \ln y^{-1} + V_0 + O(y^2), \qquad (18)$$

where the positive constant V_0 comes from the large-*m* region of the defining integral. This means $V(x) \rightarrow \infty$ as $x \rightarrow 0$ and $V^{-1}(u) \approx \exp[-\Gamma_0 \times H_{\infty}(u-V_0)]$ as $u \rightarrow \infty$. Using this information it is easy to show that $\Phi(0) = 1/\Gamma_0 H_{\infty}$ which is finite and nonzero.

To establish (14) we must, by (17), show that the integral $\int_0^{\infty} V^{-1}(u) du \equiv \Phi_{\infty}$ is finite. Convergence at the upper limit follows from (18), as already seen. Since $V^{-1}(u)$ is monotonic, only the lower limit needs study. From (16), using (6) and (8), one finds $V(y) \sim 1/H_0 A y^{(\gamma+\sigma)/\beta}$ as $y \to \infty$, so that when $u \to 0$ one has $V^{-1}(u) \sim u^{-\zeta}$ with $\zeta = \beta/(\gamma+\sigma)$. Hence $\int V^{-1}(u) du$ converges at u = 0 provided $\gamma + \sigma > \beta$, which is always so in reality.¹⁰ Finally, since $V^{-1}(u)$ is positive, the integral defining Φ_{∞} cannot vanish.

These arguments go through with Ψ replaced by another variable, say the energy E, for which the appropriate driving field, say h_E , is (i) finite on the critical isotherm, (ii) analytic across the critical isochore (small field) above T_c , and (iii) can be written in scaled form in terms of, say $\Delta E / \Delta T^{B_E}$.

Indeed a more general heuristic argument for the relation (2), not relying on the determinate Landau-Ginzburg ansatz, can be based on two assumptions: (a) that the divergence of $\tau^{(nl)}(T)$ is determined only by the long-time relaxation in the linear region; and (b) that the extent of the linear region diminishes according to $|\Psi| \leq B \Delta T^{\beta}$. The integral in (1) can then be estimated by

$$\tau^{(nl)} \approx \int_{t_1}^{\infty} dt \, \Psi(t) / \Psi_0, \qquad (19)$$

where t_1 is the time at which the system enters the linear region. Since for $t > t_1$ the system is in the linear region, the integral over $\Psi(t)/\Psi(t_1)$ gives, by definition, the linear relaxation time, so from (19) we have

$$\tau^{(nl)} \approx \tau^{(l)} \Psi(t_1) / \Psi_0.$$
 (20)

But by (b), we have $\Psi(t_1) \approx B\Delta T^{\beta}$ yielding $\tau^{(nl)}/\tau^{(l)} \sim \Delta T^{\beta}$, which implies (2). The difficulty with the argument is that the time t_1 also diverges when $T \rightarrow T_c$ so the contribution to the integral for $t < t_1$ cannot be bounded; indeed according to scaling it contributes equally strongly to the divergence.

When a system must be described by a non-Her-

mitian Liouville operator there may be oscillatory modes and the situation is more complex, since the coupling between modes plays an essential role in developing the singularities in dynamical quantities.¹¹ We have not devised general arguments for nonlinear phenomena in such situations but the following simple model serves to demonstrate that the relation between $\Delta^{(n1)}$ and $\Delta^{(1)}$ can be different from (2) in a system with a non-Hermitian Liouville operator. To this end assume the order parameter does not couple to other quantities and that its motion is described by the nonlinear oscillator equation¹²

$$\frac{\partial^2 \Psi}{\partial t^2} = -\Omega^2 \frac{\partial F}{\partial \Psi} , \qquad (21)$$

where $\partial F/\partial \Psi$ is again given by (4) except that Ψ is replaced by $|\Psi|$ and a factor sgn $\{\Psi\}$ is required. The kinetic coefficient is assumed to vary as $\Omega(\Psi, T) \approx \Delta T^{\omega} W(\Delta T/\Psi^{1/\beta})$ with $W(\infty) = W_{\infty} \neq 0$ and $W(x) \approx Cx^{-\omega}$ as $x \to 0$.

The characteristic time of this system is the period of oscillation τ . Analysis of (21) shows that τ depends on the amplitude Ψ_0 through the scaling relation

$$\tau(\Psi_0, T) = \tilde{\Phi}(\Psi_0 / \Delta T^{\beta}) / \Delta T^{\omega + \gamma/2}$$
(22)

where the scaling function is given by

$$\tilde{\Phi}(x) = 4 \int_0^x \left[Y(x) - Y(y) \right]^{-\frac{1}{2}} dy, \qquad (23)$$

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- ²The relaxation time depends not only on the initial value $\Psi_0 = \langle \psi(0) \rangle$ but also on the initial distribution of microscopic states. We believe, however, that in ergodic systems this dependence is irrelevant from the point of view of critical dynamics.
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- ⁷See, e.g., M. E. Fisher (a) Rep. Prog. Phys. <u>30</u>, 615

$$Y(y) = 2 \int_0^y m^{\delta} \tilde{K}(m^{-1/\beta}) \, dm \,, \qquad (24)$$

with $K(x) = W^2(x)H(x)$. Using the asymptotic properties of H(x) and W(x) one finds $\tilde{\Phi}(0) = \text{const}$ and $\tilde{\Phi}(x) \approx \tilde{\Phi}_{\infty}/x^{(\omega+\gamma/2)/\beta}$ as $x \to \infty$. These results allow us to conclude from (22) that

$$\Delta^{(1)} = \omega + \frac{1}{2}\gamma, \quad \Delta^{(n1)} = 0,$$
 (25)

which evidently differs from the result (2) for simple dissipative systems. We stress, however, that owing to the complexities of systems with interacting oscillatory modes this last relation may well be of restricted applicability.

It may also be remarked that a crucial feature of the model equation of motion (12) is that the "energy"

$$\frac{1}{2}\dot{\Psi}^2 + \Omega^2 \int \left(\frac{\partial F}{\partial \Psi}\right) d\Psi$$

is a conserved quantity. If, as might be more realistic, a dissipative term $\partial \Psi / \partial t$ is introduced which violates this conservation law then, close to the critical point, a crossover to purely dissipative behavior occurs and the system may be described asymptotically again by the Landau-type equation (3).

We are indebted for support to the National Science Foundation, in part through the Materials Science Center at Cornell University, and to the National Research Council of Canada.

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