Nonlinear relaxation near the critical point: Molecular-field and scaling theory

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We examine the critical-point singularity of the nonlinear relaxation of purely dissipative systems. Applying the molecular-field approximation to the time-dependent Ginzburg-Landau model, it is shown that the critical exponent of the nonlinear relaxation time ($\Delta^{(nl)}$) can be different from that of the linear one ($\Delta^{(l)}$) even in ergodic systems. From the assumptions that scaling applies and that only the near-equilibrium fluctuations significantly affect the relaxation time, a scaling law $\Delta^{(nl)} = \Delta^{(l)} - \beta$ is derived (β is the critical exponent of the order). On the base of this scaling law we reanalyze the available Monte Carlo calculations, high-temperature series, and the critical-slowing-down experiment on Ni_3Mn.

I. INTRODUCTION

The renormalization-group formalism has proved to be very useful in calculating not only the static[1] but also the dynamic critical properties.[2] By this method the critical-point singularity of the linear dynamic response has been found for several models.[2-4] The linear-response theory, however, describes the reaction of the system to an infinitesimal external disturbance, while in experiments[5] and computer simulations[6-8] it is often much easier to realize nonlinear-response situations, i.e., it is much easier to investigate the response of the system to finite changes in the thermodynamic variables. A natural question is whether the critical-point singularity of the two responses is the same.

Since in nonergodic systems the initial condition plays an important role, the linear and nonlinear responses are expected to have different singularities. In fact, the one-dimensional X-Y model,[9] which is nonergodic and exhibits a critical field at *zero* temperature, can be solved exactly; the divergence of the nonlinear relaxation time of the order $\tau^{(nl)}$ is shown[10] to be different from that of the linear one ($\tau^{(l)}$).

Ergodic systems, on the other hand, reach their equilibrium independently of the initial conditions. The assumption that the initial and intermediate stages of the relaxation do not affect the divergence of the relaxation time, since the critical fluctuations appear only very close to equilibrium,[10] led to the belief that in ergodic systems $\tau^{(nl)}$ and $\tau^{(l)}$ diverge with the same critical exponent. This view seemed to be supported by Monte Carlo calculations[7] and by high-temperature series expansion[10] on the two-dimensional one-spin-flip kinetic Ising model.[11]

Recently, it has been shown in a short communication[12] that the singularity of $\tau^{(nl)}$ and $\tau^{(l)}$ can be different even in ergodic systems. In the present

paper we discuss the problem in more detail and give a careful analysis of the assumption that the initial and intermediate stages of the relaxation do not critically affect the relaxation time. This leads to the conclusion that the singularity of $\tau^{(nl)}$ should be weaker than that of $\tau^{(l)}$.

In order to illustrate the point, first the time-dependent Landau-Ginzburg equation[13-15] is treated in the molecular-field approximation. In Sec. II we examine both the cases of conserved and nonconserved order parameter. Section III contains the calculation of the relaxation time for the case when the thermodynamic force restoring the equilibrium is assumed to be the derivative of the scaled free energy. In Sec. III we also explore the implications of the scaling assumption as applied to nonlinear relaxation, and a scaling law connecting the critical exponents of $\tau^{(l)}$ and $\tau^{(nl)}$ is derived. Finally, in Sec. IV the above mentioned Monte Carlo calculations[7,8] and high-temperature-expansion series[10] are analyzed in the light of the new scaling law, and a new interpretation is given to the critical-slowing-down experiment[5] in Ni_3Mn.

II. MOLECULAR-FIELD THEORY

The simplest and best understood systems exhibiting critical slowing down are the kinetic Ising models[11,16,17] and their continuum analogs, the time-dependent Ginzburg-Landau (TDGL) models.[3,13-15] The static properties of these systems are known and they have no complications other than the diverging relaxation time near the critical point.

The calculation of their nonlinear response involves, in an essential way, not only the small eigenvalues of the Liouville operator of the system but also the large ones, which makes the problem difficult. Thus, as a first step toward an understanding of the difference between linear and nonlinear relaxation, we shall examine the TDGL

models in a molecular-field approximation.

TDGL theory describes the time evolution of the order parameter $\psi(\vec{x}, t)$ toward the minimum-free-energy configuration by an equation of purely dissipative form

$$\frac{\partial \psi(\vec{x}, t)}{\partial t} = -\Gamma \frac{\delta F(\psi, T)}{\delta \psi(\vec{x}, t)} , \qquad (1)$$

where $F(\psi, T)$ is the standard Landau expression for the free energy of a system with a real, one-component order parameter

$$F(\psi, T) = \int d\vec{x} \{\tfrac{1}{2} r \psi^2(\vec{x}, t) + \tfrac{1}{4} u \psi^4(\vec{x}, t)$$

$$+ \tfrac{1}{2} [\nabla \psi(\vec{x}, t)]^2 \} , \qquad (2)$$

with $r = 1 - T_c/T$. T_c is the critical temperature in the molecular-field approximation and u is a positive constant.

If the order parameter is not a conserved quantity then Γ in Eq. (1) is just a constant Γ_0 setting the time scale, while if it is conserved then $\Gamma = -\lambda_0 \nabla^2$ in order to ensure that $\int d\vec{x} \psi(\vec{x}, t)$ does not depend on time. Thus after Fourier transformation, (1) takes the form

$$\frac{d\psi_q(t)}{dt} = -\Gamma_q \Big((r + q^2) \psi_q(t)$$

$$+ u \sum_{q_1 q_2} \psi_{q_1}(t) \psi_{q_2}(t) \psi_{q-q_1-q_2}(t) \Big) , \qquad (3)$$

where

$$\psi(\vec{x}, t) = \sum_q e^{i\vec{q} \cdot \vec{x}} \psi_q(t) \qquad (4)$$

and $\Gamma_q = \lambda_0 q^2$ or $\Gamma_q = \Gamma_0$, depending on whether ψ_0 is or is not conserved.

Usually a Langevin noise term and an infinitesimal field conjugate to the order parameter are added to the right-hand side of (3), the former to imitate the equilibrium fluctuations, the latter to calculate the linear response to the field. We can omit both terms since in the molecular-field approximation the Langevin noise term would disappear and we want to investigate the nonlinear relaxation, so for our purposes it is more convenient to determine the relaxation time of the order parameter in the following way. For $-\infty < t < 0$ a field is switched on so that an initial state is prepared in which

$$\delta \langle \psi_k(0) \rangle = \delta \langle \psi_{-k}(0) \rangle \neq 0 \quad \text{and} \quad \delta \langle \psi_{q \neq k}(0) \rangle = 0 , \qquad (5)$$

where

$$\delta \langle \psi_k(0) \rangle = \langle \psi_k(0) \rangle - \langle \psi_k(\infty) \rangle , \qquad (6)$$

the brackets $\langle \rangle$ being the equilibrium average in the initial ensemble with the field switched on. At $t = 0$ the field is switched off, and since the TDGL models are ergodic by construction

$$\langle \psi_k(\infty) \rangle = \langle \psi_k \rangle_{\text{eq}} , \qquad (7)$$

where the brackets $\langle \rangle_{\text{eq}}$ denote the equilibrium average in the system without the field. Throughout this paper we shall consider only the high-temperature phase where $\langle \psi_k \rangle_{\text{eq}} = 0$ for every k; thus,

$$\delta \langle \psi_k(t) \rangle = \langle \psi_k(t) \rangle . \qquad (8)$$

The systems described by TDGL models are purely dissipative systems, so having the solution of (1) for $\langle \psi_k(t) \rangle$ satisfying the initial condition (5), the relaxation time of the order parameter can be defined[10] as

$$\tau_k = \int_0^\infty \frac{\langle \psi_k(t) \rangle}{\langle \psi_k(0) \rangle} dt . \qquad (9)$$

For small initial deviation from equilibrium, τ_k goes over to the definition of the relaxation time in linear regime

$$\tau_k^{(l)} = \lim_{\langle \psi_k(0) \rangle \to 0} \tau_k$$

$$= \int_0^\infty \frac{\langle \psi_k(t) \psi_k(0) \rangle_{\text{eq}}}{\langle \psi_k^2 \rangle_{\text{eq}}} dt , \qquad (10)$$

while for finite $\langle \psi_k(0) \rangle$, (9) is the definition of the nonlinear relaxation time

$$\tau_k^{(nl)} = \tau_k \big|_{\langle \psi_k(0) \rangle, \text{finite}} . \qquad (11)$$

In the molecular-field approximation the higher-order correlation functions are decoupled so, averaging Eq. (3) over the initial ensemble, one finds that the equation of motion for $\langle \psi_q(t) \rangle$ is Eq. (3), only $\psi_q(t)$ has to be substituted by $\langle \psi_q(t) \rangle$. With the initial conditions (5), $\langle \psi_{q \neq k}(t) \rangle = 0$ and $\langle \psi_k(t) \rangle = \langle \psi_{-k}(t) \rangle$ for all t, so the equation for $\langle \psi_k(t) \rangle$ reduces to

$$\frac{d}{dt} \langle \psi_k(t) \rangle = -\Gamma_k [(r + k^2) \langle \psi_k(t) \rangle + 3u \langle \psi_k(t) \rangle^3] , \qquad (12)$$

where the factor 3 in front of u is replaced by 1 in case the uniform mode $k = 0$ is considered.

The solution of this equation satisfying the initial condition $m_0 = [3u/(r + k^2)]^{1/2} \langle \psi_k(0) \rangle$ is easily found:

$$\frac{\langle \psi_k(t') \rangle}{\langle \psi_k(0) \rangle} = [(1 + m_0^2) e^{2t'} - m_0^2]^{-1/2} , \qquad (13)$$

where $t' = \Gamma_k (r + k^2) t$.

For the case $k = 0$ and $\Gamma_k = \Gamma_0 = 1$, the solution (13) has been discussed in connection with the molecular-field approximation to both the kinetic Ising model[18] and the TDGL model.[19] In these works, however, the linear relaxation time and the lifetime of the metastable states were the main con-

cern. Regarding the nonlinear relaxation time, the authors satisfied themselves with the observation that for large t' (13) shows the same exponential decay as the linear relaxation ($m_0 \to 0$). This observation led to the erroneous conclusion[10] that the state with uniform order parameter is an asymptotic eigenstate of the Liouville operator of the system, thus $\tau^{(l)} \sim \tau^{(nl)}$.

The weakness of the above argument lies in overlooking the fact that $\tau^{(nl)}$ is determined by an integral (9), (11). The system starting with a finite deviation from equilibrium arrives at equilibrium via near-equilibrium ("linear") states which decay with the linear relaxation time; however, since the linear regime is confined near the critical point to smaller and smaller deviations from equilibrium, the linear part of the relaxation has such small weight in the integral determining $\tau^{(nl)}$ that the singularity of $\tau^{(nl)}$ becomes weaker than that of $\tau^{(l)}$. This phenomenon can be illustrated by calculating $\tau^{(l)}$ and $\tau^{(nl)}$ explicitly. Substituting (13) into (9), one finds

$$\tau_k = \frac{1}{\Gamma_k(r+k^2)} \phi\left(\left(\frac{3u}{r+k^2}\right)^{1/2} \langle \psi_k(0)\rangle\right) , \tag{14}$$

where

$$\phi(x) = (1/x)\arcsin[x/(1+x^2)^{1/2}] . \tag{15}$$

The linear relaxation time (10) is determined by putting $\langle \psi_k(0)\rangle = 0$ in (14). Since $\phi(0)=1$, we have

$$\tau_k^{(l)} = 1/\Gamma_k(r+k^2) . \tag{16}$$

If $\langle \psi_k(0)\rangle$ is kept finite when approaching T_c ($r \to 0$), then the argument of ϕ becomes large for small k, and since

$$\lim_{x \to \infty} \phi(x) = \tfrac{1}{2}\pi(1/x) , \tag{17}$$

the nonlinear relaxation time is different from $\tau^{(l)}$:

$$\tau_k^{(nl)} = A/\Gamma_k(r+k^2)^{1/2} , \tag{18}$$

where $A = \pi/[2(3u)^{1/2}\langle\psi_k(0)\rangle]$.

In the molecular-field approximation the correlation length scales as $\xi \sim r^{-1/2}$, so both $\tau_k^{(l)}$ and $\tau_k^{(nl)}$ have the form required by dynamic scaling,[20,21]

$$\tau^{(l)} = k^{-z^{(l)}}\Omega_1(k\xi) \tag{19}$$

and

$$\tau^{(nl)} = k^{-z^{(nl)}}\Omega_2(k\xi) , \tag{20}$$

but the dynamic critical exponents $z^{(l)}$ and $z^{(nl)}$ are not equal. They of course depend on whether ψ_0 is or is not conserved.

In the case when the order parameter is not conserved ($\Gamma_k = \Gamma_0 = \text{const}$),

$$z^{(l)} = 2 \text{ and } z^{(nl)} = 1 , \tag{21}$$

while in the case of conserved order parameter ($\Gamma_k = \lambda_0 k^2$),

$$z^{(l)} = 4 \text{ and } z^{(nl)} = 3 . \tag{22}$$

Since it is easily measured, special attention is usually paid to the uniform mode ($k = 0$) of the nonconserved order parameter. The critical exponents of its relaxation time are defined by the relations

$$\tau_0^{(l)} \sim r^{-\Delta^{(l)}} \text{ and } \tau_0^{(nl)} \sim r^{-\Delta^{(nl)}} , \tag{23}$$

and their value can be read from (16) and (18), putting $k = 0$ in these formulas:

$$\Delta^{(l)} = 1 \text{ and } \Delta^{(nl)} = \tfrac{1}{2} . \tag{24}$$

Since the molecular-field theory is valid for systems with infinite-range forces, the conclusion to be drawn from the above calculation is that the singularity of the linear and nonlinear response can be different even in ergodic systems.

We conclude this section with the remark that from the functional form of $\tau_{k=0}$ [Eqs. (14) and (15)] one can see that, if the initial deviation is finite but small enough ($x \ll 1$) then, on approaching the critical point, a relaxation time diverging with the linear exponent is observed first. However, close enough to T_c ($x \gg 1$) there is a crossover to the nonlinear exponent. The crossover region ($x \sim 1$) is scaled by the critical exponent of the order $\beta = \tfrac{1}{2}$:

$$\langle \psi_0(0)\rangle/r^{1/2} \sim \text{const} . \tag{25}$$

III. SCALING THEORY

In systems with short-range interactions the molecular-field theory breaks down near the critical point. As a more realistic treatment of these systems, we adopt the following approach[22]: only the uniform mode ($\psi = \langle \psi_0\rangle$) is considered and the equation of motion is assumed to be that of the TDLG theory (1), but the Landau free energy (2) is replaced by the scaled free energy[23]

$$F(\psi, r) = r^{\gamma + 2\beta} f(\psi/r^\beta) , \tag{26}$$

where now $r = 1 - T_c/T$, with T_c being the real critical temperature and γ and β are the critical exponents of the susceptibility and the order, respectively. Also, Γ is assumed to be the renormalized kinetic coefficient with temperature dependence $\Gamma = \Gamma_0 r^\sigma$.

The above approach is certainly phenomenological but its justification can be found in the Kadanoff-type cell analysis extended to time dependent phenomena.[24]

The calculation of the relaxation time is parallel to that of the previous section. Introducing scaled variables

$$t' = \Gamma_0 r^{\gamma + \sigma} t \tag{27}$$

and

$$m(t') = \psi(t')/r^{\beta} , \tag{28}$$

the equation of motion can be written

$$\frac{\partial m}{\partial t'} = -\frac{\partial f}{\partial m} . \tag{29}$$

Having the solution $m(t')$ of (29) satisfying the initial condition $m_0 = \psi(0)/r^{\beta}$, the relaxation time (9) is expressed in scaled form

$$\tau_0 \equiv \tau = (1/\Gamma_0 r^{\gamma + \sigma}) \, \phi_s[\psi(0)/r^{\beta}] , \tag{30}$$

with

$$\phi_s(m_0) = \int_0^{\infty} \frac{m(t')}{m_0} dt' . \tag{31}$$

In order to find $\tau^{(l)}$ and $\tau^{(nl)}$, the small- and large-argument asymptotes of ϕ_s are needed. The problem is that $f(m)$ is not known in general, so the solution of (29) is not available. What is known are the asymptotes of[23] $\partial f/\partial m$:

$$\lim_{m \to 0} \frac{\partial f}{\partial m} \sim m \tag{32}$$

and

$$\lim_{m \to \infty} \frac{\partial f}{\partial m} \sim m^{1+\gamma/\beta} , \tag{33}$$

so the solution of (29) can be obtained if $m(t') \gg 1$ or $m(t') \ll 1$. We shall assume that these solutions can be extended to the region where $m(t') \sim 1$ and they match there continuously.

If $m_0 \ll 1$ then the matching condition is not needed since $m(t') \ll 1$ for all t', so we are in the linear regime (32) and the solution of (29) is a simple exponential [the proportionality constants in (32) and (33) are set to 1 since they are irrelevant]:

$$m(t') = m_0 e^{-t'} ; \tag{34}$$

thus,

$$\phi_s(m_0 \to 0) = 1 , \tag{35}$$

which determines the linear relaxation time

$$\tau^{(l)} \sim r^{-\gamma - \sigma} , \tag{36}$$

so

$$\Delta^{(l)} = \gamma + \sigma . \tag{37}$$

If the $m_0 \gg 1$, then the solution of (29) for $t' < t_1$ when $m(t') \gg 1$ is given by

$$m_>(t') = m_0/[1 + (\gamma/\beta)m_0^{\gamma/\beta} t']^{\beta/\gamma} , \tag{38}$$

while for $t' > t_2$ when $m(t') \ll 1$ the solution is again an exponential,

$$m_<(t') = B e^{-(t'-t_2)} . \tag{39}$$

The assumption that the solutions (38) and (39) can be extended to the boundary between the linear and nonlinear regime $(m \sim 1)$ implies that $B \sim 1$ and $t_2 = t_1$. The matching condition $m_>(t_1) = m_<(t_1) \sim 1$ gives t_1. From (38) it is easily seen that for $m_0 \gg 1$, $t_1 \sim 1$.

Now the integral (31) can be calculated using (38) for $t' < 1$ and (39) for $t' > 1$. Elementary integration gives

$$\lim_{m_0 \to \infty} \phi_s(m_0) \sim \frac{1}{m_0} , \tag{40}$$

which implies for the nonlinear relaxation time

$$\tau^{(nl)} \sim r^{-\gamma - \sigma + \beta} , \tag{41}$$

i.e., the critical exponent of $\tau^{(nl)}$ is different from that of $\tau^{(l)}$. Comparing (36) and (41), one finds

$$\Delta^{(nl)} = \Delta^{(l)} - \beta . \tag{42}$$

Now we give a more general argument supporting this scaling law. It is based on two assumptions:

(i) The relaxation time τ has the scaled form (30). Scaling does not give any relationship between $\Delta^{(l)}$ and $\Delta^{(nl)}$, since it does not specify the large-x behavior of $\phi_s(x)$. If, for large x, $\phi_s(x) \sim x^{-a}$, then $\Delta^{(nl)} = \Delta^{(l)} - a\beta$. The value $a = 1$ is favored by observing that there is an explicit x^{-1} dependence in the definition of τ [Eq. (9)]. This will be the large-x behavior of $\phi_s(x)$ if the second assumption is valid.

(ii) The initial and intermediate stages of relaxation do not critically affect the relaxation time. Then, since ergodic systems are being considered, starting far enough from equilibrium the system forgets its initial condition when it arrives at the near-equilibrium states, so $\langle \psi(t) \rangle$ does not depend on $\langle \psi(0) \rangle$. Thus the critical contribution to the integral determining τ will depend on $\langle \psi(0) \rangle$ only through the denominator, implying that $\phi_s \sim x^{-1}$. Of course, the validity of the above assumption remains to be checked by microscopic calculations and experiments.

IV. SEARCH FOR DIFFERENCE BETWEEN $\Delta^{(l)}$ AND $\Delta^{(nl)}$

Since there is no microscopic calculation at hand, the ideas developed in the previous sections and the scaling relation (42) can be compared only with high-temperature series, Monte Carlo (MC) calculations, and with the only available experiment on Ni_3Mn.

A. MC calculations and high-temperature series

Because of computer storage and time problems, the MC calculations are mainly confined to two-

dimensional systems. Since, in the two-dimensional Ising model, β is a small number ($\beta = 0.125$) equal to the accuracy of the individual determination of $\Delta^{(l)}$ and $\Delta^{(nl)}$, the difference between them, (42), is easily overlooked, especially if one believes that they must be equal.

The first attempt to calculate the relaxation time of the two-dimensional one-spin-flip kinetic Ising model is due to Ogita et al.[6] Their MC result $\tau^{(nl)} \sim r^{-1.75}$ has been interpreted as $\Delta^{(nl)} = \Delta^{(l)} = \gamma$. In fact, this estimate comes from the temperature region $0.2 < r < 1$, where the effective exponent of the susceptibility ($\chi \sim r^{-\gamma_{eff}}$ varies between 1 and 1.5, indicating that $\Delta^{(nl)} > \gamma$. Later, it was realized[7] that the corrections to scaling are much smaller if the relaxation time is considered as a function of the natural high-temperature variable $v = \tanh(2J/kT)$. Plotting Ogita's result as a function of v one finds $\Delta^{(nl)} \approx 2.1$, which is close to the high-temperature-series estimate[25-27] of the linear relaxation time exponent $\Delta^{(l)} \approx 2.0$.

Stoll et al.[7] extended the MC calculations to the temperature region $0.02 < r < 0.2$. Their result $\Delta^{(nl)} = 1.85 \pm 0.1$ would be in good agreement with the scaling law (42) and, combining it with the previous result $\Delta^{(nl)} \approx 2.1$ from the temperature range $0.2 < r < 1$, it would even demonstrate the ideas discussed at the end of Sec. II about the crossover from $\Delta^{(l)}$ to $\Delta^{(nl)}$ as T_c is approached. The problem, however, is that they have found the same singularity for the linear relaxation time $\Delta^{(l)} = 1.85 \pm 0.1$.

High-temperature series[10] for $\tau^{(nl)}$ do not help to sort out this problem. Only six terms have been calculated and, applying the ratio method to the series[28]

$$\frac{\tau^{(l)}}{\tau^{(nl)}} = \sum_k \frac{a_k^{(l)}}{a_k^{(nl)}} V^k , \tag{43}$$

where $a_k^{(l)}$ and $a_k^{(nl)}$ are the expansion coefficients for $\tau^{(l)}$ and $\tau^{(nl)}$, one finds the following estimates for the difference between $\Delta^{(l)}$ and $\Delta^{(nl)}$:

$$\Delta^{(l)} - \Delta^{(nl)} \cong -0.25, \ 0, \ 0, \ 0.21, \ 0.11, \ -0.04 . \tag{44}$$

Since the oscillation is of order β, the question whether $\Delta^{(l)}$ and $\Delta^{(nl)}$ are equal or not remains unanswered.

Clearly, a more precise MC calculation is needed which would cover the whole temperature region $0.02 < r < 1$ for both $\tau^{(l)}$ and $\tau^{(nl)}$, and also the high-temperature series has to be extended to higher-order terms.

The qualitative picture of crossover from $\Delta^{(nl)} = \Delta^{(l)}$ to $\Delta^{(nl)} < \Delta^{(l)}$ is apparent in the dynamics of the kinetic Ising model with vacancy mechanism of

relaxation[8] (Fig. 1). The MC calculation shows that far from the critical temperature $\tau^{(l)} \sim \tau^{(nl)}$, but around $r \cong 0.2$ the effective exponent of $\tau^{(nl)}$ becomes smaller than that of $\tau^{(l)}$. Unfortunately, the two points where the deviation is observed have large error bars, so it is impossible to draw any numerical conclusion with respect to $\Delta^{(nl)}$.

A crossover can be also observed in the energy relaxation. We have not discussed this question, but it is clear from the previous sections that the nonlinearity of the relaxation affects the critical exponent of the energy relaxation (Δ_E), too. For example, in the molecular-field approximation $\Delta_E^{(l)} = 1$, while $\Delta_E^{(nl)} = 0$ (logarithmic singularity).[12] In case of the one-spin-flip kinetic Ising model high-temperature-series estimates,[27] MC calculations[7] and renormalization-group arguments[3] suggest that $\Delta_E^{(l)} = 2.0$. At the same time, the result of Ogita et al.[6] on the nonlinear relaxation of the energy (Fig. 2) shows that $\Delta_E^{(nl)} \approx 2$ only far from T_c, while around $r \approx 0.2$ there is a crossover to $\Delta_E^{(nl)} \approx 0.3$. Probably the value 0.3 does not have to be taken seriously, but it is evident that $\Delta_E^{(nl)} \neq \Delta_E^{(l)}$.

B. Dynamics of Ni$_3$Mn

To our knowledge the only experiment concerned with nonlinear critical slowing down is that of Collins and Teh[5] on the order-disorder transformation of Ni$_3$Mn. The ordering in Ni$_3$Mn is a first-order phase transition,[29] but the discontinuity of the order parameter is so small that it is undetectable by neutron-scattering experiment, and approaching
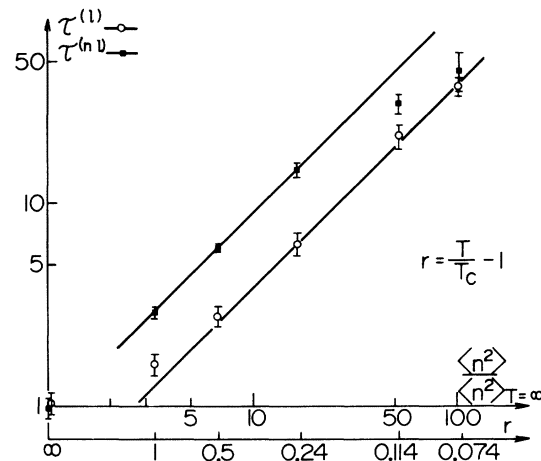

FIG. 1. Dynamical response in the kinetic Ising model with vacancy mechanism of relaxation. The linear ($\tau^{(l)}$) and nonlinear ($\tau^{(nl)}$) relaxation time of the order is plotted vs the fluctuation of the order ($\langle n^2 \rangle / \langle n^2 \rangle_{T=\infty}$). The normal temperature scale is also indicated. In the region $r \gtrsim 0.2$ the effective exponent of $\tau^{(nl)}$ becomes smaller than that of $\tau^{(l)}$.
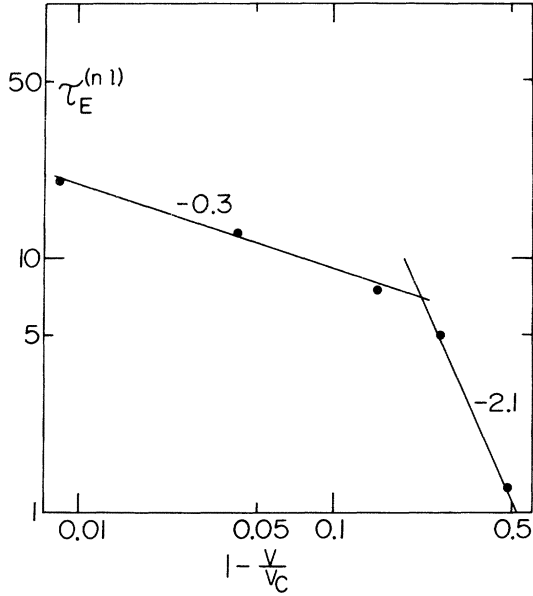
FIG. 2. Relaxation time of the energy of the one-spin-flip kinetic Ising model in nonlinear regime. $v = \tanh(2J/kT)$ is the natural high-temperature variable. The effective exponent changes from 2.1 to 0.3 on approaching T_c.

T_c one observes critical fluctuations.

The relaxation time of the order has been measured below T_c by neutron-scattering techniques. Making a steplike change in temperature ($\Delta T \approx 7°$ typically), the crystal was put into a nonequilibrium state and then the time of relaxation to the new equilibrium state was measured by observing the change in the intensity of the scattering. The resulting time τ was analyzed as a product of Ising time τ_I (ordering time) and diffusion time τ_D (unit of time for the ordering process):

$$\tau = \tau_I \tau_D . \tag{45}$$

The diffusion time was assumed to be an exponential

$$\tau_D \sim e^{-E_a/kT} , \tag{46}$$

and the activation energy $E_a = 3.0 \pm 0.2$ eV was chosen so that the ordering time had a simple power-law behavior,

$$\tau_I \sim |r|^{-\Delta} . \tag{47}$$

In this way, $\Delta = 1.04 \pm 0.09$ was found.

This result is surprising at first sight, since the binary alloys are supposed to be good examples of Ising systems. The vacancy mechanism of relaxation might change Δ as compared to the one-spin-flip model,[8] but because of the purely dissipative character of the system one still expects[30] $\Delta \geq \gamma$

= 1.25. One explanation[31] of the $\Delta \simeq 1$ result is that Ni_3Mn, being a metallic alloy, has long-range forces; thus the mean-field theory applies which predicts $\Delta = 1$ for the linear relaxation time.

It can be questioned, however, whether $\tau^{(l)}$ or $\tau^{(nl)}$ was measured in this experiment. If the change of temperature ΔT is kept constant when approaching T_c then at one point the initial deviation of the order $\delta m(0)$ becomes comparable with the equilibrium value $m \approx |r|^\beta$, so we arrive to the nonlinear regime $\delta m(0)/|r|^\beta > 1$. If this happens, then a change of the critical exponent should be observed. It can be observed, if one notices that the log-log plot of τ_I vs $|r|$ is a straight line only for the special value of the activation energy $E_a = 3.0$ eV. The activation energy of the vacancy migration in Ni_3Mn, however, is not a known quantity. Allowing other values $3.0 < E_a < 3.6$ eV (see Fig. 3 for $E_a = 3.4$ eV), the straight line breaks into two. The slope of the line in the temperature region $0.001 < |r| < 0.01$ is -1.0, and it is independent of E_a. The other part $(0.01 < |r| < 0.08)$, which is also a straight line, has a slope varying from -1.0 to -1.5 as E_a is changed from 3.0 to 3.6 eV. The location of the changeover is independent of E_a and, within a factor of 2, it coincides with the location
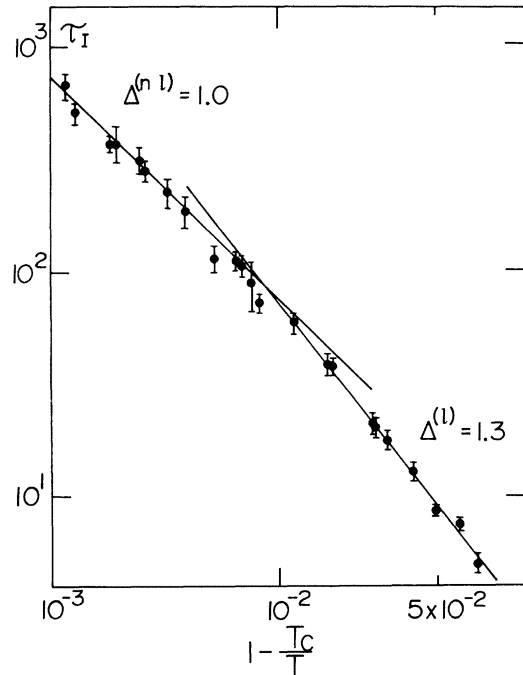

FIG. 3. Relaxation time of the order in Ni_3Mn. The activation energy of the vacancy migration E_a is set arbitrarily at 3.4 eV. The slope -1.0 in the region $0.001 < |r| < 0.01$ is independent of E_a, but the slope of the tail changes from -1.0 to -1.5 as E_a is changed from 3.0 to 3.6 eV.

of the point where the $\delta m(0)/|r|^\beta = 1$ condition is satisfied (β is allowed to vary between 0.3 and 0.5). This strongly suggests that $\Delta = 1.0$ is the critical exponent of the nonlinear relaxation, and to find $\Delta^{(l)}$ one should determine E_a from an independent experiment. If $E_a \simeq 3.4$ eV, then the scaling law $\Delta^{(nl)} = \Delta^{(l)} - \beta$ would be satisfied with $\beta \approx 0.3$ near the Ising value.

V. FINAL REMARKS

The examples described in Sec. IV demonstrate quite well that $\Delta^{(nl)} \neq \Delta^{(l)}$, but they are not sufficient to verify the scaling prediction $\Delta^{(nl)} = \Delta^{(l)} - \beta$. Although a final verification of this scaling law remains the task of a microscopic theory, we summarize what could be done by other means.

(a) In the MC calculations, $\Delta^{(l)}$ and $\Delta^{(nl)}$ have to be determined with accuracy much better than β. With improving computer techniques this does not seem to be impossible.

(b) The high-temperature series for $\tau^{(nl)}$ has to be calculated further until the oscillation in $\Delta^{(l)} - \Delta^{(nl)}$ becomes much less than β.

(c) The activation energy of the vacancy migration in Ni_3Mn has to be determined independently of the critical-slowing-down experiment. It would be also worthwhile to investigate the initial-condition dependence of the relaxation time in the nonlinear region. In the original Ni_3Mn experiment, only two points were investigated in the linear region and, of course, the relaxation time was insensitive to small variations of the initial condition.

Finally, we mention that although the arguments of the present paper apply only to purely relaxational systems, a similar changeover from linear to nonlinear behavior is expected in systems with oscillating modes. We refer here to the very simple Alben model[32] which is exactly described by the Landau theory and the equation of motion, for the order parameter is that of an anharmonic oscillator,[33]

$$\frac{d^2\psi}{dt^2} = -r\psi - u\psi^3 .$$ (48)

If the initial deviation from equilibrium is infinitesimal, the characteristic time of the system is the period of the linear oscillator which diverges as we approach T_c ($r \to 0$):

$$\tau^{(l)} \sim \omega^{-1} \sim r^{-1/2} .$$ (49)

On the other hand, if the initial deviation is finite, the period of the motion goes to a finite value at T_c:

$$\lim_{r \to 0} \tau^{(nl)} = \text{const} .$$ (50)

The difference between the exponents is again $\frac{1}{2}$ as for the TDGL model in mean-field approximation.

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