

## Scaling of thermal hysteresis with temperature scanning rate

Zhong Fan and Zhang Jinxiu

*Department of Physics, Zhongshan University, Guangzhou 510275, People's Republic of China*

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We address several problems concerning thermal hysteresis and its characterization. Based on the time-dependent Ginzburg-Landau theory of the  $(\Phi^2)^3$  model with  $O(N)$  symmetry in the large- $N$  limit, we obtain numerically familiar spindle-shaped thermal hysteresis loops under linearly varying temperature. These hysteresis loops can be presented in the coordinate plane formed by the reduced temperature and its conjugate variable, so that the enclosed areas  $A$  of the loops represent directly the dissipation in the cycles. Moreover, this dissipation scales with the rate of temperature scanning  $R$  as  $A = A_0 + aR^n$ , with  $n$  approaching two-thirds, universal for both the mean-field and field theoretical models concerned. The scaling results from the nature of the equilibrium transition point involved.

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There is recent interest in phase transitions driven by time-dependent external fields [1–6], and a scaling law of the areas of the hysteresis loops with the amplitude  $H_0$  and frequency  $\Omega$  of the applied field has been obtained to be

$$A \approx H_0^\alpha \Omega^\beta. \quad (1)$$

Rao and co-workers [1,2] used a time-dependent Ginzburg-Landau (TDGL) theory of a nonconserved  $N$ -component order parameter in large- $N$  limit with a sinusoidal magnetic field to obtain hysteresis loops and proposed the area scaling with  $\alpha \sim 0.66 \pm 0.05$  and  $\beta \sim 0.33 \pm 0.3$ , universal for both  $(\Phi^2)^2$  and  $(\Phi^2)^3$  models. Monte Carlo simulation of the two-dimensional (2D) Ising model by Lo and Pelcovits [3] confirmed the scaling and produced the exponents  $\alpha \sim 0.46 \pm 0.05$  and  $\beta \sim 0.36 \pm 0.06$ . Cell dynamical simulation of the TDGL equation for a scalar 2D  $(\Phi^2)^2$  model by Sengupta, Marathe, and Puri [4] yielded  $\alpha \sim 0.47 \pm 0.02$  and  $\beta \sim 0.40 \pm 0.01$  in accordance with Lo and Pelcovits. For the large- $N$  model, qualitative analysis by Dhar and Thomas [5] gave  $\alpha = \beta = \frac{1}{2}$ . Using the singular perturbation method, Somoza and Desai [6] showed analytically that for small field the full dynamical evolution of the order parameter, not only the exponents, is universal; this is independent of the particular form of the free energy and  $N$  for  $N \geq 2$ . Their results are based on the instability in the transverse direction of the system under the application of the external field opposed to the magnetization. For small field amplitude and frequencies, sinusoidal and sawtooth varying fields give rise to the same exponents  $\frac{1}{2}$ , which have also been confirmed by direct numerical integration [5,6] of the same equation as used by Rao and co-workers [1,2].

In addition to the spin systems, nonequilibrium transitions also involve time-dependent bifurcation parameters driving the systems to switch between the bistable states. The evolution of some of these systems, including nonequilibrium chemical reactions and laser, can be reduced to a single mode governed by a dynamic equation corresponding to the mean-field model of the spin systems.

For small frequencies of the driving field, the solution of the equation by Jung and co-workers [7] gave the exponents  $\alpha = \beta = \frac{2}{3}$ , different from Rao and co-workers, which has been a subject of debate [8].

Besides magnetic hysteresis, thermal hysteresis has also been investigated preliminarily by Rao and Pandit [2] in the  $(\Phi^2)^3$  model. The areas of the loops, as measured in the coordinate system of magnetic moment  $M$  and the reduced temperature  $r$ , also scale with the amplitude for low values of the periodically oscillating temperature, but with an exponent equal to one [2]. Although they claimed that their results were consistent with existing experimental data on ferroelectrics and charge-density waves, the fingerlike shape of their thermal hysteresis loops did not resemble at all any familiar spindle-shaped ones. The hysteresis loops saturate only in the paramagnetic phase, but are rounded in the ferromagnetic one. Moreover, the areas of the loops in the  $M$ - $r$  frame have no direct physical meaning.

Experimentally, a more easily realizable way of varying the temperature is by changing it linearly, rather than periodically. A common realization is by thermal analysis, the distinctive mode of which, among others, is changing temperature linearly with prescribed rates. These nonisothermal methods have the advantage of rapidity and effectiveness for those too slow and rapid transient transformations. A long standing question of these methods, however, concerns the rate of temperature scanning [9]. As hysteresis is frequently observed, nonequilibrium prevails and dissipation always comes into play. Consequently, hysteresis should depend on the rate of the process. As no work from basic principles, to our knowledge, has dealt with the relationship between hysteresis and temperature scanning rate, we address it here with a twofold purpose from prototype models familiar in the dynamics of first-order transitions, to see whether these models can tackle this problem and to provide theoretical insight for the experiment.

Thus, we will only concentrate on the case of linearly varying temperature. We show that the same  $(\Phi^2)^3$  model can give the familiar hysteresis loops. Furthermore, in the plane of the reduced temperature and its conjugate

variable, the areas of the hysteresis loops, a direct measure of energy dissipation per sweep cycle, scale with the rate of temperature varying  $R$  as

$$A = A_0 + aR^n, \quad (2)$$

where  $n$  approaches two-thirds and  $A_0$  and  $a$  are constants. The scaling is universal for the thermal hysteresis loops studied here.

As a preliminary, we consider first the mean-field models that have temperature driven transitions. Consider a general Landau free-energy

$$F - F_0 = \frac{1}{2}rM^2 + \frac{1}{3}bM^3 + \frac{1}{4}uM^4 + \frac{1}{5}cM^5 + \frac{1}{6}vM^6 + \dots + HM, \quad (3)$$

where, as usual,  $r$  stands for the distance from the mean-field temperature:  $r = a'(T - T_c)$ ,  $a'$ ,  $b$ ,  $c$ ,  $u$ , and  $v$  constants.  $H$  is the conjugate field, and  $F_0$  is the background free-energy. When no inversion symmetry exists, the first three terms in the expansion with  $b < 0$  suffice for a first-order temperature driven transition ( $M^3$  model). Otherwise  $b = c = 0$  and expansion to the sixth order is needed ( $M^6$  model). Dynamics is governed by the purely relaxational TDGL equation without noise. Generic numerical results of the  $M^6$  model with  $H = 0.1$  are shown in Fig. 1 for several temperature change rates. Also shown in the figure are the equation of state with different value of  $H$  (solid lines) and the spinodal points (dashed lines). It is clear that the temperature driven transitions with hysteresis take place when  $H < H_c$ , the critical point.

To obtain dissipation during one cycle, like the enclosed areas of magnetic hysteresis loops, we have to switch from moment versus temperature hysteresis loops to the reduced temperature  $r$  and its conjugate variable, so that from thermodynamic relations the area enclosed represents free-energy dissipation per cycle. Here the variable  $s$  conjugate to  $r$ , due to ordering is simply

$$s = - \left. \frac{\partial(F - F_0)}{\partial r} \right|_M = -\frac{1}{2}M^2. \quad (4)$$

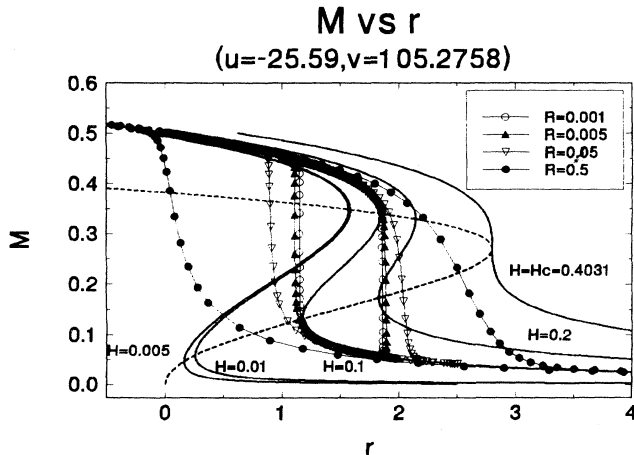


FIG. 1. Magnetic moment  $M$  vs reduced temperature  $r$ . Solid lines, state equation  $H = rM + uM^3 + vM^5$  for different  $H$  indicated; dashed line, spinodal points; thin lines with symbols, dynamic curves for different scanning rate  $R$  with  $H = 0.1$ .

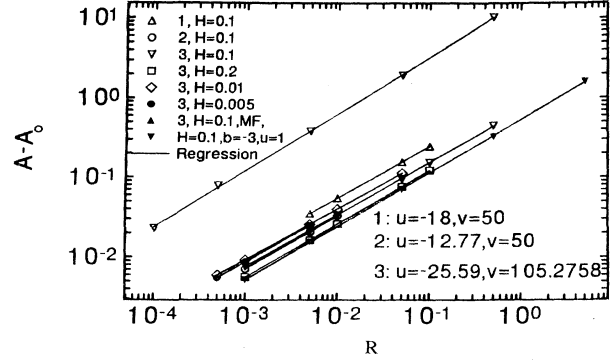


FIG. 2. Log-log plot of dissipation  $A$  vs temperature scanning rate  $R$ . Numbers in the upper-right legend after the symbols represent different choices of parameters given in the lower-left legend. MF means mean-field result of  $M^6$  model.  $M^3$  model is represented by the parameters  $b = -3$ ,  $u = 1$ , and  $H = 0.1$ . Note that the constant term  $A_0$  has been subtracted to give straight lines.

Dissipation obtained in this way scales with the rate of temperature sweeping as Eq. (2), with  $A_0$  the area formed by the static curve between the spinodal points and  $n$  equal to two-thirds, as shown in Fig. 2. The result of the  $M^3$  model is also given and also manifests similar behavior.

This scaling behavior can readily be confirmed following the theory of Jung and co-workers [7], although it is  $r$ , not the field  $H$ , that is varied. Since the transitions take place near the spinodal point, we can expand  $s$  and  $M$  at it, resulting in a Riccati equation, second order in  $s$  and  $M$ . This leads to the scaling behavior obtained numerically here, which is universal both for  $M^3$  and  $M^6$  models.

Having considered the mean-field models, we now proceed to the field theoretic  $O(N)$ -symmetric  $(\Phi^2)^3$  model with  $N$  approaching infinity, described by the Ginzburg-Landau free-energy functional

$$F[\Phi] = \frac{1}{2} \int d^d x \left[ (\nabla \Phi)^2 + r\Phi^2 + \frac{u}{2N} (\Phi^2)^2 + \frac{v}{3N^2} (\Phi^2)^3 - 2\sqrt{N} H \cdot \Phi \right]. \quad (5)$$

This is the same model as used by Rao and Pandit [2]. In the limit  $N \rightarrow \infty$ , the dynamic equations of the system are reduced to a set of integro-differential equations that can be solved numerically. Given appropriate parameters  $u$ ,  $v$ , and  $H$ , to obtain hysteresis loop under linearly varying temperature of rate  $R$ , we start from an appropriate initial condition and drive the system to the paramagnetic phase, then cool to the ferromagnetic one and finally heat back to reach a closed cycle. The initial condition is irrelevant to the results: Steady state can be easily reached from an arbitrary initial condition. Also irrelevant is the direction of changing temperature. Generic hysteresis loops are shown in Fig. 3. It is found that as  $H$  becomes smaller, the hysteresis loops become more asymmetric

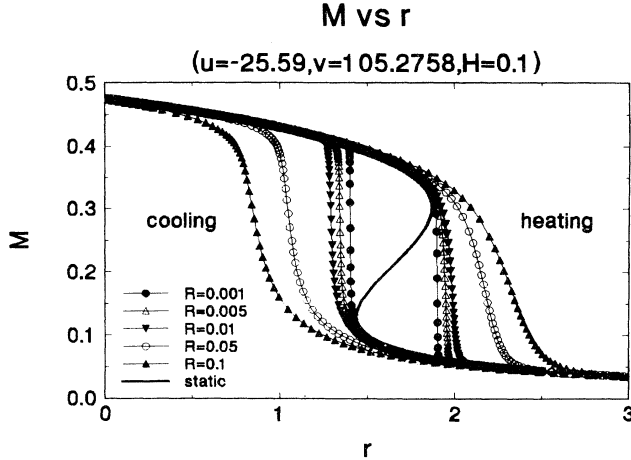


FIG. 3. Generic hysteresis loops of magnetization  $M$  vs temperature  $r$  for different temperature scanning rates  $R$ .

and shift to lower temperatures. This is the reason why saturation in the order phase could not be obtained in the previous work.

It can be seen that, like the mean-field cases, the transition temperature has to go beyond a certain spinodal point as indicated by the static curve. This is different from the magnetic hysteresis loops that can go inside the spinodal point. This essentially different feature results from the nature of the equilibrium first-order phase transition point. In the magnetic hysteresis, there are continuous paths, i.e., the transverse instability [5], due to the continuum symmetry, connecting the two phases with opposite magnetization, which are degenerate when no external magnetic field is applied. In the thermal hysteresis loops considered here, no such paths exist to circumvent the barrier; the transition can only take place at the spinodal point, where the barrier vanishes, as in the case of mean-field theories. Therefore, it is expected that the thermal hysteresis here belongs to the class of Eq. (2), with a finite zero-rate area  $A_0$  like the mean-field case above.

In order to obtain the dissipation per cycle, note that the equilibrium distribution takes the form  $\exp(-F)$ ; accordingly, the variable  $s$ , conjugate to  $r$  is given by

$$s = \frac{\partial \ln Z}{\partial r} = -\frac{1}{2Z} \sum_{\phi} \Phi^2 \exp(-F), \quad (6)$$

where  $Z$  is the partition function  $\sum \exp(-F)$ . Therefore, in equilibrium each component has

$$s = -\frac{1}{2}(M^2 + \hat{S}), \quad (7)$$

with  $\hat{S}$  the integrated structure factor of the transverse correlation function  $C_{\perp}(q, t)$  [2], i.e.,

$$\hat{S} = \frac{1}{2\pi^2} \int_0^1 dq q^2 C_{\perp}(q, t). \quad (8)$$

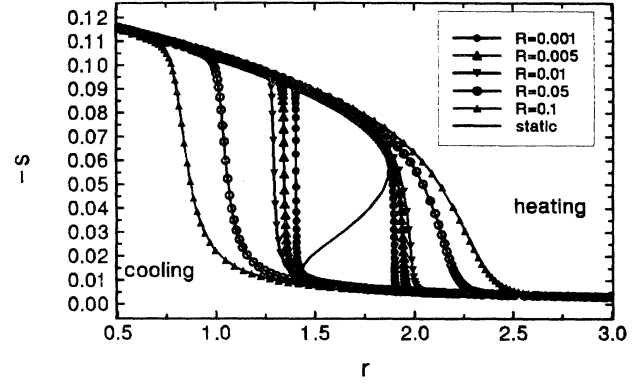


FIG. 4.  $s$  vs  $r$  of the generic hysteresis loops in Fig. 3. Note that  $s$  is given in  $-s$ .

We extend to nonequilibrium this definition of  $s$ , as it reduces naturally to the mean-field result, since  $\hat{S}=0$  when fluctuations are ignored.

The results of  $s$  versus  $r$  are shown in Fig. 4 for those loops in Fig. 3. Areas of the hysteresis loops increase with the rate of temperature scanning as expected, and scale as Eq. (2) like the mean-field results: This is different from the magnetic hysteresis of the same model, albeit fluctuations have been included. The scaling for various choices of parameters is presented in Fig. 2. It shows clearly that all have nearly the same slopes of two-thirds. It should be emphasized that exponents for the scaling with the rate of the areas of loops in the  $M$ - $r$  frame, however, varied with the external magnetic field applied. Therefore, our characterization of hysteresis loops is more meaningful and fruitful.

In summary, we have attained familiar thermal hysteresis loops under linearly varying temperature on the basis of the time-dependent Ginzburg-Landau theory of a  $(\Phi^2)^3$  model with continual symmetry. We have also cast the hysteresis loops in the frame of the reduced temperature and its conjugate variable, so that the enclosed areas of the loops represent directly the dissipation in the cycles. The dissipation so obtained increases with the scanning rate of temperature and exhibits a scaling of Eq. (2) with an exponent equal to two-thirds and a finite zero-rate dissipation for both mean-field and field theories. Furthermore, the exponent is independent of the applied field, which is an important consequence of our representation of hysteresis loops, and makes the results more meaningful. This scaling is different from the magnetic hysteresis one that has no  $A_0$  and one-half power exponent [5,6]. The difference results from the nature of the equilibrium transition point.

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