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Phenomenological framework for fluctuations around steady state

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A phenomenological framework to describe fluctuations around steady states is formulated. The framework is illustrated for a magnetic system maintained at a nonequilibrium steady state by an oscillating magnetic field, modeled at the mesoscopic level by a Langevin dynamics. The large deviation formalism along the time axis is employed to construct a generalized entropy to describe the fluctuations in the steady state for time averaged observables (state variables). We propose a phenomenological postulate that the fluctuations about the steady state can be obtained from the response of the state variables to ''thermodynamic conjugate forces'' (fluctuation-response relation), as in the ordinary thermodynamic fluctuation theory. An experimentally realizable method to study the linear response about the steady state against state variable perturbations is proposed, and illustrated for the driven magnetic system. The notion of a proper state space to describe nonequilibrium steady states is discussed, and to this end, we introduce a dissipation variable to extend the state space for our model system. In the extended state space, we elucidate and study various stability and Maxwell-type relations that follow from our *local* phenomenological (thermodynamic) framework. Some relevant issues regarding a more general thermodynamic framework are also discussed. [S1063-651X(97)04301-8]

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I. INTRODUCTION

There has been little progress in establishing general phenomenological principles for systems away from equilibrium, even if they are in steady states. There is no accepted nonequilibrium thermodynamic framework for steady states tantamount to equilibrium thermodynamics. The theory of linear irreversible thermodynamics (LIT) [1] can cover a wide class of physical phenomena that on a global scale can still be far from equilibrium, but it cannot cope with systems far away from (local) equilibrium, especially systems that are locally driven by large external forces, such as the one studied in this work. The situation does not improve very much with some attempts to extend the phenomenology to nonlinear regimes [2].

In this paper we construct a phenomenological framework to describe fluctuations around steady states. Here we mean by the phenomenological framework a theoretical framework that describes a system in terms of a few (coarse-grained) state variables and which introduces the (generalized) deviational (or fluctuational) entropy to describe the fluctuation distribution. As can be seen from the thermodynamic fluctuation theory around equilibrium states [3], phenomenological fluctuation theory should shed some light on the phenomenological (or thermodynamic) framework (if any) to describe macroscopic and/or long time behaviors of steady states. This is the main motivation to study the fluctuations in a driven system in this paper. We analyze a model of a magnetic system that is driven by a large oscillating magnetic field to illustrate the phenomenological framework.

Macroscopic or phenomenological variables are defined by an appropriate averaging. Depending on the averaging method, we have different phenomenological frameworks (even for equilibrium systems). Equilibrium thermodynamics chooses ensemble averaging. In this paper, we choose time averaging for technical simplicity. Our model system is periodic in the steady state, so defining our (macroscopic) observables as time-averaged quantities is natural. Such an approach is now standard in the thermodynamic formalism of dynamical systems initiated by Sinai [4–8]. Furthermore, if a system consists of a few small systems, as in the case of nanobiological examples, time averaging is perhaps the only realistic method to study its long time behavior phenomenologically. A phenomenological framework for ensemble averaged fluctuations will be discussed in a subsequent paper.

Just as equilibrium thermodynamic quantities of a small volume fluctuate, time averaged observables deviate (that is, fluctuate) from their long-time averages when they are considered on a small time span. Equilibrium thermodynamic fluctuation theory postulates a phenomenological postulate (the Boltzmann-Einstein relation), which allows us to relate fluctuations and responses of the equilibrium states to perturbations (the fluctuation-response relation). The postulate is empirically amply verified and is a natural consequence of statistical thermodynamics.

The fluctuation-response relation analogous to that in equilibrium holds in the steady states of our model driven system. The validity of the fluctuation-response relation allows us to study Maxwell's relations and Le Chatelier-Braun principles, etc. The reader may doubt the existence of a general phenomenological framework for steady states. We argue that the state space for arbitrary nonequilibrium states of a system is not finite dimensional (even in the steady state) in general. Therefore, a general theory for any nonequilibrium steady state, even if exists, would be too general to be useful. However, our results suggest that if we restrict our attention to *simpler* systems, the existence of a phenomenological (or thermodynamic) framework describing them is conceivable.

There are dynamical and kinetic approaches to study fluctuations around nonequilibrium steady states [9–11]. Some of these theories are beyond local equilibrium assumptions, and quite general, but still the distance from local equilibrium cannot be large due to its perturbative nature. Our ultimate aim is to relate macroscopically experimentally observable quantities with each other as in the standard equilibrium thermodynamics. As far as macro-observables are definable (as, e.g., time averaged quantities), no particular closeness to equilibrium is required.

The paper is organized as follows. In Sec. II, after a summary discussion of relevant backgrounds, the generalized fluctuation entropy function is introduced to characterize the fluctuations around the steady state. Its existence is guaranteed by large deviation theory, which allows us to construct the entropy function from the nonequilibrium entropy for Langevin equations, which was studied long ago by Graham [12]. Our model system, a coarse-grained description of a uniform magnetic particle under an oscillating magnetic field, is introduced in Sec. III. Its generalized fluctuation entropy is computationally constructed in Sec. IV. The model system can be thought of as a model of a magnetic domain, or a small magnetic particle, dispersed in an inert solid under an oscillating magnetic field. Both should be realized experimentally without difficulty. In Sec. V, we discuss the linear response about the steady state. This furnishes an experimentally accessible method to observe the generalized entropy function for fluctuations. The dissipation variable and its relation to the state space for the driven system are discussed in Sec. VI A. Various susceptibilities are introduced in Sec. VI B, followed by a discussion of Maxwell relations and Le Chatelier Braun principles in Sec. VI C. Section VII discusses the relevance of the asymptotically computed generalized entropy to nonasymptotic realistic situations (finite observation time, not small noise). We summarize our results in Sec. VIII, which also contains some discussions on the prerequisite to construct the full thermodynamic theory.

II. LARGE DEVIATION FRAMEWORK

The fundamental relation for the probabilities of fluctuations in equilibrium statistical thermodynamics is the Boltzmann-Einstein relation [13]

$$P \sim \exp\left[\frac{V}{k_B}\,\delta s\right], \tag{2.1}$$

where δs is the increase of entropy density (note that $\delta s \leqslant 0$) due to fluctuation, V is the volume where fluctuations occur, and k_B is the Boltzmann constant. The above relation is an example of a large deviation (LD) principle as pointed out by Lanford [14]; an elementary discussion of the principle is given below for convenience. The LD principle is the mathematical essence of the probability-entropy relation for equilibrium fluctuations (however, see the comments below).

A dynamical analog of the relation (2.1) for linear irreversible processes *close* to global equilibrium was introduced by Hashitsume [15] and also by Onsager and Machlup [16]. Graham later generalized the theory for arbitrary nonlinear processes far from equilibrium [12]. A general LD theoretical interpretation of these nonequilibrium generalizations was proposed in [17]. Markov processes satisfy a LD principle [7,18,20,29]. Hence a wide class of nonequilibrium systems that are modeled as Markov processes can have a statistical framework analogous to the Boltzmann-Einstein relation for time-averaged quantities. For spatially extended systems in local equilibrium, Eyink [19] recognized the relation of Graham's work to (ensemble theoretical) LD, and proposed a minimum excess work principle for the probability of spontaneous fluctuations in the ensemble theory of steady states. An analogous principle has been observed by Ross and co-workers [21].

A LD principle arises, e.g., in the following simple context [20]. Consider N independent and identically distributed (iid) random variables X_i, $i=1,\ldots,N$. Let the mean $\langle X_i\rangle = m$, and define the empirical average as $y_N = N^{-1}\sum_{i=1}^{N} X_i$. The weak law of large numbers [22] tells us (if, for example, $\langle X_i^2\rangle$ is finite) that $y_N \to m$ as $N\to\infty$ in probability. Cramér [24] proved the following refinement of the law:

$$P(y_N \in A) \sim \exp[-NI(A)] \quad \text{as} \quad N\to\infty, \tag{2.2}$$

where $I(A) = \inf_{x\in A} I(x)$. The function $I(x)$ is called the *rate function* (generalized entropy function) or the Cramér function (as proposed by Frisch [25]), which is a non-negative convex function with global minimum at $x=m$ [i.e., $I(m) = 0$]. Roughly speaking, we say the LD principle holds for X_i if (2.2) is valid.

Thus the relation of (2.1) and the LD principle for space average (which is actually the ensemble average for macroscopic systems) is explicit. However, it should be clearly recognized that purely phenomenologically, we cannot assert that the rate function for equilibrium fluctuations can be computed with the aid of equilibrium thermodynamics as (negative) deviational entropy δs, because the underlying statistical model (equilibrium statistical thermodynamics) is lacking. Therefore, (2.1) is a phenomenological postulate [3].

There is an analogous theorem (Sanov's theorem [26]) for the empirical distribution of y_N (the so-called level-2 LD principle [18,20]). In this case the LD principle reads

$$P(f\in B) \sim \exp[-NI(B)] \quad \text{as} \quad N\to\infty, \tag{2.3}$$

where $I(B) = \inf_{f\in B} I(f)$. The rate function(al) $I(f)$ is given by the Kullback-Leibler entropy

$$I(f) = \int dx\; f \ln\left(\frac{f}{f_0}\right), \tag{2.4}$$

where f_0 is the true (density) distribution function.

Consider a stochastic field $\phi(x,t)$ with a sampling measure W on the path space (i.e., the probability of a bundle (or, more precisely, a cylinder set) A of paths (histories) is given by $W([A])$. A functional version of Sanov's theorem tells us that the LD rate function(al) I characterizing the fluctuations of the path probability P (i.e., the empirical probability of a bundle of paths A, $P[A]$) is given by

$$I[P] = \mathrm{tr}\; P\; \ln(P/W), \tag{2.5}$$

where tr (trace) denotes the sum over all the histories of the process.

Suppose the stochastic process is described by the Langevin equation of the form

$$\dot{\phi}(x,t) = b(\phi,t) + \epsilon\sigma(\phi)\,\eta(x,t), \tag{2.6}$$

where $\phi(x,t)$ is a stochastic (vector) field, $b(\phi,t)$ a time-dependent (vector-valued) function of $\phi(x,t)$, $\sigma(\phi)$ a (matrix-valued) function, $\eta(x,t)$ the zero mean Gaussian white noise with $\langle \eta(x,t)\eta(x',s)\rangle = \epsilon^2\delta(t-s)\delta(x-x')$, and the parameter ϵ is the overall strength of the noise. The true distribution function(al) is known to be asymptotically in the small ϵ limit [12,27,28]

$$W[\phi(\,)] \approx \exp\left[-\frac{1}{\epsilon^2}S_0 + S_1 + O[\epsilon^2]\right], \tag{2.7}$$

where $\phi(\,)$ denotes a (space-time) history (path) of the process defined by the Langevin equation, $S_1 = \int \partial_{x_i} b_i[\phi(x,t)]dx\,dt$, and

$$S_0 = \frac{1}{2}\int \sum_{ij} a_{ij}\{\dot{\phi}_i(\,) - b_i[\phi(\,)]\}\{\dot{\phi}_j(\,) - b_j[\phi(\,)]\}dx\,dt, \tag{2.8}$$

with $a_{ij} = (\Sigma_k \sigma_{ik}\sigma_{kj})^{-1}$. The rate function for an observable $\langle f(\phi(\,))\rangle_c$, where $\langle\,\rangle_c$ denotes a functional of f [which may be the mere expectation value of f, or may become another function of (x,t)], is obtained by minimizing $I[P]$ over all P under the constraint $\mathrm{tr}\{P[\phi(\,)]f(\phi(\,))\} = \langle f(\phi(\,))\rangle_c(x,t)$. The result is the rate function, given as the Legendre transform of the generating functional $\Psi[\lambda(\,)]$:

$$I[\langle f(\phi(\,))\rangle_c] = \Psi[\lambda(\,)] - \int dx\,dt\,\lambda(x,t)\langle f(\phi(\,))\rangle_c(x,t), \tag{2.9}$$

where

$$\Psi[\lambda(\,)] = -\ln \mathrm{tr}\exp\left[-\frac{1}{\epsilon^2}S_0 + S_1 + O[\epsilon^2] - \int dx\,dt\,\lambda(x,t)\langle f(\phi(\,))\rangle_c(x,t)\right] \tag{2.10}$$

and $\lambda(x,t)$ is determined by

$$\frac{\delta}{\delta\lambda(x,t)}\Psi[\lambda(\,)] = \langle f(\phi(\,))\rangle_c(x,t). \tag{2.11}$$

If spatial fluctuation may be ignored, we may suppress the spatial coordinates in the above formalism. For time-dependent steady states (e.g., periodic states in our system) of a small particle, it is natural to characterize its state with time-averaged quantities. Hence, it is also natural to study the fluctuations of time-averaged quantities. In this case the Lagrange multiplier function λ becomes a constant. If we are interested in small noise systems, we may rely on the lowest order result of Eq. (2.7) (a saddle point approximation for the limit $\epsilon \to 0$ to evaluate rate functions). Thus the formulas corresponding to Eq. (2.9) read in the large observation time τ limit

$$I(\overline{f}) = \frac{1}{\tau}\inf\left[S_0 + \lambda\int_0^\tau dt\, f(\phi(t))\right] - \lambda\overline{f}, \tag{2.12}$$

$$P(\overline{f}) \approx \exp\left[-\frac{\tau}{\epsilon^2}I(\overline{f})\right], \tag{2.13}$$

where $\overline{f}$ denotes the time average of the observable over τ, $\overline{f} = \tau^{-1}\int_0^\tau dt\, f(\phi(t))$. The computation of the generalized entropy I for a fluctuation $\overline{f}$ involves minimizing the action $S_0(\phi(t))$ over all paths $\phi(t)$ under the constraint $\overline{f(\phi(t))} = \overline{f}$. The *optimal* path (or path of least action) for a given λ satisfies the Euler-Lagrange equation

$$\frac{d}{dt}\partial_{\dot{\phi}}L = \partial_\phi L, \tag{2.14}$$

where

$$L = \frac{1}{2}\sum_{ij} a_{ij}[\dot{\phi}_i(t) - b_i(\phi(t))][\dot{\phi}_j(t) - b_j(\phi(t))] + \lambda\overline{f(\phi(t))}. \tag{2.15}$$

The rate function I is usually referred to as an entropy function(al) for the system. However, the word must be understood just as used in the thermodynamic theory of fluctuation. As is clearly explained in Callen [3] and above, the entropy governing fluctuations is *not* interpreted as a state function defined on the thermodynamic state space without a phenomenological postulate, although statistical thermodynamics justifies this postulate. Without any postulate we cannot directly construct equilibrium thermodynamics purely phenomenologically from the study of equilibrium fluctuations alone [30]. As was first noted by Takahashi [7], the so-called thermodynamic formalism for dynamical systems [4–6] is a LD principle for time-averaged quantities. We must clearly recognize that the so-called thermodynamic formalism for dynamical systems has been used only to describe fluctuations, and not to understand the difference between different dynamical systems.

III. A MODEL SYSTEM

A driven system for which we wish to apply a general theoretical framework is a magnetic spin system under the influence of a time-dependent magnetic field. The system was considered by Zimmer [32] using a time-dependent Ginzburg-Landau equation to model the dynamics of the spatially coarse-grained magnetization. We study the model at the mean field level, but with a small noise. The model takes the following form:

$$\dot{\phi} = -\gamma_0[-r_0\phi + u_0\phi^3 + h_0\cos(\omega t)] + \epsilon\eta(t), \tag{3.1}$$

where the field $\phi(t)$ denotes the (space-averaged) magnetization, $h(t) = h_0\cos(\omega t)$ is the external magnetic field, u_0 is a positive constant, γ_0 is the kinetic coefficient, and r_0 is the temperature parameter [proportional to $(T_c - T)$ with T_c being the unrenormalized critical temperature, if T is close to T_c; constant if T is sufficiently away from T_c]. $\eta(t)$ is a white noise source $\langle\eta(t)\rangle = 0$, $\langle\eta(t)\eta(t')\rangle = \epsilon^2\delta(t-t')$.

If the spatial average of the magnetization is taken over an infinite volume, then there should not be any noise. We assume that our system is not (macroscopically) very large. Therefore, there is a residual noise. In the following we rescale the field ϕ and time to express the dynamics in terms of the minimal set of parameters, $r = r_0\gamma_0/\omega$, $h = h_0(u_0\gamma_0^3/\omega^3)^{1/2}$:

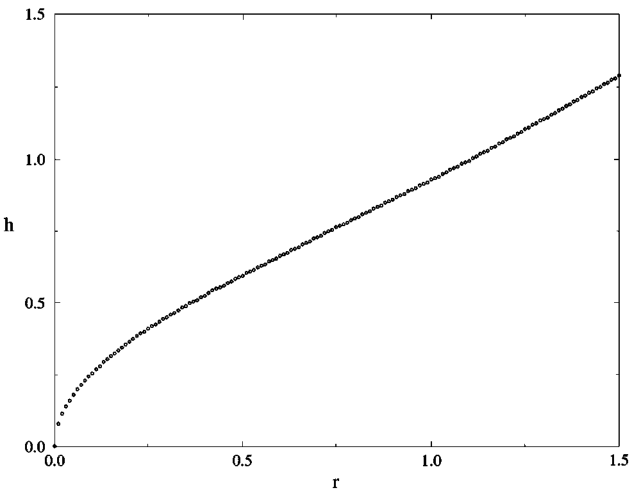


FIG. 1. Plot of the zero-noise phase boundary separating the $M=0$ (Z phase) and the $M\neq 0$ (NZ phase) phases. The upper region (i.e., large h) is the Z phase, and the lower region the NZ phase. The transition is continuous throughout the phase diagram.

$$\dot{\phi}=r\phi-\phi^3+h\cos(t)+\epsilon\left(\frac{u_0\gamma_0}{\omega^3}\right)^{1/2}\eta(t). \tag{3.2}$$

Notice that the noise strength effectively decreases with increasing frequency, so that the small noise theory [i.e., the saddle point approximation used to obtain Eq. (2.12)] should be more accurate for larger ω. For $2\pi/\omega$ smaller than the typical relaxation time of the locally course-grained magnetization, the system cannot (locally) equilibrate over the time scale of the field. Consequently, the system is intrinsically away from local equilibrium, and there is no phenomenology available.

In the zero noise limit, the system undergoes a phase transition from a zero time-averaged magnetization phase (Z phase) to a nonzero time-averaged magnetization phase (NZ phase). Its phase diagram is illustrated in Fig. 1 for convenience [33]. The transition between the Z and the NZ phases is *a continuous* (second order) phase transition for all field amplitudes. For $r<0$ (i.e., $T>T_c$) there is only the Z phase.

IV. GENERALIZED FLUCTUATION ENTROPY FOR TIME-AVERAGED MAGNETIZATION

We wish to study the probability of fluctuation around the steady state of the driven magnetic model (3.2). Here we mean by ''steady state'' any state that has well-defined long-time averages. Steady states of our model are periodic. In the small noise limit, the rate function for the long-time average magnetization of Eq. (3.2) is given by [see Eqs. (2.8) and (2.12)]

$$I(\overline{M})=\frac{\omega^3}{\gamma_0 u_0}\frac{1}{2\pi}\inf_{\overline{M}} S_0, \tag{4.1}$$

where $\inf_{\overline{M}}$ implies to search the infimum over $\phi(t)$ whose time average is equal to M, and

$$S_0=\int_0^{2\pi}dt\,\frac{[\dot{\phi}-r\phi+\phi^3-h\cos(t)]^2}{2}. \tag{4.2}$$

In this calculation, for simplicity, we look for the infimum among the set of functions periodic in 2π. This does not give the true infimum, but the discrepancy for not extremely large deviations is very small. This is the reason why the integration range of Eq. (4.2) is $(0,2\pi)$. The parameters ω,γ_0,u_0 are set to unity in what follows.

For $h=0$ (i.e., the equilibrium state) the infimum is realized by a constant path $\phi(t)=M$, yielding the rate function $I(M)=(rM-M^3)^2/2$ in the Z phase, and in the NZ phase outside the coexistence region (see below). For nonzero driving fields, the Euler-Lagrange equation for the optimal path cannot be analytically solved. One way may be to solve the equation numerically, but here a direct method is used, a deterministic steepest descent method [23]. The field $\phi(t)$ ($t\in[0,2\pi]$) was sampled along the time axis with a time spacing $\delta t=0.01$, and with periodic boundary conditions. From the most probable state [$\lambda=0$, $\dot{\phi}(t)=r\phi-\phi^3+h\cos(t)$], the Lagrange multiplier λ was slowly increased above zero. The optimal state $\phi(t)$ corresponding to the potential S_0 minimum for a given λ was then used as the initial state for the next trial with a slightly larger λ. The resultant rate function $I(M)$ is given in Fig. 2.

In the Z phase, the flatness of the rate function near its optimal value indicates the importance of correlation along the time axis. Thus the rate function becomes flattened as h moves from larger values toward its critical value $h_{cr}\simeq 0.93$. Compare $h=1.2$ and $h=0.975$. As in equilibrium, the rate function equal to the generalized entropy function, becomes flat at the symmetry breaking point.

Even in the symmetry-broken phase (in our case the NZ phase), I must have the convex shape analogous to the free energy of coexisting phases in equilibrium: $I=0$ in the coexistence region between the steady states (for example, $M\approx\pm 0.35$ for $h=0.9$ in Fig. 2). For ordinary Markov processes, such as in our case, the convexity of the rate function is guaranteed by the LD principle. However, just as the free energy in equilibrium statistical mechanics with a finite averaging volume, if the averaging time τ is not infinite, then the rate function for time-averaged quantities can be nonconvex. In the infinite time limit the optimal path for any fluctuation in the coexistence region involves (multi) instanton paths connecting the two attracting states $\pm M_0$, which yield zero action as $\tau\to\infty$. Note that the potential I here is computed in the limit $\epsilon\to 0$ first, and then $\tau\to\infty$. In this limit, the states $-M_0<M<M_0$ are realized as an average over an ensemble of multi-instanton paths; for a given sample we only observe one of the ''pure'' states $\pm M_0$. In the reverse limit, $\tau\to\infty$ first, then $\epsilon\to 0$, I would always have the unique minimum at the zero state.

As is discussed in the preceding paragraph, if the rate function is computed for not very long time, then it is not convex in the NZ phase, and behaves just as the local equilibrium free energy for the ferromagnetic phase. If the averaging time span τ is not significantly longer than the transition time to jump between the attractors, then we have a nonconvex short-time (local in time) rate function J resembling a Landau-type function with a nonconvex portion con-

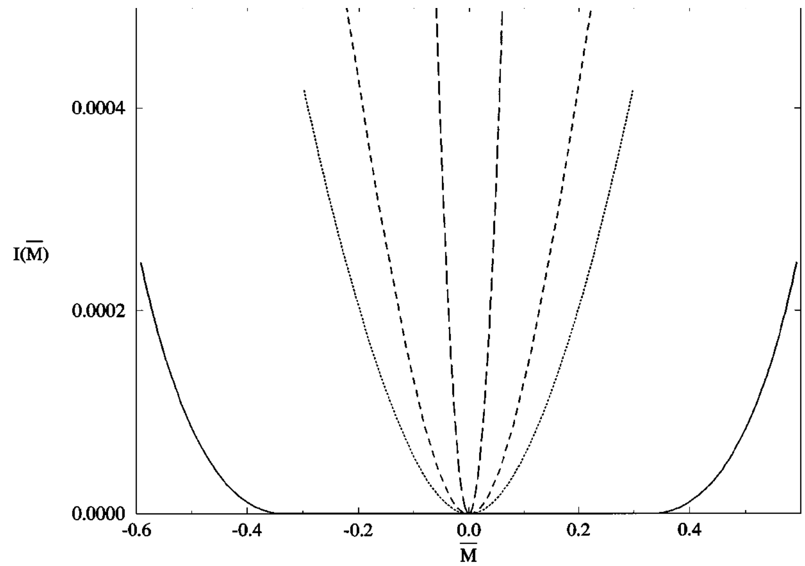


FIG. 2. $I(\overline{M})$ computed with a steepest-descent method, for $r=1$. The solid line is $h=0.9$, the dotted line $h=0.975$, the dashed line $h=1$, and the long dashed line $h=1.2$.

necting the two states $\pm\overline{M}$, but practically identical to the true rate function (for long-time) outside the coexistence region.

Let Δt be the averaging time to obtain the ''local'' rate function $J(m)$ for the ''order parameter'' m defined as the time average over Δt. Then, the probability of observing m is estimated as $P(m) \sim \exp[-\Delta t J(m)]$. We may consider $-\ln P(m)$ as an equivalent of the free energy (chemical potential), which governs the dynamics of m at the time scale of Δt. Hence, just as in the modeling of dynamics, as the (time-dependent) Ginzburg-Landau equation, we may write

$$m(t+\Delta t)-m(t) \propto \frac{\partial}{\partial m} \ln P(m) = -\Delta t \frac{\partial}{\partial m} J(m), \tag{4.3}$$

which has an indistinguishable form from the usual (near) equilibrium Ginzburg-Landau equation based on the local free energy (under local equilibrium assumption). Here the proportionality constant is related to the noise level, i.e., an analogue of the fluctuation-dissipation theorem holds.

When a coarse-grained model is used in statistical mechanics to describe nonequilibrium phenomena such as phase transition kinetics [34], often a question is raised whether we can use the (Landau-Ginzburg type) ''free energy density'' in nonequilibrium cases in which the deviation from equilibrium is sometimes not necessarily small. The above consideration tells us that the so-called Landau-Ginzburg free energy density need not be interpreted as the conventional free energy density. It is legitimate to use this type of function in much wider contexts as long as we study (space-time) coarse-grained observables.

V. FLUCTUATION-RESPONSE RELATION

Suppose an actual small magnetic particle is given. Experimentally, it is not easy to observe the rate function. This is true even in the computer simulation of the model being discussed here. The difficulty of experimentally obtaining rate functions for large fluctuations was clearly recognized in [17]. However, if an analogue of thermodynamic fluctuation theory is valid, then we can experimentally easily determine the rate function at least for small fluctuations.

In equilibrium states, statistical thermodynamics tells us that the susceptibility is related to the variance of equilibrium fluctuations as

$$\langle \delta X_i \delta X_j \rangle = -k_B \left.\frac{\partial X_i}{\partial F_j}\right|_{\{F_k\}}, \tag{5.1}$$

where $\{X_i\}$ is the set of fluctuating extensive quantities, $\{F_i\}$ the set of corresponding thermodynamically conjugate intensive quantities with respect to entropy [3], and the subscripts $\{F_k\}$ indicate that all intensive parameters except F_j are held constant. The fluctuation contribution to the entropy change can be written as

$$-\delta S = \frac{1}{2} \sum s_{ij} \delta X_i \delta X_j , \tag{5.2}$$

where s_{ij} can be computed thermodynamically according to Eq. (5.1). The essence of the phenomenological postulate of equilibrium fluctuation theory is that the Taylor expansion coefficients of the LD rate function for fluctuations are determined by the thermodynamic responses of the equilibrium state around which fluctuations are being studied.

Mimicking this equilibrium postulate, we formulate our phenomenological postulate as follows. The Taylor expansion coefficients of the rate function around the mean can be obtained from the response of the phenomenological variables to ''thermodynamic'' conjugate forces. Implicit in the postulate is the existence of a proper state space for the steady states (about which we give a detailed discussion in Sec. VI). The postulate guarantees that at least locally in the state space, we have a generalized entropy Σ and its variation $\delta\Sigma$ around a given steady state is the (negative) rate function. That the susceptibility can be determined by a response is, as we shall see soon, certainly correct. However, the postulate also requires that the conjugate force that elicits the correct response does not perturb the system out of the state space. The latter condition is less trivial, and will be discussed in Sec. VI.

Let us analyze the process of computing the rate function I for the time average of $f(\phi)$, where ϕ obeys Eq. (3.2). The Euler-Lagrange equation for the variational problem $\inf_{\overline{f(\phi)}} S_0(\phi)$ with the Lagrange multiplier λ reads [we consider the simple case of additive noise with $\sigma=1$, and a scalar drift $b(\phi,t)$ in Eqs. (2.14) and (2.15), as in our model magnetic system]

$$\ddot{\phi}=b(\phi,t)\partial_\phi b(\phi,t)+\partial_t b(\phi,t)+\lambda\partial_\phi f(\phi). \quad (5.3)$$

One can rewrite this as two first order equations:

$$\dot{\phi}=b(\phi,t)+\lambda g, \quad (5.4)$$

$$\dot{g}=-g\partial_\phi b(\phi,t)+\partial_\phi f(\phi). \quad (5.5)$$

Since S_0 is the generalized entropy functional on the path space, a natural interpretation of λ is the (''thermodynamic'') conjugate force of the ''thermodynamic'' variable $\overline{f(\phi)}$. Note that $\lambda=0$ for the most probable path (steady state), so that we shall sometimes write $\lambda=\Delta\lambda$ to emphasize that λ is the external field for a *deviation* from the steady state (i.e., λ is a generalized affinity)

Looking at Eq. (5.4), we may interpret λg as the external force we can experimentally impose to produce the desired deviation. Since λg perturbs the steady state, the deviation corresponding to the one in Eq. (5.1) can be computed from the linear response of the most probable value $\overline{f(\phi)}$ to λg. Thus the postulate of the fluctuation-response relation is almost vacuously true for the Langevin model.

The shape of the rate function near the most probable value is quadratic, that is, the distribution of small fluctuations is roughly Gaussian (away from critical points). The quadratic approximation to the rate function is obtained easily from the linear response. The Euler-Lagrange equation, or equivalent Eqs. (5.4) and (5.5), linearized about the most probable path $\dot{\phi}_0(t)=b[\phi_0(t),t]$ reads

$$\Delta\dot{\phi}=\Delta\phi\partial_{\phi_0}b(\phi_0,t)+g, \quad (5.6)$$

$$\dot{g}=-g\partial_{\phi_0}b(\phi_0,t)+\partial_{\phi_0}f(\phi_0), \quad (5.7)$$

where $\phi=\phi_0+\lambda\Delta\phi$. We introduce χ through writing the rate function for $f\equiv\overline{f(\phi)}$ to the quadratic order as $I(f)=[f-\overline{f(\chi_0)}]^2/2\chi$. Notice that $\lambda=\Delta\lambda$ is the conjugate parameter for f when we compute the rate function. Hence, χ may be computed as the response of the system to a force $\Delta\lambda g(t)$, $\chi=-\lim_{\Delta\lambda\to 0}\Delta f/\Delta\lambda$, where Δf is the deviation of f due to the perturbation in Eq. (5.6).

In the limit of zero noise, $\partial_{\phi_0}b[\phi_0(t),t]$ is given once we know the model and consequently the attractor path $\phi_0(t)$. Operationally, however, we do not know (observe) the functional form of the model [i.e., $b(\phi_0,t)$]. Therefore, a more direct procedure accessible experimentally is desirable. We now outline this procedure to determine χ, which involves the following three steps.

(1) Determination of $\partial_{\phi_0}b[\phi_0(t),t]$: The direct method to obtain $\partial_{\phi_0}b[\phi_0(t),t]$ is to slightly perturb the system and measure the relaxation of the response $\Delta\phi(t)$. We accumulate results from many runs to obtain the average, which is very close to the most probable response when noise is small, as in our case. From this, we obtain

$$\partial_{\phi_0}b[\phi_0(t),t]=-\frac{d}{dt}\ln\left[\frac{\langle\Delta\phi(t)\rangle}{\langle\Delta\phi(0)\rangle}\right], \quad (5.8)$$

where $\langle\ \rangle$ denotes the average over all samples. Depending on the noise level in the experimental sample, one would have to perturb the system periodically, and measure the average response. Perturbing the system every couple of periods is sufficient here as we consider the case $r=r_0\gamma_0/\omega=1$, which implies that the time scale for the magnetization relaxation ($\approx r_0\gamma_0$) and the period of the external magnetic field are of the same order (in this example the period is 2π). Figure 3(a) exhibits a typical sample of $\phi(t)[=M(t)]$ for the system parameters $r=1$, $h=1$, and a noise level $\epsilon=0.05$. Upon perturbing the system (with a small force ≈ 0.001 in magnitude), the measurement of the relaxation yields $\langle\Delta M(t)\rangle/\langle\Delta M(t=0)\rangle$, illustrated in Fig. 3(b). The relaxation rate $\partial_{\phi_0}b[\phi_0(t),t]$ is then obtained easily from Eq. (5.8).

(2) Computation of $g(t)$: Given $\partial_{\phi_0}b[\phi_0(t),t]$, one has to solve Eq. (5.7) for $g(t)$. We need a steady $g(t)$ (that is, the long time solution). Notice that because of the negative sign in the linear term, the equation for $g(t)$ is intrinsically unstable. To obtain the steady state solution for $g(t)$, one can expand $g(t)$ (and $\partial_{\phi_0}b[\phi_0(t),t]$) in harmonics of the frequency of the magnetic field (here set to unity), and solve the resulting linear matrix equation. As mentioned earlier, for small fluctuations, restricting the infimum to periodic functions should be a very good approximation, so that the periodic ansatz to solve Eq. (5.7) is reasonable.

(3) Determination of the response: The obtained force $g(t)$ is scaled with λ and applied to the original system. The response Δf yielding $\chi=-\lim_{\Delta\lambda\to 0}\Delta f/\Delta\lambda$ is measured; or more directly, one can solve the linear equation (5.6) for $\Delta\phi(t)$, in the same manner as described in step (2), and obtain $\Delta f=\overline{\partial_{\phi_0}f(\phi_0)\Delta\phi(t)}$.

The zero noise susceptibility for the magnetization computed from the linear response theory agrees reasonably well (as expected) with the steepest-descent optimization results. The agreement improves as I becomes narrower, that is, as the time correlation diminishes.

In the NZ phase, we can study the fluctuation around a given phase, say, the up-spin phase. The rate function for small fluctuations becomes $I=\frac{1}{2}\chi^{-1}(M-M_0)^2$, where M_0 is the time average magnetization in the steady up-spin state. The susceptibility χ defined in this way both in the NZ and Z phases behaves as shown in Fig. 4. The magnetization susceptibility diverges at the transition point ($h\approx 0.93$). Near the transition $g(t)$ (and χ) becomes large, so the first order expansion is suspect to break down. Hence, the data near the phase transition $h=0.93$ are not likely to be very accurate.

In Fig. 5, we illustrate the difference between the ''thermodynamic'' susceptibility χ defined above and the usual response to a constant field $dM/dH|_{H\to 0}$. The discrepancy is quite large near the transition; note the log-log plot near the transition. Note that even in the case of zero oscillating field, the susceptibilities are different. This is due, of course, to the fact that we are considering the fluctuations of the time-

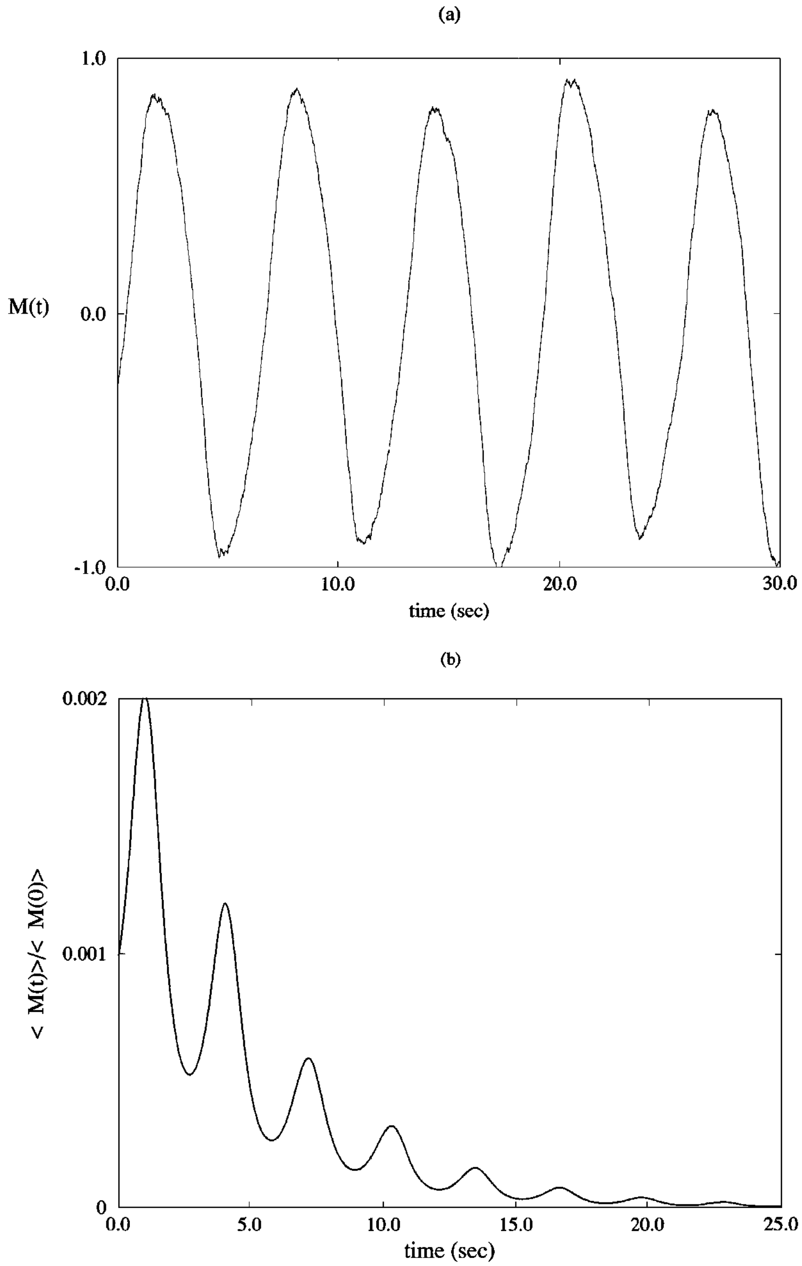


FIG. 3. For the system parameters $\gamma=\omega=u_0=r=h=1$: (a) a sample trajectory of $M(t)$ for a noise level $\epsilon=0.05$. From the average of $M(t)$ over many samples, 1000–4000 in this case, one obtains $\langle M(t)\rangle$ and (b) the response function $\langle \Delta M(t)\rangle / \langle \Delta M(t=0)\rangle$.

averaged magnetization. In the $h\to 0$ limit, $g=1/b'(M_0)=-1/2rM_0$, with M_0 being the equilibrium magnetization. The thermodynamic susceptibility in the zero oscillating field limit becomes $\chi=\{1/\partial_{\phi_0} b[\phi_0(t),t]\}^2$, the square of the usual *single-time* susceptibility. More generally, for a time-independent symmetric regression matrix $\partial_{\phi_j} b_i(\phi_0,t)\equiv K_{ij}$, and a constant diffusion matrix $a_{ij}=(\Sigma_k \sigma_{ik}\sigma_{kj})^{-1}=1$ [see Eq. (2.8)], the long-time susceptibility is given by the square of the single time susceptibility [35].

The fluctuation-response relation discussed above is for the fluctuation of time-averaged observables. The fluctuation-response relation in instantaneous observables can be studied analogously, although considering instantaneous quantities in the present context is somewhat less natural. For a given fluctuation at some time, e.g., $t=0$ [for this case $\lambda(t)=\lambda\,\delta(t)$], the solution for $g(t)$ at the linear level becomes $g(t)=-\partial_{\phi_0} f(\phi_0)|_{t=0}\ \exp\{-\int_0^t dt \partial_{\phi_0} b[\phi_0(t),t]\}$ for $t<0$ ($g=0$ for $t>0$). For gradient dynamics ($\dot{\phi}=-\partial_\phi H$), the application of $g(t)$ to produce the right fluctuation becomes superfluous; knowledge of the potential $H=I$, which defines the dynamics, automatically dictates the correct coupling. However, without knowledge of the steady state measure, we cannot prescribe the needed force to perturb the system. Therefore, one must proceed by measuring the (natural) relaxation $\partial_{\phi_0} b[\phi_0(t),t]$ to the steady state, and compute $g(t)$ via Eq. (5.7).

VI. THERMODYNAMIC FRAMEWORK FOR FLUCTUATION

A. State space

The fluctuation formula (5.2) tells us that to have a full thermodynamic fluctuation theory we must first set up the state space properly even in equilibrium. For nonequilibrium thermodynamics obviously we must have a larger state space

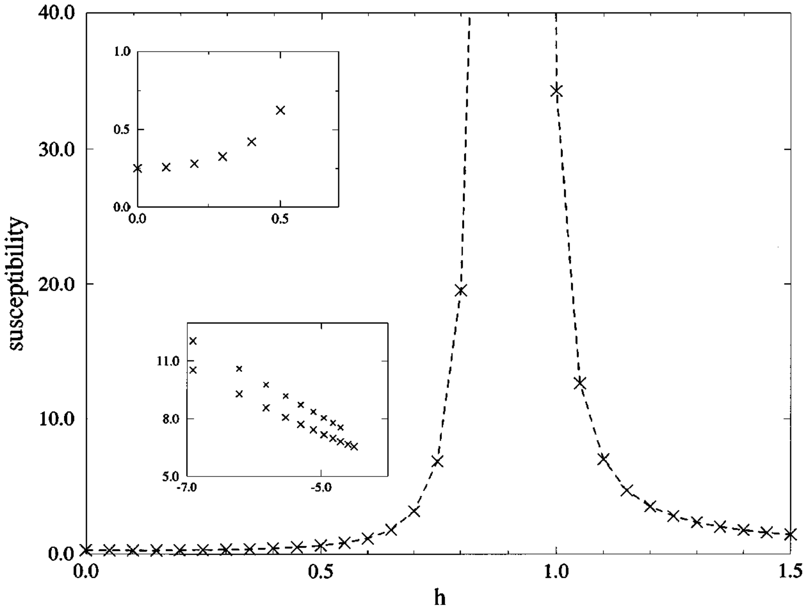


FIG. 4. The susceptibility as a function of h, for $r=1$. The inset on the upper left side is a blowup for $h \leqslant 0.5$. The inset on the lower left side is a log-log plot of the region $h=0.92$ to $h=0.94$, in intervals of $\Delta h=0.02$; the upper line of points is for $h>h_{\mathrm{cr}}$, lower line for $h<h_{\mathrm{cr}}$. $h_{\mathrm{cr}}=0.931$. The slopes of the upper line is about 2, and the lower line around 2.5.

than the equilibrium counterpart. For example, in the extended thermodynamics [36] not only the extensive quantities but also their time derivatives (or the corresponding fluxes) are regarded as state variables. Here we will not discuss in detail the condition for the state space to satisfy in order to support a thermodynamic or phenomenological theory. It is clear, however, that a point in the state space must reasonably uniquely specify the macroscopic state of the system. Operationally, the requirement is that there is a one-to-one correspondence between the set of macroscopically controlled parameters and the macroscopic states.

If we allow arbitrary (but periodic) perturbation, memory effects, and other complicated nonlinear phenomena must be taken into account, and even if we declare that our observables are time-averaged (or ensemble-averaged) quantities, the state space is generally infinite dimensional to accommodate the whole variety of changes of the state. It is obvious that the extended thermodynamics does not have a properly set up state space (however, if confined to systems slightly away from local equilibrium, the state space in this theory may be properly set up).

The above consideration tells us that it is not wise to try to construct a general thermodynamic framework for a system under an arbitrarily general nonequilibrium condition. Thus in this paper we confine ourselves to the variation of h (amplitude of the oscillating field) and a static field H, under fixed frequency and fixed noise. For our magnetic system, its time-averaged state without perturbation (i.e., the equilibrium state) is characterized by the time-averaged energy and the time-averaged magnetization. They are identical to the equilibrium internal energy and magnetization, respectively.

Let us turn on the sinusoidal perturbation. We change only its amplitude. In the Z phase, M is zero independent of h (if $H=0$). However, the various steady states for $h>h_c$ should be considered macroscopically distinct; the dissipation (heat flowing out of the magnetic system into the heat bath) in-

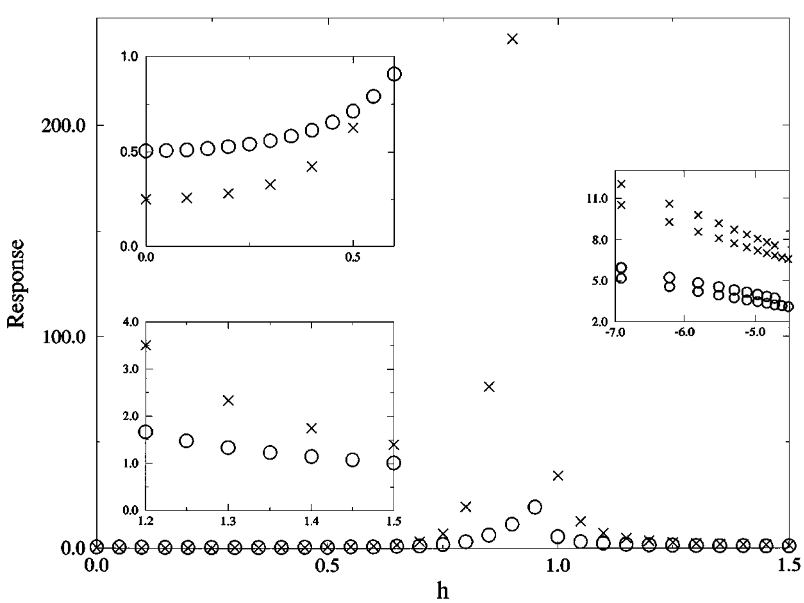


FIG. 5. A comparison between the system susceptibility χ, and the response to a constant field$=dM/dH$, for $r=1$. The cross symbol denotes χ, and the circles denote dM/dH. The insets on the left side are a blowup of the small and high field region, and the inset on the right hand side is a log-log plot as in Fig. 5. The slopes of the upper and lower lines through the circle data points in the log-log plot are about 1 and 1.5, respectively.

creases monotonically with h and is macroscopically observable. Hence, it is clear that more variables (related to the dissipation) must be included for a complete macroscopic description. We believe in order to specify a steady state, one more state variable is needed that does not have any equilibrium counterpart. In this paper we choose it to be the time-averaged energy dissipation rate Q defined by

$$\overline{Q}=\frac{h}{2\pi}\int_0^{2\pi}dt\ \sin(t)\phi(t). \tag{6.1}$$

One may conclude that the state space of our magnetic system driven by a fixed frequency sinusoidal magnetic field is spanned by M, Q, and E, the time average of the energy in the system. In the following discussion of the fluctuation, we allow M and Q to fluctuate under the condition that the conjugate variable ζ to the third state variable E is fixed. The rate function $I(M,Q)$ we study below is the Legendre transform of the full rate function $I(M,Q,E)$ with respect to E. Since ζ is fixed to its steady state value ($\zeta=0$), $I(M,Q)$ is the generalized entropy for the marginal distribution for M and Q. The rate function $I(M)$ studied in Sec. V is that of the marginal distribution for M.

B. Various susceptibilities

In the extended space $(\overline{M},\overline{Q})$, the rate function at the quadratic level is given as

$$I(\overline{M},\overline{Q})=\frac{1}{2\chi_{\overline{Q}}}(\overline{M}-\overline{M_0})^2+\frac{1}{2\chi_{\overline{M}}}(\overline{Q}-\overline{Q_0})^2 +\frac{1}{\chi}(\overline{M}-\overline{M_0})(\overline{Q}-\overline{Q_0}), \tag{6.2}$$

where $\overline{M_0}$, $\overline{Q_0}$ are the most probable values of $\overline{M}$,$\overline{Q}$, and the susceptibilities $\chi_{\overline{Q}}$ and $\chi_{\overline{M}}$ refer to fluctuations sampled under the condition of fixed Q and M, respectively; that is,

$$\chi_{\overline{Q}}=\langle(\overline{M}-\overline{M_0})^2\rangle_{\overline{Q}=\overline{Q_0}}, \tag{6.3}$$

$$\chi_{\overline{M}}=\langle(\overline{Q}-\overline{Q_0})^2\rangle_{\overline{M}=\overline{M_0}}. \tag{6.4}$$

In the above two formulas we keep the other state variable fixed when computing the susceptibility of either M or Q. However, as in the usual equilibrium theory, the susceptibilities under the constraint that the *conjugate* forces of the other variables are fixed [as in Eq. (5.1)] are the more useful and experimentally accessible ones. This latter case arises in our theory in the following way.

In Sec. V, the susceptibility of the magnetization was studied as a response to a perturbation $\lambda g(t)$ (which we shall write as $\lambda g_{\overline{M}}$); in this case there is no constraint on the variable Q. λ is interpreted as a conjugate generalized force (or affinity) to the state variable M. Similarly, the fluctuation of the dissipation Q can be studied as a response to the force $\nu g_{\overline{Q}}$. Here ν is a scale factor that is the conjugate force to Q, and $g_{\overline{Q}}$ is given by Eq. (5.7) with $f=h\sin(t)\phi(t)$. We shall denote the susceptibilities computed with the forces $\lambda g_{\overline{M}}(t)$ or $\nu g_{\overline{Q}}(t)$, respectively, by

$$\chi_\nu=-\lim_{\Delta\lambda\to 0}(\Delta\overline{M}/\Delta\lambda)_{\nu=0}=\langle(\overline{M}-\overline{M_0})^2\rangle_{\nu=0}, \tag{6.5}$$

$$\chi_\lambda=-\lim_{\Delta\nu\to 0}(\Delta\overline{Q}/\Delta\nu)_{\lambda=0}=\langle(\overline{Q}-\overline{Q_0})^2\rangle_{\lambda=0}. \tag{6.6}$$

The susceptibilities χ_ν,χ_λ refer to a passive (unconstrained) sampling of the fluctuations M,Q, as in any experimental measurement of the susceptibility by observing the natural fluctuations.

The relation between the two sets of susceptibilities discussed above follows easily from the Legendre transform of the rate function, $\Psi(\lambda,\nu)=I(M,Q)+\lambda M+\nu Q$ (a generalized Massieu function):

$$\chi_{\overline{Q}}=\frac{-\overline{\chi}^2+\chi_\nu\chi_\lambda}{\chi_\lambda}, \tag{6.7}$$

$$\chi_{\overline{M}}=\frac{-\overline{\chi}^2+\chi_\nu\chi_\lambda}{\chi_\nu}, \tag{6.8}$$

$$\chi=\frac{\overline{\chi}^2-\chi_\nu\chi_\lambda}{\overline{\chi}}. \tag{6.9}$$

Operationally, the susceptibilities $\chi_{\overline{Q}}$, $\chi_{\overline{M}}$, χ, and $\overline{\chi}$ are defined as

$$\chi_{\overline{Q}}=-\lim_{\Delta\lambda\to 0}(\Delta\overline{M}/\Delta\lambda)_{\overline{Q}=\overline{Q_0}}, \tag{6.10}$$

$$\chi_{\overline{M}}=-\lim_{\Delta\nu\to 0}(\Delta\overline{Q}/\Delta\nu)_{\overline{M}=\overline{M_0}}, \tag{6.11}$$

$$\chi=-\lim_{\Delta\lambda\to 0}(\Delta\overline{Q}/\Delta\lambda)_{\overline{M}=\overline{M_0}}=-\lim_{\Delta\nu\to 0}(\Delta\overline{M}/\Delta\nu)_{\overline{Q}=\overline{Q_0}}, \tag{6.12}$$

$$\overline{\chi}=-\lim_{\Delta\lambda\to 0}(\Delta\overline{Q}/\Delta\lambda)_{\nu=0}=-\lim_{\Delta\nu\to 0}(\Delta\overline{M}/\Delta\nu)_{\lambda=0}. \tag{6.13}$$

C. Maxwell's relation and Le Chatelier–Braun principle for steady states

Various "Maxwell-type relations" [such as the second equality in Eqs. (6.12) and (6.13)] follow from the integrability conditions of I, and its associated Legendre transforms. To interpret these relations as Maxwell relations in the conventional sense, we must use the phenomenological postulate that asserts the existence of a state function $\Sigma(M,Q)$, for which $\delta\Sigma$ due to fluctuation is I.

To make contact with the actual steady states, the mathematical conjugate parameters (λ,ν) must be related to the physical fields that naturally perturb the system. From the experimental or operational point of view, the natural perturbation variables in the space of steady states for our model magnetic system are (H,h). Locally, the relation between these two sets is given by the matrix $C=-A^{-1}B$ as $(\lambda,\nu)^T=C(\Delta H,\Delta h)^T$, where

$$A=\begin{pmatrix}\chi_\nu & \overline{\chi}\\ \overline{\chi} & \chi_\lambda\end{pmatrix},\quad B=\begin{pmatrix}(\partial\overline{M}/\partial H)_h & (\partial\overline{M}/\partial h)_H\\ (\partial\overline{Q}/\partial H)_h & (\partial\overline{Q}/\partial h)_H\end{pmatrix}. \tag{6.14}$$

As mentioned in Sec. V, the phenomenological postulate is valid if the conjugate forces (affinities) λ,ν perturb the steady state to a new state that remains in the state space. Thus we should require that the inverse of the matrix C^{-1} exists, which makes the map between (λ,ν) and $(\Delta H,\Delta h)$ one to one. Any path in the state space induced by (λ,ν) can then be mapped uniquely to a path in the (H,h) space. The convexity of the rate function automatically implies that the matrix A is invertible. The existence of B^{-1} follows from the requirement of a proper state space. This means, from the local relation $(\Delta M,\Delta Q)=B(\Delta H,\Delta h)$, that B should be an invertible matrix for each steady state (i.e., one-to-one correspondence between the system parameters and macroscopic states). Of course, one can never verify the existence of B^{-1} for every steady state. Nonetheless, it does seem likely that our state space is properly set up for the following reason.

The behavior of the state variables $\overline{M},\overline{Q}$ in the space of steady states defined by variations in h,H is typically the following: M increases with H, and decreases with h; Q decreases with H, and increases with h. Therefore, an increase in M should usually result in a decrease in Q, and vice versa. Furthermore, the change of M with H (h fixed) does tend to be larger than the magnitude of the change of M with h (H fixed), and similarly with the Q case [i.e., $|(\partial Q/\partial h)_H|\geq|(\partial Q/\partial H)_h|$]. Thus, it is likely that the condition $\partial(M,Q)/\partial(H,h)\neq 0$ is satisfied, which implies the existence of $C^{-1}=-B^{-1}A$. The existence of B^{-1} was confirmed numerically for our model Eq. (3.2), at least for the finite number of steady states that were checked (roughly the range $0<h,\ H<2$, in increments of $\Delta h,\ \Delta H=0.1$).

From the above discussion, it seems plausible that the phenomenological postulate is valid for our system. Various Maxwell relations and stability conditions automatically follow, the most typical ones being the aforementioned relations in Eqs. (6.12) and (6.13). From an appropriate Legendre transform of I, we obtain other relations, such as

$$(\partial\overline{M}/\partial\overline{Q})_{\lambda=0}=-(\partial\nu/\partial\lambda)_{\overline{Q}}=\frac{\overline{\chi}}{\chi_\lambda}, \tag{6.15}$$

or

$$(\partial\overline{Q}/\partial\overline{M})_{\nu=0}=\frac{\overline{\chi}}{\chi_\nu}. \tag{6.16}$$

Using the stability condition (i.e., the stability condition of the steady state) $\overline{\chi}^2-\chi_\lambda\chi_\nu<0$, which follows from the convexity property of I, the above two relations can be rewritten as the inequality

$$(\partial\overline{M}/\partial\overline{Q})_{\nu=0}>(\partial\overline{M}/\partial\overline{Q})_{\lambda=0}. \tag{6.17}$$

More typical consequences of stability are Le Chatelier–Braun principles, such as

$$\chi_\nu>\chi_{\overline{Q}}, \tag{6.18}$$

$$\chi_\lambda>\chi_{\overline{M}}. \tag{6.19}$$

In general, the Maxwell relation and the Le Chatelier–Braun Principles (6.17)–(6.19) are somewhat tedious to apply, as the constraints on λ,ν must be mapped to the corresponding changes in $\Delta H,\Delta h$ using the information contained in the matrix B and A, both of which vary as a function of the steady state. The usefulness and practical implications of the above relations remain to be seen. However, we can easily understand the main features of (6.17)–(6.19) in the following fashion.

Consider first the two relations (6.18) and (6.19). If we change (decrease) λ to increase M, Q should decrease. Thus, if we wish to keep Q fixed for the same change λ, M should change by less. Similarly, for a change in ν, keeping M fixed should result in a smaller change in Q than if we hold λ constant.

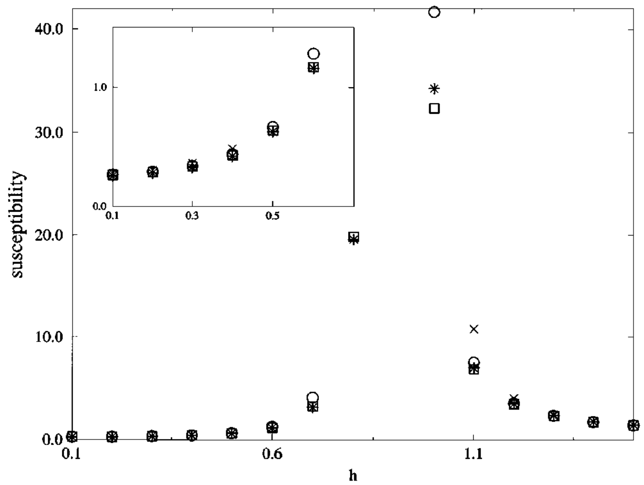
The relation $(\partial\overline{M}/\partial\overline{Q})_{\nu=0}>(\partial\overline{M}/\partial\overline{Q})_{\lambda=0}$ is slightly more awkward to explain, as the effect of the process $\lambda=0$ or $\nu=0$ is less intuitive. Generally, however, since λ is the conjugate force to M, it is reasonable to expect a smaller change in ΔM under the condition $\lambda=0$ than for the process $\nu=0$.

VII. FINITE AVERAGING TIME AND FINITE NOISE

We have been mainly concerned with the long-time small noise limit form of the rate function which is equal to the generalized fluctuation entropy. It corresponds to the equilibrium entropy in the thermodynamic limit. In the equilibrium thermodynamic theory of fluctuations, it is postulated that the macroscopic entropy function can be successfully used to describe the spatially (macroscopically) small scale fluctuations. If the postulate were significantly at variance with reality, then thermodynamic fluctuation theory would have had little practical relevance. Thus, it is a practically important question to ask how reliable our limit rate function is for shorter times with larger noises.

In order to study the reliability of the limit rate function, we studied the behavior of the susceptibility from real time simulations of the stochastic dynamics Eq. (3.2). The susceptibilities were studied for small nonzero noise levels, and for the averaging time τ a finite multiple of the basic period 2π. This stochastic dynamics was explicitly solved with the aid of a fourth order Runge-Kutta scheme with a typical time increment $dt=0.005$. After an initial relaxation time for the system to settle down to a steady state, statistics of the n period average were taken (i.e., $\tau=2n\pi$). The sampling of the fluctuations M,Q corresponds to a passive sampling (in the sense explained in Sec. VI B). Hence we are measuring χ_ν,χ_λ. In order to compare different data sets, the scaled susceptibilities $(\tau/\epsilon^2)\langle(M-\overline{M}_0)^2\rangle|_{\nu=0}$, $(\tau/\epsilon^2)\langle(Q-\overline{Q}_0)^2\rangle|_{\lambda=0}$ are plotted for finite noise and finite averaging period. Each data set involved the sampling of about 10^4–10^5 runs (n period averages). Some cases were also checked for a sampling of 10^6 runs. At least not very close to the transition, we believe enough statistics were taken to get an accurate value of the susceptibility.

The behavior of the susceptibility χ_ν,χ_λ for three different noise levels ($\epsilon=0.01$, 0.05, 0.1), with an averaging period of 10 ($n=10$), is shown in Fig. 6. As is evident in the figure, the noiseless theory is fairly accurate (at least not too close to the



(b)

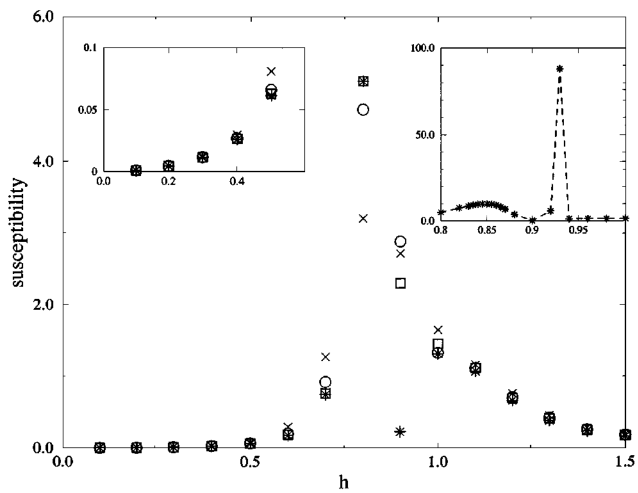


FIG. 6. The scaled susceptibility (a) χ_ν and (b) χ_λ; for $\epsilon=0,0.01,0.05,0.1$, and averaging period $n=10$. The system parameters are $H=0,\gamma=\omega=u_0=r=1$. The star symbol is the zero-noise result, the square $\epsilon=0.01$, the circle $\epsilon=0.05$, and the cross symbol the $\epsilon=0.1$ data. The inset on the right side of (b) illustrates the behavior of the zero-noise long-time χ_λ near the transition, which does seem to diverge.

transition) for noise levels $\epsilon\leqslant0.05$. For $\epsilon=0.1$ there is a shift in the bifurcation of the dynamics to smaller h, and the data begin to deviate significantly from the zero-noise theory.

For a fixed noise strength, the scaling of χ_ν for different averaging periods is illustrated in Fig. 7. In general, the data for the different averaging periods ($n=10,5,3,1$) superimposes well. In particular away from the transition there is a very good collapse of the data. Close to the transition the susceptibility for larger n moves towards the zero-noise long-time limit value.

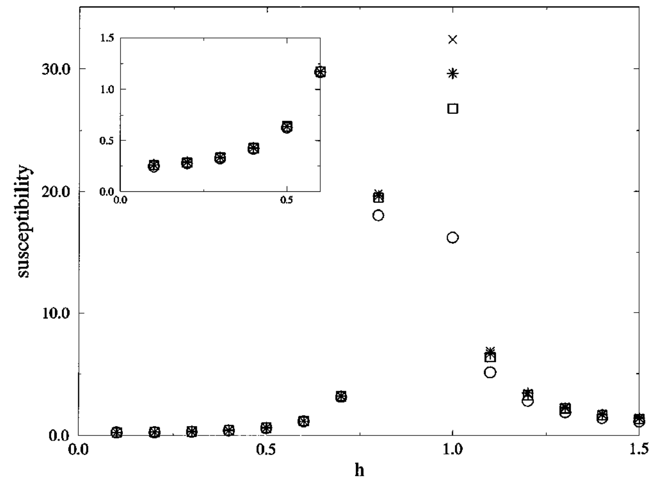


FIG. 7. The scaled susceptibility χ_ν for different averaging periods at a noise level $\epsilon=0.01$. The circles denote the $n=1$ data, the squares $n=3$, the stars $n=5$, and the crosses the $n=10$ data. The system parameters are $H=0$, $\gamma=\omega=u_0=r=1$. Similar behavior is observed for χ_λ and for $\epsilon=0.05$.

VIII. DISCUSSION

In order to have some clues for a phenomenological or thermodynamic framework for nonequilibrium systems, we have studied the fluctuation around ''steady states.'' We mean by steady state the state whose long-time average is well defined. In this paper we have discussed periodic states, but the state may be chaotic or turbulent. Large deviation theory tells us that there is a generalized entropy function(al) (rate function) that governs the fluctuation probability of time-averaged quantities.

We have proposed a phenomenological postulate that the Taylor expansion coefficients of the rate function are determined by the response of the steady state to perturbations exactly as in the equilibrium thermodynamic theory of fluctuation. For models described by Langevin equations, a fluctuation-response relation follows trivially. The way to compute the rate function tells us what the natural conjugate variables are for the observables whose fluctuations we study. This point has been illustrated by a model of a magnetic system under a periodic magnetic field.

Although the phenomenological postulate allows us to glimpse a possible (local) thermodynamic framework, still we cannot make a global framework. We have clearly recognized that the difficulty lies already in equilibrium states; no phenomenology of time-averaged quantities has been constructed, even for equilibrium states. Here we briefly discuss some relevant basic questions.

A natural question is the choice of the macroscopic observables (macro-observables) to specify the macroscopic steady state. In other words, the question is: What is the proper ''thermodynamic'' state space? This question seems to have been paid no serious thought in nonequilibrium. For example, Keizer [37] introduces the concept of a physical ensemble that may be understood as an equivalence class of microscopic states according to the macro-observable values. The idea sounds natural, but we must note that there is no guarantee that the choice of the macro-observables being used for this classification is a good macrovariable set to allow a phenomenological description such as thermodynamics (as intended in [37]). As pointed out in Sec. VI the state space of the extended irreversible thermodynamics is also insufficient, although its state space may be sufficient for linear processes (i.e., systems close to local equilibrium). In the ordinary equilibrium thermodynamics, the concept of state variables and state space itself should be understood as primitive concepts such as points and lines in Euclidean geometry. The crucial point is that the state space allows the principles of thermodynamics to hold; we cannot arbitrarily choose a set of macrovariables and construct a thermodynamic framework.

We must point out that even for equilibrium systems different kinds of thermodynamic formalisms are possible according to the averaging method to define macro-observables. The following observation sheds some light on the importance of the ensemble concept in statistical mechanics (contrary to the claim of Ma [38]). Let us consider a system under equilibrium condition. We may define the average by a time average over a chunk of material (even a small cluster of spins should do), spatial average at one instant, or ensemble average; any linear combination of different averaging methods is a respectable means to define macro-observables. Independent of the method of calculating the averaged (thermodynamic) quantities, the values of averaged energy and magnetization are intact, and equal to the internal energy and magnetization, respectively, of the standard equilibrium thermodynamics. Hence, the standard thermodynamic relation under the adiabatic condition

$$d\overline{E}=\sum_i x_i d\overline{X_i} \tag{8.1}$$

is true independent of the method of averaging denoted by the overline, where X_i are extensive observables and x_i their conjugate variables with respect to energy. Even the ordinary Gibbs relation

$$d\overline{E}=TdS+\sum_i x_i d\overline{X_i} \tag{8.2}$$

holds if S is computed as the ordinary entropy of the function of extensive quantities. T is identical to the absolute temperature.

However, fluctuations do depend on how the variance is computed, because the results depend on correlations among sampled values. The ensemble average is strictly over independent samples, but time averages of a single specimen crucially depend on the time correlation as illustrated in the context of the rate function in [17]. Perhaps the existence of the gross discrepancy in fluctuations calculated by these two averaging methods may be a good characteristic of glassy states. The discrepancy exists even in true thermodynamic equilibrium states: the rate functions for time-averaged fluctuations and ensemble-averaged fluctuations are distinct. This implies that even if the state space is spanned by E and $\overline{X_i}$, the ''entropy'' needed to write down the generalized Gibbs relation corresponding to Eq. (8.2) ought to be dependent on the choice of the averaging method.

It is now clear that the ordinary fluctuation-response relation (5.1) and the standard Gibbs relation (8.2) are compatible only when the average is understood as the ensemble average as in the ordinary statistical thermodynamics. In other words, if we choose, e.g., the time averaging, then the fluctuation theory and the usual Gibbs relation (8.2) are incompatible. However, this is not surprising, because the rate function in this case is, as we have seen, not $(-)\delta S$. The actual form corresponding to Eq. (8.2) must be the following generalized Gibbs relation:

$$d\overline{E}=\theta\, d\Sigma+\theta\sum_i y_i d\overline{X_i}, \tag{8.3}$$

where y_i is the thermodynamic conjugate variable for $\overline{X_i}$ with respect to the generalized entropy (whose deviation is the generalized affinity discussed in Secs. V and VII). Here θ is the conjugate variable of the generalized entropy Σ with respect to energy, and the rate function I for fluctuations is given by $-\delta\Sigma$. However, we have not yet succeeded in making this framework operationally meaningful.

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$$a_{ij}\int_I b_i d\phi_j = a_{ij}\int_S b_i d\phi_j - \frac{1}{2}\int \partial_i b_i dt.$$

Here the irrelevant spatial integral is suppressed, and the subscript I denotes the Ito integral and S the Stratonovitch integral.
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