Nonlinear susceptibilities and the measurement of a cooperative length

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We derive the exact beyond-linear fluctuation-dissipation relation, connecting the response of a generic observable to the appropriate correlation functions, for Markov systems. The relation, which takes a similar form for systems governed by a master equation or by a Langevin equation, can be derived to every order in large generality with respect to the considered model in equilibrium and out of equilibrium, as well. On the basis of the fluctuation-dissipation relation, we propose a particular response function, namely the second-order susceptibility of the two-particle correlation function, as an effective quantity to detect and quantify cooperative effects in glasses and disordered systems. We test this idea by numerical simulations of the Edwards–Anderson model in one and two dimensions.

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A central phenomenon in the statistical mechanics of interacting systems is the onset of long-range order when approaching phase transitions, specifically second-order ones such as the paraferromagnetic or gas-liquid transition. The coherence length ξ expressing the range of correlations is disclosed by the knowledge of an appropriate (two point) correlation function C_{ij}, as is $C_{ij}=\langle\sigma_i\sigma_j\rangle-\langle\sigma_i\rangle\langle\sigma_j\rangle$ for the prototypical Ising model. The divergence of ξ induces the scaling symmetry when the critical point is neared. In this framework, equilibrium linear-response theory, relating C_{ij} to its conjugate susceptibility χ_{ij} (and more generally two-time correlations $C_{ij}(t_1,t_2)=\langle\sigma_i(t_1)\sigma_j(t_2)\rangle-\langle\sigma_i(t_1)\rangle\langle\sigma_j(t_2)\rangle$ and susceptibilities $\chi_{ij}(t_1,t_2)$) through the fluctuation-dissipation theorem (FDT), has proved to be of the uppermost importance both theoretically and experimentally, allowing the alternative determination of correlations and, hence, of ξ through linear-response functions.

These concepts are not restricted only to equilibrium states but inform nonequilibrium statistical mechanics, as well. For example, in a broad class of aging systems, the kinetics is characterized by the growth of a characteristic length $L(t)$, determining a dynamical scaling symmetry in close analogy to what happens in static phase transitions. In view of these and related issues, increasing interest has been recently devoted to the generalization of linear-response theory to out of equilibrium systems: a research subject originating from the recognition that the relation between $\chi_{ij}(t_1,t_2)$ and $C_{ij}(t_1,t_2)$ may be used to define an *effective* temperature,[1] and to be a bridge between equilibrium and nonequilibrium properties.[2] Although a theorem of such a generality as the FDT cannot be derived off equilibrium, in the case of Markov processes, a natural generalization in the form of a fluctuation–dissipation relation (FDR) between $\chi_{ij}(t_1,t_2)$, $C_{ij}(t_1,t_2)$, and a correlator $D_{ij}(t_1,t_2)$ involving the generator of the stochastic process has been obtained.[3,4] This result could open the way, in principle, to measurements of $C_{ij}(t_1,t_2)$ and, hence, of $L(t)$ from nonequilibrium susceptibilities, provided the properties of D_{ij} are known.

This whole approach cannot be straightforwardly applied to the case of glasses, of spin glasses, and, in several instances, of disordered systems because their unusual type of long-range order is not captured by linear-response functions or even by two point correlators. These quantities remain short ranged, even when some long-range order appears in the system. This is because ordered patterns are randomized by the quenched disorder so that, for instance, $\overline{\langle\sigma_i\sigma_j\rangle}$ (where the overbar denotes the average over the disorder) vanishes even when $\langle\sigma_i\sigma_j\rangle\neq 0$. To circumvent this problem, one has to consider higher-order (nonlinear) response functions or equivalently, n-spin ($n>2$) correlation functions $C^{(n)}$. Along this line, recently, a measure of cooperativity has been proposed,[5] relying on a four-point correlation function as

$$C_{ij}^{(4)}(t,t_w)=\overline{\langle\sigma_i(t)\sigma_i(t_w)\sigma_j(t)\sigma_j(t_w)\rangle} - \overline{\langle\sigma_i(t)\sigma_i(t_w)\rangle\langle\sigma_j(t)\sigma_j(t_w)\rangle}. \quad (1)$$

The idea is that, while C_{ij} is annihilated by the disorder average, the variance of $\sigma_i\sigma_j$ survives, possibly providing information on cooperativity. $C_{ij}^{(4)}$ has been proved to be effective in numerical simulations[6,7] but its direct experimental investigation remains a challenge,[7] as in general multipoint correlators. A natural way out of this deadlock is to measure responses to external perturbations, namely susceptibilities, as suggested by Bouchaud and Biroli[8] and done experimentally in (Ref. 9). In order to make sure what actually do the nonlinear susceptibilities probe, however, it is crucial to establish their relationship with multipoint correlators. Some specific aspects of this issue have been considered recently,[8,10] limited to the case of systems governed by a Langevin equation but a general formulation is presently lacking.

In this Brief Report, we present the exact derivation of the FDR beyond-linear order for spin models evolving with Markovian dynamics. The systematic approach we use is quite general, allowing one to derive the response function of an arbitrary observable to every order in the external perturbation and to relate it to correlation functions of the unperturbed system in equilibrium and out of equilibrium as well, for generic spin models (e.g., Ising, clock, Heisenberg models, etc…) in full generality with respect to the Hamiltonian and the evolution rules. We show that the FDR takes the same form for hard spins, whose kinetics is ruled by a master equation, and for soft spins systems governed by a Langevin equation, further supporting the generality of our result. This relation shows that, already in equilibrium and beyond-linear order, the susceptibility is related not only to multispin correlations $C^{(n)}$ but also to the D correlators, much like in linear theory out of equilibrium. This feature loosens the relation between response and multispin correlations, raising the question of which response function is best suited to detect cooperative effects. We argue that a particular susceptibility $\chi^{(c,2)}$, basically the second-order response of the correlation function C is well fit to this task and bears information on the correlation length. We complement this idea by numerical simulations of disordered spin models, showing how the existence of a growing length can be detected using $\chi^{(c,2)}$.

Let us sketch the derivation of the FDR for hard spins.[11] Using the operator formalism, we consider for simplicity a system of Ising spins (but the result holds more generally) whose state is described by the vector $|\sigma\rangle = \otimes|\sigma_i\rangle$ $(i=1,N)$ on a lattice. The stochastic evolution is characterized by the propagator,

$$\hat{P}(t|t_w) = \mathcal{T}\exp\left[\int_{t_w}^{t} ds\hat{W}(s)\right], \tag{2}$$

where $\hat{W}(t)$ is the time dependent generator of the process, which is assumed to obey detailed balance, and $\mathcal{T}$ is the time-ordering operator. The expectation $\langle O(t)\rangle$ of a generic observable O on the time dependent state $|P(t)\rangle$ is given by $\langle -|\hat{O}|P(t)\rangle$, where $\langle -| = \Sigma_\sigma\langle\sigma|$ is the flat vector. Using the propagation $|P(t)\rangle = \hat{P}(t|t_w)|P(t_w)\rangle$ of the states, this can be written as $\langle -|\hat{O}\hat{P}(t|t_w)|P(t_w)\rangle$. Switching on an external field h (perturbation) at time t_w, changing $\hat{P}$ to $\hat{P}_h$, the expectation $\langle O(t)\rangle_h = \langle -|\hat{O}\hat{P}_h(t|t_w)|P(t_w)\rangle$ can be expanded as $\langle O(t)\rangle_h = \langle O(t)\rangle_0 + \Sigma_{n=1}^{\infty}(1/n!)\Sigma_{j_1\ldots j_n}\int_{t_w}^{t}dt_1\ldots\int_{t_w}^{t}dt_n R^{(O,n)}_{j_1\ldots j_n}(t,t_1,\ldots,t_n)$ $h_{j_1}(t_1)\ldots h_{j_n}(t_n)$, where

$$R^{(O,n)}_{j_1\cdots j_n}(t,t_1,\ldots,t_n) = \left.\frac{\delta^n\langle O(t)\rangle_h}{\delta h_{j_1}(t_1)\cdots\delta h_{j_n}(t_n)}\right|_{h=0} = \langle -|O\left.\frac{\delta^n\hat{P}_h(t|t_w)}{\delta h_{j_1}(t_1)\cdots\delta h_{j_n}(t_n)}\right|_{h=0}|P(t_w)\rangle, \tag{3}$$

is the nth order response function $(t\geq t_1,\ldots,t_n)$. Let us workout $R^{(\mathcal{O},2)}$ as an illustration, the generalization to arbitrary n being straightforward.[11] From Eq. (2), one has

$$\frac{\delta^2\hat{P}_h(t|t_w)}{\delta h_{j_1}(t_1)\delta h_{j_2}(t_2)} = \hat{P}_h(t|t_1)\frac{\partial\hat{W}(t_1)}{\partial h_{j_1}(t_1)}\hat{P}_h(t_1|t_2)\frac{\partial\hat{W}(t_2)}{\partial h_{j_2}(t_2)}\hat{P}_h(t_2|t_w) + \hat{P}_h(t|t_1)\frac{\partial^2\hat{W}(t_1)}{\partial h^2_{j_1}(t_1)}\hat{P}_h(t_1|t_w)\delta_{12}, \tag{4}$$

where $t_1\geq t_2$ and $\delta_{12}=\delta_{j_1,j_2}\delta(t_1-t_2)$. We choose a perturbation entering the Hamiltonian as $-\Sigma_i h_i(t)\hat{\sigma}^z_i$, where $\hat{\sigma}^z$ is the z Pauli matrix. Assuming single spin-flip dynamics for simplicity, the generalization to multiple spin flips being straightforward, the derivative of the generator is $\partial^n\hat{W}(t_1)/\partial h^n_{j_1}(t_1) = (-\beta)^n\hat{W}_{j_1}(t_1)(\hat{\sigma}^z_{j_1})^n$. Then

$$R^{(O,2)}_{j_1j_2}(t,t_1,t_2) = \beta^2\langle -|\hat{O}\hat{P}(t|t_1)\hat{W}_{j_1}\hat{\sigma}^z_{j_1}\hat{P}(t_1|t_2)\hat{W}_{j_2}\hat{\sigma}^z_{j_2}|P(t_2)\rangle + \beta^2\langle -|\hat{O}\hat{P}(t|t_2)\hat{W}_{j_2}|P(t_2)\rangle\delta_{12}. \tag{5}$$

In order to obtain an expression involving only observable quantities (i.e., diagonal operators), we write $\hat{W}_{j_1}\hat{\sigma}^z_{j_1} = \frac{1}{2}[\hat{W}_{j_1},\hat{\sigma}^z_{j_1}] + \frac{1}{2}\{\hat{W}_{j_1},\hat{\sigma}^z_{j_1}\}$, where $[\cdot]$ or $\{\cdot\}$ denote the commutator or the anticommutator. It can be easily shown that $\hat{B}_i(t) = \{\hat{\sigma}^z_i,\hat{W}_i(t)\}$ is a diagonal operator with the property $\frac{\partial}{\partial t}\langle\sigma^z_i(t)\rangle = \langle B_i(t)\rangle$. Since the term with the commutator acts like a time derivative, the second-order FDR is obtained,

$$\begin{aligned}R^{(O,2)}_{j_1j_2}(t,t_1,t_2) = \frac{\beta^2}{4}\Bigg\{&\frac{\partial}{\partial t_1}\frac{\partial}{\partial t_2}\langle O(t)\sigma_{j_1}(t_1)\sigma_{j_2}(t_2)\rangle \\ &- \frac{\partial}{\partial t_1}\langle O(t)\sigma_{j_1}(t_1)B_{j_2}(t_2)\rangle \\ &- \frac{\partial}{\partial t_2}\langle O(t)B_{j_1}(t_1)\sigma_{j_2}(t_2)\rangle \\ &+ \langle O(t)B_{j_1}(t_1)B_{j_2}(t_2)\rangle\Bigg\} \\ &+ \frac{\beta^2}{2}\langle O(t)\sigma_{j_1}(t_1)B_{j_1}(t_2)\rangle\times\delta_{j_2,j_1}\delta(t_1-t_2).\end{aligned} \tag{6}$$

Care must be used for $t_2\to t_1$ since the product of the commutators generates a singular term.[11] In a stationary state, using Onsager reciprocity, the above result simplifies to

$$\begin{aligned}R^{(O,2)}_{j_1j_2}(t,t_1,t_2) = \frac{\beta^2}{2}\Bigg\{&\frac{\partial}{\partial t_1}\frac{\partial}{\partial t_2}\langle O(t)\sigma_{j_1}(t_1)\sigma_{j_2}(t_2)\rangle \\ &- \frac{\partial}{\partial t_2}\langle O(t)B_{j_1}(t_1)\sigma_{j_2}(t_2)\rangle\Bigg\} \\ &+ \frac{\beta^2}{2}\langle O(t)\sigma_{j_1}(t_1)B_{j_1}(t_2)\rangle\times\delta_{j_2,j_1}\delta(t_1-t_2).\end{aligned} \tag{7}$$

Let us mention that for continuous variables (soft spins) governed by a Langevin equation $\partial\sigma_i(t)/\partial t = B_i(t) + \eta_i(t)$; by taking $\hat{W}$ as the Fokker–Planck generator, we obtain[12] the same FDR Eq. (6) [and hence Eq. (7)], without the last term containing the δ functions. Since on the r.h.s. do only appear correlation functions of the unperturbed system, Eq. (7) qualifies as the beyond linear FDT while Eq. (6) as its nonequilibrium generalization. This relation can be derived for the response of an arbitrary observable to every order in the external perturbation, for hard and soft spins alike, without reference to a particular Hamiltonian or transition rates. Exactly like in the linear case,[4] the above FDR serves as the basis for the development of a no field algorithm for the fast computation of the nonlinear-response function, as it will be shown below.

The peculiar feature of the nonlinear FDR Eqs. (6) and (7) is the ubiquitous (even in equilibrium) presence of the correlators D containing the operator $\hat{B}$, which introduces a specific reference to the particular dynamical process through the generator. This hinders a direct relation between response and multispin-correlation functions, hampering the procedure to associate ξ to a susceptibility, as in the equilibrium linear theory. Despite this, we argue that a quantity related to the second-order response of the composite operator $\hat{O}=\hat{c}_{ij}=\hat{\sigma}_i^z\hat{\sigma}_j^z$,

$$-\mathcal{R}_{ij}^{(c,2)}(t,t_1,t_2)=\left.\frac{\delta^2\langle\sigma_i(t)\sigma_j(t)\rangle}{\delta h_i(t_1)\,\delta h_j(t_2)}\right|_{h=0}-R_{ii}^{(\sigma,1)}(t,t_1)R_{jj}^{(\sigma,1)}(t,t_2), \tag{8}$$

where $R_{ij}^{(\sigma,1)}(t,t_1)$ is the linear-response function of the spin σ_i(Ref. 4) or alternatively, the susceptibility

$$\chi_{ij}^{(c,2)}(t,t_w)=\int_{t_w}^{t}dt_1\int_{t_w}^{t}dt_2\mathcal{R}_{ij}^{(c,2)}(t,t_1,t_2), \tag{9}$$

is well suited to detect cooperative effects (for disordered systems, a disorder average is implicitly assumed) and may be used to determine ξ. In equilibrium systems, this is readily seen since a simple statistical mechanical calculation yields

$$\chi_{ij,eq}^{(c,2)}=\lim_{t\to\infty}\chi_{ij}^{(c,2)}(t,t_w)=\beta^2\lim_{t\to\infty}[C_{ij}(t,t)]^2=\beta^2C_{ij,eq}^2, \tag{10}$$

namely the counterpart of the standard static equilibrium relation between correlations and susceptibilities. Taking the $k=0$ component $\chi_{k=0,eq}^{(c,2)}=(1/N)\Sigma_{i,j}\chi_{ij,eq}^{(c,2)}\propto\xi^{4-d-2\eta}$, therefore, one has direct access to the coherence length. Concerning the full two-time dependence of $\chi^{(c,2)}$ in a system characterized by dynamical scaling, by virtue of Eq. (10), one expects the same scaling form, with the same exponents of C^2; hence,

$$\chi_{k=0}^{(c,2)}(t,t_w)=\xi^{4-d-2\eta}f\left[\frac{\xi}{L(t)},\frac{L(t_w)}{L(t)}\right]. \tag{11}$$

On physical grounds, one may understand why cooperativity effects are revealed by $\chi^{(c,2)}$ as follows: writing the susceptibility $\chi_{ij}^{(\sigma,1)}(t,t_w)=\int_{t_w}^{t}dt_1R_{ij}^{(\sigma,1)}(t,t_1)$ as $\chi_{ij}^{(\sigma,1)}(t,t_w)=\langle x_{ij}(t,t_w)\rangle$, where[4] $x_{ij}(t,t_w)=\frac{\beta}{2}[\sigma_i(t)\sigma_j(t)-\sigma_i(t)\sigma_j(t_w)-\sigma_i(t)\int_{t_w}^{t}dt_1B_j(t_1)]$, in view of Eq. (5), $\chi^{(c,2)}$ can be cast as $-\chi_{ij}^{(c,2)}(t,t_w)=\langle x_{ii}^{(\sigma,1)}(t,t_w)x_{jj}^{(\sigma,1)}(t,t_w)\rangle-\langle x_{ii}^{(\sigma,1)}(t,t_w)\rangle\langle x_{jj}^{(\sigma,1)}(t,t_w)\rangle$. Namely, $\chi^{(c,2)}$ is the correlation of the variable whose average yields $\chi^{(\sigma,1)}$, much in the same way as $C_{ij}^{(4)}(t,t_w)$ is the correlation of the variable $\sigma_i(t)\sigma_i(t_w)$, whose average gives C. Since $\chi^{(\sigma,1)}$ is the response function conjugated to C by the FDT, this suggests that $\chi^{(c,2)}$ may be suitable (as will be further shown numerically below) to study cooperativity analogously, and for the same mechanism of $C^{(4)}$. Despite this, $\chi^{(c,2)}$ and $C^{(4)}$ can hardly be related. Actually, although $C^{(4)}$ appears in the first term on the r.h.s. of the FDR Eqs. (6) and (7) for $\mathcal{R}^{(c,2)}$, the terms containing B spoil the relation between $\mathcal{R}^{(c,2)}$ and $C^{(4)}$. It can be shown, in fact, that in most cases these terms are comparable with the first. For example, the static relation in Eq. (10) depends crucially on the contributions of the terms containing B.

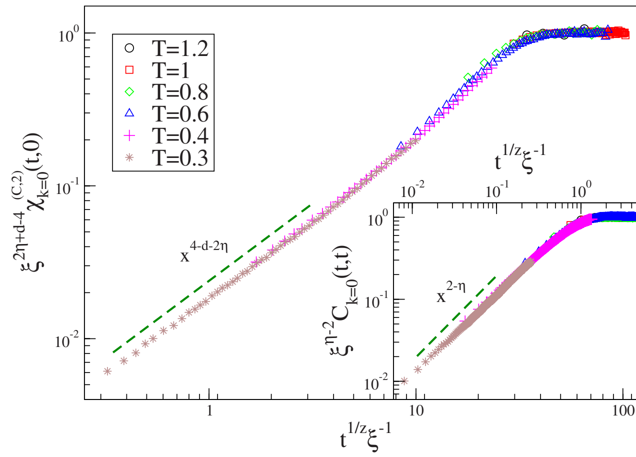

FIG. 1. (Color online) Data collapse of $\chi^{(c,2)}$ (C in the inset) for several temperatures in the $d=1$ EA model. The dashed lines are the expected power laws in the nonequilibrium regime.

An important advantage of $\chi^{(c,2)}$ with respect to multispin correlations is its fitting to experimental measurements. In fact, switching on a field h_i from t_w onwards, one has $\langle\sigma_i(t)\sigma_j(t)\rangle_h=\langle\sigma_i(t)\sigma_j(t)\rangle_{h=0}+\Sigma_{l,m}h_lh_m\int_{t_w}^{t}dt_1\int_{t_w}^{t}dt_2\delta^2\langle\sigma_i(t)\sigma_j(t)\rangle/(\delta h_l(t_1)\delta h_m(t_2))+O(h^4)$. In disordered systems, the first term on the r.h.s. vanishes and the only nonvanishing terms in the sum are those with $l=i$ and $m=j$ (or $l=j$ and $m=i$). Hence, using the definitions in Eqs. (9), (8), and (3), $\langle\sigma_i(t)\sigma_j(t)\rangle_h-\langle\sigma_i(t)\rangle_h^2=-h_ih_j\chi_{ij}^{(c,2)}(t,t_w)+O(h^4)$. Therefore, the study of a cooperative length can be reduced to the measurement of a correlation function in an external field (for instance a uniform one), as proposed by Huse.[13]

In order to check these ideas and to test the efficiency of the method to measure the cooperative length, we have computed numerically $\chi_{k=0}^{(c,2)}(t,0)$ in the Edwards–Anderson (EA) model with Hamiltonian $H=\Sigma_{ij}J_{ij}\sigma_i\sigma_j$ in $d\le 2$, simulated by means of standard Monte Carlo techniques with Glauber transition rates, where $B_i=\sigma_i-\tanh(\beta\Sigma_jJ_{ij}\sigma_j)$. The system is quenched from a disordered state at $t=0$ to different final temperature $T>0$. $\chi_{k=0}^{(c,2)}(t,0)$ is computed using Eq. (6). It must be stressed that due to the noisy nature of response functions, the advantage provided by the FDR [Eq. (6)], instead of applying an infinitesimal perturbation, is numerically unrenounceable. In fact, besides providing an incomparably better signal/noise ratio, the $h\to 0$ limit is built in the FDR. The analysis of the data proceeds as follows: from the large t value $\chi_{k=0,eq}^{(c,2)}$ of $\chi^{(c,2)}$, knowing η, ξ can be extracted for each temperature. Regarding $L(t)$ in the nonequilibrium regime $L(t)\ll\xi$, $\chi^{(c,2)}$ must be independent from ξ. Using Eq. (11) this implies $f(\xi/L(t),0)\sim(L(t)/\xi)^{4-d-2\eta}$. Hence, the nonequilibrium behavior of $L(t)$ can also be determined. With these results, one can control that the data collapse is obtained by plotting $\xi^{-4+d+2\eta}\chi_{k=0}^{(c,2)}(t,0)$ vs $L(t)/\xi$ for all the temperatures considered (see Figs. 1 and 2). We have studied first the model in $d=1$ with bimodal distribution of the coupling constants $J_{ij}=\pm 1$. This system can be considered as a laboratory since it can be mapped onto a ferromagnetic sys-

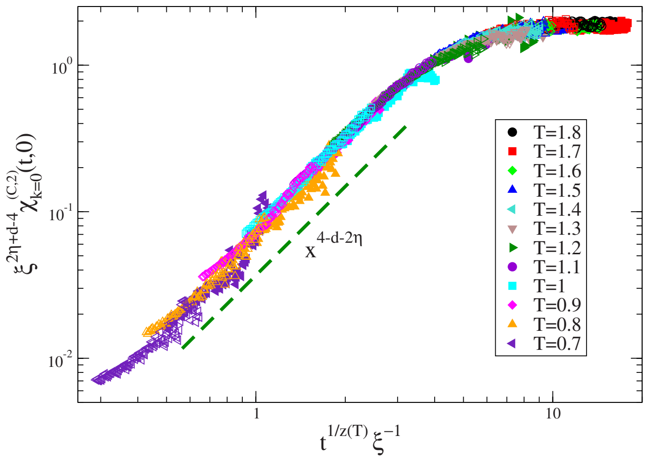

FIG. 2. (Color online) Data collapse of $\chi^{(c,2)}$ for several T in the $d=2$ EA model with bimodal (open symbols) or Gaussian (filled symbols) bond distribution, with $z(T)=4/T$. The dashed line is the expected power law in the nonequilibrium regime.

tem where $\eta=1$ and $L(t)\sim t^{1/z}$, with $z=2$, are known analytically. Moreover, besides $\chi^{(c,2)}$, one can also check the scaling of the usual correlation $C_{k=0}(t,t)$ after the mapping, and obtain another determination of $L(t)$ and ξ. In doing so, we find that the two methods to extract $L(t)$ and ξ agree within the numerical uncertainty between them, and with the analytical behaviors. The data collapse of $\chi^{(c,2)}$ and C is shown in Fig. 1. Here, one clearly observes the nonequilibrium kinetics in the early regime, characterized by a power-law behavior of $\chi^{(c,2)}$ with exponent $4-d-2\eta$, as expected, and the late equilibration with the convergence of $\chi^{(c,2)}_{k=0}(0,t)$ to $\chi^{(c,2)}_{k=0,eq}$. C behaves similarly. After this explicit verification, we turn to the $d=2$ case where the reference to C is not available. In this case, with both bimodal and Gaussian distributions of J_{ij} using $\eta=0$ (Ref. 14), we find a behavior of ξ consistent with previous results.[14,15] The nonequilibrium behavior is compatible with a power law $L(t)\sim t^{1/z(T)}$ with a temperature-dependent exponent in agreement with $z(T)\simeq 4/T$, as reported in.[16] The data collapse of $\chi^{(c,2)}$ is shown in Fig. 2. Notice also the additional collapse of the curves with bimodal and Gaussian bond distribution, further suggesting that the two models may share the same universality class at finite temperatures.[14]

In this Brief Report, we have derived the exact beyond-linear FDR. The result, which can be straightforwardly extended to every order, provides a rather general relation between the response and the correlation functions. It is satisfied by systems described by a master equation or by a Langevin equation, without reference to specific aspects of the considered model. On the basis of the FDR we argued that, providing numerical evidence, the second-order susceptibility $\chi^{(c,2)}$ is well fitted to uncover cooperative effects and to measure the coherence length in disordered and glassy systems. Importantly, this susceptibility has a simple operative definition, which might be fitted to experimental investigations.[13] Finally, we mention that the relevance of the beyond linear FDR is not restricted to the issue of cooperativity but is related to a number of open questions, among which the extension of the concept of effective temperatures beyond-linear order.

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