Observable Dependence of Fluctuation-Dissipation Relations and Effective Temperatures

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We study the nonequilibrium fluctuation-dissipation theorem (FDT) in the glass phase of Bouchaud's trap model. We incorporate an arbitrary observable m and obtain its correlation and response functions in closed form. A limiting nonequilibrium FDT plot is approached at long times for most choices of m. In contrast to standard mean field models, however, the shape of the plot depends nontrivially on the observable, and its slope varies continuously even though there is a single scaling of relaxation times with age. Nonequilibrium FDT plots can therefore not be used to define a meaningful effective temperature $T_{\rm eff}$ in this model. Consequences for the wider applicability of an FDT-derived $T_{\rm eff}$ are discussed.

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Glassy systems relax extremely slowly at low temperatures. They therefore remain far from equilibrium on very long time scales, and exhibit *ageing* [1]: the time scale of response to an external perturbation increases with the age (time since preparation) $t_{\rm w}$ of the system. Time translational invariance (TTI) and the equilibrium fluctuation-dissipation theorem [2] (FDT) relating correlation and response functions break down.

Let $C(t, t_{\rm w}) = \langle m(t)m(t_{\rm w})\rangle - \langle m(t)\rangle\langle m(t_{\rm w})\rangle$ be the autocorrelation function for an observable m, $R(t, t_{\rm w}) = \delta\langle m(t)\rangle/\delta h(t_{\rm w})|_{h=0}$ the linear response of $m(t)$ to a small impulse in its conjugate field h at time $t_{\rm w}$, and $\chi(t, t_{\rm w}) = \int_{t_{\rm w}}^{t} dt'\, R(t, t')$ the corresponding response to a field step $h(t) = h\Theta(t - t_{\rm w})$. In *equilibrium,* $C(t, t_{\rm w}) = C(t - t_{\rm w})$ by TTI (similarly for R and χ), and the FDT reads $-\frac{\partial}{\partial t_{\rm w}}\chi(t - t_{\rm w}) = R(t, t_{\rm w}) = \frac{1}{T}\frac{\partial}{\partial t_{\rm w}} C(t - t_{\rm w})$, with T the thermodynamic temperature (we set $k_{\rm B} = 1$). A parametric "FDT plot" of χ vs C is thus a straight line of slope $-1/T$. In the ageing case, the violation of FDT can be measured by a fluctuation-dissipation ratio (FDR), $X(t, t_{\rm w})$, defined through [3,4]

$$-\frac{\partial}{\partial t_{\rm w}}\chi(t, t_{\rm w}) = R(t, t_{\rm w}) = \frac{X(t, t_{\rm w})}{T}\frac{\partial}{\partial t_{\rm w}} C(t, t_{\rm w}). \quad (1)$$

In equilibrium, due to TTI, the derivatives $\frac{\partial}{\partial t_{\rm w}}$ in the FDT can be replaced by $-\frac{\partial}{\partial t}$; in the nonequilibrium definition (1) this would make less sense since only the $t_{\rm w}$ derivative of $\chi(t, t_{\rm w})$ is directly related to the impulse response $R(t, t_{\rm w})$. Values of X different from unity mark a violation of FDT and can persist even in the limit of long times, indicating strongly nonequilibrium behavior even though one-time quantities such as entropy and average energy may evolve infinitesimally slowly.

Remarkably, the FDR for several *mean field* models [3,4] assumes a special form at long times: Taking $t_{\rm w} \to \infty$ at constant $C = C(t, t_{\rm w})$, $X(t, t_{\rm w}) \to X(C)$ becomes a (nontrivial) function of the single argument C. If the equal-time correlator $C(t, t)$ also approaches a constant C_0 for $t \to \infty$, it follows that $\chi(t, t_{\rm w}) = \int_{C(t,t_{\rm w})}^{C_0} dC\, X(C)/T$. Graphically, this limiting nonequilibrium FDT relation is obtained by plotting χ vs C for increasingly large times; from the slope $-X(C)/T$ of the limit plot, an *effective temperature* [5] can be defined as $T_{\rm eff}(C) = T/X(C)$. In such a scenario, where $\chi(t, t_{\rm w})$ and $C(t, t_{\rm w})$ become simple functions of each other, either $t_{\rm w}$ or t can be used as the curve parameter for the FDT plot; the latter is the conventional prescription [3,4]. In general, however, the definition (1) ensures a slope of $-X(t, t_{\rm w})/T$ for a parametric χ-C plot *only* if $t_{\rm w}$ is used as the parameter, with t being fixed.

In the most general ageing scenario, a system displays dynamics on several characteristic time scales, one of which may remain finite as $t_{\rm w} \to \infty$, while the others diverge with $t_{\rm w}$. If, due to their different functional dependence on $t_{\rm w}$, these time scales become infinitely separated as $t_{\rm w} \to \infty$, they form a set of distinct "time sectors"; in mean field, $T_{\rm eff}(C)$ can then be shown to be *constant* within each such sector [4]. In the short time sector $[t - t_{\rm w} = O(1)]$, where $C(t, t_{\rm w})$ decays from C_0 to some plateau value, one generically has quasiequilibrium with $T_{\rm eff} = T$, giving an initial straight line with slope $-1/T$ in the FDT plot. The further decay of C (on ageing time scales $t - t_{\rm w}$ that grow with $t_{\rm w}$) gives rise to one of three characteristic shapes: (i) In models which statically show one step replica symmetry breaking (RSB), e.g., the spherical p-spin model [3], there is only one ageing time sector and the FDT plot exhibits a second straight line, with $T_{\rm eff} > T$. (ii) In models of coarsening and domain growth, e.g., the $O(n)$ model at large n, this second straight line is flat, and hence $T_{\rm eff} = \infty$ [6]. (iii) In models with an infinite hierarchy of time sectors (and infinite step RSB in the statics, e.g., the Sherrington-Kirkpatrick model) the FDT plot is instead a continuous curve [4].

$T_{\rm eff}$ has been interpreted as a time scale dependent nonequilibrium temperature and within mean field has been shown to display many of the properties associated with a thermodynamic temperature [5]. For example (within a given time sector), it is the reading which would be shown by a thermometer tuned to respond on that time

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scale. Furthermore—and of crucial importance to its interpretation as a temperature—it is independent of the observable m used to construct the FDT plot [5]. Note that the existence of such a temperature, applicable to a glassy material *as a whole,* is remarkable in the face of conventional nonequilibrium thermodynamics in which a material is considered to comprise many mesoscopic elements, each equilibrated, but at its own temperature $T(r,t)$. Nonetheless, the above picture is now very well established, at least in mean field.

Its status in nonmean field models, however, is less obvious. To check its validity, one must demonstrate that (a) a limiting FDT plot exists and that (b) it gives effective temperatures that are independent of the observable-field pair used to calculate C and χ. Depending on whether there are one or many ageing time sectors one then expects FDT plots similar to those for mean field systems.

Encouragingly, molecular dynamics and Monte Carlo (MC) simulations of binary Lennard-Jones mixtures [7], as well as MC simulations of frustrated lattice gases [8] (which loosely model structural glasses, whose phenomenology is similar to that of the p-spin model), show a limiting plot of type (i). MC simulations of the Ising model in dimension $d = 2$ and 3 show a plot of type (ii) [9]. And MC simulations of the Edwards-Anderson model in $d = 3$, 4 [10] give a plot of type (iii). The majority of existing studies, however, do not show that $T_{\rm eff}$ is independent of observable (notable exceptions are [7,8]) since they consider just one observable-field pair.

In this work, therefore, we analyze a model where FDT plots can be calculated analytically for *arbitrary* observables, allowing us to investigate in detail whether the simple mean field picture applies. Trap models [11–13] are obvious candidates for such a study. Popular as alternatives to the microscopic spin models discussed above, they capture ageing within a simplified single particle description. The simplest such model [11] comprises an ensemble of uncoupled particles exploring a spatially unstructured landscape of (free) energy traps by thermal activation. The tops of the traps are at a common energy level and their depths E have a "prior" distribution $\rho(E)$ $(E > 0)$. A particle in a trap of depth E escapes on a time scale $\tau(E) = \tau_0 \exp(E/T)$ and hops into another trap, the depth of which is drawn at random from $\rho(E)$. The probability, $P(E,t)$, of finding a randomly chosen particle in a trap of depth E at time t thus obeys

$$(\partial/\partial t)P(E,t) = -\tau^{-1}(E)P(E,t) + Y(t)\rho(E) \qquad (2)$$

in which the first (second) term on the right-hand side represents hops out of (into) traps of depth E, and $Y(t) = \langle \tau^{-1}(E)\rangle_{P(E,t)}$ is the average hopping rate. The solution of (2) for initial condition $P_0(E)$ is

$$P(E,t) = P_0(E)e^{-t/\tau(E)} + \rho(E)\int_0^t dt'\, Y(t')e^{-(t-t')/\tau(E)} \qquad (3)$$

from which $Y(t)$ has to be determined self-consistently. For the specific choice of prior distribution $\rho(E) \sim \exp(-E/T_{\rm g})$, the model shows a glass transition at a temperature $T_{\rm g}$. This can be seen as follows. At a temperature T, the equilibrium Boltzmann state (if it exists) is $P_{\rm eq}(E) \propto \tau(E)\rho(E) \propto \exp(E/T)\exp(-E/T_{\rm g})$. For temperatures $T \leq T_{\rm g}$ this is unnormalizable and cannot exist; the lifetime averaged over the prior, $\langle\tau\rangle_\rho$, is infinite. Following a quench to $T \leq T_{\rm g}$, the system never reaches a steady state, but instead ages: in the limit $t_{\rm w} \to \infty$, $P(E,t_{\rm w})$ is concentrated entirely in traps of lifetime $\tau = O(t_{\rm w})$. The model thus has just one characteristic time scale, which grows linearly with the age $t_{\rm w}$. [In contrast, for $T > T_{\rm g}$ all relaxation processes occur on time scales $O(1)$.] In what follows, we rescale all energies such that $T_{\rm g} = 1$, and also set $\tau_0 = 1$.

To study FDT we extend the model by assigning to each trap, in addition to its depth E, a value for a completely generic observable m. This is directly analogous to assigning a value m to a spin state in spin models and we thus refer to m as magnetization. In the most general case the trap population is then characterized by the joint prior distribution $\sigma(m \mid E)\rho(E)$, where $\sigma(m \mid E)$ is the distribution of m across traps of given fixed energy E. We focus on the nonequilibrium dynamics after a quench at $t = 0$ from $T = \infty$ to $T < 1$; the initial condition is thus $P_0(E,m) = \sigma(m \mid E)\rho(E)$. The subsequent evolution is governed by

$$(\partial/\partial t)P(E,m,t) = -\tau^{-1}(E,m)P(E,m,t) + Y(t)\rho(E)\sigma(m \mid E), \qquad (4)$$

where the activation times are modified by a small field h conjugate to m as $\tau(E,m) = \tau(E)\exp(mh/T)$. Other choices of $\tau(E,m)$ that maintain detailed balance are possible [13,14]; we adopt this particular one because, in the spirit of the unperturbed model, it ensures that the jump rate between any two states depends only on the initial state, and not the final one.

The autocorrelation function of m (for $h = 0$) is

$$C(t,t_{\rm w}) = \int dm\, dm_{\rm w}\, dE\, dE_{\rm w}\, m m_{\rm w} P(E_{\rm w}, m_{\rm w}, t_{\rm w}) \times [P(E,m,t \mid E_{\rm w}, m_{\rm w}, t_{\rm w}) - P(E,m,t)] \qquad (5)$$

in which $P(E,m,t \mid E_{\rm w}, m_{\rm w}, t_{\rm w})$ is the probability that a particle with magnetization $m_{\rm w}$ and energy $E_{\rm w}$ at time $t_{\rm w}$ subsequently has m and E at time t. This obeys

$$P(E,m,t \mid E_{\rm w}, m_{\rm w}, t_{\rm w}) = \delta(m - m_{\rm w})\delta(E - E_{\rm w})e^{-(t-t_{\rm w})/\tau(E_{\rm w})} + \int_{t_{\rm w}}^t dt'\, \tau^{-1}(E_{\rm w})e^{-(t'-t_{\rm w})/\tau(E_{\rm w})}P(E, t - t')\sigma(m \mid E).$$

The terms corresponds to the particle not having hopped at all since t_w (first term) or having first hopped at t' (second term) into another trap; after a hop the particle evolves as if "reset" to time zero since it selects its new trap from the prior distribution $\sigma(m \mid E)\rho(E)$, which describes the initial state of the system. We also have $P(E, m, t) = \sigma(m \mid E) P(E, t)$ since at zero field the unperturbed dynamics is recovered. Substituting these relations into (5) and integrating over m and m_w, we obtain an exact integral expression for C. This is expressible as the sum of two components: The first depends only on the mean $\overline{m}(E)$ of the fixed energy distribution $\sigma(m \mid E)$ and the second only on its variance, $\Delta^2(E)$.

To find the corresponding response function, we proved

$$T \frac{\partial}{\partial t_w} \chi(t, t_w) = \frac{\partial}{\partial t} C(t, t_w) + \frac{\partial}{\partial t} \langle m(t) \rangle \langle m(t_w) \rangle \quad (6)$$

in which $\langle m(t) \rangle = \langle \overline{m}(E) \rangle_{P(E,t)}$ is the global mean of m. Equation (6) generalizes the results of [14,15] to the case of nonzero means $\langle m \rangle$; it is exact for any Markov process in which the effect of the field on the transition rate between any two states depends only on the initial state and not the final one. Substituting our expression for C into (6), and integrating over t_w with the boundary condition $\chi(t_w, t_w) = 0$, we find a closed expression for χ. This again comprises two components which depend separately on the mean and variance of $\sigma(m \mid E)$. For convenience, we rescale the field $h \to Th$, absorbing a factor $1/T$ into the response function. The slope of the FDT plot is then $-X = -T/T_{\mathrm{eff}}$ ($= -1$ in equilibrium).

Using our exact expressions, we numerically calculated C and χ for a number of different distributions $\sigma(m \mid E)$, each specified by given functional forms of $\overline{m}(E)$ and $\Delta^2(E)$. Each form effectively corresponds to a distinct physical identity of the observable m. For example, $\sigma(m \mid E) = \delta(m - E)$ implies $m = E$, in which case $h = -\delta T/T$. For simplicity, we confined ourselves to distributions either of zero mean (but nonzero variance) or of zero variance (but nonzero mean). As expected, we find that the decay of C (and growth of χ) depends not only on the time interval $t - t_w$, but also explicitly on the waiting time t_w, consistent with the nonequilibrium breakdown of TTI. For our generic observables m, the equal-time correlator $C(t, t)$ can also depend strongly on t; this is in contrast to the spin models often considered in mean field studies, where $C(t, t)$ is automatically normalized. While from (1) a plot of χ vs C, with t fixed and t_w as the parameter, still has slope $-X$ (with t as parameter this would no longer be guaranteed *a priori*), the variation of $C(t, t)$ will prevent a limit plot from being approached as we increase t. We therefore normalize and plot $\tilde{\chi} = \chi/C(t, t)$ vs $\tilde{C} = C/C(t, t)$; the normalization factor $C(t, t)$ is independent of t_w and the same for both axes, ensuring that the slope remains $-X$ as desired.

For the first class of observables [with zero mean $\overline{m}(E)$], we considered a variance $\Delta^2(E) = \exp(En/T)$ for various values of the exponent n, generalizing the results of [14] for $n = 0$. [For the weaker power law dependence $\Delta^2(E) = E^{2p}$ we can show analytically that the limiting FDT plot reduces to that for $n = 0$.] The equal-time correlator is then $C(t, t) = \int dE\, P(E, t) \exp(nE/T)$. For large t, its scaling behavior can be found using

$$P(E, t) \sim \begin{cases} t^{T-1} \exp(E/T - E) & \text{for } \tau(E) = e^{E/T} \ll t, \\ t^T \exp(-E) & \text{for } \tau(E) = e^{E/T} \gg t \end{cases}$$

[which follows from (3), using $Y(t) \sim t^{T-1}$ [11,12]]. For $n < T - 1$, the integral for $C(t, t)$ converges in a range $E = O(1)$, giving $C(t, t) \sim t^{T-1}$: The equal-time correlator is sensitive only to shallow traps (the population of which depletes in time as t^{T-1} due to ageing), and we thus expect the two-time correlator to decay on time scales $t - t_w = O(1)$, probing only quasiequilibrium behavior. In contrast, for $T - 1 < n < T$ the integrand for $C(t, t)$ has most of its weight at energies corresponding to $\tau(E) = O(t)$, yielding $C(t, t) \sim t^n$. For such n, the two-time correlator will decay on ageing time scales $t - t_w = O(t_w)$, and we expect strong violation of equilibrium FDT. (The regime $n > T$ is meaningless: It gives $C(t, t) = \infty\ \forall\ t$.)

Our numerical results confirm the above predictions. For values of $n < T - 1$ the normalized FDT plot approaches a straight line of equilibrium slope -1 at long times $t \to \infty$. In the regime $T - 1 < n < T$ (Fig. 1), equilibrium FDT is strongly violated, as expected; a limiting *nonequilibrium* FDT plot is nevertheless approached at long times. This can be proved analytically by showing that $\tilde{C}$ and $\tilde{\chi}$ share the same scaling variable $(t - t_w)/t_w$ in this limit. The slope of each plot varies continuously with $\tilde{C}$. In contrast to mean field, this is not due to an infinite hierarchy of time sectors; the variation is in fact continuous across the single time sector $t - t_w = O(t_w)$. More seriously, different observables give nontrivially different plots: at values of $\tilde{C}$ corresponding to a fixed $(t - t_w)/t_w$, the slopes $-X$ are *not* independent of n.

Next we considered a fixed energy distribution $\sigma(m \mid E)$ with zero variance and mean $\overline{m}(E) = \exp(En/2T)$. Here, too, we found that for $n < T - 1$ the limiting FDT plot

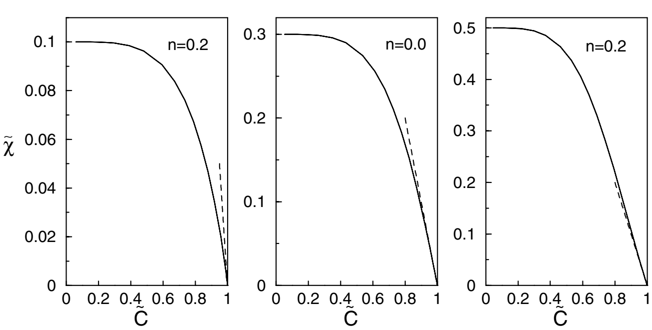

FIG. 1. FDT plots of $\tilde{\chi}$ vs $\tilde{C}$ for a distribution $\sigma(m \mid E)$ of variance $\exp(nE/T)$ (but zero mean) for $n = 0.2, 0.0, -0.2$; $T = 0.3$. For each n data are shown for times $t = 10^6, 10^7$; these are indistinguishable, confirming that the limiting FDT plot has been attained. Dashed line: The predicted asymptote $\tilde{\chi} = 1 - \tilde{C}$ for $t \to \infty$ and $\tilde{C} \to 1$.

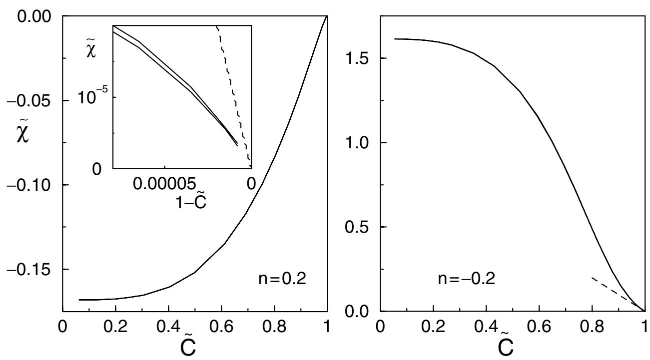

FIG. 2. FDT plots of $\tilde{\chi}$ vs $\tilde{C}$ for a distribution with mean $\exp(nE/2T)$ (but zero variance), for $n = 0.2, -0.2$; $T = 0.3$. Curves are shown for times $t = 10^6, 10^7$, but are indistinguishable except for the zoom shown in the inset (upper curve: $t = 10^7$). Dashed line: The predicted asymptote $\tilde{\chi} = 1 - \tilde{C}$ for $t \to \infty$, $\tilde{C} \to 1$.

is of equilibrium form, while for $T - 1 < n < T$ (Fig. 2) a nonequilibrium FDT plot is approached as $t \to \infty$, with a shape dependent (now very obviously) on the observable m. Finally, we considered a power law dependence of the mean, $\overline{m}(E) = E^p$; $p = 1$ corresponds to energy-temperature FDT. Interestingly, in this case no limiting plot exists. This is because the amplitude of the correlator remains finite as $t \to \infty$, while the response function, for any fixed value of $\tilde{C}$, diverges as $\ln t$.

In summary, we have studied FDT in the glass phase of the trap model, for a broad class of observables m. For observables which indeed probe the ageing regime, equilibrium FDT is violated; in most cases, a limiting nonequilibrium FDT plot is approached at long times. In contrast to mean field, this plot depends strongly on the observable. It furthermore has a slope which varies continuously across the single time sector $t - t_w = O(t_w)$. This shows that in this simple, paradigmatic model of glassy dynamics, the mean field concept of an FDT-derived effective temperature T_{eff} cannot be applied. One may dismiss the trap model as too abstract for this to be of general relevance, but by placing traps onto a d-dimensional lattice and allowing hops only to neighboring traps, a more "physical" model (of diffusion in the presence of disorder) can be obtained. Recent work [13] shows the scaling of correlation functions to be unaffected by this modification; since (6) would also continue to hold, our main results should be qualitatively unchanged.

Could the idea of a T_{eff} derived from FDT nevertheless be rescued in this and other nonmean field models? Consider first the nonuniqueness of our limiting FDT plots. One could argue that in order to probe an inherent T_{eff} characterizing the nonequilibrium dynamics, the statistical properties of the observable must not change significantly across the phase space regions visited during ageing. (In coarsening models, similar arguments have been used to exclude observables correlated with the order parameter [9].) Since in the trap model the typical trap depth E increases without bound for $t \to \infty$, a "neutral" observable would presumably require that $\overline{m}(E)$, $\Delta^2(E) \to$ const as $E \to \infty$; with this restriction we indeed find a unique limiting FDT plot. Its slope $-X$ will still vary continuously with $t - t_w$, however, excluding the link to a thermodynamically meaningful T_{eff}. Similarly "rounded" FDT plots have recently been found in coarsening models at criticality [16]; the limiting value $-X_\infty$ of the slope for $C \to 0$ was there shown to be a universal amplitude ratio. It is possible that at least this X_∞ could define a sensible T_{eff}, and in fact all our limiting FDT plots above share a common value $X_\infty = 0$. In conclusion, there is at least one class of "critical-like" models (which includes the trap model) for which a nonequilibrium T_{eff} can be defined at best from the asymptotic FDT X_∞, and possibly only for sufficiently neutral observables. It remains an open challenge to characterize this class more fully and to delineate it from those models which *appear* to exhibit mean fieldlike behavior in spite of their nonmean field nature [7–9].

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