Matching Microscopic and Macroscopic Responses in Glasses

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We first reproduce on the Janus and Janus II computers a milestone experiment that measures the spin-glass coherence length through the lowering of free-energy barriers induced by the Zeeman effect. Secondly, we determine the scaling behavior that allows a quantitative analysis of a new experiment reported in the companion Letter [S. Guchhait and R. Orbach, Phys. Rev. Lett. **118**, 157203 (2017)]. The value of the coherence length estimated through the analysis of microscopic correlation functions turns out to be quantitatively consistent with its measurement through macroscopic response functions. Further, nonlinear susceptibilities, recently measured in glass-forming liquids, scale as powers of the same microscopic length.

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Introduction.—It has long been suspected that the exceedingly slow dynamics that disordered and glassy systems (spin glasses, superspin glasses, colloids, polymers, etc.) exhibit upon cooling is due to the increasing size of the cooperative regions [1], which one would like to describe in terms of a correlation length ξ. The standard way of accessing ξ is measuring the structure factor in a neutron-scattering experiment. Unfortunately, this approach is unsuitable for experiments on glassy systems, because their structure factors show no trace of a growing length scale.

Yet, for example, for spin-glass systems, the replica method provides a microscopic approach to obtain the correlation functions of the overlap field [2–15], which decay with a correlation length ξ_{mic}. Unfortunately, these correlation functions are only easy to access through numerical simulations, since computing replicas requires direct access to the microscopic configurations.

In spite of the above difficulties it has been possible to develop effective techniques to measure ξ in real experiments. The state-of-the-art techniques are based on nonlinear responses to external perturbations. Very often these measurements are carried out in a nonequilibrium regime. If the temperature is low enough, ξ grows sluggishly but also indefinitely (unless the sample has a film geometry [16,17]). For spin glasses and superspin glasses, the magnetic response to an external magnetic field is accurately measured with a SQUID. A delicate analysis of this response yields a macroscopic correlation length, which we denote by ξ_{mac}, as a function of time. In the case of glass-forming liquids, one can study the dielectric polarizability.

Here we implement numerically, for the first time, on the Ising spin glass, the seminal experimental protocol introduced in [18], which is now a crucial protocol for spin-glass experiments [16,19]. Thanks to our dedicated computers

Janus [20] and Janus II [21], the system size and the time scales reached in our simulation allow us to assert the mutual consistency of the correlation lengths obtained through macroscopic response, $\xi_{\rm mac}$, and the length scale $\xi_{\rm mic}$ derived from the direct measurement of the overlap correlation function.

Our analysis unveils a scaling law describing how the magnetic response depends both on the applied magnetic field H and on the size $\xi_{\rm mic}$ of the magnetic domains. Remarkably, this scaling law is already very useful in the analysis of the experiment by Guchhait and Orbach described in the companion Letter [22].

The reader is probably aware of the long and ongoing controversy about the nature of the spin-glass phase. The replica symmetry breaking theory [23] predicts a spin-glass transition in a field [24], while the droplet model predicts that the magnetic field (no matter how small) avoids the transition [25–28]. In particular, the dynamics of a spin glass in a field has been analyzed within the context of the droplet model [29]. However, it has been difficult for experiments to distinguish both theories [18,30–33], because the two predict a barrier height that depends on the length scale $\xi_{\rm mic}$. Fortunately, our analysis completely avoids this controversy.

Finally, we link our results to the physics of glass-forming liquids through a study of the nonlinear susceptibilities χ_3 (see below). To date it has not been possible to reproduce the delicate experimental protocol of Ref. [18] for supercooled liquids or glasses. However, χ_3 [34] (and also χ_5 [35]) can be measured and do grow. We find that in our spin-glass simulation χ_3 has a well-defined scaling form as a power of $\xi_{\rm mic}$.

Model and protocol.—We study the Edwards-Anderson model in a three-dimensional, $D = 3$, cubic lattice of linear size L, with periodic boundary conditions. Our $N = L^D$ Ising spins, $\sigma_{\boldsymbol{x}} = \pm 1$, interact with their lattice nearest neighbors through the Hamiltonian

$$\mathcal{H} = -\sum_{\langle \boldsymbol{x},\boldsymbol{y}\rangle} J_{\boldsymbol{x},\boldsymbol{y}}\sigma_{\boldsymbol{x}}\sigma_{\boldsymbol{y}} - H\sum_{\boldsymbol{x}}\sigma_{\boldsymbol{x}}. \qquad (1)$$

The couplings $J_{\boldsymbol{x},\boldsymbol{y}}$ take the values ± 1 with 50% probability. In the absence of a magnetic field, $H = 0$, this model undergoes a spin-glass transition at the critical temperature $T_c = 1.102(3)$ [36]. The value of the dimensionless magnetic field H used in the numerical simulation can be matched to the physical one. For the Ising spin-glass $Fe_{0.5}Mn_{0.5}TiO_3$ we find $H_{\rm experimental} \approx 50\ {\rm kG} \times H$ [37]. This matching is likely to be strongly dependent on the material under consideration.

We describe succinctly our simulation protocol (for details see the analysis of the aging linear response in [40]). We consider a large system (with $L = 80$ or 160, large enough to avoid relevant finite-size effects). The initial random spin configuration is placed instantaneously at the working temperature $T = 0.7 \approx 0.64 T_c$ and left to relax for a time t_w, with $H = 0$. At time t_w, the magnetic field is turned on and we start recording the magnetization density, $m = \sum_{\boldsymbol{x}}\sigma_{\boldsymbol{x}}/N$. We write $m(t + t_w, t_w; H)$ to emphasize that the system is perennially out of equilibrium (and, hence, t_w dependent). In the following the symmetry under the inversion of the magnetic field, $m(t+t_w, t_w; H) = -m(t+t_w, t_w; -H)$, is crucial.

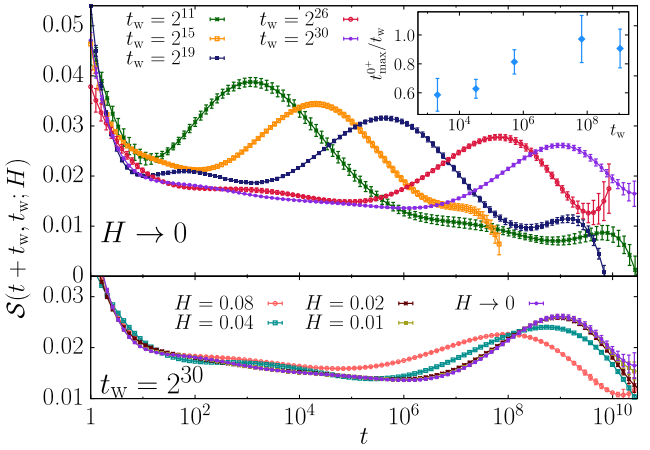

FIG. 1. The function $\mathcal{S}(t + t_w, t_w; H)$, Eq. (5), versus the time t elapsed after switching on the external magnetic field H. In the top panel we show the $H \to 0$ extrapolation for several waiting times t_w (one unit of computer time roughly corresponds to one picosecond of physical time [46]). Bottom: $\mathcal{S}(t + t_w, t_w; H)$ as a function of t for our largest waiting time $t_w = 2^{30}$ and for different values of H. Inset: The peak position ($H \to 0$), in units of t_w, depends on t_w only for $t_w < 10^6$.

Scaling.—As the system relaxes at the working temperature for a time t_w, the size of the glassy domains grows. The overlap correlation function $C_4(r, t_w)$ [41] decays with the distance r as $C_4(r, t_w) = f_c[r/\xi_{\rm mic}(t_w)]r^{-\theta}$ [3,10,11]. The cutoff function $f_c(x)$ decays faster than exponentially at large x. The exponent $\theta = 0.38(2)$ [11,42] is crucial in our analysis. The microscopic coherence length grows with time as $\xi_{\rm mic}(t_w) \propto t_w^{1/z(T)}$, with $z(T=0.7) = 11.64(15)$ [11].

In equilibrium conditions and for large $\xi_{\rm mic}$, there is a well-developed scaling theory for the magnetic response to an external field; see, e.g., [43,44]. However, dynamic scaling [45] suggests borrowing the equilibrium formulas, and replacing the equilibrium $\xi_{\rm mic}$ by the aging $\xi_{\rm mic}(t + t_w)$ (as obtained at $H = 0$). This bold approach has been successfully tested for spin glasses close to T_c [13,14] (and, to a small extent, also for glass-forming liquids [35]), thanks to the relation

$$m(t + t_w, t_w; H) = \xi_{\rm mic}^{y_h - D}\mathcal{F}(H[\xi_{\rm mic}(t + t_w)]^{y_h}, \mathcal{R}_{t,t_w}), \qquad (2)$$

where y_h is a scaling dimension that we now determine, $\mathcal{R}_{t,t_w} \equiv \xi_{\rm mic}(t + t_w)/\xi_{\rm mic}(t_w)$, and the scaling function $\mathcal{F}(x, \mathcal{R})$ is odd on its first argument for symmetry reasons. As we show below, see Fig. 1's inset, we are interested in the regime $t \approx t_w$ where the approximation $\mathcal{R}_{t,t_w} \approx 1$ is safe

[40]. Therefore, $\xi_{\rm mic}(t_w)$ is the relevant length scale from now on.

The (generalized) susceptibilities $\chi_1, \chi_3, \chi_5, \ldots$ are defined from the Taylor expansion

$$m(H) = \chi_1 H + \frac{\chi_3}{3!} H^3 + \frac{\chi_5}{5!} H^5 + \mathcal{O}(H^7), \qquad (3)$$

where we omitted the t and $t_{\rm w}$ dependencies of m and of the susceptibilities to simplify our notation. Matching Eqs. (2) and (3), we find the scaling behavior $\chi_{2n-1} \propto [\xi(t_w)]^{2y_h n - D}$. At least in equilibrium, χ_3 is connected to the space integral of the microscopic correlation function $C_4(r, t_w)$ [47]. We thus conclude that

$$2y_h = D - \frac{\theta}{2}. \qquad (4)$$

Taking θ from [11,42], we find $2y_h = 2.81(1)$. Although $2y_h$ is sometimes referred to as the fractal dimension of the glassy domains [6,19,35,48], we regard it as just a scaling dimension [49] (the droplet model prediction is $2y_h = D$).

Simulating the experiment.—The main quantity used in the experiment of [18] is

$$\mathcal{S}(t + t_w, t_w; H) = \frac{\partial}{\partial \log t} \left(\frac{m(t + t_w, t_w; H)}{H} \right). \qquad (5)$$

This quantity, shown in Fig. 1, has a local maximum at time $t^{(H)}_{\rm max}$. The time scale $t^{(H)}_{\rm max}$ was interpreted by Joh *et al.* as representative of the free-energy barriers $(t_w; H)$ that are relevant at time $t_{\rm w}$: $t^{(H)}_{\rm max} \propto \exp[\Delta / k_B T]$ [18] (see also the numerical computation in Ref. [29]).

$\mathcal{S}(t + t_w, t_w; H)$ depends on two time scales, t and t_w, as is typical of aging systems [57]. However, we want to use $\mathcal{S}$ to extract information from the single-time $\xi_{\rm mac}(t_w)$. The paradox is solved in the inset of Fig. 1, where we show that, when $t_{\rm w}$ is large enough, the ratio $t^{(0^+)}_{\rm max}/t_{\rm w}$ becomes independent of t_w: we are, in these conditions, in the asymptotic regime. This regime is also reached, at significantly shorter t_w, with Gaussian couplings [29].

The maximum $t^{(H)}_{\rm max}$ decreases upon increasing H; see Fig. 1—bottom. This reflects the lowering of the barriers Δ due to the Zeeman effect of the (glassy) magnetic domains [18]. From Eq. (2), and given the $H \leftrightarrow -H$ symmetry, it is natural to expect the Zeeman effect to be described through a smooth scaling function

$$\log \frac{t^{(H)}_{\rm max}}{t^{0^+}_{\rm max}} = F_{\rm Zeeman}(x), \qquad x = H^2 [\xi_{\rm mic}(t_w)]^{D-(\theta/2)}, \qquad (6)$$

where $t^{0^+}_{\rm max}$ is the extrapolation to $H = 0$ of $t^{(H)}_{\rm max}$. As Fig. 2 shows, this scaling holds for values of the scaling variable as large as $x \approx 6$: we have a very good scaling for close to 3 orders of magnitude. Up to that value, we find that the scaling function can be parametrized as $F_{\rm Zeeman}(x) = c_1 x + c_2 x^2$. In other words, for small H we expect the

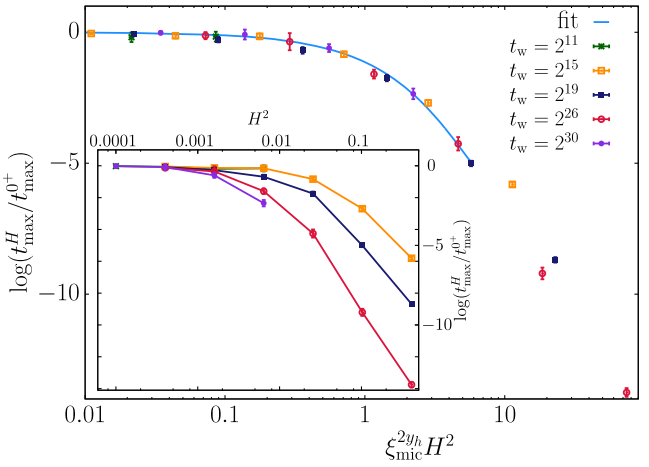

FIG. 2. The Zeeman energy follows the scaling form suggested in Eq. (6). We show a fit to $F_{\rm Zeeman}(x) = c_1 x + c_2 x^2$. Inset: the data of the main panel do not collapse when plotted as a function of H^2.

Zeeman energy to be proportional to H^2 with sizable corrections of order H^4. To the best of our knowledge, the explicit scaling form in Eq. (6) has never been used in the analysis of experimental data. Yet the authors of the original experiment [18] fitted their data at fixed $t_{\rm w}$ to

$$\log \frac{t^{(H)}_{\rm max}}{t^{0^+}_{\rm max}} = A N_f(t_w) H^2, \qquad (7)$$

where A is a $t_{\rm w}$-independent constant. $N_f(t_w)$ was interpreted as the number of spins in a correlated domain, and hence

$$\xi_{\rm mac}(t_{\rm w}) = [N_f(t_w)]^{1/D}. \qquad (8)$$

Equations (7) and (8) can be seen as the first-order expansion of Eq. (6). In fact, the smallness of exponent θ implies that the small correction $[\xi_{\rm mac}(t_w)]^{\theta/2}$ can easily go unobserved.

Figure 3 shows $\xi_{\rm mac}(t_{\rm w}) = [N_f(t_w)]^{1/(D-\theta/2)}$ [we obtained $N_f(t_w)$ from the fit to Eq. (7)]. Since different determinations of the correlation length should coincide only up to a multiplicative constant of order 1, we have not fitted for A, choosing instead $A = 1$. It is clear that $\xi_{\rm mac}(t_{\rm w})$ and $\xi_{\rm mic}(t_w)$ have the same behavior.

Finally, let us remark that in Ref. [58] it was suggested that Ising spin glasses should have a Zeeman energy of order H. On theoretical grounds, this is not possible for protocols respecting the symmetry $H \leftrightarrow -H$. However, we found that for $1 < x < 4$ a best fit to the form $F_{\rm Zeeman}(x) = d_1 + d_2 \sqrt{x}$ gives an acceptable value of χ^2, but one gets that $d_1 \neq 0$, which implies an unphysical value for the $H \to 0$ extrapolation. Only a careful control of the limit of vanishing field (see the companion Letter by Guchhait and Orbach [22]) reveals that the true behavior for small H is proportional to H^2. In practice, the transient behavior

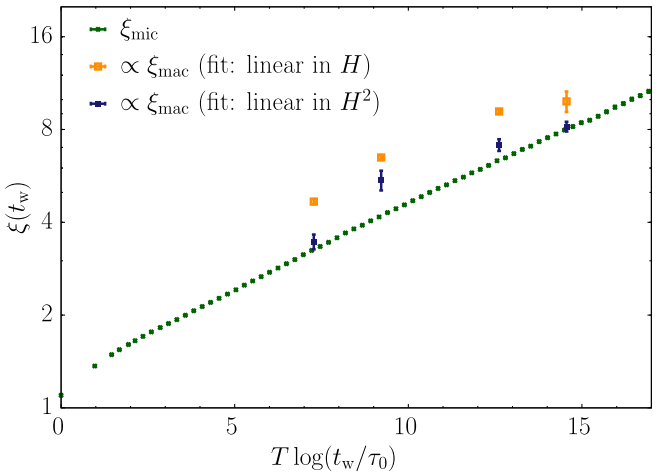

FIG. 3. The time growth of the correlation length $\xi_{\rm mic}$, as obtained from the microscopic correlation function $C_4(r, t_w)$ [10,11,40], is compared to the length $\xi_{\rm mac}$ obtained from a fit linear in H^2; see Eq. (7). The microscopic time scale $\tau_0 = 1$ corresponds to a single lattice sweep in our Monte Carlo simulation (see the caption to Fig. 1). We also show the results obtained with Eq. (9), which are sensible as well. The temperature-dependent scaling variable, $T\log(t_w/\tau_0)$, is common in the experimental literature (e.g., see Ref. [19]).

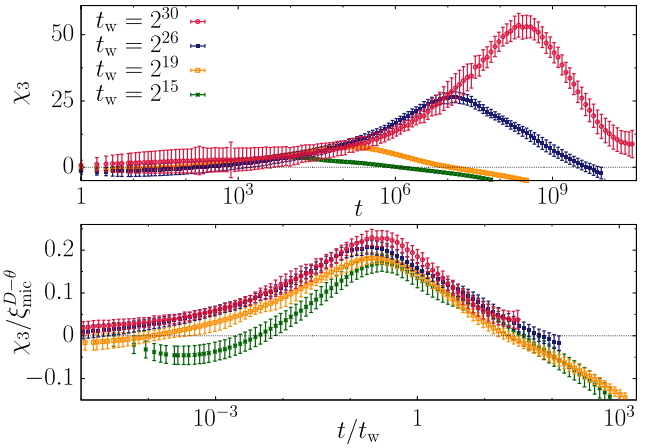

FIG. 4. The nonlinear susceptibility χ_3 is shown as a function of t for several values of t_w (top), as obtained from Eq. (3). The difficulty lies in balancing systematic errors (numerical data obtained with high fields underestimate χ_3) with statistical errors (which are larger for small values of H). Our compromise, shown here, tries to obtain statistical and systematic errors of comparable size. In the bottom panel we show the $G(t/t_w)$ scaling function (10). Note that scaling corrections are visible only for the smallest waiting times (and, even in those cases, they only appear for small t/t_w).

of $F_{\rm Zeeman}(x)$ implies that one could fit the data to the form

$$\log \frac{t_{\rm max}^{(H)}}{t_{\rm max}^{0^+}} = A' \sqrt{N_f(t_{\rm w}) H^2}, \tag{9}$$

and then extract $\xi_{\rm mac}(t_w) = [N_f(t_w)]^{1/(D-\theta/2)}$ (again, $A' = 1$). Although Eq. (9) is incorrect for small values of H, the scaling law Eq. (6) implies that one still obtains a reasonable determination of $\xi_{\rm mac}$, as we indeed find [see Fig. 3, where we also show $\xi_{\rm mac}(t_w)$ obtained from this approach].

Nonlinear susceptibilities.—At variance with spin glasses [18], the detection of a large correlation length accompanying the glass transition is still an open problem for supercooled liquids [59]. It is now clear that linear responses are not up to the task [34,60], so higher-order nonlinear responses are currently under investigation [34,35,61]. However, even in the more familiar context of spin glasses the connection between $\chi_3(t + t_w; t_w)$ and $\xi_{\rm mic}(t_w)$ needs to be clarified.

To make some progress, we extract generalized susceptibilities such as χ_3 through Eq. (3). Figure 4 (top) shows that $\chi_3(t + t_w, t_w)$ has a t_w-independent regime for $t \ll t_w$ (the time-translational invariant regime [57]; see also [62]). Yet, it displays a peak as a function of t, whose position and height are strongly t_w dependent. In fact, we empirically find [see Fig. 4 (bottom)] the following scaling behavior for large enough values of t and t_w:

$$\chi_3(t + t_w, t_w) = [\xi_{\rm mic}(t_w)]^{D-\theta} G(t/t_w). \tag{10}$$

The prefactor $[\xi_{\rm mic}(t_w)]^{D-\theta}$ follows from Eqs. (2)–(4). Deriving the details of the function $G(t/t_w)$ requires further work.

Conclusions.—Using the dedicated computers Janus and Janus II, we have studied the aging magnetic response of an Ising spin glass to an applied field. In this way, we have simulated a milestone experiment [18], and we have shown that the glassy correlation length extracted from this macroscopic response is numerically consistent with its microscopic determination from overlap correlation functions. Furthermore, we have unveiled scaling laws that relate the magnetic response to the applied field and the correlation length. We expect that this scaling analysis will be useful in future experiments on film geometry. Our scaling analysis has been relevant for the study of the experiment reported in the companion Letter [22]. The agreement with experiments is even more impressive when one notices that we are comparing numerical time scales of the order of the millisecond to experimental time scales of the order of the hour: this looks like a very nice piece of evidence for invariance in time scales.

Although the delicate experimental study of Ref. [18] has not yet been carried out for glass-forming liquids, the dielectric polarizability analogue of the nonlinear susceptibilities is measured in current experiments [34,35]. We have shown that these susceptibilities scale as powers of the microscopically determined correlation lengths.

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