# Ergodic versus nonergodic behavior in oxygen-deficient high-$T_c$ superconductors

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The oxygen-defect-induced phase transition from nonergodic to ergodic state in superconductors with intragrain granularity is considered within the superconductive-glass model. The model predictions are found to be in qualitative agreement with some experimental observations in deoxygenated high-$T_c$ single crystals.

## I. INTRODUCTION

According to recent findings (see, e.g., Refs. 1–8 and references therein), high-$T_c$ superconductors (HTS) exhibit an anomalous (nonclassical[8]) magnetic-field behavior, which has been attributed to the field-induced intragrain granularity in oxygen-deficient samples and interpreted in terms of the field-induced decoupling of regions of oxygen-rich material by boundaries of oxygen-poor material. A phase diagram $H_m(\delta,T)$, that demarcates the multigrain onset as a function of temperature and oxygen deficiency $\delta$, was found[3] to confirm that oxygen-deficient single crystals exhibit behavior characteristic of homogeneous superconductors for $H<H_m$ and inhomogeneous superconductors for $H>H_m$. The granular behavior for $H>H_m$ has been related to the clusters of oxygen defects (within the CuO plane) that restrict supercurrent flow and allow excess flux to enter the crystal. The observed $H_m(\delta,T)$ data were described by a two-dimensional (2D) percolation model for oxygen defects. It means that there exists a critical oxygen deficiency $\delta_c$ above which there are no continuous current paths. For $\delta$ greater than $\delta_c$, oxygen-rich superconducting grains are separated by oxygen-poor insulating boundaries so that there is no superconducting path through a sample. For $\delta$ less than $\delta_c$, a complete current path spans the sample and resistance measurements show metallic behavior with a superconducting transition. Since $H_m(\delta,T)$ signals the onset of granularity, a sample with $H_m(\delta,T)=0$ implies that the crystal has so many oxygen defects that it never exhibits single-grain behavior.[3]

The aim of the present paper is to show how the lack of oxygen in HTS materials inspires the phase transition from the nonergodic (in nearly fully oxygenated crystals) to the ergodic (in highly oxygen-depleted crystals) state within the so-called superconductive-glass (SG) model (see, e.g., Refs. 9–15 and references therein), and to compare the model predictions with some experimental data for deoxygenated HTS single crystals. More specifically, the nonergodic (phase-coherent) state is attributed to nontrivial equilibrium (long-time) behavior of the defect-free crystal (with $\delta\simeq 0$) [which is characterized by a nontrivial order parameter $L(\delta,T,H)\neq 0$; see below], while the ergodic (paracoherent) phase corresponds to the equilibrium state of the defected crystal (when $\delta\simeq\delta_c$) [with $L(\delta,T,H,)=0$].

## II. THE MODEL

The SG model is based on the well-known Hamiltonian of a granular superconductor which in the so-called pseudospin representation has the form[9–16]

$$\mathcal{H}_0=-\sum_{ij}^{N}J(\delta,T)\cos\phi_{ij}(\mathbf{H})\equiv-\mathrm{Re}\left\{\sum_{ij}^{N}J_{ij}S_i^{+}S_j^{-}\right\}, \quad (1)$$

where

$$J_{ij}(\delta,T,\mathbf{H})=J(\delta,T)\exp[iA_{ij}(\mathbf{H})]\,,$$
$$\phi_{ij}(\mathbf{H})=\phi_i-\phi_j-A_{ij}(\mathbf{H})\,, \quad (2)$$
$$A_{ij}(\mathbf{H})=\frac{\pi}{\phi_0}(\mathbf{H}\times\mathbf{R}_{ij})\mathbf{r}_{ij}\,,$$

$$\mathbf{r}_{ij}=\mathbf{r}_i-\mathbf{r}_j\,,\quad \mathbf{R}_{ij}=(\mathbf{r}_i+\mathbf{r}_j)/2\,. \quad (3)$$

This model describes the infinite-range interaction between oxygen-rich superconducting grains [with phase $\phi_i(t)$ or Josephson pseudospins $S_i^{+}=\exp(+i\phi_i)$], arranged in a random two-dimensional lattice (modeling the CuO plane of oxygen-depleted $YBa_2Cu_3O_{7-\delta}$, where a glasslike picture is established[1–7]) with coordinates $\mathbf{r}_i=(x_i,y_i,0)$. The grains are separated by oxygen-poor insulating boundaries producing Josephson coupling with energy $J(\delta,T)$. The system is under the influence of a frustrating applied magnetic field $\mathbf{H}$, which is assumed to be normal to the CuO plane of HTS's. The increase of the oxygen deficiency $\delta$, leads to the decrease of the Josephson energy (via the increase of the insulating layer between oxygen-rich grains). For small $\delta$ (such that $\delta\ll 1$) we can approximate the $\delta$ dependence of the Josephson energy by a linear law,[4] namely $J(\delta,T)\approx J(0,T)(1-\delta)$. The superconducting current through the Josephson junction (JJ) between grains $i$ and $j$,

$$I_{ij}^{s}(\mathbf{H})=\frac{2eJ}{\hbar}\sin\phi_{ij}(\mathbf{H})\equiv\frac{2e}{\hbar}\mathrm{Im}\{J_{ij}S_i^{+}S_j^{-}\}\,, \quad (4)$$

induces a diamagnetic moment of the weak-link network[9–13]

 

$$\boldsymbol{\mu}=\pi\sum_{ij}^{N} I_{ij}^{s}(\mathbf{H})(\mathbf{r}_{ij}\times\mathbf{R}_{ij}) . \tag{5}$$

To study dynamic (relaxation) behavior of the model (1), let us assume that in addition to the constant frustrating field $\mathbf{H}$, the superconducting grains are under the influence of a small time-varying field $\mathbf{H}_1(t)\ll\mathbf{H}$, so that

$$\cos\{\phi_i-\phi_j-A_{ij}[\mathbf{H}+\mathbf{H}_1(t)]\} \cong\cos\phi_{ij}(\mathbf{H})+A_{ij}[\mathbf{H}_1(t)]\sin\phi_{ij}(\mathbf{H}) .$$

In view of Eqs. (1)–(5), the total (perturbed) Hamiltonian can be cast into the form

$$\mathcal{H}(t)=\mathcal{H}_0(\mathbf{H})-\boldsymbol{\mu}\mathbf{H}_1(t) . \tag{6}$$

If the perturbation is applied continuously from $t=-\infty$ up to $t=0$ and is cut off at $t=0$, then the linear [with respect to the small perturbation field $\mathbf{H}_1(t)=\mathbf{H}_1\theta(-t)$] response $M(t)\equiv\overline{\langle\mu_z\rangle}/V$ will relax to its equilibrium value $M_{\rm eq}\equiv\lim_{t\to\infty}M(t)$ according to the formula[17]

$$M(t)-M_{\rm eq}=\int_{-\infty}^{t}dt'G(t-t')\mathbf{H}_1(t')=\int_{t}^{\infty}dt'G(t')\mathbf{H}_1 . \tag{7}$$

According to the fluctuation-dissipation theorem,[17] the response function $G(t)$ is related to the relaxation function $\Phi(t)$ as follows: $G(t)=-(\partial/\partial t)\Phi(t)$. Thus the above equation reads

$$M(t)-M_{\rm eq}=\frac{1}{V}H_1[\Phi(t)-\Phi(\infty)] , \tag{8}$$

where[17]

$$\Phi(t)=\beta\overline{\langle\mu_z(t)\mu_z(0)\rangle} . \tag{9}$$

Here $\beta=1/k_BT$, the bar denotes the configurational averaging over the randomly distributed grain coordinates (see Appendix A), $\langle\cdots\rangle$ means the thermodynamic averaging with the Hamiltonian $\mathcal{H}_0(\mathbf{H})$, and we have assumed that $\mathbf{H}=(0,0,H)$ and $\mathbf{H}_1=(0,0,H_1)$. Therefore the function $\Phi(t)$ describes the relaxation of magnetization $M(t)$ after removal of the outer disturbance. As a result of configurational averaging, the relaxation of magnetization can be approximated by the formula (see Appendix A)

$$M(t)=M(\delta,T,H,H_1)|D(t)|^2 , \qquad M(\delta,T,H,H_1)=\chi(\delta,T,H)H_1 , \tag{10}$$

where

$$\chi(\delta,T,H)\equiv\frac{16e^2s^2N^2J^2(\delta,T)}{k_BTV\hbar^2}\left[\frac{H}{H_0}\right]^2\left[1+\frac{H^2}{H_0^2}\right]^{-4} . \tag{11}$$

Here $H_0=\phi_0/s$ is a characteristic Josephson field with $s=\pi d^2$ an average JJ projection area, and $N$ is the number of grains. Thus, all information about the dynamic (relaxation) properties of the system is contained in the time-dependent correlator $D(t)\equiv(1/N)\sum_{ij}D_{ij}(t)$ which is defined as follows (see Appendix A):[7,12–16]

$$D_{ij}(t)=\overline{\langle S_i^{+}(t)S_j^{-}(0)\rangle} . \tag{12}$$

## III. DISCUSSION

As is well-known,[18] there can be many types of long-time behavior of $D(t)$. Two of them are of particular interest:

$$\lim_{t\to\infty}D(t)=L(T,H)\neq0 \quad (\mathrm{I}) \tag{13}$$

and

$$\lim_{t\to\infty}D(t)\propto\exp(-t/\tau) \quad (\mathrm{II}) . \tag{14}$$

A simple example of a system belonging to class I (the so-called nonergodic state) has been discussed by de Gennes and Tinkham.[19] They considered the long-time behavior of $D(t)$ for a superconducting thin film (of thickness $a$) with diffuse reflection on the boundaries, in a parallel magnetic field. When no volume defects are taken into account (pure limit), the system was found to exhibit a nonergodic behavior with field-dependent nonergodicity parameter $L(T,H)$, namely,

$$L(T,H)\cong\begin{cases}1-(\pi Ha^2/3\phi_0) , & H\ll(\phi_0/a^2) ,\\ (2\phi_0/\pi Ha^2)^2 , & H\gg(\phi_0/a^2) .\end{cases} \tag{15}$$

At the same time, the presence inside a sample of a few scattering centers (which are characterized by a mean free path $l$, $l>a$) inspires the transition of the system from the nonergodic (class I) to the ergodic (class II) state with the (inverse) relaxation time ($v_F$ is the Fermi velocity)

$$\frac{1}{\tau}=(v_Fa)\left[\frac{\pi Ha}{4\phi_0}\right]^2 . \tag{16}$$

Turning to the HTS single crystals, let us consider dynamic (relaxation) and equilibrium properties of the magnetization versus oxygen defect concentration within the SG model. By analogy with the case of slightly defected thin films, considered by de Gennes and Tinkham,[18] we assume that up to some critical value of oxygen deficiency, $\delta_g$, HTS single crystals exhibit nonergodic (phase-coherent) behavior, while for oxygen-defect concentration greater than $\delta_g$, the above-mentioned coherence (within the CuO plane) is destroyed and the crystal undergoes a phase transition to the ergodic (paracoherent) state where oxygen-rich superconducting grains are separated by oxygen-poor insulating boundaries so that there is no superconducting path through the sample. It is worthwhile to mention that the related problem of the annealed Ising magnet on percolation clusters has been recently considered by Kaufman and Touma.[20] Using the renormalization group method, three phases on the corresponding phase diagram have been identified:[20] percolating ferromagnetic, percolating paramagnetic, and nonpercolating paramagnetic.

In view of Eqs. (10)–(13), the equilibrium magnetization $M_{\rm eq}(\delta,T,H,H_1)$ is the limit

$$M_{\rm eq}(\delta,T,H,H_1)\equiv\lim_{t\to\infty}M(t)=M(\delta,T,H,H_1)L^2(\delta,T,H)\ . \tag{17}$$

Here $L(\delta,T,H)$ is the order parameter of the SG model, which is defined via the correlator $D(t)$ according to Eq. (13). To find the long-time (low-frequency) behavior of the correlator $D(t)$ [and thus of the magnetization $M(t)$], we need the equation of motion for the Josephson pseudospins $S_i^{\pm}(t)$. An approximate (valid for $N\gg1$) equation of motion reads[7,12–16] (see Appendix B)

$$\dot S_i^{+}=\beta\Omega\sum_j^N J_{ij}S_j^{+}\ . \tag{18}$$

Here $\Omega=2e^2R/\beta\hbar^2N$ is a characteristic frequency of the JJ network with $R$ being the resistance between grains in their normal state. In the so-called mode-coupling approximation,[21] $D(t)$ obeys the self-consistent master equation (see Appendix B)

$$\frac{d^2D(t)}{dt^2}+\Omega^2D(t)+\int_0^t dt'K(t-t')\frac{dD(t')}{dt'}=0\ , \tag{19}$$

with $K(t)\equiv(1/N)\sum_{ij}K_{ij}(t)$ being a memory (feedback) kernel. When there is no temporal correlation between grains (paracoherent state) the memory kernel has a white noise form $K(t)\equiv K_r(t)=2\Omega\delta(t)$, where $\delta(t)$ is the Dirac delta function. In this case the master equation results in a Debye-like decay of the uncorrelated paracoherent state, namely, $D(t)=\exp(-t/\tau)$, where $1/\tau=\Omega$. Such a situation is realized above some critical (phase-locking) temperature $T_g$ when the coherent state within the JJ network is destroyed completely, so that the order parameter $L\equiv0$. Below $T_g$, the situation changes drastically due to the superconducting correlations occurring between grains. For $N\gg1$, the coherent part of the memory kernel, $K_{ij}^c(t)$, can be approximated by the current-current correlator $K_{ij}^c(t)\cong\overline{\langle\dot S_i^{+}(t)\dot S_j^{-}(0)\rangle}$. Taking into account the equation of motion (18), the memory kernel below $T_g$ can be presented in the form (see Appendix B)

$$K(t)\equiv K_r(t)+\frac{1}{N}\sum_{ij}^N K_{ij}^c(t)=2\Omega\delta(t)+\Omega_{\rm coh}^2(\delta,T,H)D(t)\ . \tag{20}$$

Here $\Omega_{\rm coh}(\delta,T,H)=\beta\Omega J(\delta,T,H)$ and the field dependence of the Josephson energy is defined as follows (see Appendix A):

$$J(\delta,T,H)\equiv\overline{J_{ij}(\delta,T,H)}=J(\delta,T)\left[1+\frac{H^2}{H_0^2}\right]^{-1}\ . \tag{21}$$

In view of Eq. (13), a zero-frequency ($t\to\infty$) solution of the master Eq. (19) with the memory kernel (20) results in the nontrivial order parameter for the intragranular JJ network[7,12–16]

$$L(\delta,T,H)=1-\left[\frac{k_BT}{J(\delta,T,H)}\right]^2\ . \tag{22}$$

The phase-locking temperature $T_g(\delta,H)$, below which the ensemble of grains undergoes the phase transition into the coherent state, is defined by the equation $L(\delta,T_g,H)=0$, which, due to Eq. (22), gives rise to an implicit equation, viz. $T_g(\delta,H)=J(\delta,T_g,H)/k_B$. The Josephson energy depends on the temperature through the Ambegaokar-Baratoff relation, which near the single-grain superconducting temperature $T_c$ reads $J(T)\approx J(0)(1-T/T_c)$. Assuming that for high magnetic fields (when frustration is strong enough)

$$J(\delta,0,H)\ll k_BT_c\le J(\delta,0,0)\ ,$$

we get finally $T_g(\delta,H)\approx J(\delta,0,H)/k_B$. As a result, the order parameter $L=1-[T/T_g(\delta,H)]^2$ gradually changes from 0 at $T\ge T_g(\delta,H)$ to 1 at $T=0$, thus describing a continuous phase transition.

By analogy with the critical (phase-locking) temperature $T_g(\delta,H)$, we can introduce the critical field $H_g(\delta,T)$ as the solution of the equation $L(\delta,T,H_g)=0$. In view of the field dependence of the order parameter [see Eqs. (21) and (22)], the critical field reads

$$H_g(\delta,T)=H_0\sqrt{1-T/T_g(\delta,0)}\ . \tag{23}$$

Taking into account the $\delta$ dependence of the phase-locking temperature, $T_g(\delta,H)\simeq T_g(0,H)(1-\delta)$, Eq. (23) results in the following oxygen-deficiency behavior of the critical field:

$$H_g(\delta,T)=H_0\sqrt{\delta_g(T,0)-\delta}\ . \tag{24}$$

Here we have introduced the critical oxygen deficiency $\delta_g(T,H)$, which is defined as the solution of the equation $L(\delta_g,T,H)=0$ and has the form $\delta_g(T,H)=1-T/T_g(0,H)$. The physical meaning of this critical parameter is as follows. For $\delta\ge\delta_g(T,H)$ oxygen-rich superconducting grains are separated by oxygen-poor insulating boundaries so that there is no percolative path through the sample. Notice that, within the SG model, $H_g(\delta,T)$ in fact plays the role of the phase-boundary field $H_m(\delta,T)$ discussed by Osofsky *et al.*[3]

It is important to mention that the correlator $D(t)$ follows a simple Debye-like decay law only above $T_g(\delta,H)$, i.e., when the system of grains is in the ergodic state (see above). Below $T_g$ (where the order parameter $L\neq0$), relaxation of $D(t)$ [with $\mathrm{Im}\{D(t)\}=0$] can be presented in the form[13–16]

$$D(t)=L+(1-L)\tilde\Phi(t)\ . \tag{25}$$

The relaxation function $\tilde\Phi(t)$ is supposed to be normalized, viz.

$$\frac{1}{\tau}\int_0^\infty dt\,\tilde\Phi(t)=1\ , \tag{26}$$

and obeys the following boundary conditions: $\tilde\Phi(0)=1$ and $\tilde\Phi(\infty)=0$, i.e., $D(0)=1$. Of course, in principle, one can find $D(t)$ as a numerical solution of the master equation. But it seems more interesting to try and get some analytical results concerning the time behavior of $D(t)$. It is natural then to consider a simple generalization of the Debye law in the form of the so-called Kohlrausch

stretched exponential law

$$\tilde{\Phi}(t)=\exp\left[-\left[\frac{t}{\tau}\right]^{\alpha}\right], \tag{27}$$

where $\alpha(\delta,T,H)\leq 1$. Substitution of Eqs. (25)–(27) into Eq. (19) with the kernel (20) results in an implicit equation on the power exponent $\alpha(\delta,T,H)$

$$\Gamma\left[1+\frac{1}{\alpha}\right]=1+\frac{L}{2(1-L)} . \tag{28}$$

Here $\Gamma$ is the Gamma function. Near $T_g(\delta,H)$ the approximate solution of Eq. (28) gives

$$\alpha(\delta,T,H)\approx 1-L(\delta,T,H) . \tag{29}$$

It is worthwhile to mention that for the short-time limit, when $(t/\tau)^{\alpha}\ll 1$, the above Kohlrausch law (27) leads to the nonlogarithmic relaxation law for magnetization (see Appendix B)

$$M(t)=M_{\rm eq}[1-2s_{\alpha}\ln(t/\tau)+s_{\alpha}^{2}\ln^{2}(t/\tau)] . \tag{30}$$

In this approximation, the $\alpha$-relaxation rate $s_{\alpha}(\delta,T,H)$ is expressed via the Kohlrausch exponent $\alpha(\delta,T,H)$ and the order parameter $L(\delta,T,H)$

$$s_{\alpha}(\delta,T,H)=\left[\frac{1-L}{L}\right]\alpha . \tag{31}$$

In turn, $s_{\alpha}(\delta,T,H)$ is related to the activation energy $U(\delta,T,H)$ as follows: $s_{\alpha}=k_BT/U$. Thus, in view of Eqs. (29)–(31) the $\delta$, temperature, and field dependencies of the activation energy in the JJ network are effective via the corresponding dependencies of the order parameter $L(\delta,T,H)$; namely, near $T_g(\delta,H)$ the activation energy reads $U(\delta,T,H)\approx k_BTL(\delta,T,H)$. That is,

$$U(\delta,T,H)/2k_BT\approx 1-T/T_g(\delta,H)\approx\delta_g(T,H)-\delta \approx 1-H/H_g(\delta,T) ,$$

in at least qualitative agreement with what have been really observed in oxygen-depleted HTS single crystals.[4,5] It is interesting to notice that an expression similar to our Eq. (30) has been used by Sengupta *et al.*[22] to describe a nonlogarithmic relaxation in HTS single crystals.

In view of the explicit dependence of the order parameter on the oxygen deficiency, namely, $L(\delta,T,H)\approx 2[\delta_g(T,H)-\delta]$, Eq. (29) describes the restoration of the ergodic state in the system under study when the oxygen deficiency $\delta$ reaches its critical value $\delta_g(T,H)$. Indeed, when $\delta\to\delta_g(T,H)$, the order parameter $L\to 0$, and the Kohlrausch exponent $\alpha\to 1$ [see Eq. (29)] which means that relaxation becomes faster [formally, according to Eq. (31) the logarithmic relaxation rate $s_{\alpha}\to\infty$] and follows the ordinary Debye law [see Eq. (27)]. At the same time, the activation energy between grains declines (i.e., $U\to 0$) as $\delta\to\delta_g(T,H)$. To make our discussion more quantitative, let us consider some estimates of the model parameters. Using the experimental results for the phase-boundary field $H_g(\delta,T)$ obtained by Osofsky *et al.*[3] for fixed values of $\delta$ and $T$, namely,

$$H_g(\delta=0.06,\ T=60\ \text{K})\approx 1.5\ \text{T} ,$$

$$H_g(\delta=0.13,\ T=60\ \text{K})\approx 1\ \text{T} ,$$

and

$$H_g(\delta=0.13,\ T=70\ \text{K})\approx 0.4\ \text{T} ,$$

Eqs. (23) and (24) allow us to get estimates for the phase-locking temperature $T_g(\delta,H)$ and the critical value of the oxygen deficiency $\delta_g(T,H)$. The result is $T_g(\delta=0,\ H=0)\approx 75$ K, $T_g(\delta=0.13,H=0)\approx 72$ K, and $\delta_g(T=60\ \text{K},\ H=0)\approx 0.21$. Finally, using the above results, Eq. (24) brings about an estimate for the characteristic Josephson field of $H_0=\phi_0/s\approx 5$ T which gives a reasonable value of oxygen-ion scattering cross section[3] $s\approx 4\times 10^{-16}$ m$^2$. On the other hand, making use of the above-obtained estimates we can estimate the value of the activation energy

$$U(\delta,T,H)\approx 2k_BT(1-H/H_g(\delta,T)) .$$

For $H=1$ T, $\delta=0.06$, and $T=60$ K, we get $U/k_B\approx 40$ K, which reasonably agrees with the value deduced by Ossandon *et al.*[5] from $YBa_2Cu_3O_{7-\delta}$ single-crystal measurements.

In summary, the oxygen-defect-induced phase transition from the nonergodic (in nearly fully oxygenated crystals) to the ergodic (in highly deoxygenated crystals) state in HTS oxygen-depleted crystals has been considered within the superconductive-glass model. Both dynamic (relaxation) and equilibrium properties of the model magnetization were found to correlate quite reasonably with some experimental data on deoxygenated HTS's.

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## APPENDIX A

To get Eqs. (10) and (11) for the relaxation of magnetization, we have to calculate the relaxation function $\Phi(t)$. Using the so-called random-field approximation for quenched disordered systems,[10,16,23] which allows to decouple the averaging of the grain distribution (represented by the scattering potentials $J_{ij}$) from the carriers (or Josephson pseudospins), i.e., assuming that $\overline{A(r_i)B(r_j)}\cong\overline{A(r_i)}\,\overline{B(r_j)}$, we obtain from Eqs. (2)–(4) and (9)

$$\Phi(t)\cong-\frac{\pi^2e^2\beta J^2}{\hbar^2}\sum_{ij}^{N}\sum_{kl}^{N}\{\overline{\exp[i(A_{ij}+A_{kl})](x_iy_i-x_jy^i)(x_ky_l-x_ly_k)}\,\overline{\langle S_i^{+}(t)S_j^{-}(t)S_k^{+}(0)S_l^{-}(0)\rangle} -\overline{\exp[i(A_{ij}-A_{kl})](x_iy_j-x_jy_i)(x_ky_l-x_ly_k)}\,\overline{\langle S_i^{+}(t)S_j^{-}(t)S_k^{-}(0)S_l^{+}(0)\rangle}\}+\text{H.c.} \tag{A1}$$

To proceed further, we have to calculate the four-spin correlators appearing in the right-hand side of the above equation. Taking into account only pair correlations, namely, assuming that (within the mean-field approximation[10,16,23] when $N \gg 1$), e.g.,

$$\overline{\langle S_i^+(t)S_j^-(t)S_k^+(0)S_l^-(0)\rangle} \cong \overline{\langle S_i^+(t)S_l^-(0)\rangle\langle S_j^-(t)S_k^+(0)\rangle} ,$$

we can rewrite Eq. (A1) as follows:

$$\Phi(t) \cong \frac{\phi_0^2 e^2 \beta J^2}{\hbar^2} \sum_{ij}^{N} \sum_{kl}^{N} \left[ \frac{\partial}{\partial H} \overline{\exp(iA_{ij})} \right] \left[ \frac{\partial}{\partial H} \overline{\exp(iA_{kl})} \right] \times \{ D_{il}(t) D_{jk}^*(t) + D_{ik}(t) D_{jl}^*(t) \} + \text{H.c.} \tag{A2}$$

Here we have introduced the spin-spin correlator [cf. Eq. (12)]

$$D_{ij}(t) = \overline{\langle S_i^+(t) S_j^-(0)\rangle} , \quad \mathrm{Im}\{D_{ij}(t)\} = 0 , \tag{A3}$$

and made use of the fact that due to Eq. (3)

$$\overline{(x_i y_j - x_j y_i)\exp(iA_{ij})} = \left[ \frac{i\phi_0}{\pi} \right] \frac{\partial}{\partial H} \overline{\exp(iA_{ij})} . \tag{A4}$$

The frustration-field dependence of the magnetization essentially depends on the choice of a random distribution function $P(\mathbf{r}_i)$ as well as on the type of disorder.[9–15] To obtain the explicit form of the field dependence of magnetization given by Eq. (11), we have assumed, for simplicity, a site-type positional disorder allowing for weak displacements of the grain sites from their positions on the original 2D lattice, i.e., within a radius $d$ the new position is chosen randomly according to the normalized separable Gaussian distribution function $P(\mathbf{r}_i) = P(x_i)P(y_i)$, where

$$P(x) = \frac{1}{\sqrt{2\pi d^2}} \exp\left[ -\frac{x^2}{2d^2} \right] . \tag{A5}$$

Using the above distribution function, we can calculate the configurational averages appearing in Eq. (A2). In particular, the average value of the Josephson energy [see Eq. (2)] reads

$$J(\delta, T, H) \equiv \overline{J_{ij}(\delta, T, H)} = J(\delta, T)\overline{\exp(iA_{ij})} , \tag{A6}$$

where

$$\overline{\exp(iA_{ij})} \equiv \int_{-\infty}^{+\infty} d\mathbf{r}_i d\mathbf{r}_j P(\mathbf{r}_i) P(\mathbf{r}_j) e^{(i\pi H/\phi_0)(x_i y_i - x_j y_i)} = \left[ 1 + \frac{H^2}{H_0^2} \right]^{-1} . \tag{A7}$$

Here $d\mathbf{r}_i = dx_i dy_i$ and $H_0 = \phi_0/\pi d^2$. Finally, taking into account Eqs. (A3)–(A7), we arrive at Eqs. (10) and (11) for the magnetization $M(t)$.

## APPENDIX B

By accounting for the Kirchhoff law, $\sum_i I_{ij} = 0$, for the total Josephson currents, $I_{ij} = I_{ij}^s + I_{ij}^n$, where the superconducting current $I_{ij}^s$ is given by Eq. (4) and $I_{ij}^n = (\hbar/2eR)(d\phi_{ij}/dt)$ is a normal current with $R$ being the resistance between grains in their normal state, the approximate (valid for $N \gg 1$) equation of motion for the superconducting phase reads[10,14]

$$\frac{\hbar N}{2eR} \frac{d\phi_i}{dt} + \frac{2eJ}{\hbar} \sum_{j}^{N} \sin\phi_{ij} = 0 . \tag{B1}$$

Taking into account that

$$(d/dt)\exp(+i\phi_i) = i(d\phi_i/dt)\exp(+i\phi_i) ,$$

the pseudospin representation [with $S_i^+ = \exp(+i\phi_i)$] brings about the approximate equation of motion (18) for Josephson pseudospins.

In the so-called mode-coupling approximation,[21] which is based on a Mori-like projection technique,[23,24] the self-consistent master equation on the isothermal correlation function $D(t)$ can be constructed. Let us introduce the Laplace transform

$$D_{ij}(z) \equiv i \int_0^{+\infty} dt\, e^{izt} D_{ij}(t) . \tag{B2}$$

Then the continued fraction expansion for $D_{ij}(z)$ leads to the expression[14,21]

$$D_q(z) = -\left[ z - \frac{\Omega^2}{z + K_q(z)} \right]^{-1} , \tag{B3}$$

where

$$D_q(z) = \frac{1}{N} \sum_{jk}^{N} e^{iq(j-k)} D_{jk}(z) . \tag{B4}$$

Here $\Omega = 2e^2 k_B T R/\hbar^2 N$ is a characteristic frequency of the JJ network. Alternatively, using the inverse Laplace transform, Eq. (B3) can be cast into the self-consistent master equation [Eq. (19)]. Using the mode-coupling approximation scheme,[14,21] the coherent part of the memory kernel can be represented by a set of current-current correlators

$$K_{ij}^c(t) = \overline{\langle \dot{S}_i^+(t)\dot{S}_j^-(0)\rangle} + \Omega^2 \overline{\langle \ddot{S}_i^+(t)\ddot{S}_j^-(0)\rangle} + O(\Omega^4) . \tag{B5}$$

Since $\dot{S}_i^+ \propto \Omega$ [see Eq. (18)], due to a rather strong dependence of the characteristic frequency $\Omega$ on the number of grains ($\Omega \propto 1/N$), we can restrict ourselves to a linear approximation, $K_{ij}^c(t) \cong \langle \dot{S}_i^+(t)\dot{S}_j^-(0)\rangle$, assuming that $N \gg 1$. Taking into account the equation of motion (18), $K_c(t) \equiv (1/N)\sum_{ij} K_{ij}^c(t)$ can be presented in the form

$$K_c(t) \cong \frac{\beta^2\Omega^2}{N} \sum_{ij}^{N} \sum_{kl}^{N} \overline{J_{ik}J_{il}\langle S_k^+(t)S_l^-(0)\rangle} = \beta^2\Omega^2 J^2(\delta, T, H) D(t) . \tag{B6}$$

To obtain the above equation, we have used Eqs. (A3), (A6), and (A7) together with the decoupling approximations discussed in Appendix A. Using Eq. (B6), we finally arrive at Eq. (20) for the total memory kernel below $T_g$.

To get the nonlogarithmic relaxation law (30) for the magnetization, let us rewrite Eq. (10) taking into account

Eqs. (25) and (27) as follows:

$$\ln(1-Z)=\ln\left\{1-\exp\left[-\left[\frac{t}{\tau}\right]^{\alpha}\right]\right\} . \tag{B7}$$

Here

$$Z\equiv[\sqrt{M(t)}-\sqrt{M_{\rm eq}}]/[\sqrt{M_0}-\sqrt{M_{\rm eq}}] ,$$

$M_0\equiv M(\delta,T,H,H_1)$; $M_{\rm eq}$ and $M(\delta,T,H,H_1)$ are given by Eqs. (17) and (10), respectively. For the short-time limit, when $(t/\tau)^{\alpha}\ll 1$, and $Z\ll 1$, we can expand both sides of Eq. (B7) and get in the linear approximation

$$\sqrt{M(t)}-\sqrt{M_{\rm eq}}\cong-(\sqrt{M_0}-\sqrt{M_{\rm eq}})\alpha\ln(t/\tau) .$$

Finally, taking into account that $M_{\rm eq}=M_0L^0$ [see Eq. (17)], we find [cf. Eq. (30)]

$$M(t)=M_{\rm eq}[1-s_{\alpha}\ln(t/\tau)]^2 ,$$

where

$$s_{\alpha}\equiv\alpha(\sqrt{M_0}-\sqrt{M_{\rm eq}})/\sqrt{M_{\rm eq}}=\alpha(1-L)/L .$$


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