# Field-induced aging effects in inhomogeneous superconductors

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A domain theory for aging effects in inhomogeneous superconductors with oxygen-defects-induced intragrain granularity is considered within the superconducting glass model. A qualitative comparison of the model predictions with the observed memory effects in high- $T_c$  single crystals is discussed.

# I. INTRODUCTION

According to recent findings,  $1-6$  single crystals of high- $T_c$  superconductors (HTS) exhibit an anomalous behavior in applied magnetic field, 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. The granular behavior has been related to the clusters of oxygen defects (within the CuO plane) that restrict supercurrent flow and allow excess Aux to enter the crystal. At the same time, in the so-called "field jump" experiments, pronounced memory effects in magnetization of  $YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>$  (YBCO) single crystals have been observed.<sup> $7-9$ </sup> The following successive steps have been performed in the above-mentioned experiments on field-cooled (FC) relaxation of magnetization. As soon as the sample is stabilized at its measuring temperature  $T_m$  after the FC process at the initial magnetic field  $H$  through  $T_c$ , the aging time  $t_a$  of the system is measured. The system is then left in its quasiequilibrium state  $(T_m,H)$  for a certain waiting time  $t_w$  before the applied magnetic field H is increased by  $\Delta H$  to the measuring field  $H_m$ . Just at this time one can start measuring the relaxation of the trapped magnetic flux, with the total measuring time t related to the aging time  $t_a$  by  $t_a = t + t_w$ . The relaxation curves were found to be not uniquely determined: they turned out to depend drastically on the waiting time  $t_w$  that had elapsed between the moment  $t=0$  at which the temperature  $T_m$  was reached, and the moment  $t=t_w$  at which the initial field H was increased by  $\Delta H$  (in FC-type experiments). The response of the system to the field jump at  $t=t_w$  became slower as  $t_w$  was getting larger, indicating that during the waiting time the system was not in equilibrium but only slowly evolved toward it: it was aging. The main peculiarities of the memory effect observed are the appearance of a characteristic inflection point in the magnetization  $M(t)$  at  $t=t_w$  and the strong dependence of  $M(t)$  vs actual measuring time t on the waiting time  $t_w$ .

Incorporating the ideas of the domain theory in spin Incorporating the ideas of the domain theory in spin<br>glasses<sup>10,11</sup> into the so-called "superconducting glass" (SG) model,  $^{12-19}$  in the present paper the main features of the above-mentioned field-induced memory effects observed in single crystals of high- $T_c$  superconductors are discussed.

#### II. THE SG MODEL: EQUILIBRIUM RELAXATION

Let us first briefly consider the equilibrium relaxation of magnetization within the SG model. More details can be

found in Refs. 12—19. The SG model is based on the wellknown Hamiltonian of a granular superconductor which in the so-called pseudospin representation has the form

$$
\mathcal{K}_0 = -\sum_{ij}^{N} J(\delta, T) \cos \phi_{ij}(\vec{H}) = -\operatorname{Re} \left\{ \sum_{ij}^{N} J_{ij} S_i^+ S_j^- \right\},\tag{1}
$$

where

$$
J_{ij}(\delta, T, \vec{H}) = J(\delta, T) \exp[iA_{ij}(\vec{H})],
$$
  
\n
$$
\phi_{ij}(\vec{H}) = \phi_i - \phi_j - A_{ij}(\vec{H}),
$$
\n(2)

$$
A_{ij}(\vec{H}) = \frac{\pi}{\phi_0} (\vec{H} \times \vec{R}_{ij}) \vec{r}_{ij}, \quad \vec{r}_{ij} = \vec{r}_i - \vec{r}_j, \quad \vec{R}_{ij} = (\vec{r}_i + \vec{r}_j)/2.
$$
\n(3)

This model describes the interaction between oxygen-rich superconducting grains [with phases  $\phi_i(t)$  or Josephson pseudospins  $S_i^+$  = exp(+ $i\phi_i$ ), arranged in a random twodimensional (2D) lattice (modeling the CuO plane of oxygen-depleted YBa<sub>2</sub>Cu<sub>3</sub>O<sub>7- $\delta$ </sub>, where a glasslike picture is established<sup>1-6</sup>) with coordinates  $\vec{r}_i = (x_i, y_i, 0)$ . 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  $\tilde{H}$ , which is assumed to be normal to the CuO plane of HTS. 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  $\delta \leq 1$ ) we can approximate the  $\delta$  dependence of the Josephson energy by a linear law,<sup>4</sup> namely,  $J(\delta,T)$  $\approx J(0,T)(1-\delta)$ . The superconducting current through the Josephson junction  $(JJ)$  between grains i and j

$$
I_{ij}^{s}(\vec{H}) = \frac{2eJ}{\hbar} \sin \phi_{ij}(\vec{H}) = \frac{2e}{\hbar} \text{Im} \{ J_{ij} S_{i}^{+} S_{j}^{-} \}
$$
(4)

induces a diamagnetic moment of the weak-link network $12-19$ 

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

To study dynamic (relaxation) behavior of the model (1), let us assume that in addition to the constant frustrating field  $\vec{H}$ , the superconducting grains are under the influence of a

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small time-varying field  $\vec{H}_{\text{rf}}(t) \ll \vec{H}$ , so that  $\cos{\lbrace \phi_i \rbrace}$  $\{A_{ij}[\hat{H} + \hat{H}_{rf}(t)]\} \cong \cos \phi_{ij}(\hat{H}) + A_{ij}[\hat{H}_{rf}(t)] \sin \phi_{ij}(\hat{H})$ . In view of Eqs.  $(1)$ – $(5)$ , the total (perturbed) Hamiltonian can be cast into the form

$$
\mathcal{H}(t) = \mathcal{H}_0(\vec{H}) - \vec{\mu}\vec{H}_{\text{rf}}(t). \tag{6}
$$

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  $\vec{H}_{\text{rf}}(t)$ ] response of the system (magnetization)  $M(t) = \langle \mu_z \rangle / V$  will relax to its equilibrium value  $M_{\text{eq}} = \lim_{t \to \infty} M(t)$  according to the formula<sup>11,20</sup>

$$
M(t) - M_{\text{eq}} = \int_{-\infty}^{t} dt' R(t, t') \frac{dH_{\text{rf}}(t')}{dt'}.
$$
 (7)

Therefore, the function  $R(t, t')$  describes the relaxation of magnetization  $M(t)$  after removal of the outer disturbance.

ginetization  $M(t)$  after removal of the other disturbance.<br>Let us consider now the field jump experiment at constant<br>perature assuming that<sup>7,11</sup> temperature assuming that $7,11$ 

$$
H_{\rm rf}(t) = \begin{cases} 0, & t_a \le t_w, \\ \Delta H, & t_a > t_w. \end{cases}
$$
 (8)

Here  $t_a = t_w + t$ ,  $t_w$  is a waiting time,  $t_a$  is a time of aging, and  $t$  is an actual measuring time.

According to Eq. (7), the excess magnetization in this case reads

$$
\Delta M(t_a) \equiv M(t_a) - M_{\text{eq}} = \Delta HR(t_a, t_w). \tag{9}
$$

In general, for relaxation of magnetization in a single do-<br>in of size  $\sigma$  (see Sec. III) we can write<sup>10,11</sup> main of size  $\sigma$  (see Sec. III) we can write<sup>10,11</sup>

$$
R(t, t') = R_{\text{eq}}(t - t') \exp[-(t - t')/\tau_m], \quad (10)
$$

where

where

$$
R_{\text{eq}}(t) = \frac{1}{k_B T V} \overline{\langle \delta \mu_z(t) \delta \mu_z(0) \rangle}.
$$
 (11)

Here  $\tau_m(\sigma)$  is the maximum relaxation time in the relaxation spectrum of a size  $\sigma$  domain, and  $\delta \mu_z(t) = \mu_z(t) - \overline{\langle \mu_z(t) \rangle}$ . The bar denotes the configurational averaging over the randomly distributed grain coordinates (see below),  $\langle \cdots \rangle$  means the thermodynamic averaging with the Hamiltonian  $\mathcal{H}_0(H)$ , and we have assumed that  $\vec{H}=(0, 0, H)$  and  $\vec{H}_{\text{rf}}=(0, 0, H_{\text{rf}})$ . To find the equilibrium relaxation of magnetization, we have to calculate the relaxation function  $R_{eq}(t)$ . Using the so-called random-field approximation for quanched disordered systems  $^{13,16,21,22}$  which all mation for quenched disordered systems,, $^{13,16,21,22}$  which allows us 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  $A(r_i)B(r_j) \cong A(r_i) \cdot B(r_j)$ , we obtain for the equilibrium relaxation of magnetization

$$
R_{\text{eq}}(t) = \chi(\delta, T, H) [ |D_{\text{eq}}(t)|^2 - L^2(\delta, T, H) ], \qquad (12)
$$

$$
\chi(\delta, T, H) \equiv \frac{16e^2 d^4 N^2 J^2(\delta, T)}{k_B T V \hbar^2} \left(\frac{H}{H_0}\right)^2 \left(1 + \frac{H^2}{H_0^2}\right)^{-4}.
$$
\n(13)

Here we have introduced the spin-spin correlator

$$
D_{\text{eq}}(t) = \frac{1}{N} \sum_{ij} \overline{\langle S_i^+(t) S_j^-(0) \rangle}, \tag{14}
$$

the nonergodicity (order) parameter

$$
L(\delta, T, H) \equiv \lim_{t \to \infty} D_{\text{eq}}(t), \tag{15}
$$

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

$$
\overline{(x_i y_j - x_j y_i) \exp(i A_{ij})} = \left(\frac{i \phi_0}{\pi}\right) \frac{\partial}{\partial H} \overline{\exp(i A_{ij})}.
$$
 (16)

Here  $H_0 = \phi_0 / \pi d^2$  is a characteristic Josephson field with d an average grain size, and  $N$  is the number of grains.

To obtain the explicit form of the field dependence of susceptibility given by Eq. (13), we have assumed, for simplicity, a site-type positional disorder allowing for weak displacements of the grain sites from their positions of the original 2D lattice, i.e., within a radius  $d$  the new position is chosen randomly according to the normalized separable Gaussian distribution function  $\vec{P(r_i)} = \vec{P(x_i)}\vec{P(y_i)}$ , where

$$
P(x) = \frac{1}{\sqrt{2\pi d^2}} \exp\left(-\frac{x^2}{2d^2}\right).
$$
 (17)

Using the above distribution function, the average value of the Josephson energy [see Eq. (2)] reads

$$
J(\delta, T, H) \equiv \overline{J_{ij}(\delta, T, H)} = J(\delta, T) \overline{\exp(i A_{ij})},\qquad(18)
$$

where

$$
\overline{\exp(iA_{ij})} \equiv \int_{-\infty}^{+\infty} d\vec{r}_i d\vec{r}_j P(\vec{r}_i) P(\vec{r}_j) e^{(i\pi H/\phi_0)(x_i y_j - x_j y_i)} \n= \left(1 + \frac{H^2}{H_0^2}\right)^{-1}.
$$
\n(19)

By analogy with the case of slightly defected thin films, considered by de Gennes and Tinkham, $^{23}$  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 abovementioned coherence (within the CuO plane) is destroyed and the crystal undergoes a phase transition to an 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. To find the long-time (low-frequency) behavior of the correlator  $D_{eq}(t)$  [and thus of the magnetization  $M(t)$ ], we need the equation of motion for the Josephson pseudospins  $S_i^{\pm}(t)$ . By accounting for the Kirchhoff law,  $\Sigma_i I_{ij} = 0$ , 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  $N \ge 1$ ) equation of motion for the superconducting phase reads<sup>13,1</sup>

$$
\frac{\hbar N}{2eR}\frac{d\phi_i}{dt} + \frac{2eJ}{\hbar} \sum_{j}^{N} \sin \phi_{ij} = 0.
$$
 (20)

Taking into account that  $(d/dt) \exp(+i\phi_i) = i(d\phi_i/dt) \exp(+i\phi_i)$ , the pseudospin representation [with representation [with  $S_i^+$  = exp( + *i* $\phi_i$ )] brings about the approximate equation of motion for Josephson pseudospins

$$
\dot{S}_i^+ = \beta \Omega \sum_j^N J_{ij} S_j^+ \,. \tag{21}
$$

Here  $\Omega(\sigma) = 2e^2 dR/\hbar^2 \sigma \beta$  is a characteristic frequency of the JJ network, and  $\beta = 1/k_B T$ . In the so-called "modecoupling approximation,"<sup>24</sup> which is based on the Mori-like projection technique,  $t^{22}$  the self-consistent master equation on the isothermal correlation function  $D_{ij}(t)$  $=\langle S_i^+(t)S_i^-(0)\rangle$  can be constructed. Let us introduce the Laplace transform,

$$
D_{ij}(z) \equiv i \int_0^{+\infty} dt e^{izt} D_{ij}(t). \tag{22}
$$

Then the continued fraction expansion for  $D_{ij}(z)$  leads to the  $expression<sup>17,24</sup>$ 

$$
D_q(z) = -\left(z - \frac{\Omega^2}{z + K_q(z)}\right)^{-1},\tag{23}
$$
\n
$$
L(\delta, T, H) = 1 - \left(\frac{k_B T}{K_S T}\right)^{-1}
$$

where

$$
D_q(z) = \frac{1}{N} \sum_{jk}^{N} e^{iq(j-k)} D_{jk}(z).
$$
 (24)

Alternatively, using the inverse Laplace transform, Eq. (23) can be cast into the self-consistent master equation

$$
\frac{d^2 D_{\text{eq}}(t)}{dt^2} + \Omega^2 D_{\text{eq}}(t) + \int_0^t dt' K_{\text{eq}}(t - t') \frac{d D_{\text{eq}}(t')}{dt'} = 0,
$$
\n(25)

with  $D_{eq}(t) = (1/N)\sum_{ij}D_{ij}(t)$ , and  $K_{eq}(t) = (1/N)\sum_{ij}K_{ij}(t)$ being a memory (feedback) kernel. Using the mode-coupling approximation scheme,<sup>17,24</sup> the coherent part of the memory kernel can be represented by a set of "current-current" correlators

$$
K_{ij}^c(t) = \overline{\langle \dot{S}_i^+(t)\dot{S}_j^-(0)\rangle} + \Omega^2 \overline{\langle \ddot{S}_i^+(t)\ddot{S}_j^-(0)\rangle} + O(\Omega^4). \tag{26}
$$

Since  $\dot{S}_i^+ \propto \Omega$  [see Eq. (21)], 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,  $K_{ii}^c(t) \cong \langle \dot{S}_i^+(t) \dot{S}_i^-(0) \rangle$ , assuming that  $N \ge 1$ . Taking into account the equation of motion (21),  $K_c(t) \equiv (1/N) \sum_{ij} K_{ij}^c(t)$  can be presented in the form<sup>13,16,17</sup>

$$
K_c(t) \approx \frac{\beta^2 \Omega^2}{N} \sum_{ij}^N \sum_{kl}^N \overline{J_{ik} J_{jl}} \cdot \overline{\langle S_k^+(t) S_l^-(0) \rangle}
$$
  

$$
\approx \beta^2 \Omega^2 J^2(\delta, T, H) D_{eq}(t).
$$
 (27)

When there is no temporal correlation between grains ("paracoherent state") the memory kernel has a "white noise" form  $K_{eq}(t) \equiv K_r(t) = 2\Omega \delta(t)$ , where  $\delta(t)$  is the Dirac  $\delta$  function. In this case the master equation results in a Debye-like decay of uncorrelated paracoherent state, namely,  $D_{eq}(t) = \exp(-t/\tau)$ , where  $1/\tau = \Omega$ . Such a situation is realized above some critical (phase-locking) temperature  $T<sub>g</sub>$ when the coherent state within the JJ network is destroyed completely, so that the order parameter  $L=0$ . Below  $T_g$ , the situation changes drastically due to the superconducting correlations occurring between grains. Taking into account Eq. (27), the memory kernel below  $T<sub>g</sub>$  can be presented in the form

$$
K_{\text{eq}}(t) \equiv K_r(t) + \frac{1}{N} \sum_{ij}^{N} K_{ij}^{c}(t) = 2\Omega \,\delta(t) + \Omega_{\text{coh}}^{2}(\delta, T, H) D_{\text{eq}}(t).
$$
 (28)

Here  $\Omega_{coh}(\delta, T, H) = \beta \Omega J(\delta, T, H)$  and the field dependence of the Josephson energy is given by Eqs. (18) and (19). In view of Eq. (15), a zero frequency  $(t \rightarrow \infty)$  solution of the master Eq. (25) with the memory kernel (28) results in the nontrivial order parameter for the intragranular JJ network'

$$
L(\delta, T, H) = 1 - \left(\frac{k_B T}{J(\delta, T, H)}\right)^2.
$$
 (29)

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. (29), gives rise to 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)  $J(\delta, 0, H) \ll k_B T_c \ll J(\delta, 0, 0)$ , we get finally  $T_g(\delta,H) \approx J(\delta,0,H)/k_B$ . As a result, the order parameter  $L = 1 - [T/T<sub>g</sub>(\delta, H)]<sup>2</sup>$  gradually changes from 0 at  $T \geq T_e (\delta, H)$  to 1 at  $T=0$ , thus describing the continuous phase transition.

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  $\delta_g(T, H) = 1 - T/T_g(0, H)$ . 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.

It is important to mention that the correlator  $D_{eq}(t)$  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  $L \neq 0$ ), relaxation of  $D_{eq}(t)$  can be presented in the form<sup>1</sup>

$$
D_{eq}(t) = L + (1 - L)\Phi(t).
$$
 (30)



FIG. 1. The dependence of the power-law relaxation exponent,  $\alpha$ , on the oxygen-deficiency parameter,  $\delta$ , calculated according to Eq. (33).

Of course, in principle, one can find  $D_{eq}(t)$  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  $D_{eq}(t)$ .

Let us consider a powerlike law as an example of a non-Debye relaxation,

$$
\Phi(t) = \Phi(0) \left( 1 + \frac{t}{t_0} \right)^{-\alpha}.
$$
\n(31)

Substitution of Eqs. (30) and (31) into Eq. (25) with the kernel (28) results in the implicit equation on the power exponent  $\alpha(\delta, T, H)$ ,<br>  $\Gamma(1-\alpha) = \frac{1}{2} \left( \frac{2-L}{1-L} \right) \Phi(0)$ . (32) ponent  $\alpha(\delta, T, H)$ ,

$$
\Gamma(1-\alpha) = \frac{1}{2} \left( \frac{2-L}{1-L} \right) \Phi(0). \tag{32}
$$

Here  $\Gamma$  is the  $\gamma$  function, and  $\Phi(0) = \Gamma(1/2) = \sqrt{\pi}$ . Near.  $T_g$  we have approximately

$$
\alpha(\delta, T, H) \approx \frac{1}{2} + \frac{L(\delta, T, H)}{2\psi(1/2)},
$$
\n(33)

where

$$
\psi(1/2) = -2 \ln 2 + \psi(1), \quad \psi(1) = -0.577... \quad . \quad (34)
$$

Notice that in view of the explicit dependence of the order parameter on  $\delta$ , Eq. (33) suggests a rather strong dependence of  $\alpha$  on the oxygen-deficiency parameter (see Fig. 1).

#### III. THE SG MODEL: AGING EFFECTS

To describe the field-induced memory (aging) effects in deoxygenated YBCO single crystals, we will follow the socalled domain theory proposed by Koper and Hilhorst $11$  for treating the aging effects in spin glasses. Their approach is based on the following main principles:

(i) It is assumed that a thermodynamic state of a SG at a temperature  $T_1$  in magnetic field  $H_1$  is characterized by a specific set of  $(T_1,H_1)$  spin correlations (or Josephson pseudospin correlations in our case).

(ii) The correlations of two different thermodynamic equilibrium states, one at  $(T_1, H_1)$  and one at  $(T_2, H_2)$ , are nearly identical up to their overlapping length  $l(T_1 - T_2, H_1 - H_2)$  such that  $l \rightarrow \infty$  as  $(T_2, H_2) \rightarrow (T_1, H_1)$ . (iii) At any time, a nonequilibrium SG state can be analyzed with respect to its  $(T_1,H_1)$  correlations for an arbitrary choice of  $(T_1,H_1)$ . It means that the system is composed of the  $(T_1,H_1)$  domains within which there exist  $(T_1,H_1)$  correlations but beyond which these correlations are destroyed. For a system in equilibrium at  $(T,H)$  there is a single infinite  $(T,H)$  domain with a distribution of the sizes  $\sigma$  of the  $(T_1, H_1)$  domains centered around  $l(T_1 - T, H_1 - H)$ .

(iv) In the nonequilibrium system placed at a certain time in a heat bath of constant temperature  $T$  and in a constant magnetic field  $H$ , the  $(T,H)$  domains will start growing without limit. For any  $(T_1, H_1) \neq (T, H)$ , the  $(T_1, H_1)$  domains will grow until they reach the "overlapping" length  $l(T_1-T,H_1-H)$ .

(v) Magnetic relaxation within a domain is assumed to be described by a linear response theory. Small (large) domains are associated with short (long) relaxation times of magnetization.

Due to the pseudospin coherence within a domain, the maximum relaxation time  $\tau_{\text{max}}(\sigma)$  will increase with the domain linear size  $\sigma$ . In view of Eqs. (10) and (21),

$$
\tau_m(\sigma) \equiv \Omega^{-1}(\sigma) = t_1(\sigma/d), \quad t_1 = \beta \hbar^2 / 2e^2 R. \tag{35}
$$

Here  $t_1$  is a first microscopic time of the system, and we have assumed that  $\sigma \simeq dN$ , with d an average grain size and  $N$  the number of phase-correlated (oxygen-rich) regions within a domain. According to the mesoscopic domainlike theory, $^{11}$  when the domain size varies with time, the magnetic response of a pseudospin  $S_i^{\pm}(t)$  is still determined by linear response law (9) but with a relaxation function which is a functional of the time-dependent size  $\sigma(t')$  of the  $(T,H(t_a))$  domain (with  $t_w \le t' \le t_a$ ). As a result, a small (large) domain is associated with short (long) relaxation time of the magnetization. This leads in turn to a natural generalization of the relaxation function (10) as a weighted sum over all possible  $(T, H(t_a))$  histories of the Josephson pseudospin (i.e., grain phase) in the system

$$
R(t_a, t_w) = \int d\sigma(t')g(T, H(t_a); [\sigma(t')])R(t_a, t_w; [\sigma(t')]).
$$
\n(36)

Notice that a relationship between magnetization and magnetic field, suggested by Eqs. (9) and (10), is linear only in appearance since the nonlinearity comes in explicitly via the mechanism which governs the evolution of the domain size which in turn is field dependent (see below).

In analogy to the equilibrium case  $(10)$  we put<sup>10,11</sup>

$$
R(t_a, t_w; [\sigma(t')]) = D_{\text{eq}}(t_a - t_w) F(t_a, t_w; [\sigma(t')]),
$$
\n(37)

where

$$
F(t_a, t_w; [\sigma(t')] ) = \exp \left\{ - \int_{t_w}^{t_a} dt' \tau_m^{-1} [\sigma(t')] \right\}.
$$
 (38)

According to the experimental observations in HTS single crystals,  $25,26$  the characteristic size of a growing domain (in-



FIG. 2. Semilogarithmic plot of normalized magnetization,  $\Delta M(t)/\Delta M(0)$ , vs reduced time  $t/t_0$  in a field of frustration  $H/H_0 = 0.5$  for a field step  $\Delta H/H_0 = 1$  for three different waiting times (indicated by vertical arrows): (1)  $t_w/t_0 = 5$ , (2)  $t_w/t_0 = 10$ , and (3)  $t_w/t_0 = 15$ .

fluenced by the oxygen-ordering process in the basal CuO plane) increases as a power of time,

$$
\bar{\sigma}(t) \approx d(t/t_2)^n. \tag{39}
$$

Here  $t_2$  is a second microscopic time,  $n = 0.5$  for the ortho-I type ordering, and  $n = 0.25$  for the ortho-II type ordering.<sup>25</sup> In turn,  $t_2$  can be related to the oxygen diffusion process (with the diffusion coefficient  $D_{ab}$ ) as  $t_2 = d^2/D_{ab}$ . In order to proceed further we assume that the weight function  $g$  is centered around an average domain size  $\bar{\sigma}(t)$ , i.e.,  $g(T, H_m; [\sigma(t')] ) \propto \delta(\sigma(t') - \bar{\sigma}(t'))$ . According to the domain dynamics, in the interval  $(0,t_w)$  the domain size  $\bar{\sigma}(t)$ cannot grow larger than the "overlapping" length  $l_{\Delta H}$ . Within the SG model, the role of  $l_{\Delta H}$  is played by the socalled magnetic length, i.e.,  $l_{\Delta H} = \sqrt{\phi_0 / \pi \Delta H}$ . The interplay between these two length scales results in either linear or<br>nonlinear aging effects.<sup>11</sup> Let us consider first the case of small field jumps, when  $\bar{\sigma}(t_w) < l_{\Delta H}$ . After the quench to  $T < T_g$  in a frustrating field H, the characteristic size  $\bar{\sigma}(t)$ will begin to increase according to Eq. (39). For  $t > t_w$  the growth law (39) will continue to hold, and from Eqs. (9) and  $(36)$ – $(39)$  we get that the excess magnetization (near  $T_g$ where  $L \ll 1$ ) will decay as

$$
\Delta M(t) = (1 - L^2) \chi(\delta, T, H) \Delta H [1 + (t - t_w)/t_0]^{-2\alpha}
$$
  
×
$$
\exp{-2\sqrt{t_2}(\sqrt{t} - \sqrt{t_w})/t_1}.
$$
 (40)

Figure 2 shows the semilogarithmic plot of relaxation curves of magnetization  $\Delta M(t)/\Delta M(0)$  for different waiting times  $t_w$  with  $\alpha = 0.35$  (which corresponds to the oxygendeficiency parameter  $\delta = 0.05$ ; see Fig. 1). It is clearly seen that in the logarithmic time scale the magnetization shows a characteristic inflection point at  $t = t_w$  which experiences a shift with increasing  $t_w$ , in qualitative agreement with the



FIG. 3. The rate of logarithmic relaxation,  $s(t)$ , vs reduced time  $t/t_0$  for different waiting times, (1)  $t_w/t_0 = 5$ , (2)  $t_w/t_0 = 10$ , and (3)  $t_w/t_0$  = 15, and different oxygen content (see text), (a)  $\delta$  = 0.05 and (b)  $\delta = 0.2$ .

experimental data on high- $T_c$  single crystals.<sup>7,8</sup> Thus  $t_w$  can be considered as the crossover point for  $\Delta M(t)$  between the slow power-law decay ["equilibrium" relaxation due to  $R_{eq}(t)$ ] and the more rapid (exponential) decay imposed by  $F(t)$ . Figure 3 presents the time evolution of the nonequilibrium logarithmic decay rate  $s(t) \propto d[\Delta M(t)]/d[\ln(t/t_0)]$  for different waiting times and different oxygen content: (a)  $\delta$ =0.05 and (b)  $\delta$ =0.2. As is seen,  $s(t)$  exhibits a maximum at  $t_m \approx t_w$  which shifts to larger measurement times t with increasing  $t_w$ . Notice also that this shift is less pronounced for a highly deoxygenated state [when  $\delta \approx \delta_g$ ; see Fig. 3(b)]. In the case of large field jumps, when  $\bar{\sigma}(t_w) \approx l_{\Delta H}$ , the growth kinetics of the domain will be governed by the law<sup>11</sup>

$$
\bar{\sigma}(t) = d \sqrt{\left(\frac{l_{\Delta H}}{d}\right)^2 + \frac{t - t_w}{t_2}}, \quad t > t_w.
$$
 (41)

The decay of excess magnetization is still given by Eqs. (36)–(38) but with  $\bar{\sigma}(t)$  given by Eq. (41). Since  $\bar{\sigma}(t)$  is dependent on  $\Delta H$  (via  $l_{\Delta H}$ ), this is the way the relaxation becomes nonlinear in the field jump  $\Delta H$ . Furthermore, in case of large field jumps, the logarithmic decay rate,  $s(t)$ , will exhibit a maximum at  $t_m \approx t_2 (l_{\Delta H}/d)^2 \approx (\phi_0/l)$  $\pi \Delta HD_{ab}$ ). It means that the location of the relaxation rate maximum shifts to smaller values with increase of  $\Delta H$ . And the crossover between linear and nonlinear regimes occurs at a value of  $\Delta H$  which satisfies  $\bar{\sigma}(t_w) = l_{\Delta H}$ . The latter equation implies that the larger is the waiting time, the smaller is the field jump needed to provoke nonlinear relaxation. It would be very interesting to observe experimentally the manifestation (if any) of nonlinear aging effects in HTS single crystals.

In summary, the mesoscopic domain theory of spin glasses<sup>11</sup> adapted to the description of the networks of Josephson junction arrays was shown to be quite adequate for modeling the memory (aging) effects in inhomogeneous superconductors with field-induced intragrain granularity. The model predictions were found to be in a reasonable agreement with some experimental data on high- $T_c$  single crystals.

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