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COMPARISON BETWEEN MODEL CALCULATIONS AND EXPERIMENTS

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**Time Dependence of the Paramagnetic Meissner Effect: Comparison
between Model Calculations and Experiments.**

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ABSTRACT

Experimental results of the temperature, field and time dependence of the magnetisation in high temperature superconductors displaying the Paramagnetic Meissner Effect are compared with numerical results from model calculations. In experiments the relaxation rate of the zero-field-cooled magnetisation exhibits novel field dependent properties and the field cooled magnetisation is found to increase with time. A model based on an ensemble of superconducting loops, each loop containing an ordinary Josephson junction or a π -junction, is shown to be able to account for most of the experimental results. The time dependent magnetisation is explained by thermally activated flipping of spontaneous orbital magnetic moments, a dynamical process which is fundamentally different from the flux creep phenomenon usually observed in type-II superconductors.

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I. INTRODUCTION

Some sintered Bi-based high-temperature superconductors have been found to display a positive susceptibility when cooled through T_c in low magnetic fields, contrary to the usual negative susceptibility.^{1,2} This phenomenon has been called the Anti-Meissner Effect or the Paramagnetic Meissner Effect (PME). PME samples have also been studied in microwave absorption measurements² and ac-susceptibility measurements.³ In the microwave absorption measurements one finds a maximum in the absorption at zero applied field. In the ac-susceptibility experiments, the field dependence of the modulus of the second harmonic is used to monitor the existence of internal magnetic dc fields. In samples not showing the PME, internal fields are only detected in a remanent magnetic state, whereas in samples showing the PME, internal fields exist even after cooling through T_c in zero field. More recently a positive field cooled magnetisation has also been observed in other materials.^{4,5}

The experimental results tell that the PME is closely related to the superconducting state and that spontaneous magnetic moments develop below T_c . To explain these effects theoretical models have been proposed.^{6,7} Kusmartsev⁶ introduced the concept of an orbital glass. The orbital glass is a state characterised by the existence of randomly distributed orbital currents. The spontaneously generated orbital currents are due to the existence of so called π -junctions⁸ between superconducting grains. If there is an odd number of these anomalous Josephson junctions in a closed superconducting loop, a spontaneous current will be generated giving rise to an orbital magnetic moment. Several possible origins of π -junctions have been proposed: electron tunneling with a spin flip process due to randomly distributed magnetic impurities in the junction,⁸ indirect electron tunneling through a localised impurity state in the junction⁹ and tunneling of d -wave paired electrons between grains with differently oriented interfaces.⁷ Sigrist and Rice⁷ not only interpreted the PME as due to d -wave superconductivity, they also proposed a

simple model based on an ensemble of non-interacting superconducting loops, some of which should contain a π -junction. They showed that this model could reproduce several of the experimental findings. Domínguez *et al.*¹⁰ have carried these ideas further by studying the PME in 2D and 3D networks of Josephson junctions including a random distribution of π -junctions. The importance of collective effects was emphasised by these authors by pointing out that their model is equivalent to an XY spin glass, with a quadratic coupling to the external magnetic field.

In an earlier study, Ebner and Stroud¹¹ used Monte Carlo simulations to investigate the diamagnetic response of superconducting grains weakly coupled by normal Josephson junctions, here called 0-junctions, into closed superconducting loops. Two models were investigated, one with an ensemble of independent superconducting loops and another with independent clusters of superconducting loops where the loops in each cluster were connected. The results for large clusters were shown to be quite similar to results obtained for the simple model with independent superconducting loops. In both models, the field at which the first flux slip occurs, giving time dependent effects and irreversible magnetic behaviour, is found when the flux through a loop equals $\Phi_0/2$. For fields lower than this, both models only display a reversible diamagnetic response.

In the present study, based on the model by Sigrist and Rice,⁷ π -loops as well as 0-loops are included and the magnetic flux through each loop, generated by the external field, is always such that $\Phi < \Phi_0/2$. This implies that the response of 0-loops is reversible and that the time dependent and irreversible magnetic behaviour are due to the π -loops. We investigate time dependent properties of the magnetisation by recognising that loops containing π -junctions in a magnetic field behave as an ensemble of asymmetric two-level systems. By changing the external magnetic field or by changing the temperature, the energy barriers between the states of opposite current direction will change. It will be shown that thermally activated flipping of the orbital moments can

explain the field dependence of the relaxation rate of the zero-field-cooled (ZFC) magnetisation as well as the time dependence observed for the field cooled (FC) magnetisation.^{12,13} The model is also capable of explaining the unusual field dependence observed for the zero-field-cooled susceptibility versus temperature curves.¹ Thus, even though the model is an independent loop model, it captures the essential physics in the low field regime for weakly shielding samples exhibiting PME behaviour. At higher fields and for samples displaying stronger shielding properties, more complicated models are needed.

II. THE MODEL

The model by Sigrist and Rice⁷ assumes that the system is composed of non-interacting superconducting loops, each with one Josephson junction. The Josephson junctions can either be normal 0-junctions or so called π -junctions. The ground state of a π -junction⁸ corresponds to a phase discontinuity of $\phi = \pi$ in the electron pair wave function when electrons tunnel through the junction. The free energy of one loop is given by

$$F(i, \varphi) = \zeta A \left(\frac{1}{2} \zeta i^2 - \cos[2\pi\varphi + \zeta i + p\pi] \right). \quad (1)$$

The parameters ζ , A , i and φ are defined as $\zeta = 2\pi L I_c / \Phi_0$, $A = \Phi_0^2 / 4\pi^2 L$, $i = I / I_c$ and $\varphi = \Phi / \Phi_0$ where L is the loop self inductance, I the loop current, I_c the junction critical current, Φ the applied magnetic flux and Φ_0 the flux quantum. The parameter p has the value 1 if the junction is a π -junction and 0 if the junction is a 0-junction.

For a π -junction with $\zeta > 1$, there is a twofold degenerate energy minimum state in zero applied field. This implies that a spontaneous current will circulate in the ring. Both directions of the current are equally likely to occur. For $\varphi \neq 0$ one of the two minima will be lower in energy and with increasing field the locations of the minima will be slightly

shifted (the lower (upper) energy minimum will be shifted towards smaller (larger) currents). The lower energy minimum corresponds to a current generating an orbital magnetic moment, whose projection in the direction of the applied field is parallel to the field. For $\zeta < 1$ only one minimum of $F(i, \varphi)$ exists, corresponding to a linear paramagnetic response to the external magnetic field. For a loop containing a 0-junction the response to an applied field is diamagnetic for all values of ζ .

In our study, the minima of the loop free energy were found by solving $\partial F(i, \varphi) / \partial i = 0$ numerically using a modified Newton-Raphson method. A linear temperature dependence was used for the critical current, $I_c(T) = I_{c0} (1 - T/T_c)$, and for the related parameter $\zeta(T)$. Henceforth, π -loops carrying spontaneous currents will be referred to as spontaneous moments while those giving a paramagnetic response will be referred to as paramagnetic loops. A general normal distribution $p(\zeta)$ of ζ 's was used with expectation value $\bar{\zeta}$ and variance σ_ζ^2 . In the calculations the zero temperature $\bar{\zeta}(0)$ and $\sigma_{\zeta(0)}$ were fixed and the temperature dependence was given by the temperature dependence of the critical current. The orientation of each loop with respect to the direction of the applied field, θ , was assumed to be random i.e. the distribution of loop orientations was $g(\theta) = \sin \theta$. The free parameters in the model were: the volume fraction m of π -junctions, the expectation value and variance of $p(\zeta)$ and the parameter A which sets the free energy scale of each loop.

The time dependence of the magnetisation was modelled in the following way. Consider a sub volume of the ensemble of π -loops consisting of $N_i \propto p(\zeta_j) g(\theta_k) d\zeta d\theta$ identical loops, all having the same critical current and the same orientation θ_k with respect to the applied field and let $n_i^+(t)$ denote the number of π -loops having a current in the direction giving a positive magnetisation and $n_i^-(t)$ the number of loops with a negative magnetisation. In a zero-field-cooled experiment, before applying a field, there is an equal number of positive and negative spontaneous moments, $n_i^+(0) = n_i^-(0) = N_i/2$. When a field is applied, the energy barriers for flipping of the spontaneous

magnetic moments will change in such a way as to increase (decrease) the barrier for flipping positive (negative) spontaneous moments. The result of the field application is a redistribution of the spontaneous moments among the two levels. The time dependence for this process can be found by solving the differential equation

$$\frac{dn_i^+(t)}{dt} = \left[N_i - n_i^+(t) \right] \omega_0 e^{-\beta \Delta E_{i,1}} - n_i^+(t) \omega_0 e^{-\beta \Delta E_{i,2}} \quad (2)$$

where $\beta = 1/(k_B T)$ and ω_0 a microscopic attempt time. In the above equation, the energy barrier for flipping a positive (negative) spontaneous moment is denoted by $\Delta E_{i,2}$ ($\Delta E_{i,1}$). The energy barriers were obtained after solving $\partial F / \partial i = 0$ and then calculating the energy difference between the energy maximum and the two energy minima. The solution to Eq. (2) for the initial condition $n_i^+(0) = n_i^-(0)$ is

$$n_i^+(t) = N_i \left\{ \frac{1}{1 + e^{-\beta(\Delta E_{i,2} - \Delta E_{i,1})}} + \left[\frac{1}{2} - \frac{1}{1 + e^{-\beta(\Delta E_{i,2} - \Delta E_{i,1})}} \right] \times \right. \\ \left. \times \exp \left[-\omega_0 \left(e^{-\beta \Delta E_{i,1}} + e^{-\beta \Delta E_{i,2}} \right) t \right] \right\} \quad (3)$$

The magnetisation due to the N_i spontaneous moments is then given by

$$m_i(t) \propto \left(2n_i^+(t) - N_i \right) \cdot \cos(\theta_k) \quad (4)$$

To obtain the time dependent magnetisation of all spontaneous moments at a given temperature, $\Delta E_{i,2}$ and $\Delta E_{i,1}$ need to be calculated for each set of (ζ_j, θ_k) -values, defined by the distributions of critical currents and loop orientations. The time dependent magnetisation due to the ensemble of π -loops carrying a spontaneous moment is then obtained by summing the different $m_i(t)$ contributions, and the total magnetisation is

obtained by adding the time independent magnetisation due to the paramagnetic π -loops and the 0-loops.

The procedure described above for the time dependent ZFC magnetisation can also be used to calculate the temperature dependent ZFC susceptibility. The ZFC susceptibility is then obtained by calculating the magnetisation value, at different temperatures, for a certain observation time t . The initial condition $n_i^+(0) = N_i/2$ is used at each temperature. The time dependent FC susceptibility can be calculated by stepwise cooling the system to the measurement temperature. In the cooling process, equation 2 is solved for a certain time t^* at each temperature. The initial value of $n_i^+(0)$ at the new temperature is set equal to the value $n_i^+(t^*)$ obtained at the previous temperature. Finally, when the measurement temperature is reached, the magnetisation at different times is calculated. By performing calculations for $t = \infty$, also the equilibrium susceptibility, χ_{EQ} , can be derived.

III. RESULTS AND DISCUSSION

In this chapter we make a comparison between the numerical results obtained by model calculations and experimental data. Qualitatively the results from model calculations are rather insensitive to different choices of input values for the model parameters. However, since equation 2 basically gives an exponential relaxation, it is necessary to have a reasonably broad distribution of critical currents to account for the nearly logarithmic relaxation of the zero-field-cooled magnetisation extending over many decades of time as found in experiments. We have therefore required the distribution to be wide enough to produce an essentially logarithmic relaxation in the time interval $t = 10^{-1}$ s to 10^5 s. Also $A \gg k_B T_c$ is needed in order to have energy barriers for flipping of spontaneous moments substantially larger than the thermal energy. To account for the special features found experimentally in the field dependence of $\chi_{ZFC}(T)$ it was necessary to use a small fraction of π -loops.

Measurements have been performed on numerous Bi-based samples. The experimental results presented in this paper are from measurements on one typical sample displaying the Paramagnetic Meissner Effect. The sample consisted of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ prepared by reaction sintering of Bi_2O_3 , SrCO_3 , CaCO_3 and CaO at temperatures $800^\circ\text{C} - 865^\circ\text{C}$. The measurements were performed in a non-commercial, magnetically well shielded SQUID magnetometer. The sample position in the magnetometer was fixed during the measurements. Details on the sample and the experiments will be published elsewhere.¹³

In figure 1(b) and 2(b) it is seen that both the FC and ZFC susceptibilities in this sample are relatively weak. The weak screening is typical for samples where we have found the Paramagnetic Meissner Effect and is due to low intergranular critical currents. The superconducting grains are effectively decoupled and magnetic flux penetrates the intergranular space. Furthermore, the sample remains resistive even at very low temperatures, indicating that the superconducting loops are coupled only as finite clusters. The clusters can either be formed by an assembly of superconducting grains or by the internal structure of individual superconducting grains.

In figure 1(a), the calculated equilibrium susceptibility, χ_{EQ} , is shown vs. temperature for a few applied fields. For comparison, experimental results for the field cooled susceptibility, χ_{FC} , are shown in figure 1(b). The characteristic features of the Paramagnetic Meissner Effect, e. g. a positive value of the susceptibility at low fields and low temperatures, can within the model be explained as follows. When the system is cooled below the critical temperature the parameter ζ gradually increases and spontaneous moments appear. If an external magnetic field is present there will be a predominance of spontaneous currents polarised in the positive direction, generating a magnetic moment parallel to the applied field. Close to T_c the diamagnetic response of the 0-junctions still dominates since most π -loops will show a paramagnetic response and the volume fraction

of 0-junctions is larger. As the temperature is lowered an increasing fraction of π -loops will carry spontaneous moments, which leads to a positive total magnetisation at low temperatures provided the applied field is small enough. The diamagnetic magnetisation of the 0-junctions increases linearly with field, while the polarisation of the spontaneous moments saturates. This implies that a negative magnetisation will be detected at sufficiently large fields. There are experimental results indicating that the magnitude of χ_{FC} approaches a constant positive value at the very low fields.¹² The same result is obtained for the equilibrium susceptibility in the numerical calculations where the field dependence of the magnetisation due to π -loops is linear for fields $\Phi/\Phi_0 < 0.001$, followed by a levelling off towards saturation at higher fields.

In figure 2(a), the calculated zero-field-cooled susceptibility, χ_{ZFC} , is shown vs. temperature for a few applied fields. For comparison, typical experimental data are shown in figure 2(b). This comparison shows that the model can reproduce the relevant features found experimentally. For temperatures just below T_c , experiments show that χ_{ZFC} becomes more diamagnetic with increasing field, quite the contrary to what is usually found in experiments on high- T_c superconductors. For sufficiently low fields, the numerical results even show a maximum in $\chi_{ZFC}(T)$ close to T_c . A similar maximum should also be observable experimentally at low fields and long enough observation times. However, the observation times usually used experimentally and also in this investigation are too short to reveal such a maximum. One problem encountered in experiments is also that there exists a small uncompensatable background field $H_b \neq 0$ (< 0.25 mOe in our apparatus). When performing a ZFC measurement the sample is cooled in H_b and not in zero field. Applied fields appreciably larger than the background field are needed to obtain reliable ZFC susceptibility results.

The temperature dependence of the χ_{ZFC} can in terms of the model be understood as follows. At low temperatures, after having applied an external field, n_i^+ and n_i^- do not

change significantly since the energy barriers are too large with respect to the thermal energy to polarise a significant number of spontaneous moments. This implies that the magnetic response at low temperatures is dominated by the diamagnetic 0-loops and is therefore almost linear in field. With increasing temperature, there will be an increasing number of spontaneous magnetic moments polarised along the direction of the applied field giving a positive contribution to the total susceptibility. The relative contribution from the polarisation of spontaneous moments to the susceptibility becomes larger in a weak applied field since the diamagnetic susceptibility of the 0-loops is almost constant.

The only achievable low temperature equilibrium quantity is the calculated temperature dependence of the susceptibility. All other experimental or calculated susceptibilities we discuss in this paper are non-equilibrium quantities. The temperature dependence of the ZFC-susceptibility was calculated point wise at different temperatures at a specific observation time. In experiments the temperature dependence is measured at a specific cooling rate (FC) or heating rate (ZFC) corresponding to an effective observation time. If the sample during a heating or cooling process is kept at constant temperature, the magnetisation will thus continue to relax towards the equilibrium value.

We first discuss the relaxational behaviour of the FC-susceptibility derived both from the model and from experiments. In the calculation, the FC-process is modelled by cooling the system in temperature steps of $\Delta T/T_c = 0.01$ to the measurement temperature and calculating the magnetisation value at $t^* = 100$ s at every temperature step. At the measurement temperature the calculations are then performed for different observation times. The results at $T/T_c = 0.80$ and 0.90 are plotted vs. $\log(t)$ in Fig 3(a). At both temperatures the magnetisation increases with time and the relaxation is logarithmic in the investigated time window. In experiments, the FC-susceptibility is measured by cooling the sample at a specific cooling rate to a measurement temperature, where the magnetisation is recorded vs. time spent at constant temperature. In Fig. 3(b)

the relaxation of χ_{FC} is plotted vs. $\log(t)$ at $T = 50$ K and 60 K. At both temperatures the susceptibility increases with time, which is opposite to the increasing diamagnetic signal observed for the relaxation of the FC-susceptibility in type-II superconductors not exhibiting the Paramagnetic Meissner Effect.¹⁴ The curves in Fig. 3(b) do not show a logarithmic behaviour. This obvious deviation from the calculated curves is largely explained by the continuous relaxation occurring during the cooling process and our definition of the observation time, t , starting from the moment we reach the measurement temperature. The curvature of the FC relaxation data does therefore not correspond to intrinsic physical properties of the sample. Nevertheless, the observed direction of the relaxation is significant. By introducing an effective observation time $t_{eff} = t + t_c$, where t_c is of order 100 s, we can partly account for the observation time corresponding to the cooling process. In the inset to Fig 3(b), we have plotted the relaxation vs. $\log(t_{eff})$. These curves look rather more logarithmic, in better accord with the behaviour calculated from the model.

The relaxation of the ZFC-magnetisation is measured by cooling the sample in zero field to the measurement temperature. After reaching constant temperature, the magnetic field is applied and the magnetisation is recorded vs. time after the field application. In our experiments the switch time of the magnet is of order 10^{-3} s and in the calculation the switch time is infinitely fast. This implies that in the time window explored in our experiments (0.3 s - 3000 s) and calculations, the ZFC results do not suffer from an ill-defined observation time and that the functional form of the relaxation directly relates to intrinsic physical processes of the sample or of the model system. In figure 4(a), the calculated relaxation rate of the ZFC magnetisation $S = \partial M_{ZFC} / \partial \log_{10}(t)$ is plotted vs. the applied field. The time dependence of the ZFC magnetisation is essentially logarithmic in the time window $t = 10^{-1}$ s to 10^5 s. In the model calculations, for temperatures not too close to T_c , S decreases with decreasing temperature in the investigated time window.

Experimental results also show a similar behaviour for temperatures $T/T_c < 0.7 - 0.8$.¹³ This is also valid for the FC relaxation. S depends linearly on the applied field for low fields, followed by a field-independent relaxation rate at higher fields. For comparison, typical experimental results are shown in figure 4(b). In the low field region the experimentally determined relaxation rate also displays an almost linear field dependence, $S \sim H^{0.9}$. For intermediate fields the relaxation rate levels off in agreement with model calculations, while for higher fields at lower temperatures the relaxation rate increases more strongly with field and approaches an $S \propto H^2$ dependence.¹² The $S \propto H^2$ dependence is in agreement with conventional thermally activated flux creep for fields lower than the field needed to obtain intragranular full flux penetration, where a field dependence $S \propto H^\alpha$ with $\alpha \geq 2$ is expected.¹⁵ In the model calculations, the possibility of having Abrikosov vortices inside superconducting grains is not included which implies that the H^2 -dependence of the relaxation rate will not appear in those results. In fact, the relaxation studied in the model calculations is fundamentally different from thermally assisted flux line motion. The time dependent magnetisation is in the model calculations due to thermally assisted flipping of spontaneous orbital moments (or thermally assisted reversing of orbital currents). The large resemblance between model calculations and experimental results strongly supports an interpretation of the low field relaxation of the magnetisation in terms of thermally assisted flipping of spontaneous magnetic moments.

All numerical results presented above have been obtained using the following values for the free parameters of the model: $m = 0.025$, $\bar{\zeta}(0) = 40$, $\sigma_{\zeta(0)} = 40$, $A = 15k_B T_c$ and $\omega_0 = 10^{10} \text{ s}^{-1}$. These values have been chosen to adequately reproduce the experimental results. However, the values also correspond to certain characteristics of a granular sample. The loop self inductance L in a granular sample can be estimated from the relation $L \sim \mu_0 R$, where R is the typical grain size. The used value of A ($= \Phi_0^2 / 4\pi^2 L$) then corresponds to a grain size of order 10 μm (using $T_c = 85$ K). The

critical current can be estimated from the value of $\bar{\xi}$ ($= 2\pi L \bar{I}_{c0}/\Phi_0$), giving $\bar{I}_{c0} \sim 10^{-3}$ A, which corresponds to a critical current density $\bar{J}_{c0} \sim 10^3$ A/cm². We have also performed calculations using larger values of A , corresponding to smaller grain sizes. Using $m = 0.01$, $\bar{\xi}(0) = 5$ ($\bar{J}_{c0} \sim 10^5$ A/cm²), $\sigma_{\xi(0)} = 5$ and $A = 120k_B T_c$ ($R \sim 1$ μ m) also gives results in reasonable agreement with the experimental findings.

IV. CONCLUSION

By comparing model calculations with experimental results we argue that the physical mechanism, whereby the low field magnetisation in a sample displaying the Paramagnetic Meissner Effect evolves with time, corresponds to thermally activated flipping of the spontaneously generated orbital moments. In this process, the orbital moments orient themselves to be parallel with the applied field. A redistribution of the spontaneous moments among the possible energy levels can be triggered either by a field change or a temperature change. As a consequence, the field cooled magnetisation is not an equilibrium quantity but increases with time.

The model calculations assume a system composed of non-interacting superconducting loops. It is shown that this simple model accounts for many of the experimental results obtained on PME samples, especially on samples exhibiting weak intergranular critical currents. In this case, the sample behaves more or less as a decoupled superconducting network. To account for the behaviour found in samples displaying strong diamagnetic screening, model calculations on 3D networks of Josephson junctions are needed.¹⁰

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FIGURE CAPTIONS

FIG. 1. (a) Temperature dependence of the equilibrium susceptibility for different applied magnetic fields, from model calculations. The susceptibility is given in arbitrary units. The applied field is given in units of Φ_0 . $\bar{\xi}(0) = 40$, $\sigma_{\xi(0)} = 40$, $A = 15k_B T_c$ and $m = 0.025$. (b) Experimentally observed temperature dependence of the field cooled susceptibility at different applied fields.

FIG. 2. Temperature dependence of the zero-field-cooled susceptibility for different applied magnetic fields. (a) Results from model calculations. The zero-field-cooled susceptibility is given in arbitrary units. a, b, c and d correspond to the applied fields $\Phi/\Phi_0 = 0.003, 0.01, 0.03$ and 0.1 . $\bar{\xi}(0) = 40$, $\sigma_{\xi(0)} = 40$, $A = 15k_B T_c$ and $m = 0.025$. (b) Results from experiments. a, b and c correspond to the applied fields $H = 0.01$ Oe, 0.1 Oe and 1 Oe.

FIG. 3. Time dependence of the field cooled magnetisation at different temperatures. (a) Results from model calculations. The applied field was $\Phi/\Phi_0 = 0.01$. $\bar{\xi}(0) = 40$, $\sigma_{\xi(0)} = 40$, $A = 15k_B T_c$ and $m = 0.025$. (b) Results from experiments. The inset shows the field cooled relaxation vs. $\log(t_{eff})$, where t_{eff} corresponds to the effective observation time partly accounting for the finite cooling rate in the experiment. The applied field was $H = 0.1$ Oe.

FIG. 4. Field dependence of the relaxation rate for the zero-field-cooled magnetisation at different temperatures. (a) Results from model calculations. $\bar{\xi}(0) = 40$, $\sigma_{\xi(0)} = 40$, $A = 15k_B T_c$ and $m = 0.025$. (b) Results from experiments. The value of $\partial M_{ZFC}/\partial \log_{10}(t)$ is taken at $t = 100$ s.

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Fig. 1a

Fig. 1b

Fig. 2a

Fig. 2b

Fig. 3a

Fig. 3b

Fig. 4a

Fig. 4b