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Time dependence of the magnetization of $Bi_2Sr_2CaCu_2O_8$ displaying the paramagnetic Meissner effect

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The time dependence of the magnetization of a $Bi_2Sr_2CaCu_2O_8$ sample displaying the paramagnetic Meissner effect has been studied as a function of field, temperature, and age of the system using superconducting quantum interference device magnetometry. The field-cooled magnetization is found to increase with time at low fields, while at higher fields the relaxation is negative. The zero-field-cooled relaxation shows an anomalous field dependence at low magnetic fields. The age of the system does not yield an observable influence on the time dependence of the magnetization. The results are discussed in terms of a model based on so called π junctions.

I. INTRODUCTION

When cooled in a low magnetic field, some granular samples of Bi-based high-temperature superconductors have been found to show a positive susceptibility below T_c.[1–3] This phenomenon has been called the anti-Meissner effect or the paramagnetic Meissner effect (PME), in order to distinguish it from the expected negative susceptibility of a superconductor displaying the ordinary Meissner effect. The effect was first observed in the Bi-2212 system, which is the most extensively studied system. The PME has also been observed in the Bi-2223 phase. The effect has been found both in sintered, weakly screening samples[1] and in melt-textured samples that show a zero-field-cooled susceptibility close to $-1/4\pi$.[2,3] More recently, a positive field-cooled magnetization has been reported in other superconductive materials as well. Riedling *et al.*[4] have observed a positive magnetization at low fields in single crystals of $YBa_2Cu_3O_{7-\delta}$. They found that the PME was only observable when the field was applied along the c axis of the crystal, and not with a field along the a-b plane. A positive magnetization has also been found in a disk-shaped sample of niobium.[5]

Bi-based samples displaying the PME have been found to show anomalous characteristics when studied in microwave absorption experiments[2,3] as well as in measurements of the ac susceptibility.[6] In the microwave absorption measurements one finds a maximum of the absorption at zero applied field followed by a minimum as the field is increased, contrary to the monotonically increasing absorption with increasing field found in non-PME samples. 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 PME samples, internal fields exist even after zero-field cooling through T_c.

The experimental results tell that the PME is directly related to the superconducting state and that spontaneous magnetic moments develop below T_c. These spontaneous moments have been attributed to anomalous Josephson junctions, so-called π junctions,[7] between superconducting grains.[2,8,9] When a Cooper pair tunnels through a π junction it acquires a phase shift of $\Phi \cong \pi$. The free energy F of a ring with one Josephson junction, either a π junction or a normal junction (here called a 0 junction), is composed of the magnetic field and current energy together with the Josephson junction energy and is given by

$$F=\frac{1}{2}LI^2-\frac{I_c\Phi_0}{2\pi}\cos\left[\frac{2\pi}{\Phi_0}(\Phi_{ex}+LI)+p\pi\right], \qquad (1)$$

where I is the current in the loop, L is the loop inductance, Φ_{ex} is the flux threading the ring due to an external field, and I_c is the critical current of the junction. The parameter p has the value 0 for a 0 junction and 1 for a π junction. For a π loop in zero external field and with $\zeta=2\pi LI_c/\Phi_0>1$ there is a twofold degenerate energy minimum state. This implies that a spontaneous current will circulate in the ring, with equal probability for the two circulation directions. When applying an external field, the energy minimum corresponding to a current generating an orbital momentum whose projection in the direction of the applied field is parallel to the field will be lower in energy. For a π loop with $\zeta<1$ only one minimum of $F(I,\Phi_{ex})$ exists, corresponding to a linear paramagnetic response to the external field. For a 0 loop the response to an applied field is diamagnetic for all values of ζ. When several π junctions coexist in a loop, a spontaneous current is only generated if the number of π junctions in the loop is odd.

Several possible origins of π junctions have been proposed, e.g., electron tunneling with a spin-flip process due to randomly distributed magnetic impurities in the junction,[7] indirect electron tunneling through a localized impurity state in the junction[10] and tunneling of d-wave paired electrons between grains with differently oriented interfaces.[9] Kusmartsev[8] and Sigrist and Rice[9] independently pointed out that π junctions can explain the positive magnetic moments of high-T_c superconductors. Kusmartsev introduced the concept of an orbital glass state, characterized by the existence of randomly distributed orbital currents. Sigrist and Rice interpreted the PME as due to π junctions originating from d-wave superconductivity. Furthermore, they proposed a simple model which they showed could reproduce several of the experimental findings. This model assumes a system which is composed of noninteracting superconducting loops, each with one Josephson junction, where the Josephson junction is either a π junction or a normal Josephson junction. A model of a network of 0 and π junctions, including collective effects, was studied by Domínguez, Jagla, and Balseiro.[11] This model was also shown to qualitatively reproduce the experimental magnetization versus temperature results, and it exhibited some glassy properties.

In a recent paper, Magnusson et al.[12] have used the noninteracting π-loop model of Sigrist and Rice to study time-dependent effects. By introducing thermally activated flipping of the spontaneous moments generated by the π loops, the general features of the experimental results of the time dependence of the PME presented below could be reproduced.

The time dependence of the PME has until now not been thoroughly investigated experimentally. Braunisch et al.[3] reported an observation of a change in the zero-field-cooled (ZFC) magnetization with time, while they could not detect any change in the field-cooled (FC) magnetization, thus concluding that the paramagnetic FC magnetization corresponds to an equilibrium state. ZFC relaxation results have been presented by Niskanen et al.,[13] where it was shown that the relaxation rate of the ZFC magnetization has a field dependence that differs from that predicted by the critical-state model.[14] In this paper we report results from measurements of the time dependence of the FC and ZFC magnetization of a sintered sample of Bi-2212 as function of temperature, magnetic field, and age of the system. The measurements were performed in a noncommercial superconducting quantum interference device (SQUID) magnetometer. For the lowest fields applied, the FC magnetization relaxes towards more positive values, contrary to what is found in samples not showing the PME.[15] The field dependence of the relaxation rate of the ZFC relaxation shows a behavior consistent with that earlier reported by Niskanen et al.[13] No dependence of the response to a magnetic field upon the age of the system could be detected. The results are discussed in terms of a π-junction model for the paramagnetic Meissner effect.

II. EXPERIMENTAL

A. Sample preparation and characterization

The investigated $Bi_2Sr_2CaCu_2O_{8+\delta}$ sample was prepared by reaction sintering of Bi_2O_3, $SrCO_3$, $CaCO_3$, and CuO. The sample was ground and several times resintered at temperatures 790–860 °C, and then annealed at 850 °C in an atmosphere of 8% oxygen followed by annealing in 100% Ar atmosphere at 670 °C. X-ray powder analysis indicated a single-phase sample with the following cell parameters of an incommensurate structure: $a=5.406(1)$ Å, $b=24.976(2)$ Å, and $c=37.118(4)$ Å. Scanning electron microscopy revealed a rather flaky grain structure with an average platelet area of 1–4 μm^2. The critical temperature was determined magnetically to be $T_c=87$ K. The magnetization measurements also revealed traces of the Bi-2223 phase with a critical temperature of 110 K. A scanning electron micrograph of the sample is shown in Fig. 1.

B. Experimental procedure

The measurements were performed in a noncommercial SQUID magnetometer with a third-order gradiometer pickup coil configuration, the distance between two neighboring pickup coils being 5 mm. The sample, which was cut as a parallelepiped of volume $2.5\times2.1\times1.7$ mm^3, was kept fixed in one of the middle pickup coils during measurement. The sample rod was designed to eliminate displacement of the sample due to thermal expansion, the effective sample rod being as short as 10 cm. The change in magnetization was always measured relative to a point well above T_c. A constant magnetic field was produced by a small superconducting solenoid working in persistent mode, the small size allowing for switching times of the order of 10^{-3} s. The sample space was screened by μ metal and superconducting shielding. Furthermore, the residual background field H_b in the direction of the applied field was compensated for so that $H_b<0.25$ mOe. To get an absolute value of the mea-

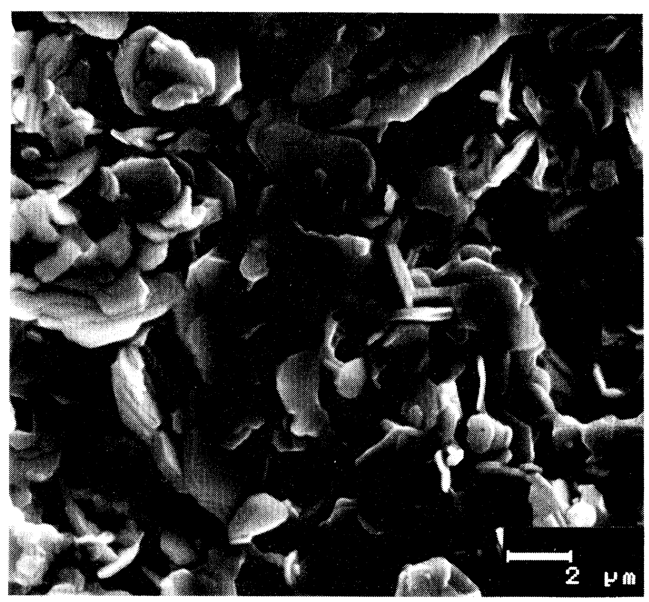

FIG. 1. Scanning electron micrograph of the studied Bi-2212 sample, seen in the direction of pressure applied during preparation.

sured magnetization, a calibration measurement was performed in a Quantum Design SQUID magnetometer at 10 Oe.

Before each measurement run, the sample was heated to 112 K. The sample was then cooled, either in zero field (ZFC) or in an applied field (FC) using a cooling rate of approximately 0.05 K/s. In the magnetization versus temperature measurements, the sample was cooled in zero field to a temperature well below T_c, 50 K or lower, where the field was switched on. The sample was then heated, the heating rate being of the same order of magnitude as the cooling rate, and the ZFC magnetization was recorded. After reaching 112 K, the sample was cooled without changing the field, giving the FC curve. At low temperature the field was removed, and measuring while heating the sample yielded the thermoremanent magnetization (TRM) curve. The sample was always allowed to attain constant temperature before a value of the magnetization was recorded.

In the relaxation measurements the change in the magnetization with time was recorded. The time window investigated in these measurements was 0.3–3000 s. When performing the ZFC relaxation measurements the sample was cooled to the measurement temperature in zero field and the sample was allowed to reach constant temperature before the field was switched on, the switching on of the field defining zero time. Since the switching time of the magnet is much shorter than the smallest time studied, the observation time in the ZFC relaxation is well defined. On the other hand, when performing the FC relaxation measurements the sample was cooled in a field and hence a continuous relaxation occurred during cooling. Since the effective cooling time was of the order of the measurement times, it is difficult to define zero time for the relaxation at the temperature of interest. Thus the functional form of the FC relaxation data does not immediately relate to intrinsic physical properties of the sample. Nevertheless, the observed direction and magnitude of the relaxation are significant. We have used the moment when the measurement temperature was reached as definition of zero time in the FC relaxation experiments.

The quantity measured in all the discussed experiments is the magnetization of the sample, M. In the discussion below, the magnetization normalized to the applied field, M/H, is often (somewhat unstringently) referred to as the susceptibility, χ.

III. RESULTS AND DISCUSSION

A. Field and temperature dependence of the magnetization

The FC and ZFC susceptibilities, χ_{FC} and χ_{ZFC}, are plotted versus temperature for fields in the interval 0.003–3 Oe in Fig. 2. The low value of the ZFC susceptibility, only about a tenth of $-1/4\pi$ even for the lowest fields, shows that the intergranular critical currents in the sample are rather weak. This is characteristic of the samples where we have detected the PME. Thus, for all the fields used in this study, one expects the magnetic field to have penetrated the intergranular regions of the sample. Since the size of the grains is comparable to the penetration depth, one can assume that most of the grains are also largely penetrated by the field. The first critical field of Bi-2212 is normally considered to be larger than the fields used in these experiments.[16] However, considering local demagnetizing effects, one should not exclude the possibility of having vortices inside the grains, especially not for the larger fields used. Intergranular vortices will exist in regions of densely packed grains. Such vortices are first created when the applied field is of the order $B \sim \Phi_0/2d^2$, where d is a typical grain diameter. Since the average grain diameter in our sample is a few microns, intergranular vortices are expected to occur for fields in the order of 1 Oe.

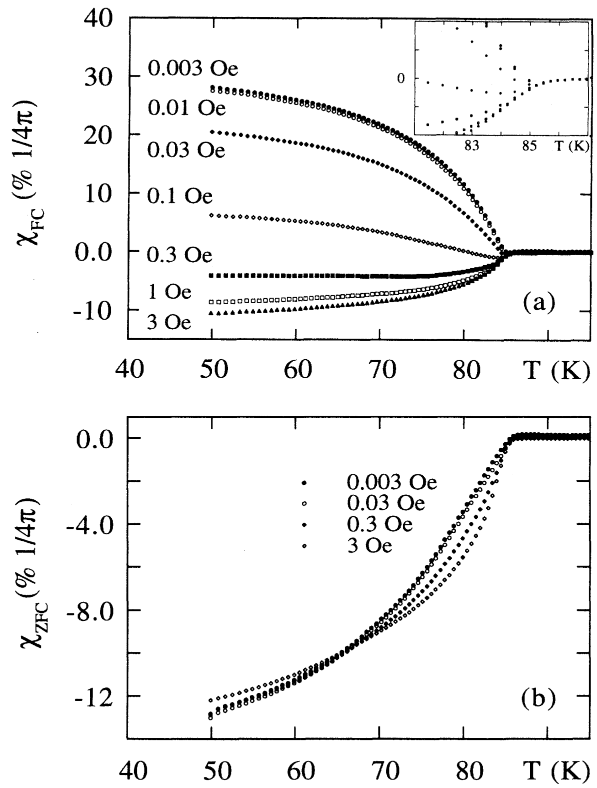

FIG. 2. Field-cooled (a) and zero-field cooled (b) susceptibilities versus temperature for applied fields in the range 0.003–3 Oe. The inset of (a) shows an enlargement of the region close to T_c, where the FC susceptibility is negative for all fields.

The most striking feature of the PME, the positive FC susceptibility at low fields, is clearly seen in Fig. 2(a). The positive susceptibility becomes larger with decreasing field. In the low-field limit, experiments indicate that χ_{FC} reaches a constant value. However, since the uncertainty in the lowest field (0.003 Oe) is approximately 10%, the true asymptotic behavior remains uncertain. For fields 0.3 Oe or larger χ_{FC} becomes negative, but, contrary to what is found in non-PME samples, the absolute value of the diamagnetic susceptibility increases with increasing field in the field range shown. For even higher fields, this behavior will inevitably change into a decreasing susceptibility with increasing field due to vortices penetrating the sample. In the temperature region just below T_c, χ_{FC} is negative and rather field independent, as can be seen in the inset of Fig. 2(a).

The ZFC susceptibility, shown in Fig. 2(b), also displays an anomalous behavior. For the low temperatures, χ_{ZFC} at constant temperature shows a decrease in magnitude with increasing field, which is to be expected from a vortex picture. However, at around 70 K, i.e., $T/T_c \approx 0.8$, the susceptibility for different fields cross each other and above this temperature, the magnitude of χ_{ZFC} increases with increasing field. The decrease of χ_{ZFC} with decreasing temperature, starting from T_c, is less sharp in low fields than in higher. If the field is increased even further, the magnitude of the ZFC susceptibility will eventually decrease with increasing fields at all temperatures due to vortex penetration.

The above results can be understood in terms of an independent π-loop model combined with Abrikosov vortices. The contributions to the magnetization in samples containing π loops are of several different origins: Positive spontaneous moments from π loops with large enough value of the parameter ζ, a positive paramagnetic response from π loops with $\zeta < 1$, a diamagnetic response from loops containing 0 junctions or an even number of π loops, a diamagnetic Meissner response from the bulk material, and possibly also a positive contribution from vortices. In the case of zero-field cooling, the spontaneous moments of the π junctions will not show any preferred direction until the field is switched on at low temperature. After the field is applied and the energy degeneracy of the two states of a π loop is broken, the thermal energy governs how efficiently the energy barrier between the two states has been overcome in the time scale of the experiment. Thus, the importance of positive magnetization from the π loops will increase with increasing temperature. The fact that the penetration depth increases with temperature further emphasizes the effect of temperature on the polarization of spontaneous moments, since at high temperature, a larger part of the sample will be penetrated by a polarizing field. In addition, the energy barrier of the π loops will decrease with increasing temperature, since the parameter ζ is dependent on the critical current. However, this also means that the number of π loops generating spontaneous moments, instead of displaying a paramagnetic behavior, will decrease as the temperature is increased. At the higher temperatures, where the π loops play a larger role in determining χ_{ZFC}, the susceptibility becomes less negative as the field is decreased. This implies that the contribution from the spontaneous moments is more important relative that from vortices and diamagnetic currents the lower the field is. Hence the increase of the number of polarized moments with increasing field has a weaker field dependence than the competing effect of a linear diamagnetic response.

A similar discussion can be applied to the χ_{FC} results. Here, the sample is penetrated by the magnetic field during cooling, giving rise to a splitting of the energy levels for the two directions of the spontaneous currents at high temperatures where the thermal energy is comparatively large, and hence to a polarization of the moments. As in the ZFC case, the relative importance of the spontaneous moments is largest for the smallest fields. The initial drop in χ_{FC} just below T_c can be accounted for by the low value of ζ near T_c. When the temperature is lowered, the critical current and hence ζ increases, causing an increased number of the π loops to generate spontaneous moments. The experimental observation that χ_{FC} approaches a constant value as the low-field limit is approached is also found from calculations using the simple model by Sigrist and Rice.[9,12]

In Fig. 3 the field dependence of χ_{ZFC}, χ_{FC}, and M_{TRM}/H is plotted at 70 K. It is worth noting that the fundamental relation $M_{TRM}/H = \chi_{FC} - \chi_{ZFC}$ is valid also for a PME sample. The main contribution to M_{TRM} at low fields is from polarized π loops.

B. Time dependence of the magnetization

Below the critical temperature, the results presented in the previous section do not reflect equilibrium properties of the sample, but all discussed magnetic properties show a dependence upon time, which in the $\chi(T)$ experiments appears as a dependence upon cooling and heating rates. This time dependence of the magnetization will be further investigated in the following.

The relaxation of the ZFC magnetization is always towards a less diamagnetic behavior. This is to be expected both from an ordinary vortex description of the process and from a π-loop model. The ZFC susceptibility versus time at 60 and 70 K is plotted in Fig. 4. At the temperatures and fields and in the time window investigated the relaxation rate, $S_{ZFC} = \partial M_{ZFC} / \partial \log_{10}(t)$, is almost time independent, i.e., the relaxation is close to logarithmic. As can be seen in the figure, the deviation from a pure logarithmic behavior is largest for small fields at 60 K and for large fields at 70 K. At 60 K the curvature of χ_{ZFC} versus $\log(t)$ is more positive the smaller the field is, and at 70 K it is negative for the larger fields. Ascribing the main relaxation at low fields to the polarization of π loops and making the realistic assumption that the distribution of relaxation times of the spontaneous currents in the sample is broad and display a maximum for some relaxation time $\tau_{max}(T,H)$, one can understand the trends in the deviation of S_{ZFC} from a constant value. At low temperatures and low fields, the observation times are smaller than τ_{max} yielding an increase of the relaxation

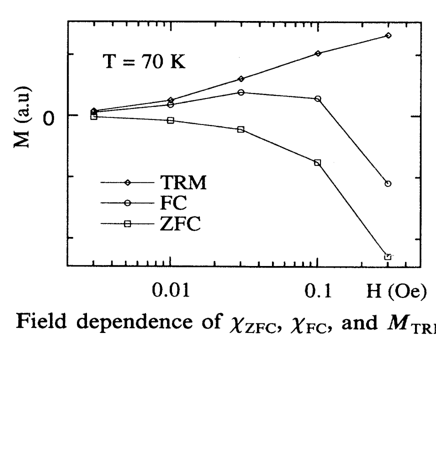

FIG. 3. Field dependence of χ_{ZFC}, χ_{FC}, and M_{TRM}/H at 70 K.

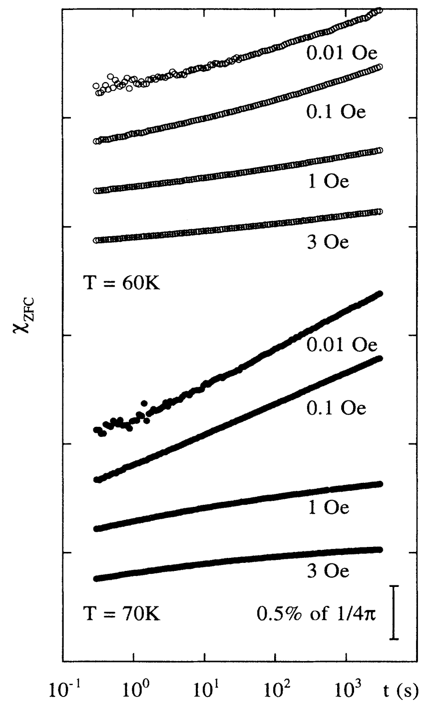

FIG. 4. Time dependence of the zero-field-cooled susceptibility for fields in the range 0.01–3 Oe at 60 and 70 K.

rate with time in the experimental time window, while at high temperatures and high fields τ_{max} is smaller than the observation times resulting in a corresponding decrease of the relaxation rate. At even higher fields, the relaxation due to vortices will dominate.

In Fig. 5, $(1/H)S_{ZFC}$ is shown versus temperature for three different fields, where S_{ZFC} has been determined at 100 s. The normalized relaxation rate exhibits a maximum, which is positioned at higher temperature the smaller the field is. A maximum in the temperature dependence of the relaxation rate is consistent with both the π-loop model and with a vortex description. At low temperatures, the thermal energy is small yielding a slow relaxation, whereas a high temperatures, the relaxation becomes faster. On approaching T_c, a major part of the relaxation has occurred at times shorter than the time window investigated, implying a decrease of the measured relaxation. In a π-loop description, the number of loops giving spontaneous currents decreases with increasing temperature, adding to the decreased relaxation rate observed. A shift of the maximum towards lower temperatures as the field is increased is consistent with a critical-state model of vortex penetration.

In Fig. 6, the FC susceptibility at 60 K is plotted versus observation time for fields in the region 0.01–3 Oe. One clearly sees that the relaxation is positive for the small fields, even for some of the fields where the measured magnetization is negative. For field as large as $H=3$ Oe, the relaxation has become negative. The magnitude of the normalized relaxation rate, $1/H|S_{FC}|$, is smallest for fields around 1 Oe. S_{FC} is defined as $S_{FC}=\Delta M_{FC}/\Delta\log_{10}(t)$, where the difference is taken be-

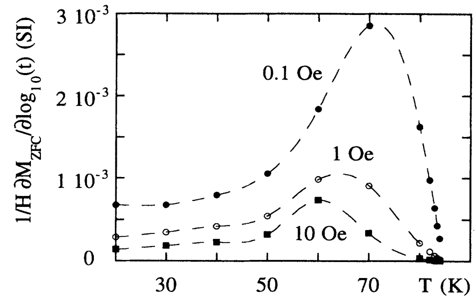

FIG. 5. Temperature dependence of the relaxation rate of the zero-field-cooled magnetization for fields 0.1, 1, and 10 Oe.

tween 300 and 3000 s. The temperature dependence of $(1/H)S_{FC}$ is shown in Fig. 7. $1/H|S_{FC}|$ versus T shows a maximum, although in the field region investigated the maximum is positioned at higher temperatures the higher the field is, contrary to that of $(1/H)S_{ZFC}$.

A positive field-cooled relaxation cannot be explained within a vortex picture. Since H_{c1} is smaller the higher the temperature, one will, in a field-cooled experiment on a type-II sample containing pinning sites, get trapped vortices that are not in an equilibrium state. Hence the number of vortices will decrease with time, yielding a relaxation towards a more diamagnetic state. On the other hand, for an assembly of π loops, the relaxation is expected to be positive in both field-cooled and zero-field-cooled experiments. In this case, the relaxation of the magneti-

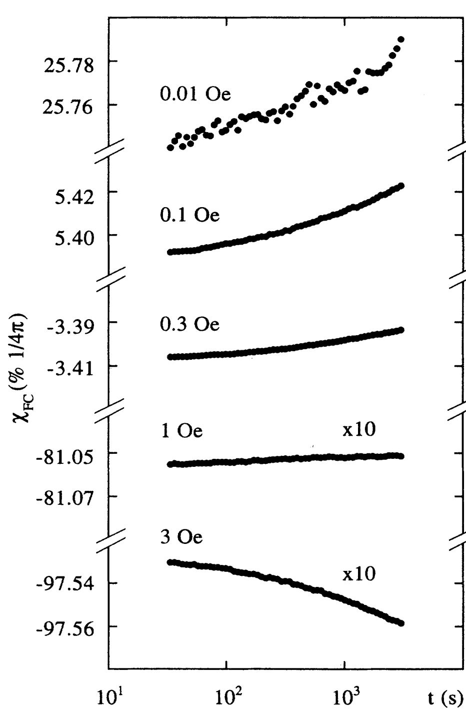

FIG. 6. Field-cooled susceptibility versus observation time for fields 0.01–3 Oe at 60 K. The susceptibility for the two largest fields is enlarged 10 times.

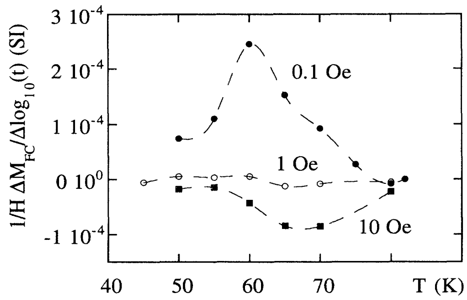

FIG. 7. Temperature dependence of the relaxation rate of the field-cooled magnetization for fields 0.1, 1, and 10 Oe.

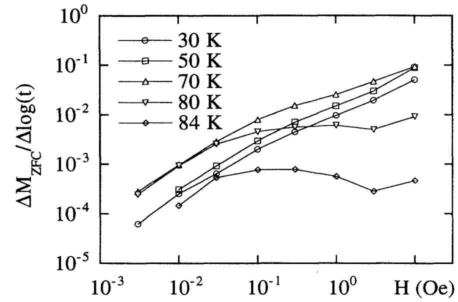

FIG. 8. Zero-field-cooled relaxation rate versus magnetic field at 30 (○), 50 (□), 70 (△), 80 (▽), and 84 K (◇). In the low-field regime the relaxation rate depends almost linearly on the applied magnetic field.

zation is due to thermally activated flipping of the spontaneous orbital moments. Thus, in samples containing π loops, the field-cooled relaxation is governed by two competing effects. In the low-field region, where there are few vortices, the relaxation is dominated by the positive π-loop relaxation. At larger fields, the negative vortex relaxation gives the dominant contribution since it has a stronger field dependence than the contribution from the π loops. Concerning the temperature dependence of $S_{\rm FC}$, a similar discussion as that for $S_{\rm ZFC}$ is relevant. However, in addition to the dependence on thermal energy, experimental time window and the value of ζ, the recorded temperature (and field) dependence of $S_{\rm FC}$ also depends on how fast the sample was cooled to the measurement temperature, since part of the relaxation takes place during cooling.

The positive field-cooled relaxation of the magnetization implies that there is a positive contribution to the magnetization of the ground state. In the π-junction picture, this means that the spontaneous currents are stable. Additionally, the expulsion of vortices causes a negative contribution to the relaxation. The time scales for these two relaxation processes might differ considerably. However, at low enough fields, the positive contribution from spontaneous currents dominates the diamagnetic response due to Meissner currents and currents in 0 loops ($\chi_{\rm FC} > |\chi_{\rm ZFC}|$) and one can conclude that the ground state has a paramagnetic magnetization.

The field dependence of the ZFC relaxation rate is shown in Fig. 8. For low fields, $S_{\rm ZFC}$ increases almost linearly with field at all temperatures. At higher temperatures, the relaxation rate levels off when the field is increased and at the two highest temperatures it even starts to decrease. At the highest fields, there is a tendency to an upwards bending of the relaxation rate curves. In an earlier investigation by Niskanen *et al.*[13] reaching higher fields, the initial linear increase of $S_{\rm ZFC}$ with field was at lower temperatures shown to be followed by an H^2 dependence.

Numerical results of Magnusson *et al.*,[12] obtained by recognizing that the π loops behave as thermally activated asymmetric two-level systems and using a broad distribution of relaxation times, also show a linear field dependence of $S_{\rm ZFC}$ at low fields. The experimentally observed linear field dependence of $S_{\rm ZFC}$ at low fields indicates that the relaxation is dominated by the polarization of π loops in this field region. However, with increasing field, the energy difference between the two current directions increases and the energy barrier for flipping moments to the favorable direction decreases. The relaxation time spectrum is thus displaced towards shorter times and, at high enough fields, there will eventually remain very few spontaneous moments that have not already flipped at a time shorter than those in the experimental time window. This explains a weaker field dependence, or even a decrease, of the relaxation rate with increasing field. With decreasing temperature the relaxation times increase and at the lower temperatures, the field region where a leveling off of the relaxation rate occurs is not reached at our field strengths. At these lower temperatures and with increasing fields, a competing relaxation mechanism appears: ordinary flux creep. The usual flux-creep and critical-state models predict a stronger field dependence of $S_{\rm ZFC}$ for fields between H_{c1} and the field yielding full flux penetration: $S_{\rm ZFC} \propto H^{\alpha}$, where $\alpha \geq 2$ (the value of α depends on the field dependence of the critical current).[14] For fields lower than H_{c1}, no relaxation is to be expected from a model based on flux creep. Thus, the tendencies of the current investigation and the results by Niskanen *et al.* indicate that the ZFC relaxation is dominated by polarization of π loops in the low-field region and by flux creep at high fields.

We have discussed the unusual magnetic properties of our PME sample in terms of an assembly of noninteracting π loops. However, if the π-loop concept is correct, there will inevitably be dipole-dipole forces between the different π loops. It is also possible that one has a network of connected π and 0 loops in the sample, which may result in collective effects implying spin-glass-like properties of the sample.[11] One signifying property of slowly relaxing systems with random interactions is the existence of an ageing phenomenon. Spin glasses inherently possess ageing properties.[17] Small particle systems, only interacting via dipole-dipole forces, show ageing properties when the particles are close enough, while ageing is not resolved for more dilute particle systems.[18] Ageing in magnetic systems manifests itself through a dependence of the response to a magnetic field upon wait time t_w, t_w being the time elapsed between the points of time when the measurement temperature was reached and a probing field was applied. Ageing experiments were performed on the PME sample at temperatures in the range 22–78 K using wait times $t_w = 30$, 300, and 3000 s. The sample was either cooled in a field (0.1–1 Oe) or in zero field. At the measurement temperature, a small probing field (0.003–0.1 Oe) was applied, and the magnetization versus time was recorded. Within our experimental resolution, no dependence upon wait time was detected in the magnetization of the sample. This indicates that the collective network effects are weak and that the dipole-dipole interaction energy between the different spontaneous π-loop moments is small compared to the thermal energy and to the energy barrier between the two different states of a π loop. A numerical estimation of the relative importance of the dipole-dipole interaction can be made using the relation $E_{d-d} = (\mu_0 / 4\pi k_B)(\bar{\mu}^2 / r^3)$ for the interaction energy. Using the maximum measured value of the field-cooled magnetization at 70 K (4×10^{-1} A/m at 0.03 Oe) and assuming the size of the loops to be of the order 1 μm and j_c of the junction to be 10^4 A/cm^2 yield an interaction energy corresponding to a temperature of the order of 1 K.

IV. CONCLUSIONS

The above presented positive relaxation of the FC magnetization in low fields can be explained by thermally activated flipping of spontaneous moments generated by loops containing an odd number of π junctions, while a vortex description is not capable of explaining these results. In higher fields, the FC relaxation becomes negative, indicating that the relaxation due to vortices dominates that due to π loops. The relaxation rate of the ZFC magnetization shows an anomalous field dependence at low fields, which is consistent with an earlier numerical study[12] of a model based on π loops. The relaxation of the measured magnetization in the particular sample studied does not depend on the age of the system, which indicates that the interaction between the different spontaneous moments is weak compared to the energy barrier and the thermal energy.

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[1]P. Svedlindh, K. Niskanen, P. Norling, P. Nordblad, L. Lundgren, B. Lönnberg, and T. Lundström, Physica C **162-164**, 1365 (1989).

[2]W. Braunisch, N. Knauf, V. Kataev, S. Neuhausen, A. Grütz, A. Kock, B. Boden, D. Khomskii, and D. Wohlleben, Phys. Rev. Lett. **68**, 1908 (1992).

[3]W. Braunisch, N. Knauf, G. Bauer, A. Kock, A. Becker, B. Freitag, A. Grütz, V. Kataev, S. Neuhausen, B. Boden, D. Khomskii, D. Wohlleben, J. Bock, and E. Preisler, Phys. Rev. B **48**, 4030 (1993).

[4]S. Riedling, G. Bräuchle, L. Lucht, K. Röhberg, H. v. Löhneysen, H. Claus, A. Erb, and G. Müller-Vogt, Phys. Rev. B **49**, 13 283 (1994).

[5]M. S. M. Minhaj, D. J. Thompson, L. E. Wenger, and J. T. Chen, Physica C **235-240**, 2519 (1994).

[6]Ch. Heinzel, Th. Theilig, and P. Ziemann, Phys. Rev. B **48**, 3445 (1993).

[7]L. N. Bulaevskii, V. V. Kuzii, and A. A. Sobyanin, Pis'ma Zh. Eksp. Teor. Fiz. **25**, 314 (1977) [JETP Lett. **25**, 290 (1977)].

[8]F. V. Kusmartsev, Phys. Lett. A **169**, 108 (1992); Phys. Rev. Lett. **69**, 2268 (1992).

[9]M. Sigrist and T. M. Rice, J. Phys. Soc. Jpn. **61**, 4283 (1992); J. Low. Temp. Phys. **95**, 389 (1994).

[10]B. I. Spivak and S. A. Kivelson, Phys. Rev. B **43**, 3740 (1991).

[11]D. Domínguez, E. A. Jagla, and C. A. Balseiro, Phys. Rev. Lett. **72**, 2773 (1994).

[12]J. Magnusson, J.-O. Andersson, M. Björnander, P. Nordblad, and P. Svedlindh, Phys. Rev. B **51**, 12 776 (1995).

[13]K. Niskanen, J. Magnusson, P. Nordblad, P. Svedlindh, A.-S. Ullström, and T. Lundström, Physica B **194-196**, 1549 (1994).

[14]Y. Yeshurun, A. P. Malozemoff, F. Holzberg, and T. R. Dinger, Phys. Rev. B **38**, 11 828 (1988).

[15]P. Norling, P. Svedlindh, P. Nordblad, L. Lundgren, and P. Przyslupsky, Physica C **153-155**, 314 (1988).

[16]D.-X. Chen, A. Hernando, F. Conde, J. Ramíres, J. M. González-Calbet, and M. Vallet, J. Appl. Phys. **75**, 2578 (1994).

[17]L. Lundgren, P. Svedlindh, P. Nordblad, and O. Beckman, Phys. Rev. Lett. **51**, 911 (1983).

[18]T. Jonsson J. Mattsson, C. Djurberg, F. A. Khan, P. Nordblad, and P. Svedlindh (unpublished).

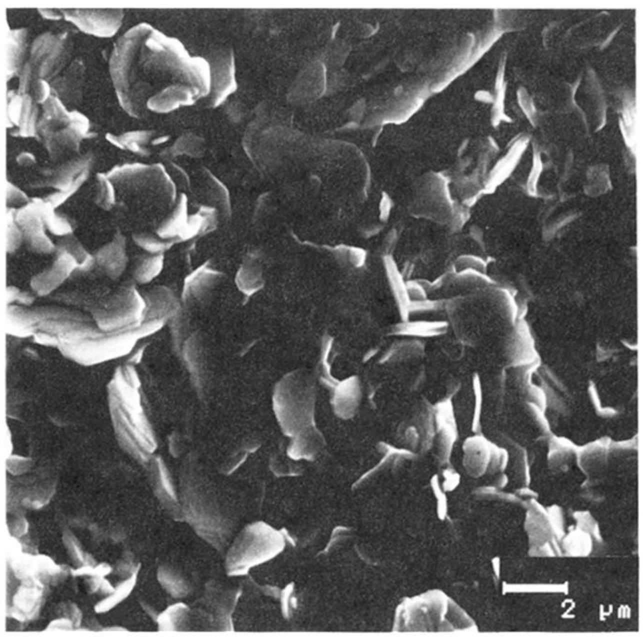

FIG. 1. Scanning electron micrograph of the studied Bi-2212 sample, seen in the direction of pressure applied during preparation.