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Ageing, rejuvenation and memory phenomena in a Bi-2212 superconductor showing the paramagnetic Meissner effect

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Abstract. A melt-cast $Bi_2Sr_2CaCu_2O_8$ sample showing the paramagnetic Meissner effect (PME) and an ageing phenomenon has been studied by magnetic relaxation and ac-susceptibility experiments. A memory behaviour is observed in the low frequency ac-suscpetibility and in the magnetisation *vs.* temperature curves measured on heating after certain cooling protocols. It is also found that large enough temperature shifts and positive temperature perturbations cause rejuvenation of the ageing system. All these observations show striking similarities with the ageing behaviour of spin glasses and indicate the existence of a low temperature glassy phase in this PME material.

PACS. 74.25.Ha Magnetic properties – 74.72.Hs Bi-based cuprates

1 Introduction

The paramagnetic Meissner effect (PME) is inherent to certain $Bi_2Sr_2CaCu_2O_8$ (Bi-2212) high temperature superconductors [1]. This PME effect is likely to originate from the existence of π-junctions and the possibility to form narrow current loops containing an odd number of such junctions [2–4]. The magnetic moments associated with these current loops experience an energy barrier for flipping from an 'up' to a 'down' state, which can be overcome thermally or magnetically. A wide distribution of such energies causes the system to slowly relax when a magnetic field is applied or removed from the sample [5]. In addition, it has been shown from experiments [6] and simulations [7] that under certain circumstances the current loops (magnetic moments) interact with each other by a mechanism that allows random sign and magnitude. This interaction gives rise to a glassy low temperature state that exhibits an ageing behaviour with similarities to the ageing observed in spin glasses [8].

In this experimental study, it is found that there also exist rejuvenation and memory phenomena of the ageing states that have been imprinted in the PME system at specific temperatures. These phenomena show similarities with the corresponding phenomena occurring in spin glasses [9,10].

2 Sample and experiments

The sample investigated is a melt-cast Bi-2212 bulk sample, which has been extensively studied in earlier reports [6,11,12]. The ordering temperature, T_c, is 87 K and the sample exhibits a positive field cooled magnetisation (PME) in low enough magnetic fields ($H < 0.3$ Oe). Transmission electron microscopy and electron diffraction studies of Bi-2212 samples have revealed profound microscopic characteristics that distinguish samples showing the PME from non-PME samples [13]. To introduce the sample, we show in Figure 1 the temperature dependence of

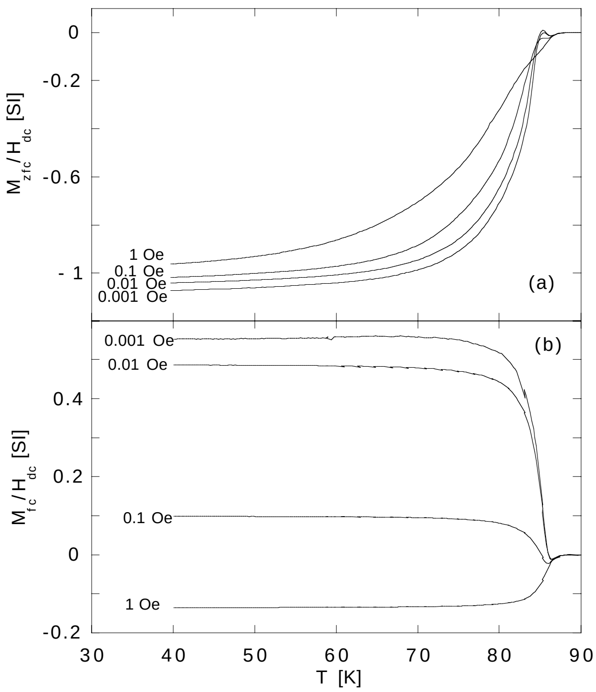

Fig. 1. M_{ZFC}/H (a) and M_{FC}/H (b) of the melt-cast Bi-2212 sample at different applied dc fields as indicated in the figure. From reference [4].

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the field cooled (FC) and zero field cooled (ZFC) 'susceptibility' (M/H) demonstrating a strong PME in the weak field FC curves and an almost complete shielding in the ZFC curves at low temperatures [6].

The experiments were performed in a non-commercial SQUID magnetometer [14]. In this equipment, measurements of the magnetisation *vs.* temperature or time are always performed while keeping the sample fixed in the centre of one of the two middle pick-up coils of a third order gradiometer. The procedures employed to investigate non-equilibrium (ageing) dynamics of the current PME sample are inherited from corresponding studies of spin glasses and other disordered magnets. The relaxation of the ZFC magnetisation is measured by cooling the sample rapidly from a temperature above T_c to the measurement temperature T_m, where the sample is kept a wait time t_w before the weak magnetic field is applied and the magnetisation is recorded as a function of the time elapsed, t, after the field application. In an ageing system, the relaxation depends on the wait time. The ageing process is also reflected in a decay of the low frequency ac susceptiblity with time at constant temperature. The correspondence between the time scales in the two procedures is that $t=1/\omega$ (where ω is the angular frequency of the applied ac field) and the age, t_a, of the system is given by the time the sample has been kept at constant temperature.

3 Results

In Figures 2a–c results from ZFC magnetisation relaxation measurements are displayed. In Figure 2a, the relaxation of the ZFC magnetisation measured after different wait times at 82 K is shown. The three curves look different, showing a decreased relaxation rate ($S = \partial M/\partial \log(t)$) with increasing age and exhibiting an inflection point at an observation time closely equal to the wait time. A behaviour that already was demonstrated in reference [6] for the same melt-cast Bi-2212 sample. Such an influence of age on the dynamics is common to many disordered magnetic systems [8]. In Figures 2b and 2c, the ZFC relaxation has been recorded after a wait time of 3000 s, but just prior to the field application, the sample is subjected to a temperature cycling of magnitude ΔT. Positive cyclings are shown in Figure 2b and negative temperature cycles in Figure 2c. The two curves labelled $\Delta T = 0$ and inf. (infinity) are ordinary ZFC experiments measured with $t_w = 3000$ s and 0 s, respectively, without any temperature perturbation, and are used as reference curves. It is seen from the figures that positive temperature cyclings of large enough magnitude rejuvenate the system to respond to the field application as if it had been immediately cooled from a temperature above T_c. On the other hand, the system is indifferent to negative temperature cycles, *i.e.* the system responds as if the temperature disturbance had not been enforced. These relaxation experiments show that the system, when kept at constant temperature, spontaneously reorganises itself towards a more favourable configuration. They also suggest that the

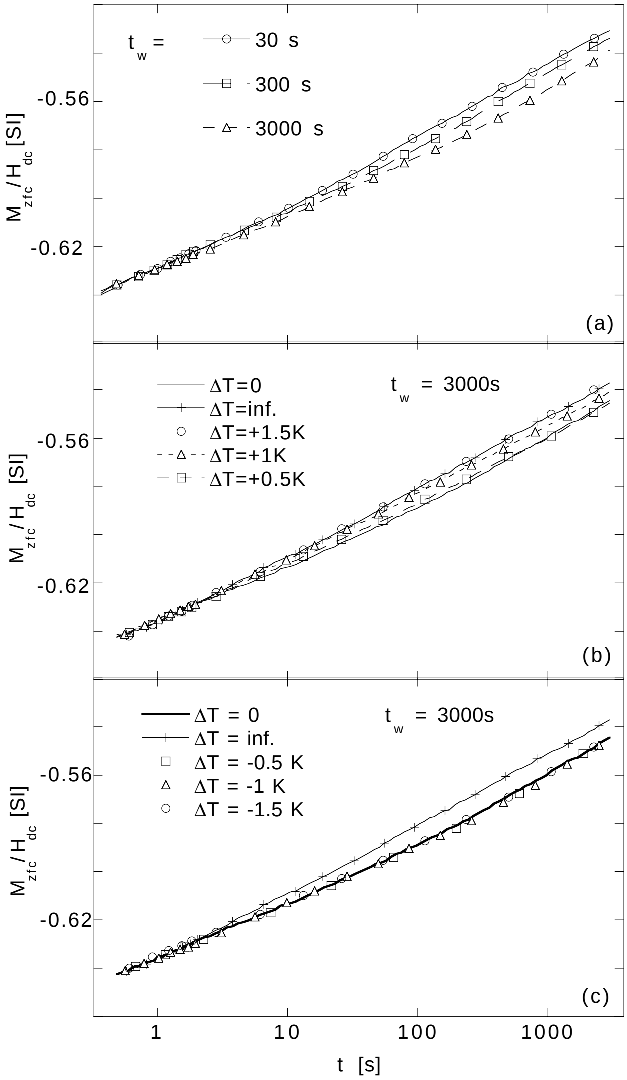

Fig. 2. $M_{\rm ZFC}/H$ *vs.* $\log(t)$ measured at 82 K in an applied field of 0.01 Oe. (a) different wait times 30, 300, 3000 s, (b) $t_w = 3000$ s and different magnitudes of positive temperature cycles ΔT, (c) $t_w = 3000$ s and different magnitudes of negative temperature cycles ΔT. $\Delta T =$ inf. has been measured by cooling the sample to T_m and then immediately applying the magnetic field; *i.e.* a $t_w = 0$ curve. $H_{dc} = 0.01$ Oe.

achieved state is disrupted by a positive temperature perturbation, but that a memory of the ageing state is imprinted in the system if the sample is cooled down and this state is retrieved when the sample is heated back to the temperature where it was aged.

To further elaborate on this subject, we have performed experiments where the sample is cooled in zero (or a weak) applied field to a low temperature according to a certain protocol and then re-heated at a constant rate in a weak (or zero) applied field, a procedure adopted from reference [10]. Figure 3 shows the results of experiments according to two simple cooling protocols: (i) a constant cooling rate is employed (reference curve) and (ii) the same cooling rate is employed, but the cooling is halted for about 3 h at 82 K after which the cooling

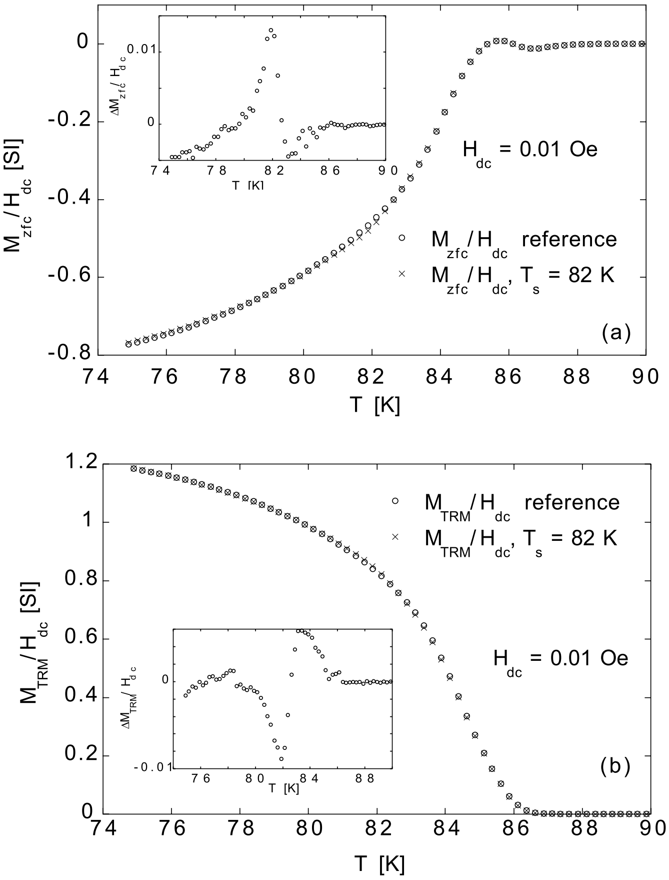

Fig. 3. The ZFC (a) and TRM (b) magnetisation measured *vs.* temperature. The different curves are measured without a halt (reference) and with a halt of about 3 h at $T_s = 82$ K. In the insets the difference between the reference and the 'stop' curves ($\Delta M/H_{dc}$) in the main frames are plotted.

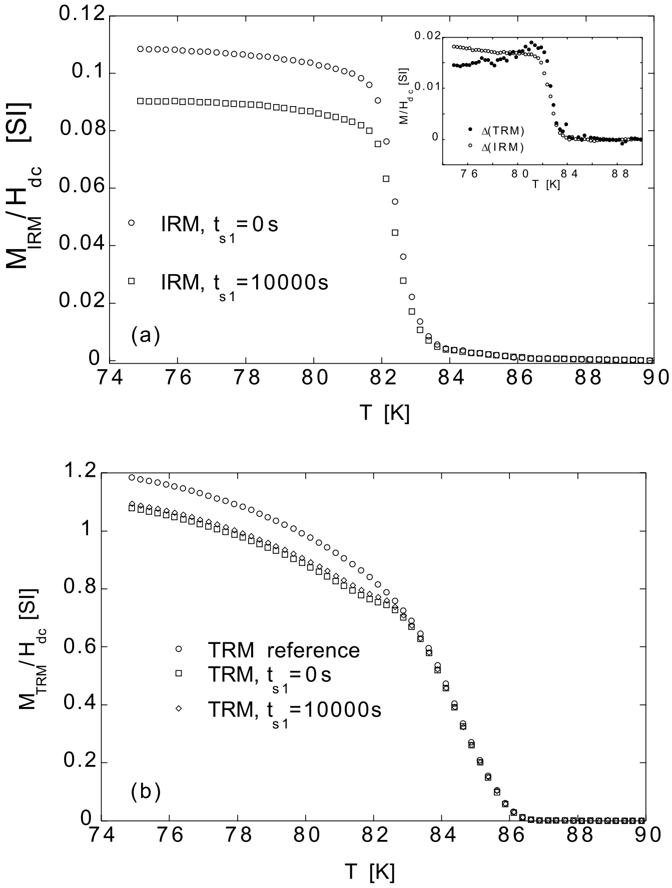

Fig. 4. IRM (a) and TRM (b) measured according to the procedure described in the main text. In the inset to (a), the difference between the two curves recorded after $t_{s1} = 10\,000$ and 0 s in the main frames are depicted. The reference curve in (b) is the ordinary TRM curve measured on heating after continuous cooling. $H_{dc} = 0.01$ Oe.

is resumed. At the lowest temperature the weak field is switched on (ZFC) or turned off (thermoremanent magnetisation (TRM)) and the sample is heated at a constant heating rate while the magnetisation is recorded. Figure 3a shows the results of the ZFC procedure and Figure 3b for the TRM procedure. In the insets of the figures the difference between the reference and the 'halt' curves is shown. As is seen from the curves there is a memory of the halt at constant temperature imprinted in the system on re-heating the sample. The influence of the halt is largely confined to a rather narrow temperature region surrounding the halt temperature. During the halt at constant temperature, the system has aged and the configuration imprinted yields a somewhat more robust system at temperatures surrounding T_s enforcing a weak anomaly – dip (bump) – in the ZFC (TRM) curve relative to the reference curve.

The ageing-memory behaviour may also be illustrated by the isothermal remanent magnetisation (IRM) and TRM curves shown in Figures 4a and b. The experimental protocol is the following: the sample is cooled in zero field (or in a weak dc field) to T_s, where the sample is kept a time t_{s1} (0 or 10 000 s); then a weak magnetic field is applied (turned off) during 10 000 s; the field is again turned off (switched on) and the sample is immediately cooled to the lowest temperature (where the field is turned off for the TRM case) after which the sample is re-heated while the remanent magnetisation is recorded. The resulting curves for the IRM procedure are shown in Figure 4a. The profound difference between the two curves reflects the difference in the relaxation of the magnetisation after after a wait time of 0 s and 10 000 s at T_s (*cf*. Fig. 2). The corresponding two curves for the TRM protocol are shown in Figure 4b, where also the ordinary TRM curve without any extra field switches or halts is shown for comparison. The difference between the two TRM curves employing field switches is clear but relatively smaller than in the IRM case. However, in the inset, the difference between the two IRM and the two TRM curves are shown, yielding a quite similar magnitude and temperature dependence.

An experimental procedure that for spin glasses has turned out to effectively expose ageing, rejuvenation (chaos) and memory behaviour of the dynamics is certain low frequency ac-susceptibility experiments. If the system is rapidly cooled from above T_c and the cooling is halted at

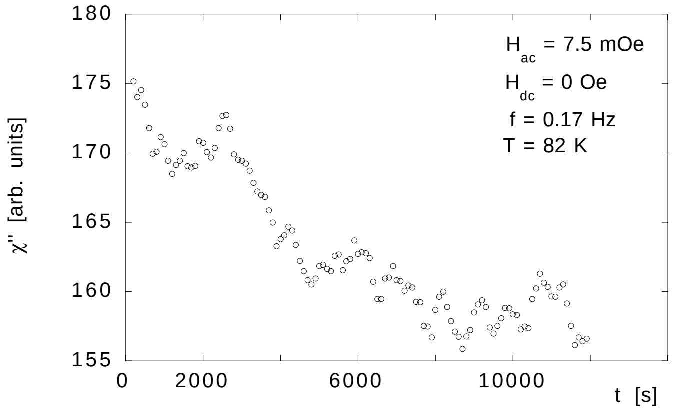

Fig. 5. The relaxation of the out-of-phase component of the 0.17 Hz ac susceptibility at 82 K. This decay of χ" was recorded during the stop at constant temperature in the curve denoted 'stop' in Figure 6.

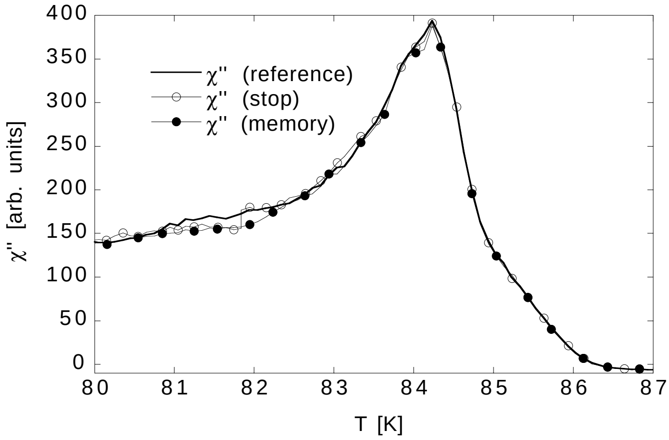

Fig. 6. χ"(T) measured on heating after cooling the sample at a constant cooling rate. The curve 'reference' was recorded after cooling without a halt and the curve 'memory' was recorded after a halt of about 3 h has been imposed at 82 K during cooling. For comparison, also the curve denoted 'stop' is plotted, which was measured during cooling including the stop at 82 K. $f = 0.17$ Hz, $H_{\rm ac} = 7.5$ mOe.

a certain temperature, the magnitude of the low frequency ac susceptibility decays over extended time scales. An example of such a behaviour for the current PME sample is shown in Figure 5, where the out-of-phase component of the ac susceptibility ($f = 0.17$ Hz) is plotted *vs.* time spent at 82 K. There is a significant decay of χ" with time. To further explore this signature of ageing, we have employed the same temperature protocol as in Figure 3 for the ZFC and TRM magnetisation and also a procedure that has been used to demonstrate the memory behaviour of spin glasses [9]. The sample is cooled at a constant rate to a lowest temperature and the ac susceptibility (0.17 Hz) is recorded on continuously heating the sample. Two experiments are performed one without a halt (reference) and one with a halt at constant temperature for about 3 h (memory). Figure 6 shows the results of this experiment. The reference curve shows a smooth behaviour, whereas the memory curve shows a shallow dip around the halt temperature. Also plotted in the figure is the cooling curve including the halt at 82 K (denoted 'stop'). The behaviour reveals a rejuvenation phenomenon, *i.e.* the ageing at $T_{\rm s}$ causes an equilibrated state that is only relevant in a rather narrow temperature region around $T_{\rm s}$. When the temperature is shifted away from this temperature, the dynamics is similar to a situation when the sample has been directly cooled to this temperature and indistinguishable from the dynamics governing the reference curve. However, the memory of the equilibration at $T_{\rm s}$ is retrieved when this temperature is recovered on heating.

4 Discussion and conclusions

Earlier studies on the dynamics of sintered Bi-2212 samples have shown that the PME gives rise to relaxation phenomena on all time scales covered by ordinary susceptibility and magnetisation experiments (10^{-4} to 10^4 s) immediately at temperatures below $T_{\rm c}$. However, no indication of a collective nature of the relaxation processes was observed [5]. The current melt-cast sample has earlier been shown to exhibit an ageing phenomenon [6] and indications of critical slowing down [12] that suggest the existence of a low temperature spin glass-like phase. Assuming that the PME arises from interaction between the spontaneous current loops containing π-junctions, it has been shown from Monte Carlo simulations that frustration and ageing are inherent properties of a 3-dimensional network of Josephson junctions containing 0 and π-junctions [7].

The empirical results presented above are intended to further explore the collective nature of the PME state of a melt-cast Bi-2212 sample by allowing direct comparisons with the behaviour of *e.g.* ordinary spin glasses. The behaviour observed regarding non-equilibrium dynamics shows all the characteristics of a spin glass phase, albeit of a significantly less pronounced magnitude. In our interpretation of the results, we attribute the ageing behaviour to originate from the PME state and being decoupled from the dynamics caused by thermally generated vortices, which contribute substantially to the relaxation and the dissipation in the sample [11]. The part of the observed relaxation and dissipation that is to be related to stationary processes (non-ageing) is large and thus it explains the weak ageing effect relative to the magnitude of the measured relaxation. It is however quite striking that the PME dynamics owns all the characteristic features of the non-equilibrium dynamics of spin glasses. The results are thus indicative of the formation of a low temperature glassy phase in a system showing a pronounced PME, a phase that resembles the chiral glass phase promoted by Kawamura [15].

A positive fc magnetisation has also been observed in measurements on other systems, such as disks of Nb [16–18] and Al [18]. Since these are conventional low-$T_{\rm c}$ superconductors, a mechanism based on the existence of

π-junctions can not explain their positive fc magnetisation. However, several other models have been developed, attributing the positive fc magnetisation to flux trapped in the bulk of the material due to inhomogeneous cooling [19] or to the persistence of a giant vortex formed in the bulk due to finite size effects [20]. The mechanisms behind these models all assume that the sample is cooled in a magnetic field through the transition temperature, T_c. The signatures assigned to the PME in this paper all rest upon a spontaneous formation of magnetic moments (in zero as well as finite applied magnetic field). Also, most of the experiments in the current study are performed after cooling the sample in zero applied magnetic field and probing the system at low temperatures with a weak dc or ac-field. *I.e.*, models requiring a finite cooling field to create the magnetic moment responsible for the PME are not applicable to our results. Indeed, the observed results can only be understood in terms of models prescribing the appearance of spontaneous magnetic moments below T_c. It is, in this context, important to note the direct observation by Kirtley *et al.* [21] of spontaneous magnetic moments on a Bi-2212 sample by SQUID-microscopy.

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