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Time dependence of mixed state magnetisation in $La_{1.8}Sr_{0.2}CuO_4$

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Abstract. The results of temperature and time (at $T = 4.2$ K) dependent magnetisation (M) measurements on the polycrystalline high-T_c superconductor $La_{1.8}Sr_{0.2}CuO_4$ at different magnetic fields H, $4\,\text{kOe} \leq H \leq 35\,\text{kOe}$, are reported. The diamagnetic shielding and Meissner effect are studied. Like other authors, we found a glass-like time dependence of the shielding effect. A new phenomenon is found in the Meissner effect: the inversion of the sign of the magnetisation from negative to positive during a time in which other conditions are unchanged. The differential susceptibility remains diamagnetic in the state with positive M. It is shown that there exists a temperature $T_0 < T_c$ such that when $T < T_0$ the usual mixed state with $M < 0$ for the field cooled regime is metastable and evolves rapidly to a new metastable mixed state with $M > 0$ and $dM/dH < 0$. Such a phenomenon is not observed in traditional type II superconductors.

1. Introduction

In this paper we report the results of temperature and time dependent magnetisation (M) measurements on the high-T_c superconductor (HTS) $La_{1.8}Sr_{0.2}CuO_4$ in magnetic fields H in the range $4\,\text{kOe} \leq H \leq 35\,\text{kOe}$. A single-phase sample with the superconducting temperature $T_c = 36$ K and mass density $\rho = 6\,\text{g cm}^{-3}$ was prepared using standard ceramic technology reported elsewhere [1]. According to the results of AC susceptibility measurements [1] the sample maintains 100% of the superconducting phase. The sample was of cubic form of dimensions $4 \times 4 \times 4\,\text{mm}^3$.

The magnetic measurements were performed using a string magnetometer [2]. String magnetometer operation is based on the Faraday method. The magnetic force $\boldsymbol{F}$ acts on the sample placed in a non-uniform magnetic field $\boldsymbol{H}$: $\boldsymbol{F} = (m/\mu)\chi \boldsymbol{H} \cdot \nabla \boldsymbol{H}$ (here χ is magnetic susceptibility, m and μ are respectively sample mass and molecular weight). This force is transmitted by a long quartz rod to a fine (40 μm) tungsten wire (string) and the degree of tension, $T = P + F$ (here P is the weight of the quartz rod with the sample on its lower end), of the string undergoes changes as a function of the force applied, $\boldsymbol{F}$. The tension determines the string's natural oscillation frequency $f = \frac{1}{2}(T/ml)^{1/2}$ (here m is the string mass and l is the string length). For measuring f, the string is placed into the bore of a permanent magnet and connected to an amplifier feedback circuit, so that the string and the amplifier act as a self-generator. The frequency of that generator is determined by the natural oscillation frequency of the string. Measuring f permits

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determining the sample's susceptibility χ or magnetisation $M = \chi H$. The string generator stability is 10^{-6}; the long-time apparatus zero stability (for 10 h) was better than 0.005%.

By the excitation conditions of string oscillation, the attaching point of the quartz rod's upper end to the string is the vibration node. Therefore the string oscillations are not transmitted to the rod, and the sample does not oscillate relative to a magnetic field source. A superconducting solenoid was used as the magnetic field source. High self-induction of the solenoid ($L = 2.82$ H) circuited on the stabilised power supply (better than 10^{-4}) excludes magnetic field pulsations. The uncontrolled long-time deviation of H was less than 0.4 Oe. These conditions exclude the possibility of dynamic magnetisation caused by changing of the magnetic flux $\partial\Phi/\partial t$ through the sample due to its movement in the internal non-uniform magnetic field, or due to pulsation of $\boldsymbol{H}$. Use of such a technique allows us to conserve the strictly stationary conditions for the sample, and as regards H and ∇H.

As was shown in [3], the measured quantity in our experiment is the magnetisation M averaged over the sample volume, and not a combination $(M + H\,\mathrm{d}M/\mathrm{d}H)$ as in the case of the sample interacting with the magnetic field of a short circuited solenoid or permanent magnet (as in [4]).

The measurements were performed in two regimes:

(i) diamagnetic shielding (ZFC)—the sample was cooled at $H = 0$ down to $T = 4.2$ K, then a magnetic field was applied and $M(t)$ was measured (t is the time);

(ii) Meissner effect (FC)—the sample was cooled from room temperature at non-zero H down to $T = 4.2$ K, then exposed for a long time and heated up to $T > T_c$, and H was kept constant (the magnetometer readings were recorded through the whole 'cooling–exposure–heating' cycle).

The measurements at the different cooling rates were carried out in the FC regime from 1 to 20 K min^{-1}; no appreciable difference in the $M(t)$ dependences was found.

2. Results and discussion

2.1. Diamagnetic shielding

At $T = 4.2$ K, χ_{ZFC} (measuring the shielding effect) was near 20% of ideal value $-1/4\pi$ at $H = 1$ kOe. Like other authors [5] we observed a slow decay of χ_{ZFC} with the time. The time dependence is approximately logarithmic in agreement with the results reported earlier [5] for a La–Ba–Cu–O compound, and after 4 h of exposure χ_{ZFC} was near 80% of its initial value. Such behaviour of χ_{ZFC} is typical for HTSs and agrees with Anderson's predictions [6] for a glass-like state. A bulk of literature now exists on such magnetic relaxation effects in HTSs as well as in traditional superconductors (see [7, 8]).

2.2. Susceptibility and Meissner effect measurements

The temperature dependences of the FC magnetisation for $La_{1.8}Sr_{0.2}CuO_4$ are shown in figure 1 for a few values of H (all the data are presented in the dimensionless units $M/H = \rho\chi$ in which the value $-1/4\pi$ corresponds to the ideal diamagnetism). These data agree qualitatively with the results reported in [5] for the analogous compound La–Ba–Cu–O. It will be noted that according to our data, the temperature at which the diamagnetic contribution to χ appears is practically independent of H up to $H = 35$ kOe.

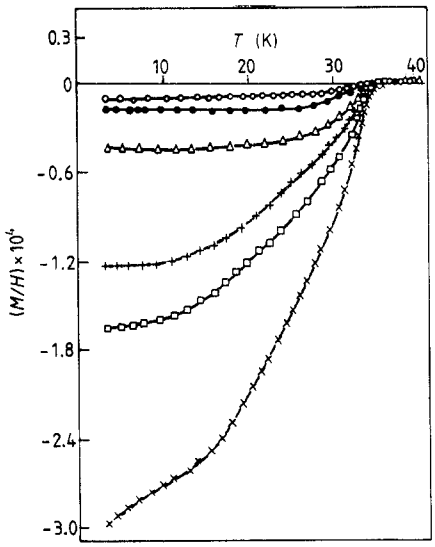

Figure 1. Temperature dependences of FC magnetisation of HTS $La_{1.8}Sr_{0.2}CuO_4$ cooling at H = 4 kOe (×); 6 kOe (□); 7.5 kOe (+); 10 kOe (△); 20 kOe (●) and 35 kOe (○).

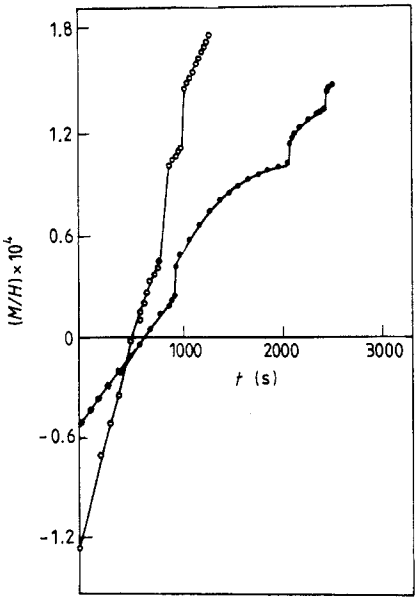

Figure 2. Time dependences of the magnetisation of FC $La_{1.8}Sr_{0.2}CuO_4$ at T = 4.2 K after cooling in the magnetic fields H = 7.5 kOe (○) and 10 kOe (●).

At $T > T_c$, $\chi = 0.25 \times 10^{-6}\,\mathrm{cm^3\,g^{-1}}$ ($=1.5 \times 10^{-6}$ in dimensionless units) and is independent of up to $T = 300$ K. The small value of the normal state susceptibility and absence of its temperature and magnetic field dependences exclude the possibility of the presence of any ferromagnetic or paramagnetic inclusions in quantities that can appreciably influence the measured results at $T < T_c$.

A quite unusual time dependence of the magnetisation, $M(t)$, is found for the FC sample. Two examples of such $M(t)$ behaviour are shown in figure 2 for HTS $La_{1.8}Sr_{0.2}CuO_4$ after cooling at H = 7.5 kOe and H = 10 kOe. As can be seen from figure 2, the magnetisation of the FC sample decays rapidly down to zero with time, then changes its sign to positive and then grows in the positive values region. The $M(t)$ dependence takes the form of quasi-logarithmic segments between which $M(t)$ changes by 'jumping' (a similar $M(t)$ behaviour for Y–Ba–Cu–O single crystals was found in [9] but without the sign inversion of M).

It is important to emphasise that in the positive-M region (for $t > 10$ min, see figure 2) the differential susceptibility, i.e. the sample response to small variations of H (in the limits of 20 Oe), is diamagnetic: $\chi \equiv \mathrm{d}M/\mathrm{d}H < 0$. In other words, the sample remains superconducting. Diamagnetic response to the small changes of H in the state with $M > 0$ (M_+-state [3]) indicates also that positivity of M is not connected with ferromagnetic or paramagnetic impurities but rather has the same origin as a mixed state with $M < 0$, i.e. is caused by the non-vanishing superconducting currents. It is interesting that, if large enough changes of H ($\Delta H > 100$ Oe) or mechanical action on the sample in the M_+-state are made, a state forms immediately with $M < 0$ which is similar to the ZFC state. This means that the M_+-state is evidently metastable. The measurements carried out by

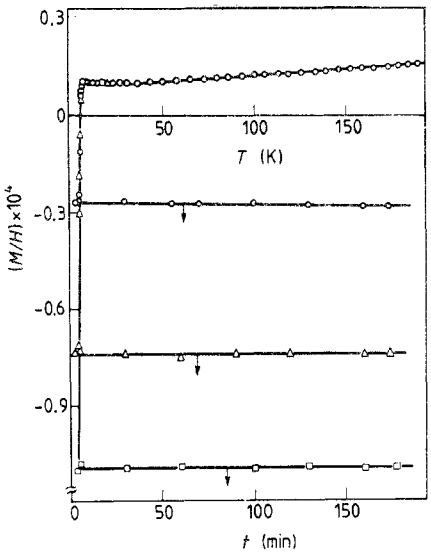

Figure 3. Temperature and time (at $T = 4.2$ K) dependences of magnetisation of the traditional type II superconductor VN during and after cooling in the magnetic fields $H = 5$ kOe (□), 10 kOe (△) and 20 kOe (○).

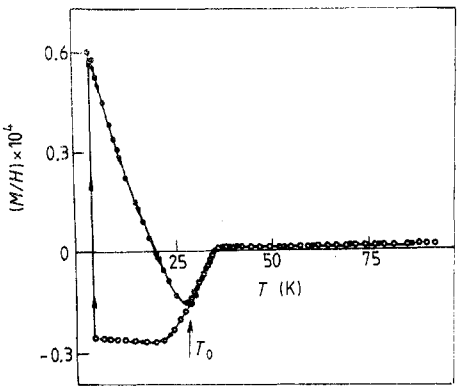

Figure 4. The results of measurements of the whole 'cooling (○)–exposure for $t = 50$ min (——)–heating (●)' cycle for $H = 20$ kOe.

us on samples of other forms and sizes show that the results are independent of these factors.

Thus it follows from our results that the usual mixed state with $M < 0$ in HTS $La_{1.8}Sr_{0.2}CuO_4$ turns out to be metastable, and evolves rapidly to a new mixed state with $M > 0$, $dM/dH < 0$ which is metastable too. We found a similar $M(t)$ behaviour also for poly- and mono-crystalline $YBa_2Cu_3O_{7-\delta}$ [3] and for other known high-T_c superconducting compounds ([10], [14]). However, analogous investigations of the traditional type II superconductors (namely Nb, Nb_3Al, VN, Ti–V alloys) do not show any sign of time dependence of the magnetisation in either FC or ZFC regimes. As an example, in figure 3 the results of such measurements are shown for a VN sample with $T_c = 12$ K. The mixed state magnetisation of this compound after cooling in the different magnetic fields remains constant within the limits of measurement accuracy on the same timescale (figure 3 shows also that the surprising $M(t)$ behaviour of HTS is not connected with a drift of the apparatus zero). Therefore, we conclude that the FC mixed state evolution with the alteration of magnetisation sign is an intrinsic property of HTSs that is characteristic of high-T_c superconductors but not of traditional superconductors.

It is well known (see for example [11]) that the metastable mixed state with $M > 0$ and $dM/dH < 0$ may be reached in the imperfect type II superconductors with developed vortex pinning by decreasing the applied magnetic field (trapped flux state). The results of suspending the samples under the permanent magnet (which were described recently for both HTSs and the traditional type II superconductor Nb_3Sn [12]) are clear illustration of that.

We do not think that the M_+-state discovered in our experiments has the same origin. As can be seen from the results of figure 2, in the positive region magnetisation values

are of the same order or even greater than the initial values in the 'usual' mixed state. To trap such a flux, magnetic field decrease must be of the order of H itself. At the same time, as was pointed out above, we performed our measurements at constant H with an accuracy better than 0.4 Oe, i.e. 10^{-5} H. Moreover, we should have observed analogous effects for traditional type II superconductors in which the vortex pinning is developed strongly; however, we did not (see above).

Therefore, we believe that the origin of the M_+-state in HTSS is not simply reduced to the appearance of the redundant vortex concentration, but is rather caused by the unusual nature of the vortexes themselves in HTSS. In our opinion, such surprising FC mixed state magnetisation behaviour of HTSS is caused by the existence of a developed structure of Josephson junctions in these compounds (in a single crystal [3] their role may be played by twin boundaries). A model explaining the appearance of the M_+-state is suggested by Druzhinin in [3].

The measured results for the whole 'cooling–exposure–heating' cycle in the FC regime at $H = 20$ kOe are shown in figure 4. It can be seen that there exists a temperature $T_0 < T_c$ (generally dependent on H [3]) such that at $T > T_0$ the results of the measurements are reversible. In other words, in the region $T_0 < T < T_c$ the usual mixed state with $M < 0$ is stable. At $T < 20$ K, some increase of $M(T)$ takes place with decreasing temperature which we relate to the metastability of the usual mixed state for $T < T_0$ and its evolution to the M_+-state. It should be pointed out that in [13] a low temperature minimum in $M(T)$ was found for a $YBa_2Cu_3O_7$ compound cooled in an external magnetic field. We believe that such minima may be caused by the phenomena discussed here.

In conclusion, the data presented in this paper, in combination with the results of [3, 10], show that the mixed state of HTSS turns out to be very unusual. In the ZFC regime, the mixed state evolution may be explained in terms of a superconducting glass state [5, 6]. In the FC regime, the usual mixed state for $T < T_0$ is essentially unstable and evolves rapidly to a metastable mixed state of a new kind (the M_+-state); this is apparently caused by the particular nature of the vortexes in these compounds. Determination of this nature requires the following investigations.

The question remains of why the phenomenon described here was not observed by others. It is difficult to give a full reply because that would demand a full analysis of the experimental conditions and techniques for each case. However, we can formulate some requirements which must be satisfied for observation of the M_+-state:

(i) the $M(t)$ measurements must be performed for $T < T_0$;

(ii) no changes in H must be made after cooling the sample in the internal magnetic field;

(iii) a technique must be used that excludes the possibility of $\partial\Phi/\partial t$ affecting the measuring procedure because of pulsations of $\boldsymbol{H}$ or oscillations of the sample (even with a very small amplitude) in non-uniform magnetic fields (the latter may be found when the standard Faraday balance technique is used);

(iv) a short circuited superconducting solenoid cannot be used as the internal magnetic field source (see [3]).

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