

## Low-field magnetic relaxation effects in high- $T_c$ superconductors Sr-La-Cu-O and Ba-La-Cu-O

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The relaxation of the isothermal dc magnetization  $M$  in zero-field-cooled Ba-La-Cu-O and Sr-La-Cu-O has been investigated. After the external magnetic field has been changed or cycled, the resultant magnetization  $M$  decays following the law  $M \propto \ln(t/t_0)$  with great accuracy over an observation time period of four decades. At  $H=0$ , the logarithmic decay rate  $\partial M/\partial \ln t$  is proportional to the temperature  $T$  and it is also proportional to  $H_i^3$ . Here  $H_i$  is the external field which is reduced to zero before the decay rate of persistent currents is observed.

Recently, Müller, Takashige, and Bednorz<sup>1</sup> reported magnetic measurements in the Ba-La-Cu-O ceramic showing features expected for a superconducting glass. These include values of the zero-field-cooled diamagnetic susceptibility  $\chi_{dc}$  larger than the field-cooled values, and the observation of irreversibilities and metastable states created both by switching off the field after field cooling the specimens and by switching on a field after zero-field cooling. In both cases, the metastable states gave nonexponential decays as might be expected for a glassy system with many supercurrent-carrying states of nearly equal energy.

Here we report new experimental results of magnetic relaxation on macroscopic time scales when changes of the magnetic field are made at constant temperature. The measurements were done after zero-field cooling (ZFC) at temperatures well below  $T_c$ . In this metastable region the "working point" was at a nearly constant  $-M/H$  value. We observe accurately linear decays of the magnetization

as a function of the logarithm of time over a time interval of four decades. Furthermore, we find that the slope of  $M$  against  $\ln t$ , at  $H=0$ , depends strongly both on temperature and on the magnetic history of the specimen.

Three samples have been investigated:  $\text{Ba}_{0.15}\text{La}_{1.85}\text{CuO}_4$  and  $\text{Sr}_{0.2}\text{La}_{1.8}\text{CuO}_4$  in powder form, and  $\text{Sr}_{0.2}\text{La}_{1.8}\text{CuO}_4$  sintered and cut into a parallelepiped. The preparation of these samples has already been described by Bednorz, Takashige, and Müller.<sup>2</sup> Isothermal dc magnetization curves and ac susceptibility were measured with a superconducting quantum interference device (SQUID) magnetometer<sup>3</sup> in fields up to 385 Oe. The specimens were held at rigidly fixed positions in contact with helium and coupled by means of dc flux transformers to SQUID's. The components  $\chi'$  and  $\chi''$  of the magnetic susceptibility were measured using ac measuring fields lower than 0.3 mOe at frequencies between 15 and 150 Hz. Within this range of frequency we observed only a few tenths of a percent frequency dependence in the

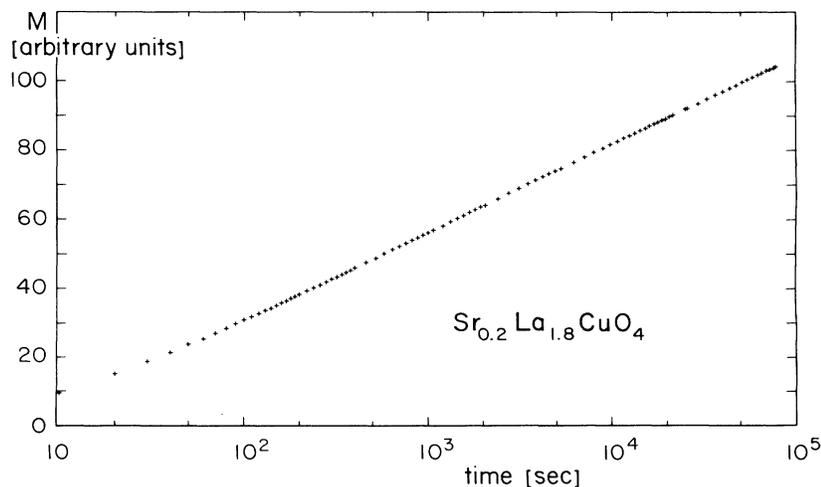


FIG. 1. Decay of the magnetization  $M$  at  $H=0$  as a function of time for powdered Sr-La-Cu-O ( $T=4.2$  K).

values of  $\chi'$  for the three specimens.

Results of the total low-field dc magnetization and susceptibility will be reported elsewhere. Here we shall mention only that at  $T \leq 9$  K both Sr-La-Cu-O specimens showed approximately 100% diamagnetic signals in  $\chi_{ac}$  and  $\chi_{dc}$  for  $H \rightarrow 0$  in agreement with Maletta *et al.*<sup>4</sup> On the other hand, the Ba-La-Cu-O specimen showed only an approximately 5% signal at  $T=9$  K, as already reported by Bednorz, Takashige, and Müller,<sup>5</sup> and about 15% at  $T=0.01$  K.

The measuring procedure for obtaining the decay rate of the isothermal magnetization consisted of zero-field cooling the sample to the desired temperature and then turning on a field of a chosen strength. For each measurement, it was essential to start with the specimen in a "virgin state," i.e., after the specimen had been heated well above  $T_c$ . The cooling time was about one to two hours in each case. By zero field we actually mean the 2 mOe residual field in our cryostat. After waiting a few seconds the field was turned off and the decay of the dc remanent magnetization recorded for about 24 h. A typical curve is shown in Fig. 1 for powdered Sr-La-Cu-O at  $T=4.2$  K after an initial field  $H_i=77$  Oe has been switched off. Here we have plotted the remanent magnetization as a function of the logarithm of time in arbitrary units. In this case the magnetization relaxed from positive values towards zero, that is, flux was expelled slowly through the sample. Similar curves were obtained for the other two specimens.

Metastable states that decayed logarithmically covering a time scale of  $10$ – $10^5$  sec were also observed when the

field was raised from zero up to a certain value and then kept constant. In this case,  $M$  decayed in the opposite direction, i.e., towards smaller absolute values of  $M$ . It is important to point out that at  $T=4.2$  K the decay of  $M$  in the first  $10^5$  sec amounts only to about 1%–2% of the total value of  $M$ .

The dependence of the decay rate at zero applied field on magnetic history is shown in Fig. 2. Here we have plotted  $-\partial M/\partial \ln t$  as a function of  $H_i$ . It should be noted that  $H_i$  is *not* the applied field during the measurement of the decay, but it is the initial field up to which the sample was cycled before the  $H=0$  measurement of  $M$  as a function of time was made. All these points were taken at  $T=4.2$  K. From the data we see clearly that for fields  $25 < H_i < 385$  Oe, the logarithmic decay rate  $-\partial M/\partial \ln t$  is proportional to  $H_i^3$ .

In Fig. 3 we show  $-\partial M/\partial \ln t$  as a function of temperature for a constant  $H_i=115$  Oe. A function of the type  $-\partial M/\partial \ln t \propto T$  describes the data well, indicating that the decay rate is indeed thermally activated.

Logarithmic time decay of the critical state in type-II superconductors is a well-known phenomenon. The first experimental evidence of decay of persistent currents due to flux creep was obtained by Kim, Hempstead, and Strnad<sup>6</sup> and the theory of flux creep in irreversible type-II superconductors was first given by Anderson.<sup>7</sup> Precise measurements by Beasley, Labusch, and Webb<sup>8</sup> confirmed the main predictions of the theory but left some uncertainties about whether the flux creep is thermally activated or not, due to the rather weak temperature dependence observed experimentally. In Anderson's theory the current decays logarithmically with time, and the rate of decay is proportional to the temperature  $T$ .

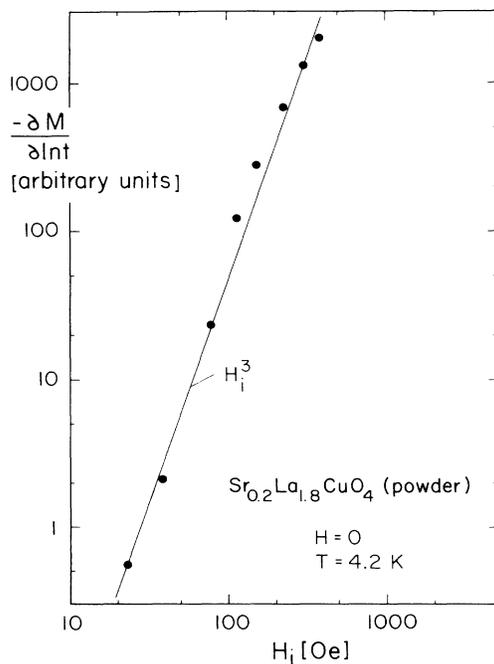


FIG. 2. Logarithmic decay rate at  $H=0$  and  $T=4.2$  K as a function of the external field  $H_i$  up to which the sample was cycled.

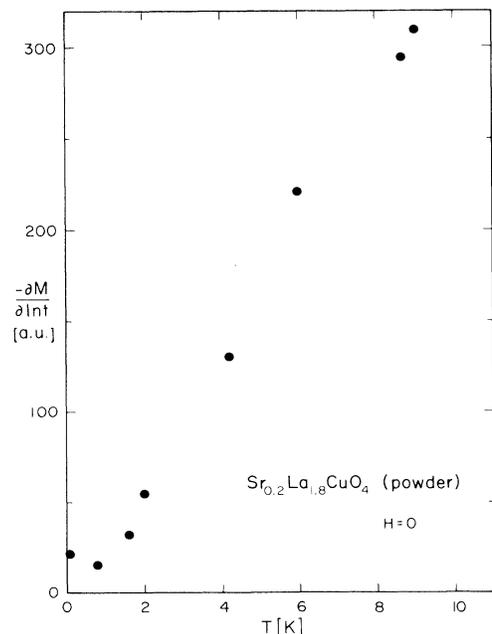


FIG. 3. Logarithmic decay rate at  $H=0$  as a function of temperature. The field  $H_i$  is in this case always equal to 115 Oe.

As a comparison for our observations on high- $T_c$  specimens we studied a bulk PbIn specimen. As already pointed out by Beasley *et al.*,<sup>8</sup> *no flux creep* in the purely diamagnetic region of the initial magnetization curve could be observed. Only by cycling the field above  $H_{c1}$  did we obtain visible flux decays. At low fields, in agreement with those authors, we observed somewhat erratic behavior of  $M$  vs  $t$  with continuous flux jumps of one or several flux quanta. A very different behavior was observed with the Sr-La-Cu-O and Ba-La-Cu-O powdered specimens. At  $H=0$ , the decay of the induced supercurrents proceeded continuously with no visible flux jumps in the scale of one flux quantum  $\phi_0$ , when fields as small as 30 Oe were cycled up and down in an otherwise virgin sample.

Recently, Ebner and Stroud<sup>9</sup> discussed the strong-field behavior of superconducting clusters and predicted various spin-glass features in the diamagnetic susceptibility of such systems. These include remanent magnetization in zero field with anomalous time dependence and strong differences between dc and ac susceptibilities starting at a field  $H$  given by  $H=\phi_0/2S$ . Here  $S$  is the area of a characteristic current loop. In our experiment we do not observe a noticeable difference between  $\chi_{ac}$  and  $\chi_{dc}$  in fields up to about 385 Oe, so we must conclude that, if that model applies to our system, the areas of the loops in-

volved are smaller than  $0.03 \mu\text{m}^2$ .

A comparison of our data with similar data for spin glasses is by no means trivial. In spite of extensive work, the low-field relaxation, as well as the time decay of the remanent magnetization in magnetic spin glasses, are still matters of controversy. Recently, Binder and Young<sup>10</sup> reviewed the experimental and theoretical situation. While some experimental work extending over one decade of time gives a relaxation of the remanent magnetization proportional to  $T \ln(t/t_0)$ , other results extending over two decades are more consistent with a power law or with a fractional exponential decay.

Recently, a large-scale 2D simulation of a superconducting glass model was carried out by Morgenstern<sup>11</sup> using the Hamiltonian  $H=J_{ij} \cos(\phi_i - \phi_j - A_{ij})$  also employed by Ebner and Stroud.<sup>9</sup> He obtained a phase diagram in the  $T$ - $H$  plane. In this diagram there exists a phase of weak randomness for small  $H$  and  $T < T_c$ , whose order parameter is the magnetic moment  $M$ . The "working point" of the present experiment would be located deep in that phase.

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