Journal of Alloys and Compounds, 195 (1993) 471–474
JALCOM 7509

Critical magnetization currents in a hydrogenated $La_{1.85}Sr_{0.15}CuO_4$ superconductor

H. Drulis[a], A. Zygmunt[a], J. Klamut[a] and N. M. Suleĭmanov[b]

[a] Institute of Low Temperature and Structure Research, Polish Academy of Sciences, P. O. Box 937, 50-950 Wrocław 2, Poland*

[b] Kazan' Physicotechnical Institute, Russian Academy of Sciences, 420029 Kazan', Sibirskiĭ Trakt 10/7, Russia

Abstract

An influence of soft hydrogenation on the superconducting properties of the $La_{1.85}Sr_{0.15}CuO_4$ powder system has been studied. DC magnetization measurements performed at several temperatures have revealed a significant enhancement of the intragrain critical current density. A Gabay-Toulouse-like behaviour of the irreversibility lines has been found.

1. Introduction

The critical current densities of high-T_c ceramics are determined mainly by internal structural defects within the grains and by grain boundaries which occur naturally in these materials [1, 2]. However, these natural defects very often do not pin vortices strongly enough to sustain a supercurrent up to critical current density (J_c). To optimize critical transport currents one needs to add a new effective pinning centers and overcome the weak link behavior at the grain boundaries. A significant progress in enhancement of the critical current density in high-T_c superconductors has been achieved through neutron or ion irradiation [3, 4].

The another promising way to improve critical parameters of high-T_c materials might be a soft hydrogenation [5, 6]. It has been established [7-9] that hydrogen behaves in HTSCs as a proton (H^+) and that electron transferred from hydrogen can decrease the number of charge carriers. It causes a gradual disappearance of the superconducting phase and appearance of antiferromagnetic, non-superconducting clusters [10, 11]. In particular, these clusters might act as pinning centers for flux motion [12].

In the present work the influence of hydrogen on irreversibility line of $La_{1.85}Sr_{0.15}CuO_4$ powder sample is reported. The influence of hydrogenation on critical magnetization currents of that system has been published in our previous paper [13].

2. Experimental

Ceramic samples of metalloxides were prepared from mixtures of $SrCO_3$, $CaCO_3$, La_2O_3, and CuO. The mixtures were ground, heated at 900°C for 5 hours in air, and ground again. The products were pressed into pellets, then heated at 1050°C for one hour in air. The reaction of hydrogen gas with $La_{1.85}Sr_{0.15}CuO_4$ was studied by volumetric method in a sealed reaction cell. The powder metalloxide samples were initially outgassed at 110°C in vacuum of 10^{-4} Tr for period of *ca* 30 min. Then, pure gaseous hydrogen was admitted into the cell under pressure of up to 600 Tr. The absorption was performed at constant temperature, 170°C, by monitoring hydrogen pressure as a function of time. The hydrogen content in the sample was determined from the overall decrease of hydrogen pressure in the cell.

Three samples of $La_{1.85}Sr_{0.15}CuO_4H_x$ with H content $x = 0.039$, 0.043, and 0.064 were prepared by this method with accuracy of ±0.002. X-ray measurements performed on the hydrogenated sample with $x = 0.064$ showed

* This work was financed by the National Committee for Scientific Research, Grant No. KBN-2-23-62-9203.

0925-8388/93/$06.00

that there is no modification of the diffraction pattern of the initial metalloxide.

To determine the superconducting temperature, *dc* magnetization measurements were carried out with a SQUID magnetometer (SMS450-S12) in magnetic field of 1.1 Oe and at temperatures down to 4.2 K. The magnetization hysteresis loop measurements were made on powdered samples using the standard Foner vibrating magnetometer in fields up to 5 T.

3. Results and discussion

Figure 1 shows the field-cooled (FC) temperature dependence of the *dc* diamagnetic susceptibility non-hydrogenated and hydrogenated samples. The values of T_c, defined as the onset of superconductivity were determined from the intersection of two extrapolated lines of the magnetization over the temperature region of rapid changes illustrated in Fig. 1. A small increase of T_c from $T_c = 33.5$ K for initial specimen to $T_c = 35.4$ K for hydrogenated one was observed.

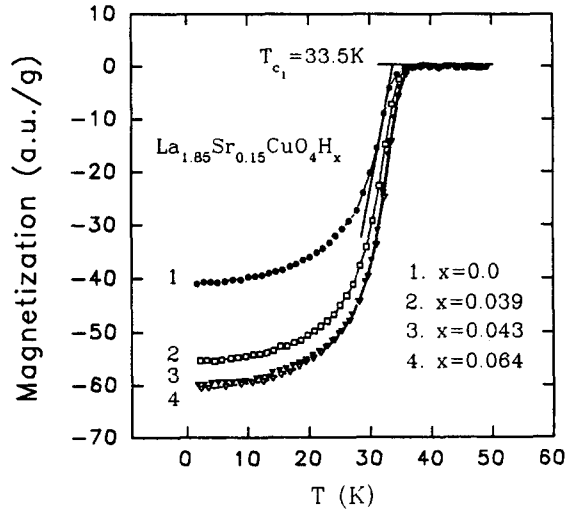

Figure 1. Temperature dependence of the *dc* diamagnetic magnetization measured at $H = 1.1$ Oe for $H_xLa_{1.85}Sr_{0.15}CuO_4$ samples.

Figure 2 shows the magnetic field dependence of $[M^+(H) - M^-(H)]$ where $M^+(H)$ and $M^-(H)$ are the values of magnetization on the ascending and descending branches of the magnetic hysteresis cycle, respectively, for two representative samples with $x = 0$ and $x = 0.064$. The accurate determination of absolute values of J_c from $M(H)$ hysteresis loop is not a light task for ceramic materials. With respect to hydrogen effects, the most reasonable way is to compare the widths of hysteresis loops before and after hydrogenation, $\Delta M_H / \Delta M_0$, which, according to the Bean's critical state model [14] is proportional to the corresponding fractional change of J_c.

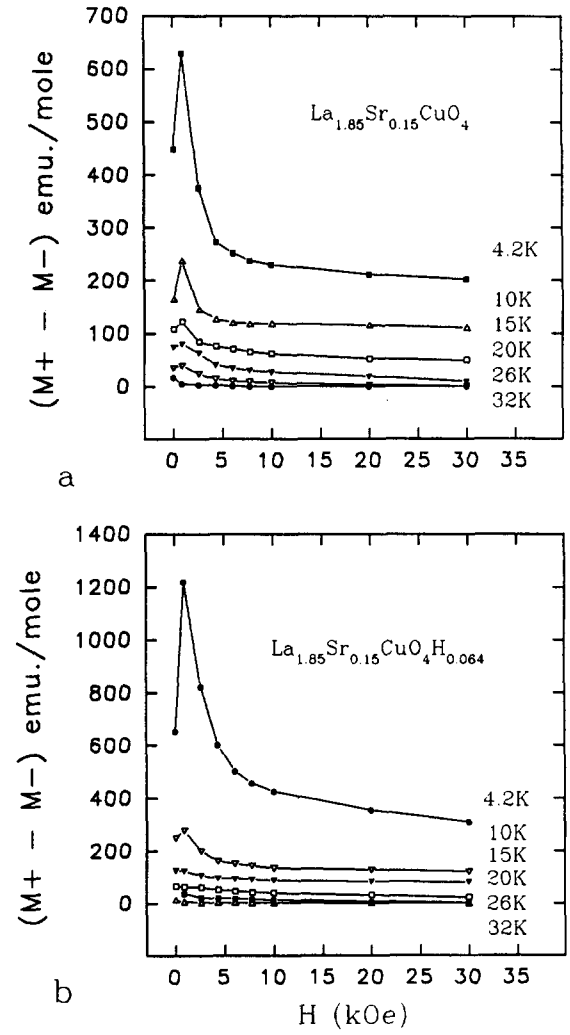

Figure 2. Magnetic field dependence of the magnetic irreversibility $[M^+ - M^-]$ for: (a) – $La_{1.85}Sr_{0.15}CuO_4$; (b) – $H_{0.064}La_{1.85}Sr_{0.15}CuO_4$ at various temperatures.

In Fig. 3 the values of enhancement factor $p = \Delta M_H / \Delta M_0$ at 4.2 K are displayed as a function of magnetic field. Experimental data presented in Figs. 2 and 3 indicate that the magnetic irreversibility $[M^+ - M^-]$ for hydrogenated sample is larger than that of the non-hydrogenated ones. Thus we can assume that introduction of small quantity of hydrogen into the lattice of $La_{1.85}Sr_{0.15}CuO_4$ creates an additional number of pinning centers or strengthens the pinning forces of the already existing ones.

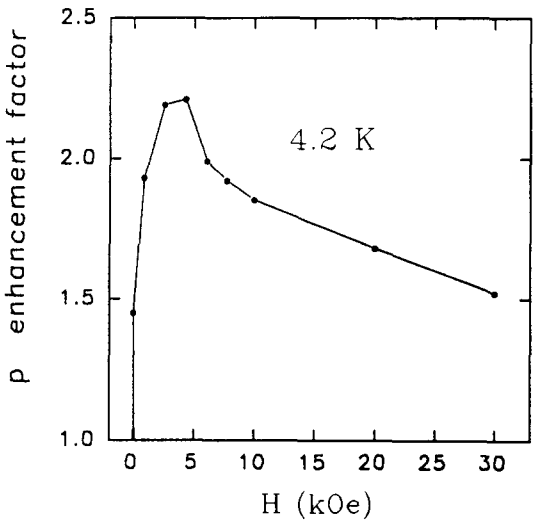

Figure 3. $H_{0.064}La_{1.85}Sr_{0.15}CuO_4$: critical magnetization current enhancement, $p = \Delta M_H / \Delta M_0$, versus applied magnetic field at 4.2 K.

3.1 Irreversibility line

The disappearance of flux pinning at certain fields and temperatures by thermal activation of the flux lines is often referred to as the irreversibility or depinning line. The irreversibility line marks the boundary between reversible and irreversible magnetization and a passage into a "viscous flow" regime for unpinned flux line.

In this work the irreversibility field H_{irr} values were determined from the disappearance of magnetization hysteresis at fixed temperatures. The experimental errors in determining H_{irr} came mainly from the fact that ΔM curves in Fig. 1 reached zero line tangentially. We found also that near the reversible regime the signal to noise ratio was marginal, making difficult to pick out the transition accurately.

All these obstacles allowed us to estimate only few experimental points on irreversibility lines of both studied specimens. Nevertheless, trend of the data collected in Fig. 4 over the entire temperature and magnetic field ranges shows roughly a downward curvature. This curvature can be fitted to a dependence $1 - t = aH^2$, where t is the reduced temperature T_{irr} / T_c^* relative to the zero-field transition temperature.

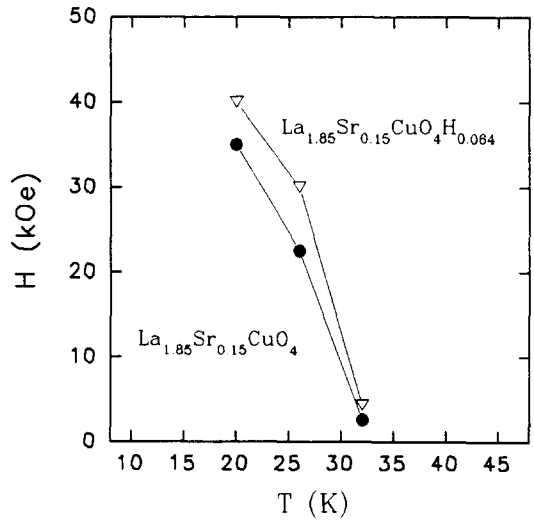

Figure 4. Irreversibility lines versus temperature for non-hydrogenated and hydrogenated $La_{1.85}Sr_{0.15}CuO_4$ sample.

The overall picture looks qualitatively very similar to the "Gabay-Toulouse irreversibility line" in spin glasses [15], which is also often observed on H-T diagrams of superconducting materials in the range of high magnetic fields [16, 17].

Unfortunately, due to the small sensitivity of our magnetometer, we were unable to investigate more accurately the low field part of H-T diagram near T_c where one could expect to find the so-called irreversibility line with de Almeida-Thouless-like behavior ($H^{2/3}$- dependence).

4. Conclusion

In summary, our experiments reveal a Gabay-Toulouse-like behavior of irreversibility curves in $La_{1.85}Sr_{0.15}CuO_4H_x$ superconducting system. After soft hydrogenation the curve turned out to be shifted towards higher temperatures and magnetic fields.

With increased proton concentration in the sample, the density of superconducting carriers n_s decreases because the protons reduce the number of holes in CuO_2 planes. As a consequence, the non-superconducting, probably antiferromagnetic clusters are formed within the material [10-12]. If these micro-nonconducting areas have a size, d, smaller or comparable to the coherence length, ξ, they will play the role of pinning centers.

Acknowledgements

We thank Mr.Mr. M. Działak, Z. Chrapkiewicz and E.F. Kukovitskiĭ for their technical assistance.

References

1 P.H. Kes and J. van den Berg in: *Studies in High Temperature Superconductors, vol. 5,* ed. by A. Narlikar, Nova Sci. Publ., Commack, NY, 1990, p. 83.

2 M. Murakami, M. Morita, K. Doi and K. Miyamoto, *Jpn. J. Appl. Phys., 28* (1990) 1189.

3 K. Ozawa, *A Bibliography of Irradiadion Effect on High-T_c Superconductors,* 1990, Energy Res. Lab., Hitachi, Japan.

4 M.P. Siegal, R.B. van Dover, Julia M. Phillips, E.M. Gyorgy, A.E. White and J.H. Marshall, *Appl. Phys. Lett., 60* (1992) 2932.

5 J.J. Reilly, M. Suenaga, J.R. Johnson, P. Thompson and A.R. Moodenbaugh, *Phys. Rev. B, 36* (1987) 5694.

6 T. Ekino, K. Matasukuma, T. Takabatake and H. Fujii, *Physica B 165 & 166* (1990) 1529.

7 H. Fujii, H. Kawanaka, W. Ye, S. Orimo and H. Fukuba, *Jpn. J. Appl. Phys., 27* (1988) L525

8 K. Donnelly, J.F. Lawler, M.J.D. Coey, D. Fruchart, S. Miraglia and J.L. Soubeyroux, *Physica C, 172* (1990) 207.

9 N.M. Suleĭmanov, H. Drulis, G. Chądzyński, A.D. Shengelaya, E.F. Kukovitskiĭ, R.G. Mustafin and J. Janczak, *JETP Lett., 51* (1990) 422.

10 N.M. Suleĭmanov, H. Drulis, V.E. Kataev, E.F. Kukovitskiĭ and G.W. Chądzyński, *J. Magn. Magn. Mat., 90* (1990) 635.

11 H. Glückler, U. Binninger, Ch. Niedermayer, E. Recknagel and A. Weidinger, *Z. Phys. Chem. - Neue Folge* (to be published).

12 J. Gross and M. Mehring, *Physica C* (to be published).

13 H. Drulis, J. Klamut, A. Zygmunt, N.M. Suleĭmanov and E.F. Kukovitskiĭ, *Solid State Commun.* (in print, 1992).

14 C.P. Bean, *Rev. Mod. Phys., 36* (1964) 31.

15 M. Gabay and G. Toulouse, *Phys. Rev. Lett., 47* (1981) 201.

16 P. Pureur and J. Schaf, *Solid State Commun., 78* (1991) 723.

17 J. Schaf, P. Pureur and J.V. Kunzler, in *Progress in High Temperature Superconductivity, vol. 25,* ed. by R. Nicolsky, World Sci., Singapore 1990, p. 262.