Journal of Alloys and Compounds, 195 (1993) 455–458
JALCOM 7505

Reversibility of the mixed state of the 90K $Tl_2Ba_2CuO_{6+x}$ superconductor

C. Opagiste[a], M. Couach[a], A.F. Khoder[a], F. Monnier[a], A. Junod[b], G. Triscone[b], J. Muller[b], J.L. Jorda[c], T.K. Jondo[c] and M.Th. Cohen-Adad[c].

[a]Centre d'Etude Nucléaire de Grenoble / SPSMS / LCP, BP 85 X, F-38041 Grenoble Cedex (France).

[b]D.P.M.C., Université de Genève, CH-1211 Genève 4 (Switzerland).

[c]Laboratoire de Physico-Chimie Minérale II, Université Claude Bernard Lyon I, F-69622 (France).

Abstract

The reversible mixed state of the $Tl_2Ba_2CuO_{6+x}$ high Tc superconductor with Tc of 90K has been investigated by DC SQUID measurements of the magnetization M(H, T) and by direct measurement of the AC differential susceptibility χ'_H which reduces to the equilibrium dM/dH when, as it is actually the present case in the reversible regime, the flux flow viscous contribution to χ'_H could be neglected.

The two methods give quite similar results: a strong (field enhanced) contribution from diamagnetic fluctuation above Tc, while below Tc one observes deviation from the Abrikosov mean field theory of the mixed state which is presumably generated by thermal fluctuation enhanced effect on the vortex density and distorsions in the layered structure. This behavior appears to be common to all the high Tc superconductors as we have shown in an earlier work [1, 2, 3] for the $YBa_2Cu_3O_{7-x}$, 2223 Tl and 2212 Bi based compounds.

Moreover, these deviations prevent any reliable determination of the H_{C2} critical field line by the standard method of linear extrapolation of M(H, T) curves or by the step anomaly on χ'_H expected to occur at Tc(H) if the mean field theory was to be followed in the mixed state.

1. Introduction

Much experimental work has been and is still devoted to the irreversible behavior of the vortex state of the high Tc superconductors because of its evident practical implications on the critical currents enhancement and more important the usability of these compounds at high temperature, say liquid N_2 temperature, by the control of the location of the so-called irreversibility line H*(T) (by irradiation, or chemical and metallurgical defects). Meanwhile, irreversibility in the mixed state makes too much complicated the study of the basic parameters of the superconductivity which can be extracted from equilibrium thermodynamical properties such as magnetization M(H, T) and specific heat Cp(H, T). In this paper, we will focus on the equilibrium magnetic properties of the mixed state, accessible in the "reversible" regime of the $Tl_2Ba_2CuO_{6+x}$ 90K superconductor and will show that, like in the other high Tc layered superconductors [1, 2, 3, 4, 5], the mixed state magnetic behavior of these compounds deviates seriously from what can be expected theorically within the framework of the Abrikosov mean field theory both in the linear (close to Tc) regime as well in the London regime ($H_{C1} << H << H_{C2}$).

2. Considerations on measurements on ceramics ($H_{C1} << H << H_{C2}$)

The investigated $Tl_2Ba_2CuO_{6+x}$ samples were prepared by using a high pressure, high temperature route as described in [6, 7]. As this compound belongs to a class of highly anisotropic materials, our measurements give an angular average of M and dM/dH. Micrograhs of the investigated samples revealed no preferable texturation [6], thus a random polycrystalline approximation within the framework of an effective mass model to account for anisotropy allows the calculation of an angular

0925-8388/93/$06.00

averaging factor $g(\gamma)$ which gives the ceramic to single crystal (H ∥ c axis) ratio of the measured M or dM/dH where γ is the anisotropy ratio ($(mc/ma)^{1/2} = \lambda c/\lambda a$). The expression of $g(\gamma)$ is that [8]:

$$g(\gamma) = 1/2 \, [\, 1+\gamma^{-1} (\gamma^2 - 1)^{-1/2} \ln[\, (\gamma^2 - 1)^{1/2}+\gamma \,] \,]$$

which reduces to 1 for the isotropic case ($\gamma = 1$) and to 0.5 for $\gamma >> 1$, which is the adequat limit for all the high Tc's. Thus, a multiplying factor of 2 is necessary to scale our results relatively to the single crystal ones measured with H ∥ c axis [3].

We should finally insist on the fact that owing to the relatively low level of M in all the investigated reversible regime, demagnetizing effects could be neglected and that our results are free from geometrical considerations (sample and/or grains size and shape).

3. Experimental results and discussion

The magnetization M of several $Tl_2Ba_2CuO_{6+x}$ samples have been measured with a r.f. SQUID magnetometer in the field range of 0.05 to 5.5 T. Typical results on the sample 12E-A0 are shown in figure (1) for different magnetic field values (we use the MKSA system where $B = \mu_0 (H + M)$).

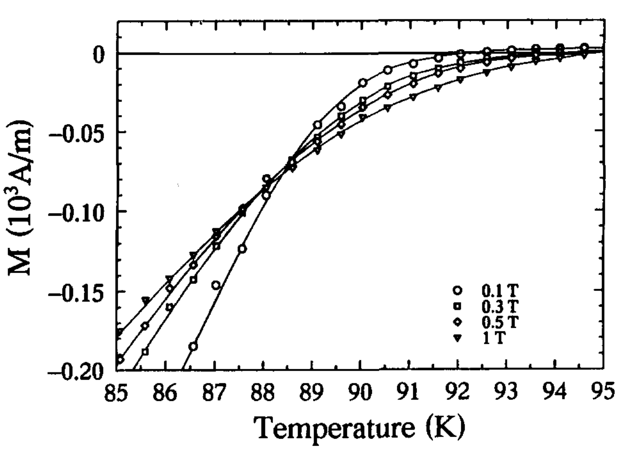

Figure 1: *Magnetization as a function of temperature for different fields.*

A remarkable behavior around Tc (~90K) is apparent which consists of a rounding of M extending to higher temperature with increasing H. The different curves intercept each others at lower temperatures and recover a somewhat less anomalous behavior for lowest temperatures. Linear extrapolation of M(H, T) to determine $Tc_2(H)$ gives a "critical" temperature which increases with H [9] as it has been shown in the case of $Bi_2Sr_2CaCu_2O_8$ single crystals, first by Kritscha et al [4] and by Kes et al [5], and in the case of $YBa_2Cu_3O_{7-d}$ by Junod et al [10] and Welp et al [11]. This field enhancement of diamagnetism is predicted by the theory of the diamagnetic fluctuations [12]. Welp et al [11] and Kes et al [5] have shown respectively the adequacy of a scaling approach to account for such behavior.

Thus, experimentally, the linear regime of M(H, T) close to Tc (although Tc can not be unambiguously determined) does not seem to exist or at least gives a rather surprising result. The same kind of behavior is obtained with very precise and sensitive measurements of the differential susceptibility χ'_H (=dM/dH as the flux flow contribution could be neglected [3]) which goes through a negative minimum around Tc before recovering the positive values at lower temperatures. Comparison of the AC determined χ'_H = dM/dH and the DC $\Delta M/\Delta H$ for $\mu_0 H = 0.1$ T is given in the figure (2) showing the excellent agreement between the two measurements techniques.

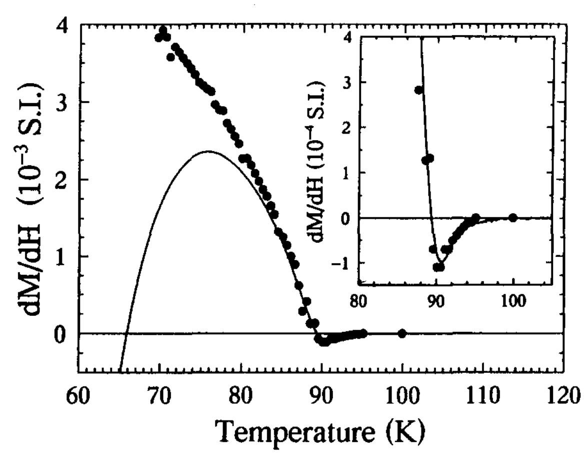

Figure 2 : *Comparison of dM/dH as determined by DC (full circles) and AC (continuous line) measurements at 0.1 Tesla.*

Deviations of the AC results from the DC ones observable at lowest temperature are essentially due to the different time-scale of the two methods ($\tau_{AC} \sim 10^{-2}$ sec while $\tau_{DC} \sim 10^{2-3}$ sec) which makes "irreversibility" appears at highest temperature for the shorter experimental time scale. Actually the rounding of χ'_H and its subsequent decreasing while the DC determined $\Delta M/\Delta H$ is still increasing, reflects the fact that the flux flow contribution is becoming important and can no more be neglected [3].

As the χ'_H negative minimum at Tc reflects the field enhanced diamagnetic fluctuations, we should refear to the first observation of this behavior in Tl-2223 and Bi-2212 ceramics [1, 2, 13] and also in the oxygen off-stiochiometric $YBa_2Cu_3O_{7-x}$ (with x~0.5) single crystal [3].

The expected behavior of the differential susceptibility χ'_H is to show a step increasing at $Tc_2(H)$ given by $1/2\kappa^2$ where κ is the Ginzburg-Landau parameter. This expected step is apparently washed out by an unusually extended fluctuation regime and presumally by large effect of thermal fluctuation on the vortex density [14].

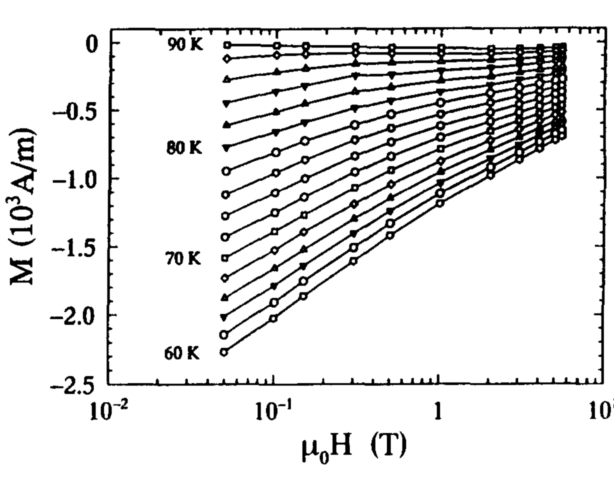

Figure 3: *Magnetization as a function of lnH for different temperature.*

Another regime, the London regime, is also expected to be followed by M(H, T) (or equivalently by dM/dH) immediately below Tc(H) because the condition $H_{C1} << H << H_{C2}$ is fullfilled for any finite value of H in the case of the extreme type II superconductors (large κ). In this regime, M has a lnH dependance or equivalently dM/dlnH is field independant.

$$M = -\frac{\Phi_0}{8\pi\lambda^2}\ln\frac{H_{C2}}{H} \quad \text{and} \quad \frac{dM}{d\ln H} = \frac{\Phi_0}{8\pi\lambda^2} = \frac{H_{C1}}{2\ln\kappa}$$

If these relationships are to be verified measurement of M or of χ'_H allow direct measurement of $\lambda(T)$. As it is shown in figure (3) for M and in figure (4) for $H_*\chi'_H$ (i-e dM/dlnH) the above relationships are not exactly followed and deviations from the lnH dependance of M, although not strong, are clearly observed. If we neglect these deviations, and correct for the actual estimated superconducting volume [6], an estimation of λ (0) can be made and this gives 3300 Å (for λ_{ab}).

In a recent work, Bulaevskii et al [14] suggest by analogy to the Kosterlitz-Thouless transition in layered superconductors, to take into account the entropy contribution to the free energy of the mixed state, which originates from thermal distortions of vortices. This entropy contribution results in a possible spontaneous nucleation of vortex lines at a temperature Ts which would be the analog of Kosterlitz-Thouless transition temperature. Experimentally, Ts will correspond to the temperature where χ'_H goes through zero passing from positive to negative value close to Tc (see figure (4)). The model predicts also that below Ts the magnetization will recover a "normal", although renormalised by Ts replacing Tc, logarithmic dependance on H.

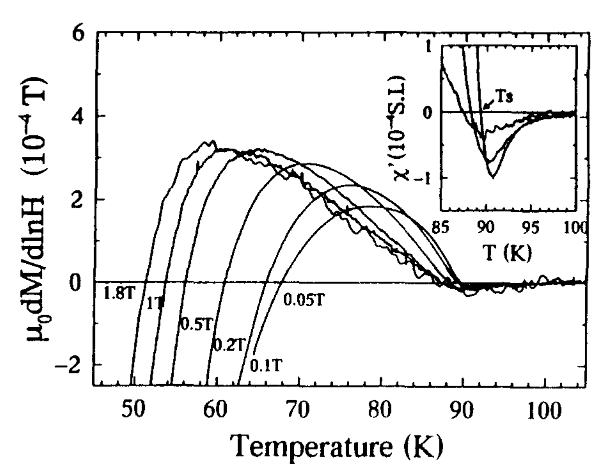

Figure 4: *The curves dM/dlnH (= $H_*\chi'_H$) should superpose if the lnH dependance of M is to be verified. Insert: from right to left 0.1, 0.2 and 0.5T. Ts decreases with increasing field.*

Our experimental observations could be considered as in agreement with this model with some reserve based on the fact that the $\chi'_H(Ts)=0$ criteria gives no unique Ts (equivalently the curves M(H, T) for different H do not intercept in a unique point Ts) and more important that deviations from the lnH relationship are clearly present.

4. Conclusion

Basically the $Tl_2Ba_2CuO_{6+x}$ 90K superconductor behaves magnetically in the mixed state as the Bi based cuprates and the $YBa_2Cu_3O_{7-\delta}$ (δ>0.2), i-e, around Tc thermal fluctuations control strongly the diamagnetic contribution as well as the vortex density. Meanwhile deviation in the reversible mixed state from the Abrikosov mean field theory as evidenced by the direct measurement of the magnetization seems to be more pronounced in the case of the 2201 Tl compound than for other cuprates.. As it has been shown, more effort on pure ceramic compound with detailed studies in the reversible mixte state, could lead to a better understanding of these phenomena.

5. Acknowledgments

The Region Rhône Alpes is greatly acknowledged for supporting this work by the program Eurodoc. The authors are grateful to J.A. Fernandez, F. Liniger and A. Naula for their technical assistance.

References

1 Couach, A.F. Khoder, F. Monnier, J.L. Jorda, M.Th. Cohen-Adad and R. Abraham, High Temperature Superconductors in Materials Science Monographs Vol.70 p353-362, Elsevier Sci. Publ., Amsterdam 1991.

2 A.F. Khoder, M. Couach and J.L. Jorda, Phys. Rev. B 42 (1990) 8714.

3 A.F. Khoder, M. Couach, F. Monnier and J.Y. Henry, Europhys. Lett. 15 (1991) 337.

4 W. Kritscha, F.M. Sauerzopf, H.W. Weber, G.W. Crabtree, Y.C. Chang and P.Z. Tiang, Physica C 179 (1991) 59.

5 H. Kes, C. J. Van der Beek, M. P. Maley, M.E. Mc Henry, D.A. Huse, M.J.V. Menken and A.A. Menovsky, Phys. Rev. Lett. 67 (1991) 2383.

6 C. Opagiste, M. Couach, A.F. Khoder, T.K. Jondo, J.L. Jorda, M.Th. Cohen-Adad, A. Junod, G. Triscone and J. Muller, these proceedings.

7 C. Opagiste, M. Couach, A.F. Khoder, T.K. Jondo, J.L. Jorda,.M.Th. Cohen-Adad, G. Triscone, A. Junod and J. Muller, to be published in physica C.

8 V.G. Kogan, M.M. Fang and S. Mitra, Phys. Rev. B 38 (1988) 11958.

9 G. Triscone unpublished work.

10 A. Junod unpublished work.

11 U. Welp, W.K. Kwok, G.W. Crabtree, K.G. Vandervoort and J.Z. Liu, Phys. Rev. Lett. 16 vol.62 (1989) 1908.

12 For a simple view see Tinkham book on superconductivity and references therein.

13 M. Couach and A.F. Khoder, in Magnetic Susceptibility of Superconductors, an Other Spin Systems, Ed. R.A. Hein, T.L. Francavilla and D.H. Liebenberg Plenum Press (New York 1991) p25.

14 L.N. Bulaevskii, M Ledvig and V.G. Kogan, preprint.