Bull. Mater. Sci., Vol. 14, No. 4, August 1991, pp. 931–935. © Printed in India.

DC magnetization studies of $(Pr_xY_{1-x})Ba_2Cu_3O_{7-y}$

L S VAIDHYANATHAN, C K SUBRAMANIAM and
G RANGARAJAN
Physics Department, Indian Institute of Technology, Madras 600036, India

Abstract. Magnetization was measured on Pr-doped $YBa_2Cu_3O_{7-y}$ for fields up to 5·5T. The paramagnetic behaviour is correlated to the free ion values of Pr^{3+} and Pr^{4+}. Hysteresis experiments were done for superconducting composition for fields up to 1100 Gauss. The intragrain critical current density is calculated using Bean's formula.

Keywords. DC magnetization; paramagnetic behaviour.

1. Introduction

Substitution of Y^{3+} by a rare earth ion in $YBa_2Cu_3O_{7-y}$ does not in general affect the superconducting properties; Pr is an exception to this. In the case of $(Pr_xY_{1-x})Ba_2Cu_3O_{7-y}$, it is known that for $x \geqslant 0{\cdot}6$ no superconductivity occurs above 2 K. The role of Pr ion in suppressing the superconductivity has been attributed to band-filling (Goncalves *et al* 1988) and pair-breaking (Kebede *et al* 1988). Hybridization of Pr-4*f* orbitals with the Cu–O orbitals has also been inferred (Kang *et al* 1989). The possible existence of Pr in 3+ and 4+ valence states has also been discussed earlier (Dalichaouch *et al* 1988).

In previous publications, heat capacity and magnetic susceptibility data have been reported and interpreted in terms of possible crystal field effects giving rise to a Schottky anomaly in the former case (Sankar *et al* 1989). The data also suggested mixed valence for the Pr-ion. However, a serious limitation to these data was the existence of a large diamagnetic moment below T_c for $x < 0{\cdot}5$. Magnetization measurements in the presence of an applied magnetic field in the paramagnetic state may therefore be expected to throw additional light on the problem, since at relatively high fields, the diamagnetic response would be negligibly small in comparison with the paramagnetic response due to the Pr-ion. Magnetization measurements at low fields below T_c may also be expected to provide information on the superconducting state e.g. critical current density as a function of applied field (H), temperature (T) and the composition (x). In this paper, preliminary results of such measurements are reported.

2. Experimental

The $(Pr_xY_{1-x})Ba_2Cu_3O_{7-y}$ samples were prepared by following standard procedures and used in the form of polycrystalline pellets (Natarajan *et al* 1988). Magnetization was measured between 5 K and 90 K in applied magnetic fields of up to 5·5 T, using a quantum design SQUID magnetometer.

3. Results

A typical magnetization curve for $x = 0{\cdot}5$ at 5 K is shown in figure 1. It is clear that for fields up to about 0·1 T, there is a hysteresis response characteristic of a type-II superconductor while for higher fields, the dominant contribution arises from the paramagnetic Pr-moment. The average magnetization of the Pr-ions is shown for $x = 0{\cdot}5$ in figure 2 as a function of H/T along with the theoretically expected free ion values for the Pr^{3+} and Pr^{4+} ions with ground states of $^3H_4$ and $^2F_{5/2}$respectively, calculated from the standard expression

$$M = NgJ\mu_B B_J(gJ\mu_B H/kT),$$

where g is the Lande g factor, J the total angular momentum, μ_B the Bohr magneton, k the Boltzmann constant, N the number of ions, M the magnetization and $B_J(x)$ is the Brillouin function.

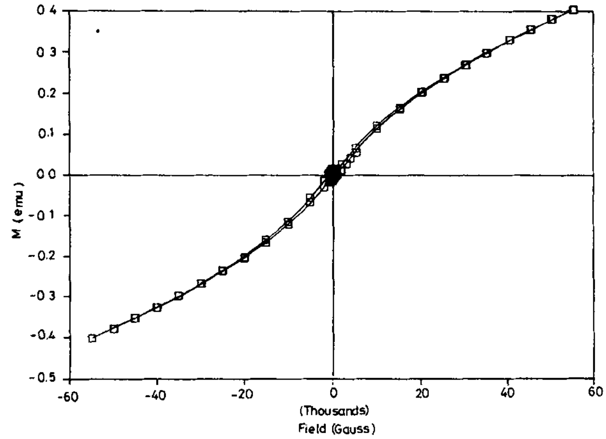

Figure 1. Magnetization vs applied magnetic field for $Pr_{0{\cdot}5}Y_{0{\cdot}5}Ba_2Cu_3O_{7-y}$ at 5 K.

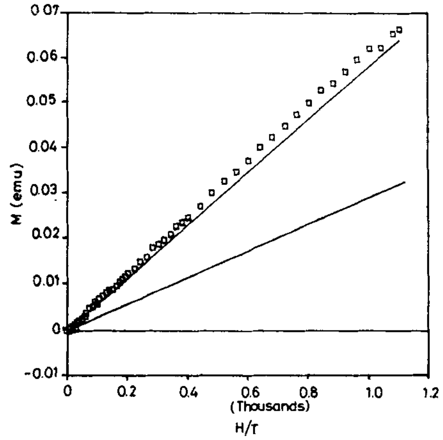

Figure 2. M vs H/T for $Pr_{0{\cdot}5}Y_{0{\cdot}5}Ba_2Cu_3O_{7-y}$ at 50 K along with free ion values for Pr^{3+} and Pr^{4+}.

It is clear from figure 2 that for $T > 20$ K, the magnetization data fit Pr^{3+} values rather closely while they do not agree with the values for Pr^{4+}. It should however be remembered that if crystal field effects are important and are taken into account, this picture may be modified. However at 5 K there is considerable decrease in the magnetization and are consistent neither with the values for Pr^{3+} nor for Pr^{4+} paramagnetic moments. It is however known that there is an antiferromagnetic ordering of Pr-ions below 20 K for $x \geqslant 0{\cdot}6$. This is also the case for samples investigated by us, for which $d\chi/dT$ data are shown in figure 3 for $x = 0{\cdot}6$ and $1{\cdot}0$. There is a clear discontinuity in $d\chi/dT$ though at somewhat lower temperatures (of 10–12 K) than reported. In the case of $PrBa_2Cu_3O_{7-y}$ a heat capacity peak at 12 K was also observed (Sankar, pers. commun.) Neutron diffraction measurements yielded a value of $0{\cdot}74\mu_B$ for the saturated magnetic moment of the ordered Pr-ions (Li *et al* 1989) and this may account for the significant decrease in the magnetization at 5 K in the present case.

Similar measurements carried out for $x = 0{\cdot}6$ are shown in figure 4. In this case however for $T > 35$ K, satisfactory agreement with the measured values could only be obtained by combining the free ion magnetization values for Pr^{3+} and Pr^{4+} in

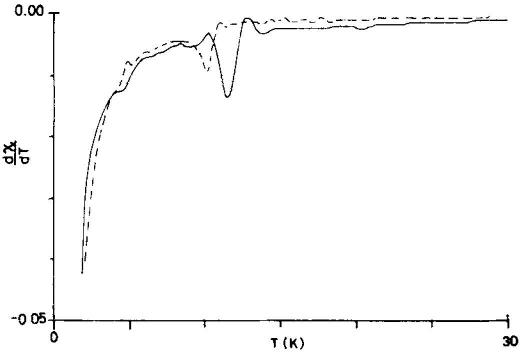

Figure 3. $d\chi/dT$ vs temperature for $Pr_{0{\cdot}6}Y_{0{\cdot}4}Ba_2Cu_3O_{7-y}$ and $PrBa_2Cu_3O_{7-y}$.

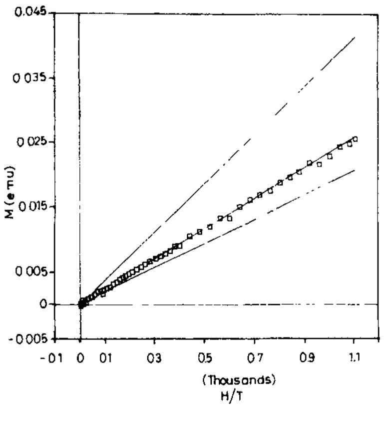

Figure 4. M vs H/T for $Pr_{0{\cdot}6}Y_{0{\cdot}4}Ba_2Cu_3O_{7-y}$ at 50 K along with free ion values for Pr^{3+}, Pr^{4+} and combination of Pr^{3+} and Pr^{4+}. The ratio is 1:3.

the ratio 1:3. At 5 K however there is a large decrease in magnetization which probably is due to the antiferromagnetic ordering of Pr moments.

Hysteresis plots were obtained for the compound with $x = 0{\cdot}5$ at various temperatures between 5 K and 100 K for fields up to 1100 G. These are shown in figures 5a and 5b. The diamagnetic moments obtained even above T_c are consistent with the presence of an impurity phase of $YBa_2Cu_3O_{7-y}$ with a concentration < 5%. We thus see that this method is capable of detecting with high sensitivity the presence of a second superconducting impurity phase with T_c greater than that of the dominant phase.

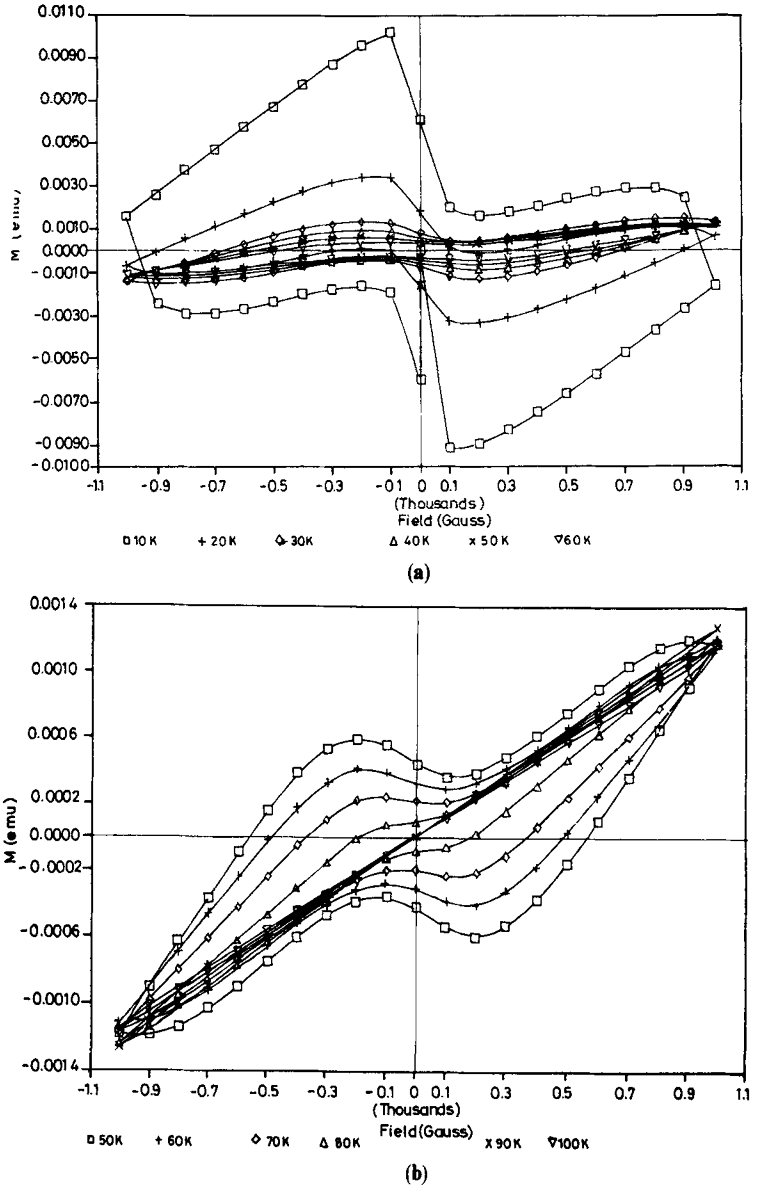

Figure 5. Hysteresis plots for $Pr_{0.5}Y_{0.5}Ba_2Cu_3O_{7-y}$ for fields up to 1100 Gauss. **a.** 10 K to 60 K. **b.** 50 K to 100 K.

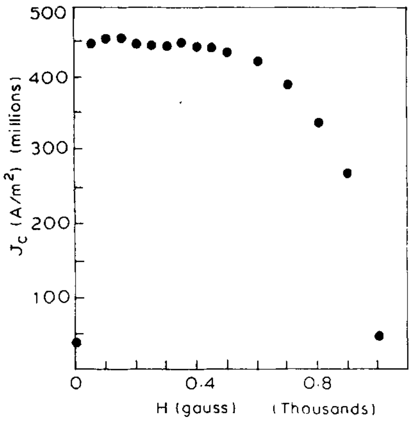

Figure 6. Critical current density vs applied magnetic field for $Pr_{0.5}Y_{0.5}Ba_2Cu_3O_{7-y}$ at $T = 5$ K.

The intragrain critical current density J_c may be calculated for the magnetization data using Bean's formula: $J_c = 15\Delta M/R$, where R is the grain size and M the difference between the magnetization values for a given positive and negative values of the applied field (Sekula *et al* 1989). The variation of J_c with field for $x = 0{\cdot}5$ and $T = 5$ K is shown in figure 6. However, the volume fraction of the superconducting phase as measured from a.c. magnetic susceptibility (diamagnetic shielding) measurements using a reference sample of lead is only 16%. When this is taken into account J_c at 5 K turns out to be 3×10^5 A/cm² which is more than one order of magnitude lower than the value for $YBa_2Cu_3O_{7-y}$. J_c begins to fall appreciably at 500 G. More detailed measurements of J_c as a function of x, T and H are now in progress. A more detailed account of the magnetization and specific heat results will be presented at a later stage.

Acknowledgements

The authors thank Prof G.V. Subba Rao of the Materials Science Research Centre for providing the samples and the Programme Management Board for superconductivity of the Department of Science and Technology for financial support.

References

Dalichaouch Y, Torikachvili M S, Early E A, Lee B W, Seaman C L, Yang K N, Zhou H and Maple M B 1988 *Solid State Commun.* **65** 1001

Goncalves A P, Santos I C, Lopes E B, Henriques R T, Almeida M and Figueiredo M O 1988 *Phys. Rev.* **B37** 7476

Kang J S *et al* 1989 *J. Less Common Metals* **148** 121

Kebede A *et al* 1988 *J. Magn. Magn. Mater.* **76, 77** 619

Li W H, Lynn J W, Skanthakumar S, Clinton T W, Kebede A, Jee C S, Crow J E and Mihalisin T 1989 *Phys. Rev.* **B40** 5300

Natarajan S, Chandrasekharan K, Thomas K A, Varadaraju U V, Subba Rao G V, Raju N P, Sankaranarayanan V and Srinivasan R 1988 *Physica* **C153–155** 926

Sankar N, Vaidhyanathan L S and Rangarajan G 1989 *Proc. Int. Conf. on Modern aspects of superconductivity, Paris, France* (ed.) R Suryanarayanan, p 189

Sekula S T, Brynestad J, Christen D K, Thompson J R and Kim Y C 1989 *IEEE Trans. Magn.* **25** 2266