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1997 J. Phys.: Condens. Matter 9 L525

(http://iopscience.iop.org/0953-8984/9/38/001)

J. Phys.: Condens. Matter **9** (1997) L525–L531. Printed in the UK PII: S0953-8984(97)85621-4

LETTER TO THE EDITOR

The paramagnetic Meissner effect in $Nd_{2-x}Ce_xCuO_y$ superconductors at 40 G and flux trapping

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Received 17 July 1997

Abstract. The paramagnetic Meissner effect (PME) has been observed in $Nd_{2-x}Ce_xCuO_y$ superconductors at 40 G. The origin of the effect has been considered on the basis of the local oxygen variation which implies non-superconducting or lower-T_c regions embedded inside the higher-T_c layers of superconducting (SC) grains or portions. These shell-like varying-T_c SC layers in turn apparently create 'cumulatively compressed flux trapping' and hence the PME. The exhibition of PME seemingly depends on the type of defect as well as their density.

The Meissner effect is a fundamental property of superconductors. Type I superconductors in an ideal sense exclude applied magnetic field completely if the field is below the critical field, H_c. Type II superconductors exhibit this effect below a lower critical field, H_{c1}. They, in particular high-temperature superconductors (HTSCs), mostly exclude the field partially and are also field dependent [1]. The Meissner fraction is therefore conveniently defined as the diamagnetic (dc) susceptibility, χ, multiplied by -4π. The exhibition of a partial Meissner effect was earlier ascribed to low-quality samples [2], and later on attributed to flux pinning [3]. There are however substantial reports on the observation of complete diamagnetism at low magnetic fields [4], but these experimental conditions are not applicable to all superconductors as the paramagnetic Meissner effect (PME) has been observed at fields below 1 G in some cases [5–7].

Braunisch *et al* [6] found that the PME was saturated at a certain field and that it was temperature independent for $T \ll T_c$. They attempted to explain it in terms of a certain defect structure that favours formation of spontaneous orbital currents in the ground state. Similarly, Kusmartsev [8] assumes π junctions associated with the SC loops that in turn have localized magnetic impurities. The susceptibility of the state associated with the loops may be paramagnetic even if the sample is in the SC state. However, the situation favours for certain external magnetic field, the limiting field being 2 G [8]. Of particular interest to note is that these were found in less homogeneous samples [6]. The inhomogeneity may

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¶ This form of name will henceforth be used instead of Okram G Singh; corresponding author. E-mail address: okram@nrim.go.jp comprise of stacking faults, dislocations, vacancies, grain and twin boundaries, precipitate (insulator, semiconductor, normal metal) or superconductors with lower T_c. Magnetic field sees these defects as low energy regions, thereby vortices are formed and the flux is pinned [4, 9]. Sigrist and Rice [10] argued that such a pinned flux may not be sufficient to exhibit PME and perhaps requires the superconductor to attract magnetic field. They consistently showed that the effect arose due to the presence of frustrated SC state based on d_{x2-y2} wave. However, this argument does not seem to be consistent with the observation of PME in s-wave superconductors as well (see e.g. [11, 12]). Therefore, there seem to be some other physical reasons to exhibit this property.

In addition to the inhomogeneity mentioned above, that caused by oxygen distribution can be a main cause for concern in oxide superconductors [13, 14]. Okram *et al* [13] suggested that this feature may have a bearing on the unusual properties such as low Meissner fraction, coexistence of anti-ferromagnetism and superconductivity, and anisotropic normal-state properties observed in electron-doped $R_{2-x}M_xCuO_y$ (R = Pr, Nd, Sm, Eu; M = Ce, Th) superconductors. In this scenario, if PME were observed in electron-doped superconductors, it would be very intriguing. In our effort to investigate these material properties, we conducted the zero-field- and field-cooled susceptibility (ZFC χ and FC χ) measurements on various compositions of the T′ type $Nd_{2-x}Ce_xCuO_y$ (NCCO) superconductors. The Meissner fraction was observed to decrease appreciably with decreasing field in the optimum composition ($x = 0.150$) sample and PME was observed in $x = 0.155$ and 0.165 compositions. In this letter, we discuss these features in the light of the local oxygen variation found in these superconductors [13] that perhaps may cause cumulatively compressed flux or equivalently magnetic field attraction and hence the PME.

To test this, samples of nominal compositions $x = 0.145$, 0.150, 0.155 and 0.165 were synthesized by a standard solid state reaction method. The details of the preparation procedure are given elsewhere [15]. The zero-field-cooled (ZFC) and field-cooled (FC) magnetization measurements were made on approximately tapered parallelepiped pellets using a vibrating sample magnetometer (VSM, Oxford Instruments, model VSM 3001) in the temperature range 1.8–30 K. The tapered pellet effectively reduces the demagnetization factor and possible PME due to random shape [11, 12, 16]. Further, this reduces the probability of field inhomogeneity around the sample, which may contribute to PME [17]. Also, the region of field homogeneity in the present VSM is 10 mm, the sample vibration frequency is 66.66 Hz and the amplitude is 1.5 mm. The temperature stability in the present measurements was 0.1 K. Before starting the magnetization versus field measurements at low temperatures, the optimum sample position was obtained by measuring the magnetization as a function of vertical position at 300 K. The sample, size (length) less than 2 to 3 mm, was placed at the centre. The remnant field of the superconducting magnet was about 5 G. A magnetic field of 40 G was applied parallel to the sample surface. For the $x = 0.150$ sample, the measurements were made for 10, 20, 30 and 50 G as well. By cooling the sample in nominal zero field from 30 K down to 1.8 K, the desired field was applied and the data were recorded while the sample was slowly heating up to 30 K at a temperature sweep rate of 0.5 K min^{-1} for ZFC measurements. The FC measurement was made while the sample was cooling down to 1.8 K from 30 K at the rate of 0.5 K min^{-1} in the desired applied field. The minimum possible remnant field was ensured by subjecting the magnet to decreasing cyclic magnetic field before each experiment The susceptibility (χ), multiplied by -4π, was calculated using 7.0 g cm^{-3} density for all the samples without demagnetization factor correction.

The samples were characterized using powder x-ray diffraction. The $x = 0.150$ sample was identified to be single T′ phase while the $x = 0.145$, 0.155 and 0.165 samples were of poorer quality; traces of Nd_2O_3 were detected. The FC and ZFC χ data as function of temperature at 40 G for different sample compositions are plotted in figure 1. The T_c onsets of the $x = 0.145$ and 0.150 samples are nearly the same ($\sim$21.0 K). The SC transition for the $x = 0.150$ sample is sharper and the susceptibility value saturates at lower temperature. These features are not visible in the $x = 0.145$ sample, indicating its inferior quality. It is also clear from the figure that the T_c decreases with increase in x beyond these compositions (see the phase diagram in [15] for comparison). The figure further reveals that the FC χ displays negative values (i.e. PME) for the $x = 0.155$ and 0.165 samples. The observed PME may be compared with that reported in other HTSCs wherein the applied field is below 1 G [5–7]. This perhaps shows the contrasting feature of the present observation which is at 40 G, not below 1 G.

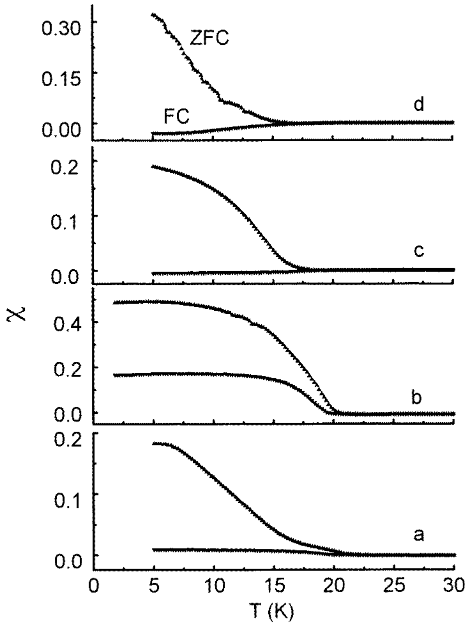

Figure 1. χ (ZFC and FC) against T of $Nd_{2-x}Ce_xCuO_y$ superconductors for (a) $x = 0.145$, (b) 0.150, (c) 0.155 and (d) 0.165 at 40 G.

The poor-quality samples have very low Meissner fraction or PME (table 1). The $\Delta\chi$, (FC $\chi -$ZFC χ), value at 5 K of the $x = 0.165$ sample is about twice that in either the $x = 0.145$ or 0.155 sample, but comparable to that of the $x = 0.150$ sample. The $\Delta\chi$ are found to correspond with the remnant field [4, 18], which may be compared with the internal field of the sample [7]. These supplement perhaps the coincidence of the FC χ and ZFC χ only at T_c. The coincidence in turn reveals the equality of the irreversible temperature, T_{irr}, and T_c, and implies large flux pinning.

Further, the FC and ZFC χ data as a function of temperature of the $x = 0.150$ sample at 10, 20, 30, 40 and 50 G are plotted in figure 2. It is apparent from the figure that there is a noticeable change in the transition region feature with field. As the field decreases, its characteristics becomes more and more steplike. This may indicate that at lower fields, the SC grains respond to the field in a more pronounced manner. Apparently, the T_c onset is somewhat higher in lower field (10 G) than in the higher field (50 G). The FC χ and ZFC χ

Table 1. χ (ZFC and FC), $\Delta\chi$ and T_c onset data of $Nd_{2-x}Ce_xCuO_y$ superconductor for different compositions at various magnetic fields.

					T_c onset (K)	
x	Field (G)	FC χ at 5 K	ZFC χ at 5 K	$\Delta\chi$ at 5 K	ZFC χ	FC χ
0.150	10	0.011	0.349	−0.338	19.0	21.0
0.150	20	0.151	0.465	−0.314	19.5	20.5
0.150	30	0.180	0.505	−0.325	19.5	20.5
0.150	40	0.171	0.490	−0.319	19.5	20.5
0.150	50	0.165	0.505	−0.340	19.5	20.5
0.145	40	0.009	0.182	−0.173	19.5	21.0
0.155	40	−0.006	0.189	−0.195	—	18.0
0.165	40	−0.030	0.321	−0.351	—	16.0

values do not change much between 50 and 30 G, while these values decrease appreciably at 10 G (see table 1). This is in contrast to the commonly observed characteristic in HTSCs that exhibit increasing susceptibility with decreasing field [4, 19, 20]. The FC χ value at 5 K drops to less than 1% at 10 G (table 1); this is suggestive of almost complete flux trapping in the sample. However, the $\Delta\chi$ values at this temperature remain almost the same at different fields. Moreover, the FC χ and ZFC χ values meet only at T_c similar to those of the above samples wherein the same field (40 G) was applied. Further, the T_c onset noted from the FC χ data is in general lower than that recorded from the ZFC χ data. While the ZFC χ and FC χ values at 5 K remain more or less the same with decreasing field down to 20 G, the latter suddenly drop for 10 G. This characteristic to our knowledge has not been observed in any of the HTSC materials [4]. These results reaffirm the contention that there is large flux pinning in this type of superconductor.

It may be noted that the present PME observations are not due to experimental artifacts. Even though our predetermined remnant magnetic field is ±5 G, consider a negative remnant magnetic field of the VSM above the highest nominal applied field (50 G) while the runs were made. Then, all the ZFC χ data observed would have been essentially FC χ data with negative sign. However, this is not true in all the ZFC χ data shown in figures 1 and 2. Therefore, there is no remnant magnetic field, greater than the nominal applied field, in the VSM while the present data were collected. Thus, the observed decrease in Meissner fraction at 10 G is notable (figure 2). Further, $\Delta\chi$ remains almost unchanged (figure 2 and table 1). This may be due to the frozen-in flux with the same internal field for any applied field. The same situation may hold true in other compositions and freezing or trapping is perhaps complete in the samples that exhibit PME.

The frozen-in flux is directed along the applied magnetic field and has been reported for many other superconductors [7, 18, 19]. Nevertheless, the present PME observations at 40 G are in contrast to those observed below 1 G earlier in other HTSCs [5–8]. Impurities in an SC sample are expected to be paramagnetic below and above the T_c. The local oxygen variation is expected to have a major contribution for a real material which can in turn cause variation of T_c [13]. A similar situation can exist around an impurity as well. This may be compared with the observation of PME in less homogeneous Bi superconductors [6].

Let us consider that the PME is due to the trapped flux created by local oxygen variation in the bulk of the sample in particular [13]. This assumption may lead to generalization of the understanding of PME as this has been understood to require defects to be present in the superconductor [6, 8]. In addition to the non-SC impurities present in the samples, how

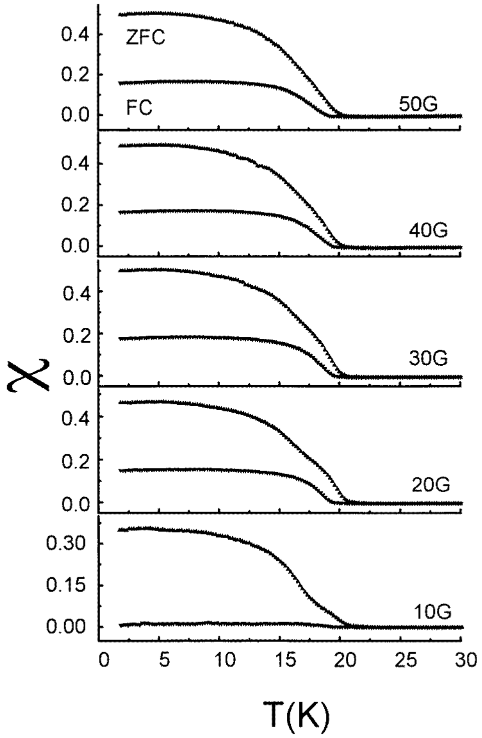

Figure 2. χ (ZFC and FC) against T of $Nd_{1.85}Ce_{0.15}CuO_y$ superconductor at different magnetic fields.

the oxygen distribution varies in general implies a gradual fall of T_c towards the centre of the grain [13]. This in turn suggests the maximum T_c near the outermost surface and the minimum value towards the centre. In other words, at the same $T < T_c$, the Meissner effect (or the supercurrent-produced field) will be stronger on the outer layer, and strength can fall monotonically in the inner layers towards the centre. Then, the applied magnetic field will be trapped inside and get force compressed successively inside, instead of expelling all of a sudden when the sample cools down below its T_c. The resultant magnetic moment due to the supercurrents flowing in the outer region and paramagnetic pinning currents flowing in the inner region turns out to be paramagnetic. The PME, normalized to the magnetization in the Meissner state at the same field, M_M, can be written as [21].

$$M/M_M = 1.44[-1 + f + ((1 - b/w)/2)(\ln(1/(1 - b/w)) - 0.31)]$$

where f is trapped flux, $2w$ is the width of the thin SC strip of thickness d and $2b$ is the width surrounded by flux free regions of width w–b.

This equation holds true under the assumption that $w - b \ll w$ as may be expected for the experimental condition in general. Further, it tells us that the moment becomes more diamagnetic with decreasing f and becomes more paramagnetic with increasing flux compression or decreasing value of b/w. For $f = 1$, the complete flux trapping, M is always paramagnetic.

It may be recalled here that, as in the present case and reported earlier [6], not all the samples with the same nominal compositions exhibit PME. This may suggest that there is a critical 'cumulatively compressed flux trapping' favourable to exhibiting PME. We, therefore, believe that present explanation based on the local oxygen variation is equally applicable in other HTSCs wherein oxygen variation is always expected in real samples. It is clear from the above argument that for the samples with PME or at the threshold of exhibiting PME, the Meissner effect is expected to be partial as was observed in NCCO samples in particular and HTSCs in general [1–3, 15]. Further, the PME observed at 40 G seemingly correlates with the presence of more prominent local oxygen variation compared to other HTSCs because of the reduction step required. This may be considered as macroscopic compared to the defects generally present in the HTSC materials: in the HTSCs where the PME is observed at fields below 1 G, the defects expected to be present are nanoscopic. Therefore, the field required for exhibiting PME is perhaps related to the defect size or type, not only the density. Furthermore, the present model is irrespective of whether the material is d or s wave [10]. This model may be more realistic, because the T_c variation due to oxygen is intrinsic. The possible PME exhibition due to sample shape, orientation and field uniformity, observed quite recently, may be considered as additional factors to take into account [11, 12, 16, 17].

In conclusion, we have studied the FC and ZFC magnetization of the NCCO materials. The Meissner fraction was observed to drop suddenly at 10 G for the $x = 0.150$ sample. This is in contrast to that observed in other HTSCs. Moreover, PME was observed at 40 G in the $x = 0.155$ and $x = 0.165$ compositions. An attempt has been made to explain the observed PME on the basis of the presence of inhomogeneous SC or non-SC regions in the bulk of the sample.

We (GSO, BDP, OP) thank P Chaddah, CAT, Indore, and A K Grover, TIFR, Mumbai, for valuable discussions, and DST, New Delhi, for financial support.

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