Interplay of Paramagnetic Signal with Superconductive Environment in a (Nd,Eu,Gd)BaCuO Single Crystal

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Abstract. In LRE-Ba2Cu3Oy (LRE-123, LRE=Nd, Eu, Sm, Gd) superconductors paramagnetic signal of LRE atoms complicates evaluation of weak superconducting signals, like of the equilibrium reversible magnetization. While above \( T_c \) the superconductor follows Curie-Weiss (C-W) law, below \( T_c \) the measured reversible response above irreversibility field is a mixture of reversible magnetization and a paramagnetic background. A way to correct for the latter is an extrapolation of a pure paramagnetic response from temperatures well above \( T_c \). We measured magnetic behavior of a weakly doped \((\text{Nd}_{0.33}\text{Eu}_{0.38}\text{Gd}_{0.28})\text{Ba}_2\text{Cu}_3\text{O}_y\) single crystal from 300 K to 80 K. The reversible magnetization was successfully extracted, which enabled evaluation of the associated thermodynamic quantities for the given compound.

1. Introduction
The family of ternary light rare earth (LRE) LRE-Ba2Cu3Oy superconductors features due an exceptionally high technical characteristics like critical current density [1], irreversibility field [2] and operating temperature [1,3]. The excellent properties originate from an exceptional flexibility of these materials with respect to creation of various types of effective pinning structures. First of all, point-like defects due to LRE/Ba substitution, but also oxygen vacancies [4] play important role, especially at moderate magnetic fields. In melt-textured materials at low magnetic fields micron and submicron particles are effectively utilized. By reducing size of such intentionally added particles (Gd-211, Zr-, Ti-, Mo- or Nb- oxides) to a few tens of nanometers, vortex pinning at low fields can be dramatically enhanced. With increasing temperature the range of such an effect extends to intermediate fields, where these defects cooperate with point-like ones. Altogether the associated critical current density nearly approaches levels met in thin films [1]. The exceptionally effective flux pinning enhanced critical current densities up to a close vicinity of the critical temperature and enabled levitation at 90.2 K, i.e. in liquid oxygen [1,3]. Ternary (Nd,Eu,Gd)-123 compounds with excess of Eu develop a specific frustrated nanoscopic substructure correlated with ordinary twin planes [2]. They appear in the form of zig-zag structures or planar nanoscopic lamellas. These structures, filling the channels between regular twin boundaries, lead to an exceptional enhancement of vortex pinning at high magnetic fields, resulting in increase of irreversibility field at 77 K by factor two [2].

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Any of the above achievements in vortex pinning efficiency has strengthened the efforts to go a step further in this direction. However, the higher was the critical current density and the associated irreversible magnetic moment, the more difficult was to identify the rather weak equilibrium reversible magnetization associated with the thermodynamic characteristics of the compound. In the present work we looked in detail at the mutual interaction of superconductivity and paramagnetism in a weakly doped (Nd$_{0.33}$Eu$_{0.38}$Gd$_{0.28}$)Ba$_2$Cu$_3$O$_y$ single crystal. We succeeded in separating the individual contributions and eventually evaluated the thermodynamic characteristics of the material.

2. Experimental details
The (Nd$_{0.33}$Eu$_{0.38}$Gd$_{0.28}$)Ba$_2$Cu$_3$O$_y$ single crystal was grown by self-melting in air of a mixture of high purity commercial powders of Nd$_2$O$_3$, Eu$_2$O$_3$, Gd$_2$O$_3$, and BaCO$_3$. The crystal was oxygenated in pure oxygen at 410 °C to reach the optimum content of oxygen, y=6.96. The dimensions were 1.35×1.42×0.53 mm$^3$, the volume and weight were 1.016 mm$^3$ and 5.9 mg, respectively.

The total magnetic moment $m$ was measured as a function of magnetic field and temperature by a MPMS SQUID with a 7 Tesla magnet. In the latter experiment both zero field cooled (ZFC) and field cooled (FC) modes were detected in the range 300 to 5 K. The full magnetization loops, $m(H)$, were measured between -1 T and 7 T in the temperature range 300 K to 80 K, with the field applied along c-axis.

3. Experimental results
The magnetization curves measured at 14 temperatures between 300 K and 80 K are presented in figure 1 (a). Above $T_c$, at 300 K, 200 K, 150 K and 100 K, only the reversible linear paramagnetic background was detected. Thanks to perfect oxygenation and good quality the crystal exhibited only a weak pinning. Therefore, the equilibrium reversible magnetic moment was dominant in the whole investigated temperature range below $T_c$ (93.8 K, 80 K). Below $T_c$ the superconducting signal mixes with the paramagnetic one in the whole field range below $H_{c2}$. The problem is that $H_{c2}$ is not a-priori known. Evolution of the magnetization loop was traced through temperatures of 93, 92, 91, 90, 89, 88, 86, 84, 82 and 80 K. Below the irreversibility field, $H_{irr}$, the mean value of magnetic moments on the ascending and descending branch of the MHL was calculated. This mean moment smoothly joined

![Figure 1](https://example.com/figure1.png)

**Figure 1.** (a) Magnetic hysteresis loops of the NEG-123 single crystal measured at 14 temperatures between 300 K and 80 K. (b) Magnetization as a function of temperature was measured between 300 and 5 K, first in zero field cooled (ZFC) mode, switching field on at 5 K and warming the sample up to 300 K. Then the sample was field-cooled back to 5 K. The dashed line indicates the best fit to Curie-Weiss law. The experiment starts to deviate from the C-W law at $T_c$. Magnetic field ranged from 1 to 7 Tesla. Panel (b) shows the case $\mu_0 H=5$ Tesla.
the mixed reversible moment at $H_{\text{irr}}$. The thermodynamic equilibrium reversible magnetization was obtained by subtracting the paramagnetic background from the measured $M(H)$ curve. A linear dependence with the slope established from the Curie-Weiss law, extrapolated from the temperatures above $T_c$ (see figure 1 (b)), was subtracted. Such a correction worked, however, only close below $T_c$, at low temperatures it failed.

The reversible magnetization obtained after the paramagnetic correction (figure 2) was analyzed in terms of the expression [5],

$$M(H) = \frac{a\Phi_0}{8\pi\mu_0^2} \ln \frac{H}{\beta H_{c2}} \approx \frac{aH_{c1}}{2\ln \kappa} \ln \frac{H}{\beta H_{c2}}$$

with field independent parameters $a = 0.77$ and $\beta = 1.44$ (valid in the range $0.02 < H/H_{c2} < 0.3$), the Ginsburg-Landau parameter $\kappa = \lambda/\xi$, where $\xi$ is the coherence length, $\lambda$ is the penetration depth, $\Phi_0$ is the flux quantum, and $\mu_0$ is the permeability of vacuum. The data were fitted by the analytical form $M(H)=k_1\ln(\mu_0 H/k_2)$ with the fitting parameters $k_1$, $k_2$. The best fit to each of the curves is in figure 2 denoted by a full line. $M$ and $k_1$ are in units of $\text{A/m}$, $k_2$ is in units of Tesla. $k_2$ gives a direct information on the upper critical field, $H_{c2}=H_{c2}/(\mu_0\beta)$. Taking into account $H_{c2}/H_{c1}=2\kappa^2/\ln \kappa$ and $H_{c1}=(2k_1 \ln \kappa)/a$, we have $\kappa = \frac{ak_1^2}{4\beta\mu_0 k_2}$. From the definition of $H_{c2} = (\frac{\Phi_0}{2\pi\mu_0^2 \xi^2})$, one receives $\xi = \sqrt{\frac{\beta \Phi_0}{2\pi k_2}}$ and then $\lambda = \xi \kappa$. At temperatures close to $T_c$ (above 90 K) equation (1) looses precision in higher parts of the curves. There, the linear dependence $M(H) \approx 0.58(H-H_{c2})/\kappa^2$ suggested by Abrikosov [6] should be considered. The results of the above evaluation are summarized in figure 3. Critical fields $H_{c1}$ and $H_{c2}$ are shown as a function of temperature in figure 3 (a), together with the irreversibility field, $H_{\text{irr}}$. In figure 3 (b) the temperature dependencies $\kappa(T)$, $\xi(T)$ and $\lambda(T)$ are presented. Assuming the temperature dependence of $\xi$ in the form $\xi(T) = \xi_0 \sqrt{1-(T/T_c)}^\gamma$, we get nearly temperature independent $\xi_0$ (except the points above 90 K – see figure 3 (b)) with the average value $1.897 \pm 0.018$ nm. In the same way we obtained $\lambda(0) \approx 120$ nm. Both $\xi_0$ and $\lambda(0)$ values agree surprisingly well with the results previously obtained by specific heat measurements on the similar material [7] and those commonly observed in YBCO [8,9]. The deviation of the $H_{c2}(T)$ from the linear dependence close to $T_c$ (figure 3 (a)) indicates that the present procedure looses precision close $T_c$, which we attribute partly to thermal fluctuations, partly to the loss of validity of equation (1) in the major part of the field range.
4. Discussion and conclusions

A strong irreversible magnetization and its planar anisotropy due to channelling effect of the twinning structure were largely suppressed in the present experiment. The only major obstacle hindering from a rigorous evaluation of the equilibrium reversible magnetization in the ternary LRE-123 superconductor was the significant paramagnetic signal from the LRE ions. Paramagnetic background was corrected using the Curie-Weiss dependence of the magnetic susceptibility evaluated at temperatures above $T_c$ ($= 93.8$ K). Such a correction failed at too low temperatures but was correct enough at vicinity of $T_c$ (up to 80 K). The rather narrow hysteresis curves produced a mean magnetization that was evidently close to the equilibrium reversible magnetization. At the irreversibility field it smoothly joined the reversible curve measured above $H_{irr}$. The analysis of the data after the paramagnetic correction produced surprisingly smooth results. This was particularly evident when trying to compare the $B_{c2}(T)$ data with those obtained from the joining point of the FC and ZFC curves. The latter data were so scattered that did not allow for a reasonable comparison. The resulting $\xi_0$ (1.897±0.018 nm) data are in very good coincidence with the values obtained in other 123 compounds and with the result obtained by means of specific heat measurements on the melt-textured NEG-123 material [7]. Interestingly, the present anomalies of $\kappa$, $\xi$ and $\xi_0$ in vicinity of $T_c$ (figure 3) well reproduce the data behaviour observed in [7].

5. References

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