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Evidence for two-dimensional superconducting fluctuations in $CaLaBaCu_3O_{7-\delta}$

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Abstract

We report measurements of the magnetization and diamagnetic susceptibility of a polycrystal of $LaBaCaCu_3O_{7-\delta}$. The sample has a zero-field transition temperature $T_{c0} = 77$ K and a transition width of 2.0 K. The results show large fluctuation effects, which can be explained by Ginzburg–Landau fluctuation theory for a two-dimensional (2D) system. The experimental data for the magnetization in high magnetic fields (20–50 kOe), were fitted by using a theoretical model based on the lowest-Landau-levels approximation, showing good agreement. The weak-field fluctuation diamagnetic susceptibility above T_c can be fitted well in terms of the 2D Lawrence–Doniach model.

1. Introduction

In the last few years, the phase transition in the vortex lattice in high-temperature superconductors (HTSCs) has been widely studied not only because of its fundamental interest, but also due to the implications for potential applications. In addition to high superconducting transition temperatures, these materials present other important characteristics such as their short superconducting coherence lengths and strongly anisotropic superconducting, as well as normal-state, properties. These characteristics lead to a behaviour in the vicinity of $H_{c2}(T)$ strongly influenced by the thermodynamic fluctuations. The effects of fluctuations in the HTSCs extend over a wide range of temperatures around $T_c(H)$, the mean-field transition temperature, bringing about changes in the phase diagram of these materials, and new phase boundary lines appear. Many experimental [1–5] as well as theoretical [6–9] results have been reported in relation to the critical behaviour arising as a consequence of the thermal fluctuations in HTSCs.

In an experiment performed by Welp *et al* [1], high-precision measurements of the magnetization and resistivity of orthorhombic $YBa_2Cu_3O_{7-\delta}$ crystals were carried out, near the

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superconducting transition, in magnetic fields applied perpendicular to the CuO_2 layers. These data showed a scaling behaviour in the variable $[T - T_c(H)]/(TH)^{2/3}$ which is consistent with the Ginzburg–Landau (GL) fluctuation theory for a three-dimensional (3D) system in a high magnetic field. However, a two-dimensional (2D) version of the scaling function in the variable $[T - T_c(H)]/(TH)^{1/2}$ was demonstrated [2] for the tetragonal $YBa_2Cu_3O_{6.6}$. The system $CaLaBaCu_3O_{7-\delta}$ (CLBCO) is one of the HTSCs exhibiting a superconducting transition temperature around 78 K. The structure of $CaLaBaCu_3O_{7-\delta}$ is analogous to that of the tetragonal $YBa_2Cu_3O_{7-\delta}$ compounds. More recently a similar scaling behaviour has been observed for various grain-aligned $HgBa_2Ca_2Cu_3O_y$ samples [4]. Schmidt *et al* [5] have studied the angular dependence of the first-order vortex-lattice phase transition in $Bi_2Sr_2CaCu_3O_8$, noting that the transition field H_m follows the 2D scaling function $H\cos\theta$, with θ being the tilt angle of the dc magnetic field with respect to the c-axis. The scaling fails for field orientations close to the ab-plane.

Tesanovic *et al* [6–8] have developed a nonperturbative theory of critical behaviour for anisotropic superconductors for both 2D and 3D systems in the presence of strong magnetic fields applied in the direction parallel to the c-axis, describing the critical behaviour by means of an interacting particle system with long-ranged multiple forces (a dense vortex plasma). The superconducting transition corresponds to the liquid–solid transition in the dense vortex plasma (DVP). This theory uses a nonperturbative approach to the GL free-energy functional in which the order parameter is expanded in terms of the lowest Landau levels (LLL). The scale invariance of the DVP leads to a universal character of the thermodynamic quantities, such as magnetization and specific heat, for both 2D [7] and 3D [8] cases.

In this paper we study the scaling behaviour of the magnetization, in the critical region near $H_{c2}(T)$, of a polycrystalline sample of $CaLaBaCu_3O_{7-\delta}$ in the presence of high magnetic fields. In section 2 we present experimental details and in section 3 the experimental data are analysed and compared with Tesanovic's theoretical model [7].

2. Experimental details

CLBCO, which is one of the $YBa_2Cu_3O_{7-\delta}$-like (Y:123-like) like compounds, is a bulk superconductor with a T_c around 78 K [10–12]. Its structure is isomorphic to that of tetragonal Y:123, with the Y^{3+} sites being occupied by the $Ca^{2+}$and La^{3+} ions and the Ba^{2+} sites occupied by the Ba^{2+}, La^{3+} and Ca^{2+} ions [13, 14]. Compared with orthorhombic $YBa_2Cu_3O_{7-\delta}$ superconductor, CLBCO presents a simpler structure and, in this material, there are no twins [15] and few oxygen vacancies [16] and spiral dislocations [17].

Samples with the nominal composition $CaLaBaCu_3O_7$ were prepared by the solid-state reaction process. The high-purity chemical constituents (purity 99.99%) La_2O_3, $BaCO_3$, $CaCO_3$ and CuO were mixed in stoichiometric ratio. The mixed powder was finely powdered and calcined at 950 °C for 12 h. The calcined material was cooled down to room temperature by air quenching and the sample was again ground. The calcination process was repeated three times. After the calcination, the material was then finely powdered and pressed into circular discs at a pressure of 5 ton cm^{-2}. These pellets were sintered at 975 °C in flowing oxygen for 24 h, then slowly cooled to 575 °C and annealed at this temperature for 24 h. Finally, the samples were slowly cooled down to room temperature over a span of 10 h.

X-ray diffraction (XRD) was used to determine the phase purity and lattice parameters of the sample. The x-ray spectra were recorded using a Siemens D-500 diffractometer with Cu Kα radiation ($\lambda = 1.5406$ Å). The XRD patterns were indexed assuming a primitive tetragonal $P4/mmm$ space group and the lattice parameters were evaluated by least-squares fitting of the data. The XRD of the sample was characteristic of a single-phase tetragonal RE:123 structure.

The temperature dependence of the magnetization was measured by using a superconducting quantum interference device (SQUID) magnetometer (MPMS, Quantum Design). The data were taken by measuring the magnetization versus temperature at various fixed magnetic fields in the temperature range from the irreversible temperature to 300 K. Scans of 3 cm were used. The normal-state magnetization (100–300 K) was fitted to a Curie law, $M = (\chi_0 + C/T)H$, and was carefully subtracted.

Meissner effect and screening ac susceptibility measurements were carried out in an ac field amplitude of 0.1 Oe and at 31 Hz, for temperatures between 100 K and 5 K.

3. Results and discussion

In figure 1 were present magnetization data $M(T, H)$ for a $CaLaBaCu_3O_{7-\delta}$ superconducting polycrystalline sample in the critical region in the presence of high magnetic fields. It is interesting to note the existence of a crossing point for various $M(T)$ curves for different fields, located at $T^* = 72.3$ K where the value of the magnetization is $M^* = -0.050$ G. This crossing point is characteristic of fluctuations and has been calculated theoretically by Tesanovic *et al* [7] and Bulaevskii *et al* [9].

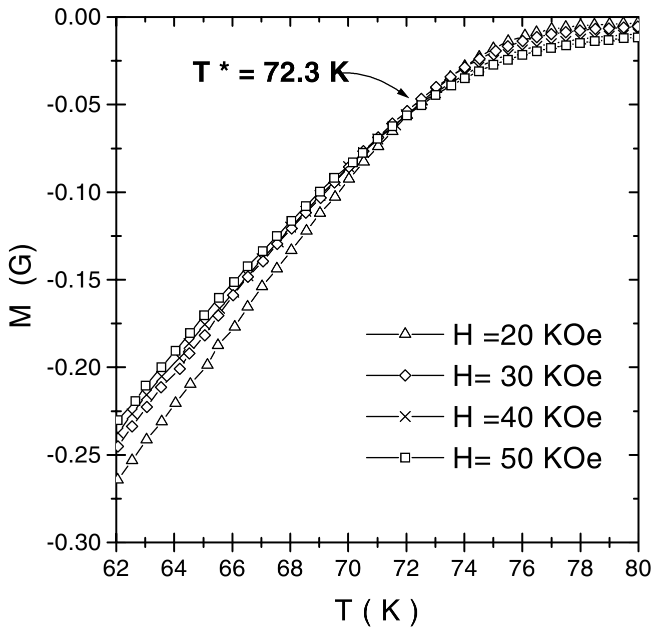

Figure 1. The temperature dependence of the reversible magnetization of polycrystalline $CaLaBaCu_3O_{7-\delta}$ in various magnetic fields.

In the case of a single crystal, the temperature T^* and magnetization $M_0^*(T^*)$ are related by the expression

$$M_0^*(T^*) = -\frac{k_B T^*}{\phi_0 s} \tag{3.1}$$

where ϕ_0 is the flux quantum $hc/2e$ and s is the effective spacing of the superconducting layers.

For high magnetic fields oriented in the c-direction, the scaling function for the magnetization $M_0(T, H)$ of a single crystal for 2D type-II superconductors in the critical region around $H_{c2}(T)$ can be expressed in the explicit form [7]

$$\frac{M_0(T, H)}{\sqrt{TH}} = \frac{k_B T^*}{2\phi_0 s(T_{c0} - T^*)} \left[t - \sqrt{t^2 - 4(T_{c0} - T^*)/(H'_{c2} T^*)} \right] \tag{3.2}$$

where T_{c0} is the zero-field transition temperature, H'_{c2} is the slope of the upper critical field $H_{c2}(T)$ at $T = T_{c0}$ and $t = [T - T_c(H)]/(TH)^{1/2}$. Equation (3.2) is derived from the GL fluctuation theory in the LLL approximation. It is supposed that higher Landau levels are taken into account by renormalization of the GL parameters, and these parameters differ from the original ones for the GL model for $H = 0$ due to the contribution of higher Landau levels. This contribution is small for $H \gtrsim \frac{1}{3}H^*$ [7], with H^* given by

$$H^* = H_{c2}(T^*) = H'_{c2}(T_{c0} - T^*). \tag{3.3}$$

For lower fields, higher Landau levels are important.

In terms of the crossing parameters, equation (3.2) can be rewritten in the form

$$\frac{M_0}{M_0^*} = \frac{1}{2}\left[1 - \tau - h + \sqrt{(1 - \tau - h)^2 + 4h}\right] \tag{3.4}$$

where $\tau = (T - T^*)/(T_{c0} - T^*)$ and $h = H/H^*$.

So far, these results assume that $\boldsymbol{H}$ is perpendicular to the layers, as in single crystals with $\boldsymbol{H} \parallel c$-axis. However, these results still apply to a polycrystal, provided that the material is sufficiently anisotropic that only the component of $\boldsymbol{H}$ perpendicular to the layers is effective [18]. The contribution of a single-crystal grain to the magnetization along $\boldsymbol{H}$ is $M_0(T, H\cos\theta)\cos\theta$, where θ is the angle between $\boldsymbol{H}$ and the c-axis. Then the magnetization $M_S(T, H)$ of the polycrystal is

$$M_S(T, H) = \langle M_0(T, H\cos\theta)\cos\theta\rangle \tag{3.5}$$

where $\langle\cdots\rangle$ denotes an angular average [18]. In the angular interval where the 2D scaling behaviour is valid [5], the scaling function for the magnetization of the polycrystalline sample can be expressed in the form

$$\frac{M_S(T, H)}{\sqrt{TH}} = \langle\cos\theta\rangle\,\frac{M_0(T, H)}{\sqrt{TH}}. \tag{3.6}$$

Therefore the value of the magnetization of the polycrystalline sample at the crossing point is $M_S^* = \langle\cos\theta\rangle\, M_0^*$.

From equation (3.1) it is possible to estimate the effective spacing of the superconducting layers; one obtains the value $\langle\cos\theta\rangle(96.45$ Å), in which $\langle\cos\theta\rangle$ can take values from 0.5, for a random polycrystal, to 1.0, for a grain-aligned sample [18]. On the other hand, the spacing between superconducting layers obtained from crystallographic analysis is 11.7 Å. As described by Kogan *et al* [19], we ascribe this difference to the presence of nonsuperconducting material in the sample. To correct for this volumetric effect, it is necessary to introduce an additional factor of (11.7 Å)/($\langle\cos\theta\rangle$96.45 Å) $= 0.12/\langle\cos\theta\rangle$. When both angular and volumetric factors are taken into account, the dependence on $\langle\cos\theta\rangle$ is cancelled, leading to an effective correction factor of 0.12.

A 2D scaling of magnetization data for different fields is shown in figure 2. The solid line corresponds to the theoretical curve of the scaling function for $M_S(T, H)$ obtained from equation (3.2) with the appropriate correction factor. The values of T^* and M_S^* were taken directly from the data for the crossing point. Two other parameters, T_{c0} and H'_{c2}, were obtained by the least-squares minimization method using equation (3.4) and the experimental data [7]. From this fit we obtain the values $T_{c0} = 74.7$ K and $H'_{c2} = 29.85$ kOe K^{-1}. The corresponding experimental values are 77 K and 28.96 kOe K^{-1}, respectively. The difference is due to the contribution of higher Landau levels mentioned above. In figure 3 we show the slope of $H_{c2}(T)$ near T_{c0}.

In order to check our results we also analyse the weak-field fluctuation diamagnetic susceptibility above T_c. In figure 4 we present a log–log plot of $-\Delta\chi/T$ versus $(T - T_{c0})/T_{c0}$

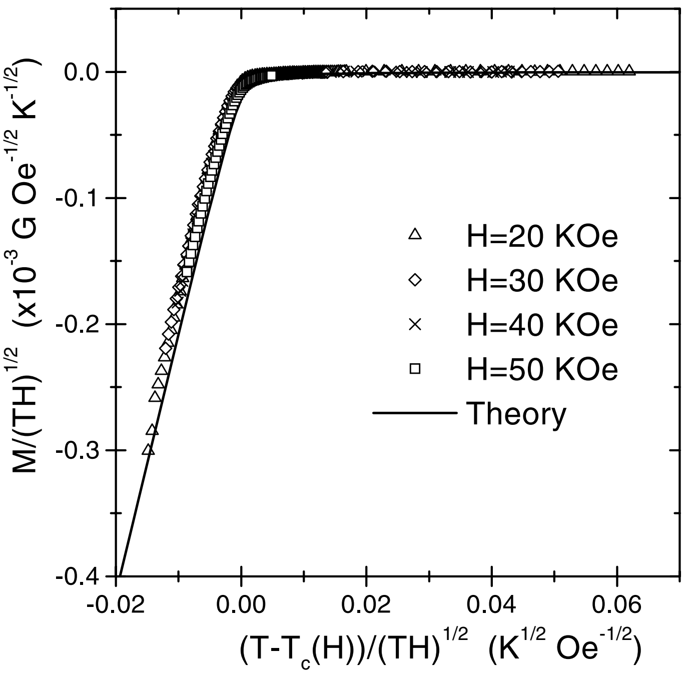

Figure 2. The 2D scaling of the magnetization data of polycrystalline $CaLaBaCu_3O_{7-\delta}$. The theoretical curve is obtained from the scaling function given by equation (3.2) with a suitable correction factor.

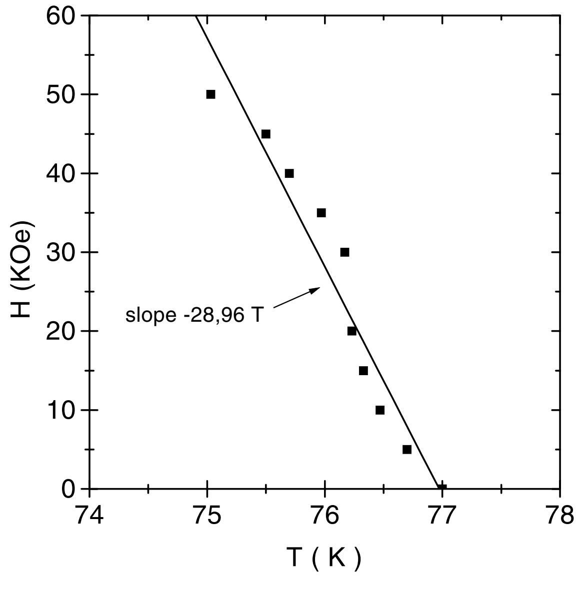

Figure 3. The temperature dependence of the upper critical field H_{c2} for polycrystalline $CaLaBaCu_3O_{7-\delta}$ near T_{c0}. We note a value of $H'_{c2} = 28.96$ kOe K^{-1} for the slope of $H_{c2}(T)$ at $T = T_{c0}$.

for the data taken at 4 kOe. The value obtained for the critical exponent is $n = -0.95$ which is in good agreement with the value $n = -1$ predicted by the Lawrence–Doniach model for a 2D system [3]. The normal-state magnetization agrees well with the Curie–Weiss law, $M = H(\chi_0 + C/T)$. The value obtained for the temperature-independent susceptibility χ_0 is

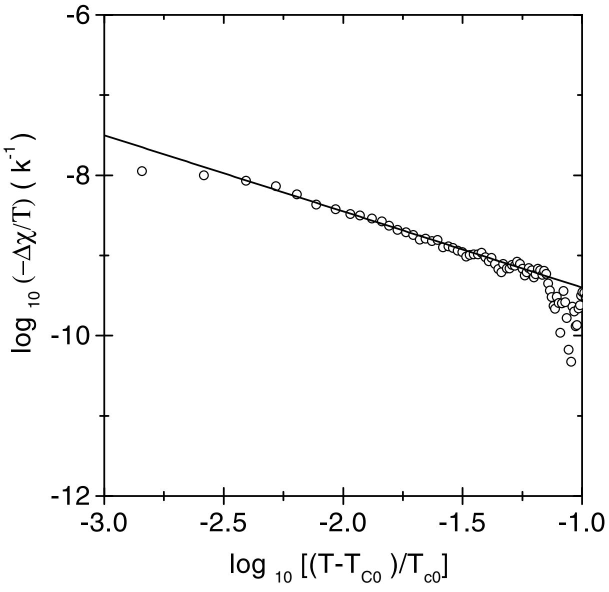

Figure 4. The log–log plot of $-\Delta\chi/T$ versus $(T - T_{c0})/T_{c0}$ for the data taken at 4 kOe for a polycrystalline sample of $CaLaBaCu_3O_{7-\delta}$.

4.0×10^{-5} emu mol^{-1}. The Curie constant estimated from the fit is 0.016 emu K mol^{-1}. The effective magnetic moment per copper atom calculated from the Curie constant is 0.183 μ_B.

In summary, we have presented reversible magnetization and diamagnetic susceptibility measurements for polycrystalline $CaLaBaCu_3O_{7-\delta}$ in the presence of high magnetic fields. The analysis of the thermal fluctuations shows an excellent 2D scaling behaviour.

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