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## LETTER TO THE EDITOR

## Magnetization scaling below $T_c$ in BiSrCaCuO and TlBaCaCuO superconducting ceramics

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Abstract. The reversible magnetization M(T, H) in the mixed state of the quasi-twodimensional superconductor Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> appears to factorize into a product f(T)g(H). It was recently argued that this implies a puzzling *temperature-independent* critical field  $H_{c2}$ , at least in the region where scaling is observed. We propose an alternative explanation in which this scaling property is approximate and results from the presence of vortex fluctuations, keeping a *conventional* temperature dependence for  $H_{c2}(T)$ . New data in the BiSrCaCuO (2212) and TIBaCaCuO (2201, 2212) systems that obey to a good approximation the factorization M(T, H) = f(T)g(H) are presented.

The critical fields of superconductors are normally expected to follow a roughly parabolic decrease with temperature. In the quasi-two-dimensional superconducting cuprates, the experimental temperature dependence of the upper critical field  $H_{c2}$  is not well established, owing to the difficulties in the determination of the transition point in non-zero fields, but nevertheless intriguing anomalies have recently been reported. For example, a resistive determination of the upper critical field in the Bi<sub>2</sub>Sr<sub>2</sub>CuO<sub>6</sub> (Bi-2201) phase shows a dramatic increase of  $H_{c2}(T)$  at low temperature [1]. In the Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> (Bi-2212) phase, Cho *et al* [2] showed scalings of the reversible magnetization M(T, H) = f(T)g(H). It was argued that the latter implies a *temperature-independent* critical field in the region where scaling takes place.

In this paper, we observe and confirm the scalings mentioned by Cho *et al*, not only in high-quality Bi-2212, but also in Tl<sub>2</sub>Ba<sub>2</sub>CuO<sub>6</sub> (Tl-2201) and Tl<sub>2</sub>Ba<sub>2</sub>CaCu<sub>2</sub>O<sub>8</sub> (Tl-2212) ceramics. The central point of this work, however, consists of showing that a *conventional* temperature dependence of the upper critical field may be recovered, if one takes into account fluctuations of vortices, a contribution that moreover can account for the crossing of magnetization curves in a single point { $T^*$ ;  $M^*$ } [3].

| Phase   | Sample code | T <sub>c</sub> <sup>onset</sup> (K) | f (%) | Space group | a (Å)     | b (Å)     | c (Å)     |
|---------|-------------|-------------------------------------|-------|-------------|-----------|-----------|-----------|
| Bi-2212 | N/-2/6      | 92,4                                | 48    | A2aa        | 5.410(1)  | 5.412(2)  | 30.88(1)  |
| Tl-2201 | 12EA0       | 92.1                                | 61    | I4/mmm      | a = b =   | 3.8700(3) | 23.226(2) |
| TI-2212 | T2-12       | 108.7                               | 45    | I4/mmm      | a = b = b | 3.8548(5) | 29.328(4) |

Table 1.  $T_c^{onset}$  is the temperature at which 0.1% of ideal Meissner susceptibility is found.



Figure 1. Scaling plot of the M(T, H) data versus T (the symbols +, \*,  $\blacksquare$ ,  $\bigcirc$ ,  $\triangle$ ,  $\times$ ,  $\diamondsuit$ ,  $\blacktriangle$  and  $\Box$  correspond to 1, 3, 5, 10, 20, 30, 40, 50 and 55 kOe, respectively). The choice of T' in the reversible regime (50 K, 70 K and 60 K for the Bi-2212, TI-2212 and TI-2201, respectively) is arbitrary.

Sample preparation and characterization are described in [4]. Note that the oxygen concentration of all phases is optimized to obtain the highest critical temperatures. Table 1 lists  $T_c$  values, crystallographic data and the fraction f of complete Meissner field expulsion at low temperature in an applied field  $H_a = 20$  Oe. The latter exceeds 45%.

In the present analysis, we use the CGS system where  $B[G] = H[Oe] + 4\pi M[G]$ and  $\chi_v = M[emu \text{ cm}^{-3}]/H = \rho[g \text{ cm}^{-3}]\chi_g[emu g^{-1}]$ . The reversible magnetization M(H, T) was measured with a SQUID magnetometer using a small scan length (3-4 cm) to minimize the variation of the magnetic field during the displacement of the sample in the detection coils. Inspection of zero-field-cooling (ZFC) measurements followed by field-cooling (FC) measurements, completed by hysteresis measurements M(H) at constant temperature, convinced us that thermodynamic relations are applicable in a large reversible temperature domain of about 30 to 40 K below  $T_c$  in sufficiently large magnetic field (3-5 kOe). All measurements presented in the following are corrected for the normal-state contribution. The latter is obtained by a fit of the quasi-field- and temperature-independent susceptibility above the region where superconducting fluctuations contribute. The volume



Figure 2. Scaling plot of the M(T, H) data versus H for the Bi-2212 phase. The choice of H' = 20 kOe is arbitrary. The same scaling is also observed for the TI-2201 and TI-2212 phases.

of these pure ceramics is considered to be 100% superconducting below  $T_{\rm c}$ .

Figures 1 and 2 show the experimental data M(T, H) in both scalings proposed by Cho et al. Figure 1 presents M(T, H)/M(T', H) versus T, and figure 2 M(T, H)/M(T, H')versus H. The scaling temperatures T' and fields H' may be chosen anywhere in the reversible regime, and the values used here for definiteness are 50 K for Bi-2212, 70 K for TI-2212 and 60 K for TI-2201. Raw data are given in [4]. Except for the fluctuation region near  $T_c$ , the data for nine different fields (1-5 kOe) merge into single curves within experimental uncertainty. It mathematically follows that M(T, H) = f(T)g(H). The implications of this factorization on the critical field curve  $H_{c2}(T)$  require however a model, which we discuss now. Note that all ( $\approx$ 300) experimental points for Bi-2212 are reported in figure 3 as lines in the curve labelled 'data'.

We now consider the reversible mixed-state magnetization of extreme type II superconductors ( $\kappa \gg 1$ ) in intermediate fields ( $H_{c1} \ll H \ll H_{c2}$ ). Cho et al [2] used a model which contains the essential physics of the London model [5], and for simplification we turn to the latter in the present discussion. The results are equivalent within numerical factors of order one. For a ceramic with a large anisotropy ratio of the critical field, the



Figure 3. Scaling plot of the experimental points M(T, H)/M(T', H) versus T (curves labelled 'data') for the Bi-2212 phase. T' is set equal to 50 K. The other curves are recalculated points obtained after fitting different models (see the text). For clarity, the temperature scale is shifted by +10, +20 and -10 K for the 'London, WHH', 'London,  $H_{c2}$  = constant' and 'London + Fluctuation, WHH' models, respectively.

averaged magnetization given by the model of effective masses becomes [4]:

$$\langle M(T, H) \rangle^{\text{London}} = -\frac{1}{2} \frac{\phi_0}{32\pi^2 \lambda_{ab}^2(T)} \ln\left(\sqrt{e} \frac{\eta}{e} \frac{H_{c2,c}(T)}{H}\right)$$
(1a)

of the form  $f(T)g(H/H_{c2})$  and (T)).  $\phi_0$  is the flux quantum,  $\lambda$  the penetration depth  $\eta/e$  a numerical factor of order one (actually set to one in the following fits). The index c or ab refers to the direction of the field with respect to crystallographic axes. We have found the factors  $\frac{1}{2}$  and  $\sqrt{e}$  that come from angle averaging. Clearly, the experimental factorization M(T, H) = f(T)g(H) and the London model imply  $H_{c2}(T) = \text{constant}$ , as deduced by Cho et al. In figure 3, the curve labelled 'London,  $H_{c2} = \text{constant}'$ , we show a fit of the Bi-2212 data in this spirit. The 287 data used are in the range 60 < T < 89 K and 10 < H < 55 kOe. The penetration depth is assumed to follow the law  $\lambda^{-2}(t) = \lambda_{ab}^{-2}(0)(1 - t^k)$  with  $t = T/T_c$ . The fitted parameters are  $H_{c2,c} = 1450$  kOe,  $\lambda_{ab}(0) = 2420$  Å,  $T_c = 93.0$  K and k = 2.00; the quality of the fit is  $\chi^2 \approx 2.2\%$ . Most of the deviations come from the region close to  $T_c$ .

It is interesting to test how bad the prediction of the London model would be if a conventional variation were imposed for  $H_{c2,c}(T)$ . In figure 3, the curve labelled 'London, WHH' we performed a fit using the same data, but enforcing for  $H_{c2,c}(T)$  the WHH curve [6] and for the thermodynamic critical field  $H_c(T)$  the parabolic law of the two-fluid model; the relative variation of  $\lambda_{ab}(T)$  and  $\kappa^2(T)$  is then uniquely defined. The fitted parameters are now  $H_{c2,c}(0) = 8140$  kOe,  $\lambda_{ab}(0) = 2920$  Å, and  $T_c = 94.4$  K; the quality of the fit is  $\chi^2 \approx 3.4\%$ . Note that this unrealistic value of  $H_{c2,c}(0)$  is caused by the approach of the crossing point where M is independent of H and consequently,  $H_{c2,c}(0)$  in this

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analysis varies with the upper limit of the fitting range in an unphysical way. Clearly, the curves depart from scaling in figure 3, but accurate data are required to ascertain this. The difference between the curves 'London, WHH' and 'London,  $H_{c2}$  = constant' is the experimental evidence actually used by Cho *et al* to establish their result  $H_{c2}$  = constant. We now proceed to show that the spread in the bundle of curves labelled 'London, WHH' may be cancelled (within experimental uncertainty) by considering an extra term in the magnetization.



Figure 4. Scaling plot of M(T, H)/M(T, H') versus H for the Bi-2212 phase; we show here the recalculated points after fitting the 'London + Fluctuation, whit' model.

Bulaevskii et al [3] showed that fluctuations of vortices give rise to a contribution that reads, after averaging for a ceramic with large anisotropy [4]:

$$\langle M(T, H) \rangle^{\text{Fluctuation}} = +\frac{1}{2} \frac{k_{\text{B}} T}{\phi_0 s} \ln \left( \sqrt{e} \frac{16\pi k_{\text{B}} T \kappa^2}{\alpha_{\text{B}} \phi_0 s H \sqrt{e}} \right). \tag{1b}$$

 $k_{\rm B}$  is the Boltzmann constant,  $\kappa^2 = \lambda^2/\xi^2 = 2\pi H_{c2,c} \lambda_{ab}^2/\phi_0$ ,  $\xi$  is the coherence length,  $\alpha_{\rm B}$ a numerical factor of order one, and s the spacing between superconducting (groups) of layers. The factors  $\frac{1}{2}$  and  $\sqrt{e}$  come from angle averaging. In figure 3, the curve labelled 'London + Fluctuation, WHH', we show a fit using again the same data and including both equations (1a) and (1b). The free parameters are those of the 'London, WHH' scenario, which was just shown to be unsatisfactory, and the single additional numerical parameter of the vortex fluctuations term,  $\alpha_{\rm B}$ . The *effective* interlayer distance s between planes or groups of superconducting planes is determined independently by using the crossing point  $\{T^*; M^*\}$  and formula  $s = -\frac{1}{2}(k_{\rm B}T^*/\phi_0 M^*)$  [3,7]. The quality of the fit is now very good with  $\chi^2 \approx 1.5\%$ . Fitted parameters are given in table 2. Clearly, the data recalculated using equations (1a) and (1b) show the scaling property within experimental uncertainty, *embody a conventional variation of*  $H_{c2,c}(T)$ , and moreover reproduce more closely the

data in the fluctuation region near  $T_c$ . Indeed, they account for the crossing point  $\{T^*; M^*\}$  where  $\partial M/\partial H|_{T^*} = 0$  in the unscaled curves. Figure 4 shows the recalculated scaling M(T, H)/M(T, H') with the same model.

Table 2. Here  $(\phi_0/4\pi\lambda^2)H_{c2} = H_c^2$ ,  $H_{c2} = H_c\sqrt{2}\kappa$ ,  $H_{c1} = \phi_0 \ln(\kappa)/(4\pi\lambda^2)$ ,  $H_{c2,c}(0) = \phi_0/(2\pi\xi_{ab}^2(0))$ .

| Phase   | Bi-2212 | <b>Tl-2201</b> | T1-2212 |  |
|---|---------|----------------|---------|--|
| M* (emu cm <sup>-3</sup> )  | -0.145  | -0.064         | -0.145  |  |
| T* (K)  | 90.5    | 88.8           | 105.0   |  |
| s (Å)   | 20.8    | 46.3           | 24.6    |  |
| $\lambda_{ab}(0)$ (Å)   | 2210    | 2630           | 1910    |  |
| $H_{c2,c}(0)(10^4 \text{ Oe})$  | 67.9    | 43.2           | 79.7    |  |
| $T_{c}^{L+B}$ (K)   | 99.7    | 94.3           | 112.6   |  |
| α <sub>B</sub>  | 1.51    | 1.38           | 1.56    |  |
| $\partial H_{c2,c}/\partial T _{T_c}$ (10 <sup>4</sup> Oe K <sup>-1</sup> ) | -0.95   | 0.64           | -0.99   |  |
| $H_{\rm c}(0)$ (10 <sup>4</sup> Oe)   | 0.479   | 0.320          | 0.601   |  |
| ĸ   | 100     | 95             | 94      |  |
| $H_{c1,c}(0)$ (Oe)  | 156     | 108            | 206     |  |
| ξ <sub>ab</sub> (0) (Å)   | 22.0    | 27.6           | 20.3    |  |

Attempts to observe high-field scaling of the form  $M/\sqrt{TH} = f([T - T_c(H)]/\sqrt{TH})$ near  $T_c$  [7] were rather unsuccessful, possibly owing to the polycrystalline nature of the ceramics.

The experimentally found scaling M(T, H)/M(T', H) versus T and M(T, H)/M(T, H') versus H in the mixed state of quasi-2D superconducting cuprates [2] may be explained either (a) by a constant upper critical field, using the London model alone [2], or (b) by a conventional variation of the upper critical field, if we additionally take into account the fluctuations of vortices. In the latter case, scaling is not exact, but experimentally indistinguishable from true scaling.

We verified that the more 3D-like YBaCuO compounds do not show this scaling property. This reflects the fact that vortex fluctuations in the small reversible regime below  $T_c$  do not contribute substantially to the magnetization, as mentioned previously by Bulaevskii *et al* [8].

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