Large effect of columnar defects on the thermodynamic properties of $Bi_2Sr_2CaCu_2O_8$ single crystals

C. J. van der Beek
Département de Physique, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

M. Konczykowski
Laboratoire des Solides Irradiés, Ecole Polytechnique, 91128 Palaiseau, France

T. W. Li and P. H. Kes
Kamerlingh Onnes Laboratorium, Leiden University, P.O. Box 9506, 2300 RA Leiden, The Netherlands

W. Benoit
Département de Physique, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

(Received 15 April 1996)

The introduction of columnar defects by irradiation with 5.8-GeV Pb ions is shown to affect significantly the reversible magnetic properties of $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals. Notably, the suppression of superconducting fluctuations on length scales greater than the separation between columns leads to the disappearance of the "crossing point" in the critical fluctuation regime. At lower temperatures, the strong modification of the vortex energy due to pinning leads to an important change of the reversible magnetization. The analysis of the latter permits the direct determination of the pinning energy. [S0163-1829(96)50926-4]

The layered structure of the high-temperature superconductor oxides, in which strongly superconducting CuO_2 planes are separated by weakly superconducting layers of thickness s, leads to a high sensitivity of the vortex lattice to both pinning and thermal fluctuations, particularly in the most anisotropic materials such as $Bi_2Sr_2CaCu_2O_{8+\delta}$.[1] At sufficiently low temperature, the vortices optimally adapt to the pinning centers, which results in a large critical current density j_c and irreversible magnetization. However, as the temperature or the field is increased, vortex positional fluctuations eventually cause the rapid drop of j_c and of the creep barriers,[2] and the irreversible part of the magnetization is suppressed. In the London limit, corresponding to fields B much below the upper critical field $B_{c2}(T)$, vortex positional fluctuations, which correspond to fluctuations of the phase of the superconducting order parameter ψ, also modify the logarithmic field dependence of the reversible magnetization.[3] As $B_{c2}(T)$ is approached, quasi-two-dimensional (2D) fluctuations of the overall amplitude $|\psi|$ become important. The interaction between amplitude fluctuations in the critical regime gives rise to the smooth behavior of the magnetization M around the field-dependent transition temperature $T_c(B)$,[4] with the notable feature that M is field independent at the temperature T^*, i.e., it shows a "crossing point" as function of temperature.[1,5]

A theory that continuously describes the superconducting fluctuations from the weak fluctuation regime above the mean-field transition temperature T_0, through the critical regime, to the vortex state below $T_c(B)$ was developed by Tešanović *et al.*[5,6] The authors derived an explicit functional form for the free energy of a 2D superconductor in a magnetic field that is in excellent agreement with experiments on $Bi_2Sr_2Ca_2Cu_3O_0$ tapes and $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals.[7] In particular, the magnetization in the critical regime obeys the 2D scaling law[5]

$$\frac{M}{\sqrt{TH}}\frac{\Phi_0 s H'_{c2}}{Ak_B}=f\left(A\frac{T-T_c(H)}{\sqrt{TH}}\right), \tag{1}$$

$$f(x)=x-\sqrt{x^2+2}, \tag{2}$$

with $\mu_0 H'_{c2}=(\partial B_{c2}/\partial T)_{T=T_c}$ and A a constant given in Ref. 5. Moreover, the function (2) reproduces the "crossing point." Equation (1) is valid for reduced fields $b\equiv B/[(T-T_c)(\partial B_{c2}/\partial T)_{T=T_c}]>\frac{1}{3}$ where superconducting electrons are confined to the lowest Landau level.

In this Rapid Communication we show *directly* that the introduction of very strong pinning centers, namely, amorphous columnar defects with radius comparable to the Ginzburg-Landau coherence length ξ, considerably modifies the free energy of the vortices in the mixed state. The measurement of the corresponding change of the equilibrium magnetization yields a direct and model-independent determination of the energy gain due to pinning in the equilibrated system. Furthermore, we will show that there is also a large effect on superconducting fluctuations, most pronounced in the critical regime, where we find a maximum in $|M|$ and the disappearance of the "crossing point" for fields $0.2B_\Phi \lesssim \mu_0 H < B_\Phi$. Here $B_\Phi=\Phi_0 n_d$ is the matching field at which the density of vortices $n_v=B/\Phi_0$ is equal to the density of defects n_d and Φ_0 is the flux quantum.

Single crystals of $Bi_2Sr_2CaCu_2O_{8+\delta}$ were grown using the traveling-solvent floating zone technique.[8] Crystal 1 had dimensions $1.2\times2.4\times0.020$ mm^3 and was annealed in air, whereas crystal 2, of dimensions $0.55\times1.45\times0.060$ mm^3, was cut out of a larger piece that had been annealed in oxy-

0163-1829/96/54(2)/792(4)/$10.00

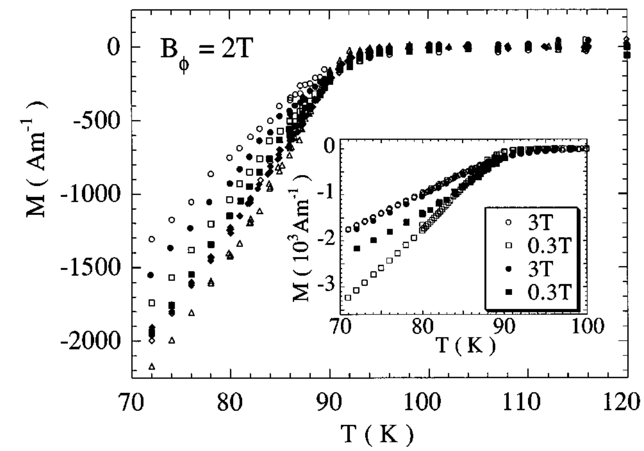

FIG. 1. The reversible magnetization of crystal 2, with $T_c \approx 89$ K and $B_\Phi = 2$ T, as function of temperature, at field values of 0.3 T (△), 0.7 T (♦), 1 T (◇), 2 T (■), 3 T (□), 4 T (●), and 5 T (○). The inset shows the same data for $\mu_0 H = 0.3$ T and 3 T (■, ●), compared to the magnetization of the unirradiated sample of Ref. 1 at those same fields (□,○).

gen for a period of three days. The crystals were subsequently irradiated at doses of 5×10^{14} m^{-2} and 1×10^{15} m^{-2} 5.8 GeV Pb ions at the Grand Accélérateur National d' Ions Lourds (GANIL) at Caen, France. The heavy ion beam was aligned parallel to the sample c axes, and produced parallel amorphous columnar defects of radius $b_0 \approx 3.5\times10^{-9}$ m, which traversed the whole sample. Defects produced in this manner are known to be insulating,[9] and thus represent regions in which superconductivity is completely suppressed. The density of columns, n_d, corresponds to the irradiation dose, and to the matching fields B_Φ of 1 T (crystal 1) and 2 T (crystal 2). Measurements of the magnetization were performed using a commercial superconducting quantum interference device (SQUID) magnetometer, using a scan length of 3.0 cm. In order to minimize the background signal, the sample was suspended between two fine quartz fibers. Nevertheless, a temperature-independent background remained; all measurements presented below have been corrected by subtracting the magnetization measured at $T = 120$ K. Approximate $T_c(0)$ values of 82.0 K (sample 1) and 88.8 K (sample 2) were obtained as the extrapolation to zero of the field-cooled magnetization as measured in the remnant field of the superconducting magnet.

Figure 1 shows the magnetization of sample 2 as a function of temperature, as measured in different constant applied magnetic fields. In the temperature and field regime shown, M was completely reversible; only the measurements taken at $\mu_0 H = 0.3$ T showed a slight irreversibility at $T < 75$ K. A comparison with the as-grown $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal of Ref. 1, which has nearly the same T_c, shows that whereas at fields above B_Φ the behavior of the magnetization is similar to that of the unirradiated sample, at fields below B_Φ its magnitude has become much smaller, especially at temperatures below 85 K. As function of field, $|M|$ first decreases, then slightly increases, before decreasing again when $\mu_0 H \geqslant B_\Phi$.

The same three field regimes are seen more clearly in Figs. 2 and 3, which show the magnetization of sample 1.

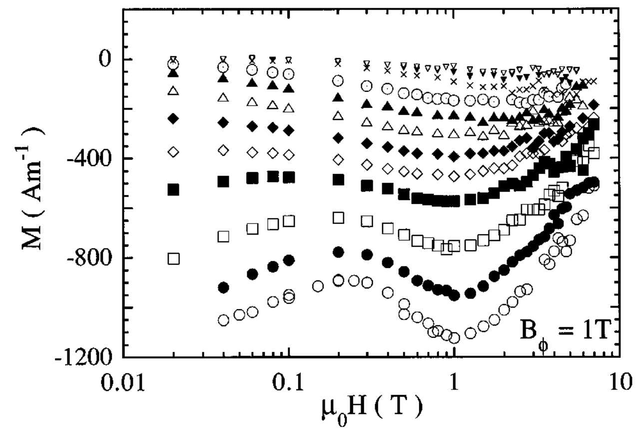

FIG. 2. The reversible magnetization of crystal 1, with $T_c(0) \approx 82$ K and $B_\Phi = 1$ T, as function of magnetic field, at temperatures of 72 K (○), 74 K (●), 76 K (□), 78 K (■), 79 K (◇), 80 K (♦), 81 K (△), 82 K (▲), 83 K (⊙), 84 K (×), 85 K (▼), and 86 K (▽).

Below 78 K, $|M|$ first decreases logarithmically as function of H, then, for $\mu_0 H \gtrsim 0.2 B_\Phi$, $|M|$ *increases* with increasing H, until it reaches a maximum at $\mu_0 H = B_\Phi$. Above B_Φ, $|M|$ again decreases proportionally to $\ln H$, but with a larger slope. Although the maximum in $|M|$ was also observed in Ref. 10, neither the clear correlation of the position of the maximum with the value of the matching field, nor the minimum in $|M|$ at lower H was reported there. The low-field magnetization is independent of H at $T_1^* \approx 78.9$ K, implying the existence of a ''crossing point'' in this field regime. Above B_Φ the magnetization becomes independent of H only at $T_2^* \approx 84$ K. For $T_1^* < T < T_2^*$, the magnetization

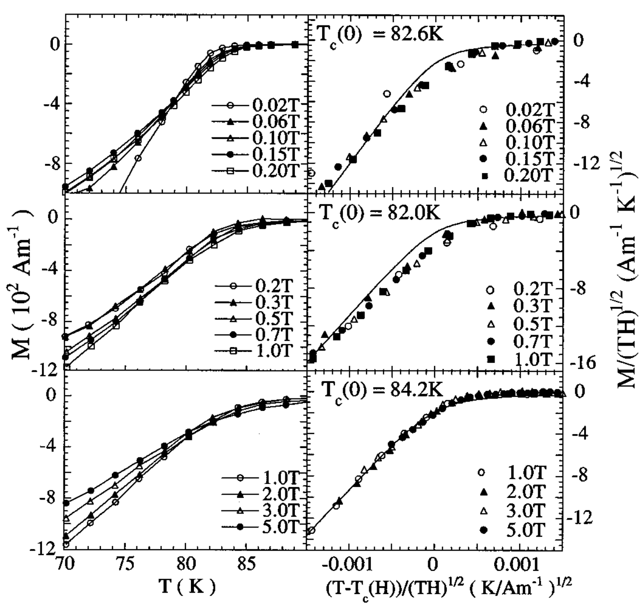

FIG. 3. The reversible magnetization of crystal 1, as function of temperature. The left-hand panels depict, from top to bottom, the magnetization in the field range 0.02 T $< \mu_0 H <$ 0.2 T, 0.2 T $< \mu_0 H < B_\Phi =$ 1 T, and 1 T $< \mu_0 H <$ 5 T. The right-hand panels show the same data, scaled according to Eq. (1). The drawn lines indicate best fits to the scaling function $f(x) = x - \sqrt{x^2+2}$.

shows a single maximum at $\mu_0 H = B_\Phi$, and above T_2^*, $|M|$ monotonically increases over the whole field range, with a linear departure from $H=0$ and saturating to a constant value at large H. The same features were found in the $M(H)$ behavior of sample 2. Summarizing, we find that at low fields the magnetization shows a crossing point at $T_1^* = 78.9$ K and 86.8 K for samples 1 and 2, respectively. Both $t_1^* = T_1^*/T_c(0) \approx 0.97$ and the value of $M_1^* \approx 400$ A m^{-1} are comparable to the values found in the unirradiated sample. At fields above B_Φ there is a crossing point at $t_2^* = T_2^*/T_c(0) \approx 1.0$, with $M_2^* \approx$ 150 A m^{-1}. In the intermediate field regime the crossing point in the critical fluctuation regime is suppressed.

In spite of the important changes with respect to the unirradiated sample, the magnetization in the critical regime still shows the 2D scaling property predicted by Refs. 4 and 5 at all fields. Figure 3 shows plots of $M/\sqrt{TH}$ versus the scaling parameter $[T - T_c(H)]/\sqrt{TH}$ for crystal 1. Here we took $T_c(H) = T_c(0) - H/H'_{c2}$, with $\mu_0 H'_{c2} = 1.15$ T K^{-1}, consistent with a $\mu_0 H_{c2}(0)$ value of 75 T, and $T_c(0)$ as the only free parameter. While the magnetization curves in each of the three field regimes $\mu_0 H \lesssim 0.2 B_\Phi$, $0.2 B_\Phi \lesssim \mu_0 H < B_\Phi$, and $\mu_0 H > B_\Phi$ obeyed the scaling law separately, with $T_c(0)$ values of 82.6 K, 82.0 K, and 84.2 K, respectively, it was not possible to scale *all* the data together in a single curve, that is, using a single $T_c(0)$ value. The curve measured in 0.02 T did not conform to the scaling plot at all, probably because the critical regime is too narrow at this field. As for the functional behavior of the magnetization, only the data measured in fields above B_Φ was well described by Eq. (2), and therefore display the same scaling as the magnetization of the unirradiated sample.

The decrease of the reversible magnetization after heavy-ion irradiation and the maximum in $|M|$ in the low-b London regime can be straightforwardly accounted for by considering the decrease of the mixed state free energy due to the localization of vortices on columnar defects. The free energy of the irradiated superconductor in a magnetic field can be expressed as

$$G(B) = G(0) + \frac{n_v \epsilon_0}{2} \ln\left(\frac{\eta B_{c2}}{B}\right) - \frac{k_B T}{s}(n_v - n_\Phi)\ln\left(\frac{B_0}{B}\right) - n_\Phi U(T,B), \quad (3)$$

where $G(0)$ is the free energy in zero field, the second and third terms on the right represent, respectively, the energy of the free vortices and the entropy gain due to their fluctuations, assuming that this is of the same form as in the unirradiated system.[3,11] The energy scale for vortex interactions $\epsilon_0 = \Phi_0^2/4\pi\mu_0\lambda^2$, with λ the penetration depth, η is a constant of order unity introduced to take the finite size of the vortex core into account, and B_0 is a scaling field of the order of B_{c2}.[11] The fourth term in Eq. (3) is the product of the density of vortices trapped by a column n_Φ and the pinning energy per unit length per trapped vortex $U(T,B)$. The magnetization $M = -\partial G/\partial B$ is equal to

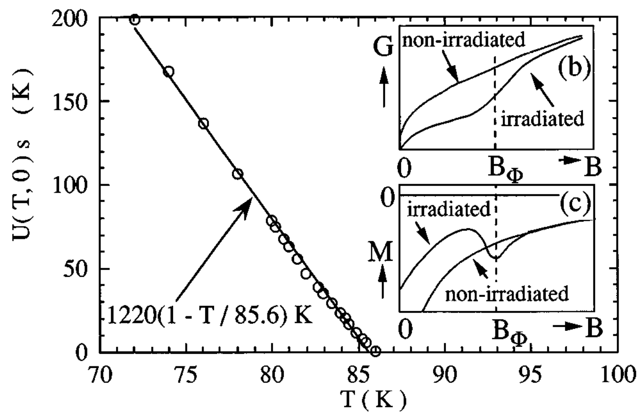

FIG. 4. (a) Gain in vortex energy due to localization on a columnar defect, as extracted from the difference in the magnetization of an irradiated and an unirradiated $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystal. (b) Qualitative behavior of the mixed state free energy G in the heavy-ion irradiated sample. (c) Corresponding behavior of the magnetization $M = -\partial G/\partial B$.

$$M \approx -\frac{\epsilon_0}{2\Phi_0}\ln\left(\frac{\eta B_{c2}}{eB}\right) + \frac{k_B T}{\Phi_0 s}\left[\left(1 - \frac{\partial n_\Phi}{\partial n_v}\right)\ln\left(\frac{B_0}{B}\right) - 1 + \frac{n_\Phi}{n_v}\right] + \frac{1}{\Phi_0}\frac{\partial n_\Phi U(T,B)}{\partial n_v}. \quad (4)$$

At fields $B \ll B_\Phi$, we expect all vortices to be individually pinned by a single column, so that the field dependence of $U(T,B)$ can be ignored and $n_\Phi \approx n_v$. The difference ΔM between the magnetization of the irradiated and the unirradiated crystal should then be directly proportional to the single-vortex pinning energy $U(T,0)$. In Fig. 4, we plot $\Delta M(\Phi_0 s/k_B) = U(T,0)s/k_B$ for $\mu_0 H = 0.3$ T. The magnitude and temperature dependence, $1220(1 - T/[\mathrm{K}]/85.6)$ K is in good agreement with the prediction for the pinning energy per 2D vortex segment (''pancake'') of length s, $\epsilon_0 s \approx 1020(1-t)$ K for $\lambda(0) = 1.7\times10^{-7}$ m.[12] At fields $B > B_\Phi$, vortices will be pinned collectively; the product $n_\Phi U(T,B)$ will then decrease as function of field. The overall behavior of the total free energy and its derivative as function of field then directly explains the presence of a maximum in $|M|$ in the London regime (Fig. 4). The crucial point of the above analysis is that vortex pinning not only affects the irreversible magnetic properties of the superconductor, but also its thermodynamic properties. Notably, the reversible magnetization is modified in the presence of pinning because of the lowering of the Gibbs free energy of the vortex state, as can be clearly seen in the inset of Fig. 1. Accordingly, when fluctuations are not dominant, the analysis of the reversible magnetization permits the determination of the energy gain due to pinning in the equilibrated system.

It follows from Eq. (4), in conjunction with the above assumptions that $U(T,B \ll B_\Phi) \neq U(B)$ and $n_\Phi(B \ll B_\Phi) \approx n_v$, that in the low field limit $\partial M/\partial \ln B \approx \epsilon_0/2\Phi_0$. At $B \gg B_\Phi$, $\partial M/\partial \ln B$ is determined by the excess vortices that are not trapped by a columnar defect, and should therefore be approximately equal to $\epsilon_0/2\Phi_0 - k_B T/\Phi_0 s$. However, Fig. 2 shows that the logarithmic field derivative of M is larger at high fields than at low fields. This means that already in the

London regime, the columnar defects modify superconducting fluctuations. The fluctuation entropy seems to become the dominant contribution to the free energy at reduced temperatures $t \approx 0.95$. Namely, Fig. 4 shows that the pinning energy vanishes around this temperature.[13] There is then no longer any reason to expect that the vortices are still localized on the columns. The fact that above $t \approx 0.95$ the data obey the 2D scaling law (1) proposed by Ullah and Dorsey and by Tešanović *et al.* then leads us to believe that near $T_c(B)$ vortices are not only delocalized from the columnar defects, but are thermally decomposed into 2D ''pancake'' vortices, as is the case in unirradiated samples at these temperatures and fields.

Our second important point concerns the persistence of the maximum in $|M|$ in the critical fluctuation regime and the disappearance of the crossing point in the magnetization at intermediate field strengths. The critical regime is theoretically defined as that portion of the (H,T) plane where $b > \epsilon_0 s/k_B T$, i.e., where the typical length scale of superconducting fluctuations $\xi > (\epsilon_0 s/2\pi k_B T n_v)^{1/2}$.[11] Experimentally, it corresponds to the temperatures and fields where the magnetization obeys the 2D scaling law (1). The large value of ξ in the critical region means that the interaction between fluctuating regions determine the behavior of thermodynamic and transport quantities.[4] The large effect of the columnar defects should therefore be interpreted in terms of an effective ''screening'' of this interaction. A comparison of the column density n_d and the quantity $\epsilon_0 s/k_B T \xi^2$ shows that such an effect may be important over a temperature span of up to 5 K around $T_c(0)$. We can then understand the peculiar field dependence of $|M|$ in terms of the distribution of distances between columns. At small fields, fluctuating regions are, on the average, far apart, and their extent is limited by the typical distance between columns. Superconducting coherence over large distances cannot set in until $\xi \ll n_d^{-1/2}$, which is reflected in the low value of $T_c(0)$ extracted from the 2D scaling procedure. At large fields $B > B_\Phi$, the fluctuations are limited by the interaction with other zeros of the order parameter, so that the qualitative behavior is similar to that of an unirradiated sample. This is in agreement with the fact that the magnetization fits the function (2). At intermediate fields, the magnetization is determined by the averaged behavior of regions of the sample in which columnar defects are either far apart or close together. The inhomogeneity introduced by the heavy-ion irradiation now separates the sample into many regions with slightly different $T_c(B)$, explaining the absence of the crossing point. In the weak fluctuation regime, we always have $\xi \ll n_d^{-1/2}$ and there is no noticeable effect of the columns. Finally we note that the large value of $\partial M/\partial \ln B$ at fields $B > B_\Phi$, and the related fact that the crossing point at high fields happens at a higher temperature and lower magnetization value than in the unirradiated sample may indicate the suppression of long-wavelength fluctuations in the vortex system, such as suggested in Ref. 14.

In conclusion, we have shown that the introduction of columnar defects significantly modifies the free energy of the mixed state and the reversible magnetization in $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals. In the London regime, the main effect is due to the large reduction of the vortex energy associated with the pinning by columnar defects. The analysis of the reversible magnetization then permits the determination of the pinning energy in a direct and model-independent way. The influence of the defects remains pronounced all the way into the critical fluctuation regime, where the suppression of fluctuations at distances larger than the separation between columns leads to the disappearance of the crossing point in the magnetization.

We gratefully acknowledge M.V. Indenbom, A. Buzdin, and L.N. Bulaevskii for discussions, J.-Ph. Ansermet and B. Doudin for access to the SQUID magnetometer, and the Netherlands Foundation FOM (ALMOS) for providing the crystals. This work was supported by the Swiss National Foundation under Grant No. 21-42015.94, and by an International cooperation Grant of the European Union under Contract No. CI-1-CT93-0069.

[1] P. H. Kes *et al.*, Phys. Rev. Lett. **67**, 2382 (1991).
[2] C. J. van der Beek *et al.*, Phys. Rev. Lett. **74**, 1214 (1995); Phys. Rev. B **51**, 15 492 (1995).
[3] L. N. Bulaevskii, M. Ledvij, and V. G. Kogan, Phys. Rev. Lett. **68**, 3773 (1992).
[4] S. Ullah and A. T. Dorsey, Phys. Rev. B **44**, 262 (1991).
[5] Z. Tešanović *et al.*, Phys. Rev. Lett. **69**, 3563 (1992).
[6] Z. Tešanović and L. Xing, Phys. Rev. Lett. **67**, 2729 (1991).
[7] Qiang Li *et al.*, Phys. Rev. B **48**, 9877 (1993).
[8] T. W. Li *et al.*, J. Cryst. Growth **135**, 481 (1994).
[9] J. Aarts *et al.* (unpublished).
[10] A. Wahl *et al.*, Physica C **250**, 163 (1995).
[11] A. E. Koshelev, Phys. Rev. B **50**, 506 (1994).
[12] D. R. Nelson and V. M. Vinokur, Phys. Rev. Lett. **68**, 2398 (1992); Phys. Rev. B **48**, 13 060 (1993).
[13] This observation is in agreement with that of Ref. 2 where no pinning contribution to the ac shielding current could be found above $t \approx 0.92$.
[14] L. N. Bulaevskii *et al.*, Phys. Rev. B **48**, 13 798 (1993).