Vortex Lattice Structure in $Bi_2Sr_2CaCu_2O_{8+\delta}$ at High Temperatures

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(Received 6 August 1996)

The microscopic structure of the magnetic flux-line lattice (FLL) in $Bi_2Sr_2CaCu_2O_{8+\delta}$ superconductors was studied at temperatures up to 77 K by decoration. Comparison of FLLs obtained at 55 and 4.2 K shows that twisted bond defects are a manifestation of thermal fluctuations at elevated temperature. Analyses of the orientational and translational correlation functions for field and zero-field cooled lattices obtained at 55 K suggest that the observed FLL is an equilibrium hexatic. These data were also used to estimate the FLL freezing temperature. [S0031-9007(96)01837-6]

PACS numbers: 74.60.Ge, 61.18.–j, 74.60.Ec, 74.72.Hs

The static and dynamic structure of magnetic flux line lattices (FLLs) in high-T_c superconductors (HTSCs) has been the focus of much research [1]. Bitter decoration can be used to probe the microscopic structure of the FLL by providing a picture of the configuration of individual vortices emerging from a sample surface. This technique has been used to elucidate a number of interesting features in HTSCs [2–5], although almost without exception these investigations have been carried out by quenching samples to 4.2 K in a magnetic field [6]. It is generally believed that the FLL probed in these experiments falls out of equilibrium close to the irreversibility temperature $T_{\rm irr}$ [2–5], and this has been supported by estimates of the effective penetration depth in several experiments [3,4]. Hence the structures probed in previous experiments at 4.2 K do not represent equilibrium configurations. To access equilibrium FLL structures, which are essential to quantitative interpretations of structural data [5] and current interest in melting, requires that measurements be carried out above the temperature T_f, where the FLL dynamics are effectively frozen.

In this Letter, we demonstrate the first successful decorations of FLLs at temperatures up to 77 K in $Bi_2Sr_2CaCu_2O_{8+\delta}$ (BSCCO) superconductors. Individual vortices have been clearly resolved in fields of 14 and 30 G and temperatures up to 77 K. Quantitative analysis of the FLL structure obtained in field-cooled (FC) and zero-field-cooled (ZFC) experiments at 55 K along with the comparison to conventional 4.2 K experiments provides microscopic insight into the topological nature of thermal fluctuations, the equilibrium FLL structure, and the freezing temperature in previous low-temperature decoration experiments.

Single crystals of BSCCO were grown and characterized as described elsewhere [7]. Experiments were carried out using pumped liquid nitrogen as the cryogen between 54 and 77 K. The helium background gas in the decoration chamber was adjusted to achieve a mean-free-path of $\sim 1\ \mu$m at these temperatures. In FC experiments, samples were cooled at a rate of 0.05 K/min to the desired temperature $T_{\rm FC}$; decorations were carried out at the specified temperature after waiting 30–60 min. In ZFC experiments, samples were cooled to the desired temperature $T_{\rm ZFC}$, the field was applied, and then decorations were carried out after waiting 30–60 min. There were no appreciable differences observed between FLL structures at the central region of samples decorated after a 30 or 60 min equilibration period. In addition, samples from the same batch were decorated at 4.2 K (LT) using the conventional field cooling and quenching process.

Figure 1 shows typical images of the decoration patterns obtained from FC and ZFC experiments at $\sim$55 K in a 14 G field. In general, we find that the positions of

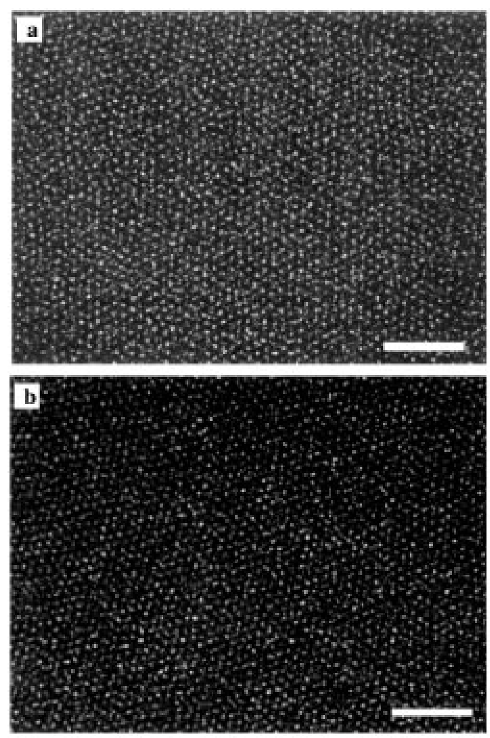

FIG. 1. Typical SEM images of decoration patterns obtained at 14 G for (a) FC ($T_{\rm FC} = 54$ K) at a rate of 0.05 K/min and (b) ZFC ($T_{\rm ZFC} = 57$ K). The samples were decorated after allowing 30 min for equilibration. Scale bar corresponds to 10 μm.

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most vortices are well-resolved in these high-temperature decoration experiments. The iron clusters decorating each vortex site are more irregular than in our previous low-temperature studies [4,5], and this irregularity leads to slightly greater uncertainty in the positions of individual vortices compared to low-temperature experiments. The irregular shape of the clusters may be due in part to thermal motion of vortex lines [8] and a reduction in the attractive interaction between vortices and magnetic clusters relative to thermal energy.

We also have been able to decorate successfully the FLL at 77 K [Fig. 2(a)]. Many individual vortices as well as lattice rows of the FLL can be resolved at 77 K, although some vortices are hard to resolve through the background iron particles on the surface. The observation of well-defined lattice rows shows that vortices are in a solid rather than liquid state. This point was confirmed through the autocorrelation function and Fourier transform of the raw data [Figs. 2(b) and 2(c)]. The autocorrelation function shows that at least fifth nearest neighbors are seen on average, and the six clear first order peaks in Fourier space show that there is a well-defined triangular FLL. These results contrast a previous decoration of BSCCO at 15 K where several blurred iron clusters were observed and attributed to a vortex liquid state [6]. Qualitatively, our results demonstrate that vortices exist in a solid state in BSCCO for temperatures up to at least 77 K for the low-field regime for our experiments, and are thus consistent with recent local Hall probe magnetization measurements [9].

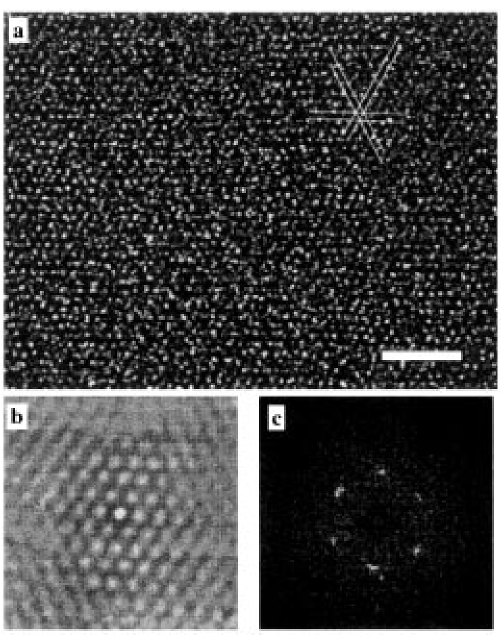

FIG. 2. (a) SEM image of a sample decorated at 77 K in a field of 14 G. The white lines at the upper right highlight the lattice rows. The scale bar corresponds to 10 μm. (b) Central portion of the autocorrelation function calculated from the data in (a). (c) Fourier transform of the data in (a).

To obtain a more quantitative picture of the high-temperature FLL structure, we have determined the nature of topological defects and the translational and orientational correlation functions. Topological defects in the FLL are readily highlighted in Delaunay triangulations of the vortex positions [2–5]. Typical results obtained for FC, ZFC, and LT experiments are shown in Fig. 3. Both isolated dislocations and defect clusters consisting of tightly bound dislocations can be seen in all of the patterns. A summary of the defects statistics obtained from several images for each experimental condition is given in Table I. This summary shows that the density of defects without a net Burgers vector (i.e., topologically uncharged) is larger in FLLs at 55 K compared to those at 4.2 K, while the densities of isolated dislocations are similar at 55 and 4.2 K [10]. Most of the uncharged topological defects consist of tightly bound dislocations called twisted bonds. Because twisted bonds can be formed by simply distorting the lattice at short range, these defects cost less energy to create than isolated dislocations. Hence we believe that the proliferation of twisted bonds at 55 K corresponds to the short-range thermal fluctuations in the equilibrium state at this temperature.

The translational $g_G(r)$ and the six-fold bond orientational $g_6(r)$ correlation functions were calculated from digitized images as described previously [2]. For all data, $g_G(r)$ is well fit by an exponential $\propto \exp(-r/\xi_T)$, where ξ_T is the translational correlation length. The $\xi_T s$ in units of the flux-line lattice constant a_0 were $\sim$6.6 (LT), $\sim$2.3 (FC), and $\sim$2.5 (ZFC) at 14 G [Fig. 4(a)], and $\sim$10 for LT, FC, and ZFC experiments at 30 G. The $g_6(r)$ calculated from these same data were well fit by a power law $\propto r^{-\eta_6}$ (vs exponential) with exponents $\eta_6 \sim 0.24$ (FC), $\sim$0.26 (ZFC), and 0.07 (LT) at 14 G [Fig. 4(a)]. The exponents for FC, ZFC, and LT data obtained at 30 G are all close to our experimental limit 0.03. The observation of long-range (i.e., power law decay) bond orientational order and short-range positional order for the FLLs obtained in both FC and ZFC experiments suggest that the high-temperature phase is a hexatic [11]. The similarity in structural parameters, including calculated structure functions [Fig. 4(b)], implies that the FLL configurations obtained from FC and ZFC samples are the same (despite different histories), and thus that the hexatic is a true equilibrium state.

Hexatic order was reported previously for FLL structures obtained in LT experiments [2]. It was suggested that the hexatic state might reflect either (1) a competition between impurity pinning and intervortex interaction [12] or (2) a frozen-in (i.e., quenched) high-temperature hexatic liquid state [13]. On the basis of our experiments it is possible to rule out that hexatic order is simply a quenched-in vestige of a high-temperature hexatic liquid state [i.e., (2) above]. It is also worth noting that pinning by point impurities is expected to be very weak at 55 K,

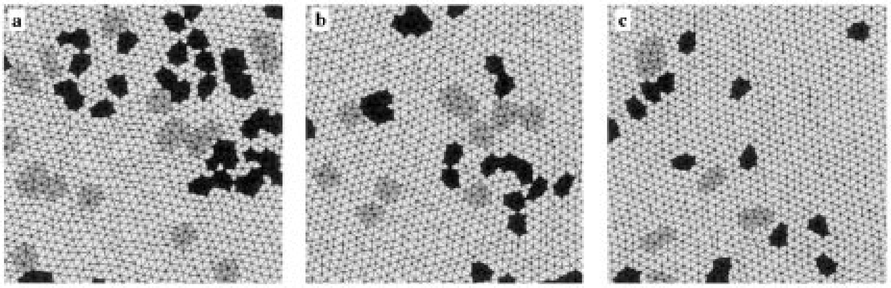

FIG. 3. Delaunay triangulation of FLL data recorded on (a) FC 54 K, (b) ZFC 57 K, and (c) LT (FC quenched to 4 K); the applied field is 14 G. Defect clusters containing an isolated dislocation are shaded black and twisted bonds defects are shaded gray.

and thus (1) may not be a viable explanation of the observed hexatic order. Studies of the temperature and field dependence of the topological defects and correlation lengths should resolve this important point.

From our data it is also possible to estimate the energy of twisted bond defects, which are a topological manifestation of thermal fluctuations, and thereby the freezing temperature in previous 4.2 K experiments. By assuming a two-state model, the fraction of twisted bonds relative to the total number of vortices is given by $f_{\mathrm{TB}} = \exp(-E_a/k_B T)$, where E_a is the activation energy for the formation of twisted bond defects and k_B is Boltzmann's constant. Analysis of the high-temperature decoration data obtained at 14 and 30 G shows that the corresponding E_a are 136 and 220 K, respectively. We can thus estimate the effective freezing temperatures T_f in LT experiments using E_a and the experimentally determined f_{TB}: T_f is ~35 K for data obtained at both 14 and 30 G. This value of T_f represents a ~lower bound to the true freezing temperature, because we have not accounted for the uncertainty in vortex positions associated with somewhat irregular iron clusters at high temperature (see above).

Hence, we have also probed T_f by analyzing the long-wavelength tail of the two-dimensional structure function $S_2(q_\perp)$, since this should be less sensitive to local uncertainties in vortex position. $S_2(q_\perp)$ derived using a hydrodynamic model [14], which is applicable to the analysis of a solid FLL away from the reciprocal lattice vectors, is

$$S_2(q_\perp) \approx \frac{n_0 k_B T}{\sqrt{c_{11}(q_\perp) c_{44}(q_\perp)}}\, q_\perp \,, \tag{1}$$

where $q_\perp$ is the in-plane wave vector, n_0 is the density of flux lines, and c_{11}, c_{44} are the compressional and tilt moduli of the FLL. Assuming that c_{11} and c_{44} have a small-temperature dependence in our experimental range [1], the slope of the long-wavelength tail of $S_2(q_\perp)$ is proportional to the temperature at a given magnetic field. The T_f estimated from the ratio of the slopes of linear fits to the long-wavelength tails [inset of Fig. 4(b)] shows that T_f is ~30 K for both 14 and 30 G experiments. These results are in rough agreement with the value of T_f obtained from the analysis of twisted bond defects. We thus believe that the actual vortex freezing temperature may close to our lower bound, although

TABLE I. The fraction of twisted bonds defects f_{TB} and fraction of isolated dislocations f_{ID} observed under different conditions. More than 4000 vortices were considered for each entry.

Field (G)	History	f_{TB}	f_{ID}
14	FC	0.080	0.010
	ZFC	0.062	0.009
	LT	0.019	0.006
30	FC	0.017	~0
	ZFC	0.014	0.002
	LT	0.002	0.001

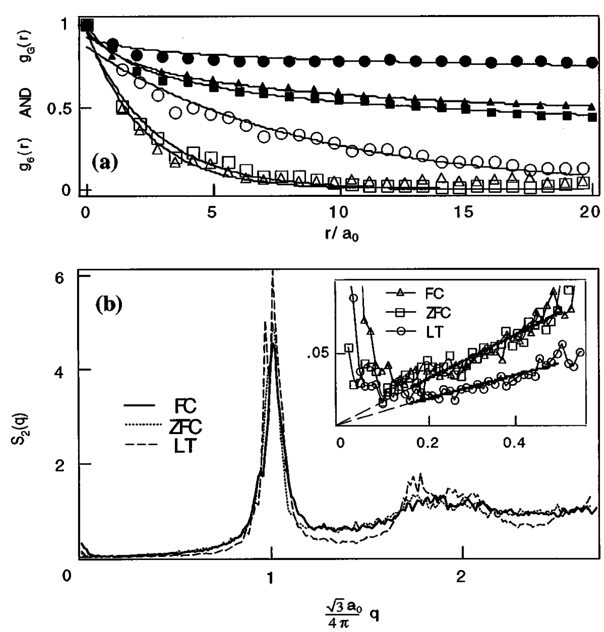

FIG. 4. (a) Translational correlation functions (empty symbols) $g_G(r)$ and bond orientational correlation functions (filled symbols) $g_6(r)$. Triangles, squares, and circles correspond to FC, ZFC, and LT experiments, respectively. The solid lines correspond to fits to exponential $[g_G(r)]$ and power law $[g_6(r)]$ decay functions. (b) Angular averages of the two-dimensional structure functions $S_2(q_\perp)$ calculated from the FLL data. The inset corresponds to blow-ups of the long wavelength tails of $S_2(q_\perp)$; the solid lines correspond to linear fits using Eq. (1). All experiments were carried out in a field of 14 G.

additional experiments are needed to define this value clearly.

These results suggest that FLL structures on a length scale of $\sim(2-10)a_0$ do not fall out of equilibrium until temperatures far below the irreversibility temperature T_{irr} that is determined from dc-magnetization measurements. A similar conclusion was reached previously in decoration studies of $YBa_2Cu_3O_{7-\delta}$ [3]. This is perhaps not surprising since the value of T_{irr} determined by magnetization measurements in a clean crystal is governed primarily by sample geometrical or surface barriers [9]. In our decoration measurements, we probe the microscopic properties of the FLL when analyzing data away from the sample edges [15]. Hence, the T_f should be associated with a microscopic phenomena related to the rapid slowing of the relaxation of individual vortices at some temperature below the first order melting line in the presence of point pinning centers. The existence of a microscopic irreversibility line well below the melting line has been recently examined theoretically [16] and is supported by our results.

In conclusion, we have reported the first successful decoration studies resolving individual flux lines above 55 K. Comparison of the vortex-resolved FLL structures obtained at 55 and 4.2 K has shown that the fraction of twisted bond defects is greater at high temperatures, while the density of dislocations is roughly independent of temperature. These results suggest that twisted bonds are a topological manifestation of thermal fluctuations in the FLL. Analyses of the orientation and translational correlation functions for FC and ZFC FLLs obtained at 55 K further show that the lattices have the same correlation lengths, and that these are typical of a hexatic state. Because the same structural parameters were obtained independent of thermodynamic histories, we suggest that the observed FLL is an equilibrium hexatic and not a vestige of a high-temperature hexatic liquid state. Lastly, analyses of the fraction of twisted bonds and the slope of the long wavelength tail of $S_2(q_\perp)$ provide a lower bound to the temperature at which vortex relaxation is frozen in the bulk.

We would like to thank D.R. Nelson for helpful discussions. C.M.L. acknowledges support of this work by the Materials Research Science and Engineering Center Program of the National Science Foundation (Grant No. DMR 9400396) and the National Science Foundation under Award No. DMR-9306684.

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