Strong Vortex Liquid Correlation from Multiterminal Measurements on Untwinned $YBa_2Cu_3O_{7-\delta}$ Single Crystals

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Multiterminal transport measurements indicating a strongly correlated vortex liquid above the first order melting transition in untwinned $YBa_2Cu_3O_{7-\delta}$ single crystals are presented. The correlation is found to be surprisingly stable for different weak variations of point disorder. We investigate the differences between intervortex and intravortex correlations in different contact geometries. The results are in agreement with the local conductivity model, with zero resistivity along straight vortex line segments.

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The question of vortex correlation above the first order melting transition (FOT) in high-T_c superconductors is of great importance for the understanding of the field-temperature (H-T) phase diagram. In particular, the existence and extension of the vortex line liquid state is still under debate. Early multiterminal measurements on $Bi_2Sr_2CaCu_2O_x$ (Bi-2212) single crystals with the pseudo flux-transformer configuration revealed no signs of vortex correlation in the vortex liquid [1,2]. Similar measurements on twinned $YBa_2Cu_3O_{7-\delta}$ (YBCO), however, showed that the low temperature part of the vortex liquid could be correlated over the sample thickness along the magnetic field direction [3–6]. For clean, untwinned, single crystals, the solid-to-liquid transition is of first order [7,8], and the phase boundary marked by a sharp step in the resistivity at the melting temperature T_m. For such YBCO crystals, López *et al.* reported that the vortex correlation along the field direction was lost simultaneously with the correlation between different vortices [9]. This led to the general observation that twin boundaries and other correlated disorder (aligned with the field direction) act as to increase the correlation along vortex lines [9–12]. Later, Righi *et al.* estimated the upper limit of the vortex correlation length in untwinned YBCO to be a few μm just above T_m [13]. The intrinsic vortex state above T_m in untwinned crystals has thus remained a matter of discussion.

The relevance of point disorder for the vortex liquid behavior is still unclear. Signs of the FOT are readily suppressed by a sufficiently large amount of point disorder, most easily introduced by electron or proton irradiation [14,15]. The use of less pure powders for the crystal growth gives a similar effect [16]. With a lower dose of electron irradiation, it was shown that point disorder favors the vortex liquid phase and thus facilitates line deformation and wandering [17]. The observation that point disorder counteracts the localization effect of twin boundaries is in agreement with this result. Such an effect of point disorder was inferred from a study of proton irradiated, twinned YBCO in the flux transformer geometry, with a loss of vortex correlation after irradiation as the result [18].

In this paper, we present multiterminal measurements on untwinned YBCO which indicate a strongly correlated vortex liquid well above the first order melting transition. We connect the vortex correlation strength to the resistivity component along the applied magnetic field. The relation between this resistivity component and various kinds of disorder is discussed.

Single crystals of YBCO were grown in Y_2O_3-stabilized ZrO_2 crucibles using a standard self-flux method [19]. Care was taken to diminish the concentration of point defects by using high-purity starting powders (99.999% $BaCO_3$, 99.999% CuO, and 99.99% Y_2O_3). After oxygenation at 400 °C for 5–6 days in O_2, around 10% of the thin crystals were naturally untwinned (as observed by polarized light microscopy) and selected for further use. Contacts were prepared with epoxy silver paint and cured for 5–30 min under similar conditions as the oxygenation, resulting in contact resistances typically below 1 Ω. The selected samples had zero resistance T_c slightly above 92 K, transition widths below 0.3 K, and thicknesses $t = 10$, 33, and 53 μm. A dc picovoltmeter (EM Electronics) with noise level below 0.2 nV was used as a preamplifier for all measurements.

Our main result is shown in Fig. 1 for the sample of intermediate thickness. The other samples showed consistent behavior, differing only in properties related to the thickness as discussed below. Each pair of curves was recorded during a single, slow temperature sweep while scanning the current between the two contact pairs. Sharp melting steps are clearly seen for magnetic fields ranging from 0.5 to 8 T. For a range above T_m no difference can be resolved in the measured voltage V_{23} for the two current paths (I_{14} and I_{58}), in clear contrast to the measurements by López *et al.* [9,10]. If we assume that vortex motion is the cause of dissipation in the vicinity of T_m, we are led to conclude that the vortices in our case are extending from top to bottom even above T_m, moving with a uniform velocity [4,6].

To investigate if the observed correlation may be caused by columnar disorder, such as microscopic twin planes, we studied the behavior as a function of magnetic field direction; see Fig. 2. It is seen that any correlated disorder

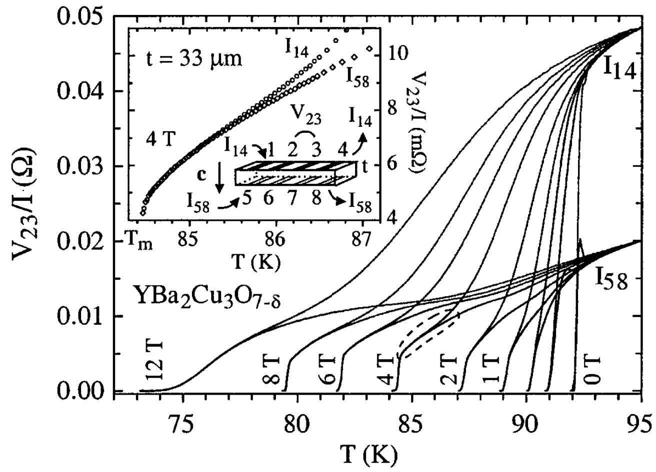

FIG. 1. V_{23}/I_{14} and V_{23}/I_{58} as a function of temperature for various magnetic fields **H** parallel to the c axis. Low-field, unlabeled curves correspond to $\mu_0 H = 0.5$ T and 0.2 T. The current $I = 0.1$ mA is flowing (mainly) along the a axis. Sample dimensions are $(l \times w \times t) = (0.45 \times 0.09 \times 0.033)$ mm^3. Inset: Contact configuration and an enlargement of the 4 T curves as indicated (only every fourth datapoint is shown).

along the c axis can be disregarded, since strong vortex correlation is still found far from the c axis. This also excludes any possible stabilizing effects from the crystal edges.

When going closer to the b axis, on the other hand, a change in the correlation characteristics seems to set in for some angle in the range $15° < \theta < 30°$. This is natural, since, for small angles, the probed correlation is mainly that between neighboring vortices. Perfect intervortex correlation could hardly be expected in a line liquid regime. Indeed, the critical angle below which vortices cannot extend from the top to the bottom surface is $\theta_{cr} = 19.7°$ for

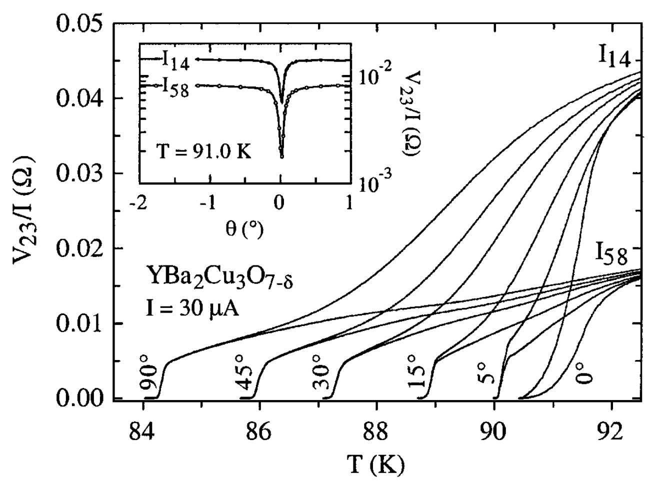

FIG. 2. V_{23}/I as a function of temperature with $\mu_0 H = 4$ T applied at different angles θ. The angle θ is defined from the b axis and lies in the bc plane. Inset: For $\theta < 0.2°$ intrinsic pinning dominates, and all signs of melting are suppressed.

the discussed sample, in good agreement with the observed behavior.

With the magnetic field applied exactly in the ab plane intrinsic pinning comes into play and the melting transition is replaced by a continuous transition. This occurs for a very narrow range of angles; see the inset in Fig. 2. It is interesting here that the top to bottom voltage ratio (i.e., with I_{14} and I_{58}) *increases* with decreasing temperature for fields in the plane ($\theta < 0.2°$), while it normally decreases with decreasing temperature. This increase is equivalent to an increased resistive ratio ρ_c/ρ_{ab} and is consistent with vortices flowing more easily along than perpendicular to the planes. (Vortex flow along the a axis is caused by the c-axis current for **H** along the b axis and affects the c-axis resistivity.)

It seems that all the above observations can be explained by simple Lorentz force arguments and local conductivity. With straight vortices, a current flowing along the field direction does not give rise to any Lorentz force, and consequently the longitudinal resistivity (the resistivity along the magnetic field) should be zero. For $\mathbf{H} \parallel c$, equal current will flow at top and bottom surfaces. With increasing temperature, the vortices soften and the longitudinal resistivity increases, causing the current to flow closer to the current surface. This effectively decreases the observed correlation.

While such a description appears to be sufficient, it has been suggested that nonlocal resistivity (the appearance of an electrical field at a point **r** caused by a current flowing at $\mathbf{r}' \neq \mathbf{r}$) has to be considered when interpreting multiterminal experiments [3,20–24]. To investigate this, we assumed local conductivity and determined the resistivity components $\rho_\parallel$ and $\rho_\perp$ (parallel and perpendicular to **H**) by solving the Laplace equation through a finite difference method (FDM) analysis [25] of the 4 T data in Fig. 1; see Fig. 3. Array sizes from 32×136 elements were used and checked to give consistent results. With nonlocal contributions, one would expect discrepancies between analyses with different current paths. We could, however, see no clear signs of nonlocality but found good agreement between the calculated voltages from the main case (solid lines) and the measurements with current I_{15}, as shown in the upper inset in Fig. 3.

A Lorentz force description is further supported by the great resemblance between the resistivities in Fig. 3 and the corresponding measurements of the Lorentz force contribution by Kwok *et al.* for **H** in the ab plane [26]. This indicates a significant flux flow contribution to $\rho_\perp$. The absence of a macroscopic Lorentz force for currents along the magnetic field does not mean, however, that the longitudinal resistivity should be zero. Instead, $\rho_\parallel$ will be closely related to the degree of *local* vortex alignment (misalignment) with the field direction, suitably expressed in some temperature dependent vortex correlation length l_c.

The temperature T_{th} at which a clear difference between voltages with current I_{14} and I_{58} can be resolved

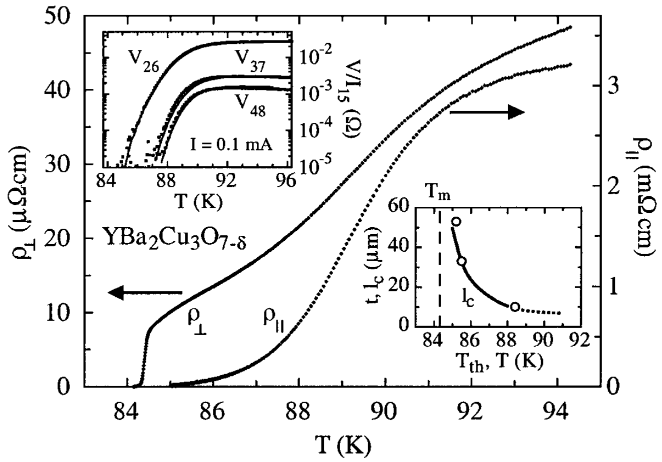

FIG. 3. Resistivities $\rho_{\parallel}$ (ρ_c) and $\rho_{\perp}$ (ρ_a) for $\mu_0 H = 4$ T along the c axis, as found from a FDM analysis of the multiterminal data in Fig. 1, assuming local conductivity. Upper inset: Measured voltages V_{26}, V_{37}, and V_{48} for current I_{15}. The solid curves show the expected response from FDM, using the already obtained resistivities from the main figure (with current I_{14}, etc.). Lower inset: Symbols mark the location of T_{th} for the investigated samples. The curve shows the calculated temperature dependence of l_c, using resistivities from the main figure. T_{th} is defined from the condition $V_{23}(I_{58})/V_{23}(I_{14}) = 0.99$ and l_c is assumed to equal t at T_{th}.

is found to be thickness dependent with correlation over the sample thickness t extending to higher temperatures for thin samples, as shown in the lower inset in Fig. 3 for $\mu_0 H = 4$ T. This clearly shows that Fig. 1 should *not* be interpreted as evidence of a line vortex liquid transforming into a decoupled pancake vortex gas at some specific transition temperature. Instead, the thickness dependence naturally arises from the combination of finite longitudinal resistivity, sample geometry, and definition of T_{th}. Recent muon spin rotation measurements on the more anisotropic Bi-2212 compound indicate a decoupling transition above T_m [27]. It is presently not clear to what extent these measurements are compatible with our results of a more continuous transition.

Since there is a close connection between $\rho_{\parallel}$ and l_c one could say that T_{th} corresponds to the temperature where l_c becomes comparable to t. Let us therefore further discuss the temperature dependence of l_c. Zero resistivity along the field direction is equivalent to $l_c \rightarrow \infty$, which could occur only below T_{th} of any chosen, finite thickness sample. Interestingly, it appears that $\rho_{\perp}$ and $\rho_{\parallel}$ may both become nonzero at T_m. This indicates that the vortex liquid is not perfectly correlated but that the difference between $V_{23}(I_{14})$ and $V_{23}(I_{58})$ goes continuously to zero when T approaches T_m. It is, however, easy to numerically calculate $l_c(T)$ by applying the resistivities in Fig. 3 to FDM models of samples with varying thickness, since each t gives a certain T_{th}. This relation is shown in the lower inset in Fig. 3.

Although l_c is useful for picturing the strength of vortex correlation, it merely describes but does not explain the longitudinal resistivity. If the vortices were all straight, parallel rods the longitudinal resistivity should indeed be zero. Thermal fluctuations, however, create vortex segments which are no longer aligned with the main field direction and thus locally feel the Lorentz force. The occurrence of such transversal vortex segments should depend on the vortex stiffness and vortex interactions but also on any presence of disorder, making the problem very complex.

To obtain a first estimate of the situation in the presence of point disorder, let us study the spatial configuration of a vortex with local tilt modulus $g \sim \varepsilon_0$, where $\varepsilon_0 \simeq \phi_0^2/4\pi\mu_0\lambda^2$ is the vortex line energy. Disregarding any confinement from vortex interactions in the liquid, the energy for a kink is $E = gr^2/2l$, which, after thermal averaging gives us $\langle r^2 \rangle = 2k_B T l/g$. Here $r(l)$ is the transverse vortex deflection for a longitudinal length l. The straightness of a vortex is often expressed in terms of the entanglement length l_z which is found by putting $\langle r^2 \rangle = a_0^2$ [28]. When taking the effect of disorder into account, however, the length l should assume the optimal value l_{opt} that minimizes the total energy. Since each kink has the freedom to thread one point defect, the optimal condition (for equally strong defects) should be given by $\pi \langle r^2 \rangle l_{\text{opt}} = 1/c$, where c is the defect concentration. The distance between kinks thus decreases with increasing c, following $l_{\text{opt}} \propto c^{-1/2}$. The occurrence of thermally induced, transversal vortex segments can finally be expressed through the average tilt angle φ;

$$\tan\varphi = \frac{\langle r^2 \rangle^{1/2}}{l_{\text{opt}}} \sim \left(\frac{k_B T}{g}\right)^{3/4} c^{1/4}. \qquad (1)$$

Thus line wandering increases with temperature and increasing amount of point disorder and diverges at T_c where g goes to zero. The effect of disorder, according to Eq. (1), is an increased resistivity $\rho_{\parallel}$. This increase should move T_{th} down in temperature and thus decrease the correlation, as was actually observed in [18].

We have inquired into the reasons for the striking differences between our observations and those of López *et al.* [9,10] by investigating samples with different weak disorder structure. Thus we attempted to reproduce a decorrelated vortex liquid in our crystals, while retaining the first order melting transition. Since oxygen vacancies are the most dominating defects in YBCO, we raised the annealing temperature to 450 °C, increasing the oxygen defect concentration. This reduced the upper critical point of the FOT from $\mu_0 H_{\text{cp}} \approx 8$ T to $\mu_0 H_{\text{cp}} \approx 5$ T, but the correlation strength remained unaffected. We also investigated the importance of high purity powders for the crystal growth by using less pure $BaCO_3$ (99% instead of 99.999%). In this case, only weak signs remained of the FOT, still without destroying the vortex correlations.

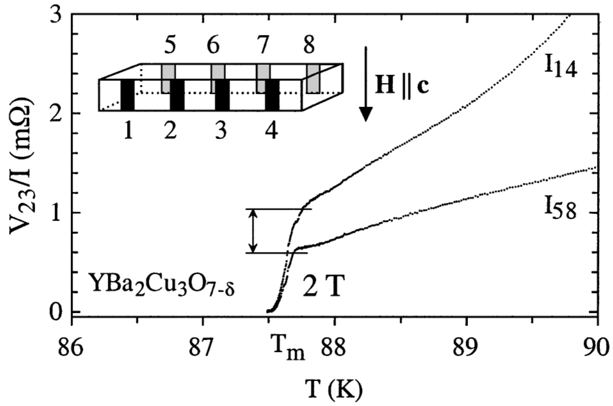

FIG. 4. V_{23}/I_{14} and V_{23}/I_{58} as a function of temperature for $\mu_0 H = 2$ T ($\mathbf{H} \parallel c$). Note the different contact geometry. No correlation is found in the liquid, and voltages even differ just above T_m as marked in the figure. This contact configuration is well suited for studying intervortex correlations and possible nonlocalities.

In a third experiment, we exposed the present samples to electron irradiation with total doses of 5×10^{16} cm^{-2} and 10^{17} cm^{-2} using a pulsed 3 MeV microtron. These doses are low enough not to destroy the FOT but, nevertheless, substantially increase the concentration of point defects, as reflected in a small decrease ($\approx$0.3 K) in T_c. Any reduction of correlation strength could not be found in this case either. A possible explanation is that only strong enough defects/clusters would facilitate a decorrelated vortex system. This possibility is supported by the observed differences in pinning characteristics between point defects from electron irradiation and clusters induced by proton irradiation [29].

We have further investigated vortex correlations with current and voltage contacts in the $ac(bc)$ planes. Keeping the field directed along the c axis, this allows intervortex correlations to be studied. It is interesting to note that the vortex correlation in this geometry is lost at the melting, and that, in addition, the "top" and "bottom" voltages differ already at T_m. The result is illustrated in Fig. 4. We explain this by the absence of current along the field direction in this geometry, so that all current components will yield nonzero Lorentz forces. Figure 4 also provides a hypothetical explanation for previous observations of an uncorrelated or just weakly correlated vortex liquid in the standard configuration. Using too wide samples may lead to unknown effective contact positions. If, e.g., the top contacts are located closer to one edge of the sample while the bottom ones are closer to the other, measurements would mimic a loss of correlation.

In summary, we have presented flux transformer measurements which indicate a highly correlated vortex liquid in clean, untwinned, YBCO single crystals. Microtwins and other correlated disorder along the c axis were ruled out by studying the vortex correlation as a function of the direction of the magnetic field. Weakly varied point disorder was found not to affect the vortex correlation. The results were discussed in terms of local conductivity, the importance of the Lorentz force, and the interplay between vortex tilt and point disorder.

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