Latent-heat and non-linear vortex liquid at the vicinity of the first-order phase transition in layered high-$T_c$ superconductors

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In this work we revisit the vortex matter phase diagram in layered superconductors solving still open questions by means of AC and DC local magnetic measurements in the paradigmatic Bi2Sr2CaCu2O8 compound. We show that measuring with AC magnetic techniques is mandatory in order to probe the bulk response of vortex matter, particularly at high-temperatures where surface barriers for vortex entrance dominate. From the $T_{FOT}$-evolution of the enthalpy and latent-heat at the transition we find that, contrary to previous reports, the nature of the dominant interlayer coupling is electromagnetic in the whole temperature range. By studying the dynamic properties of the phase located at $T \gtrsim T_{FOT}$, we reveal the spanning in a considerable fraction of the phase diagram of a non-linear vortex phase suggesting bulk pinning might play a role even in the liquid vortex phase.

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I. INTRODUCTION

One of the main features of the phase diagram of vortex matter in layered high-temperature superconductors is the occurrence of a first-order transition at $T_{FOT}$ due to the relevance of thermal fluctuations at temperatures close to the critical, $T_c$. The layered nature of vortex matter in these extremely anisotropic materials plays a key role on the location of $T_{FOT}$ and on the temperature-evolution of the main thermodynamic magnitudes at the transition. When applying a field along the sample c-axis vortices are actually a stack of pancake vortices lying in CuO planes, coupling between layers via electromagnetic and Josephson interactions. In the case of the paradigmatic layered Bi2Sr2CaCu2O8 compound the first-order transition separates a solid phase at low temperatures and a liquid or decoupled gas of pancake vortices with reduced shear viscosity at high temperatures.

The solid phase presents irreversible magnetic behavior ascribed to bulk pinning and surface barriers, each of them dominating at different temperature and measuring-time scales. Direct imaging of vortices in pristine Bi2Sr2CaCu2O8 reveals the vortex solid has quasi long-range positional order. Josephson-plasma-resonance measurements indicate the $T_{FOT}$ transition line corresponds to a single-vortex decoupling process between pancake vortices from adjacent layers within the same stack. The first-order line is then a single-vortex transition that depends, at best, on the density of the surrounding vortex matter. Therefore the relative importance of the two types of interaction between pancake vortices determines the temperature-evolution of the enthalpy and latent-heat, $\Delta s_{T_{FOT}}$ of the transition.

The main thermodynamic magnitudes entailed in this transition, namely the entropy-jump per pancake vortex, $\Delta s$, and the enthalpy that is proportional to the observed jump in local induction, $\Delta B$, have been investigated experimentally as well as theoretically. A previous report claims that for $T_{FOT} \sim T_c$ the latent-heat of the transition is not satisfactorily described by considering the electromagnetic interaction as the dominant coupling mechanism between pancakes of adjacent layers. The same work proposed then the existence of a crossover to a high-$T_{FOT}$ regime where interlayer coupling is dominated by the Josephson interaction.

Indeed, at such high temperatures the magnetic response of Bi2Sr2CaCu2O8 vortex matter is dominated by geometrical or Bean-Livingston surface barriers. Nevertheless, a previous work reported on the plausibility of bulk pinning playing a role in vortex dynamics even at temperatures larger than $T_{FOT}$, within the vortex liquid phase. Bulk pinning do play a dominant role for temperatures below 0.6$T_c$, and the first-order transition is observed as the so-called second-peak effect or order-disorder transition, $H_{SP}$, which region of the transition is detected through an increase of the width of DC hysteresis loops, or as a minimum in AC magnetization loops, both occurring at $H_{SP}$, independently of frequency.

Therefore several issues in the vortex phase diagram of layered high-temperature superconductors remain still open to discussion. Most of the attempts in order to settle down this debate suffered from comparing data obtained with experimental techniques lacking on the proper sensibility in order to ascertain the relative importance of bulk effects, surface barriers, and sample inhomogeneities. In this work, we apply AC local-magnetic-measurement techniques with the aim of providing an
Figure 1. DC and AC magnetic hysteresis loops of Bi₂Sr₂CaCu₂O₈ sample measured at 75 K. Top: ascending and descending branches of a DC hysteresis loop. Insert: zoom-in of the DC loop at the vicinity of the first-order transition entailing a B-jump (see black arrow). Red arrows indicate the ascending and descending branches. Bottom: AC transmittivity loops with paramagnetic peaks fingerprinting the first-order transition field \( H_{\text{FOT}} \). The loops were measured with ripple fields of 0.9 Oe rms in amplitude and frequencies of 7.5 and 437 Hz. Blue arrows indicate the ascending and descending branches.

accurate description of the physics entailed in the first-order vortex phase transition in Bi₂Sr₂CaCu₂O₈. We rule out the proposal of a crossover between electromagnetic to Josephson coupling taking place at low fields (high-temperatures), suggesting the dominant interaction between pancakes at \( T_{\text{FOT}} \) is always of the same nature. In addition, we report on a non-linear liquid vortex phase spanning a considerable fraction of the high-temperature region of the phase diagram. The large extent of this phase challenges the idea of a vortex liquid being a linear phase.

II. EXPERIMENTAL

The optimally-doped Bi₂Sr₂CaCu₂O₈ single-crystal studied in this work \( (T_c = 90\, \text{K}) \) was grown by means of the traveling-solvent floating zone technique. The local magnetization of the \( 220 \times 220 \times 30 \, \mu \text{m}^3 \) sample was measured with a microfabricated 2D electron gas Hall-sensor array. The eleven-probes array was photolithographically fabricated from GaAs/AlGaAs heterostructures and having each sensor an active area of \( 6 \times 6 \, \mu \text{m}^2 \).

Local AC magnetization measurements were performed applying DC and ripple magnetic fields parallel to the \( c \)-axis, \( H \) and \( H_{\text{ac}} \) respectively. DC magnetic hysteresis loops are obtained by measuring the magnetization, \( H_s = B - H \), when cycling \( H \) at fixed temperatures. In the AC measurements the ripple field has an amplitude of 0.9 Oe rms and frequencies ranging from 1 to 1000 Hz.

The first and third-harmonics of the AC magnetic induction are simultaneously measured by means of a digital-signal-processing lock-in technique. The in-phase component of the first-harmonic signal, \( B' \), is converted to the transmittivity, \( T' \), a magnitude extremely sensitive to discontinuities in the local induction associated to first-order magnetic transitions. A non-negligible magnitude of the third harmonic signal, \( |H_{\text{ac}}| \), indicates the appearance of non-linearities in the magnetic response. The onset of \( |H_{\text{ac}}| \) on cooling is considered as the irreversibility temperature or field, \( T_{\text{IL}} \) or \( H_{\text{IL}} \).

In order to track the \( H - T \) location of the first-order transition and irreversibility lines in Bi₂Sr₂CaCu₂O₈ vortex matter, we perform two types of measurements: Isothermal DC and AC hysteresis loops, and temperature-evolution of \( T' \) and \( |H_{\text{ac}}| \) on field-cooling at various magnetic fields (see Figs. 1 and 2). We discuss the magnetic response of our sample at three characteristic temperature regimes of the vortex phase diagram.

III. RESULTS AND DISCUSSION

In the high-temperature regime, \( T \gtrsim 0.83\, T_c \), the first-order transition is manifested in \( T' \) as a prominent para-
magnetic peak developing at the same $H_{FOT}$ than the jump in $B$ detected in DC hysteresis loops. The top panel of Fig. 1 shows a typical two-quadrant DC loop observed in this temperature regime indicating surface barriers dominate the vortex entrance to the sample. Closer inspection of the DC data in the vicinity of 80 Oe reveals a $H_s$ jump with similar height for both, ascending and descending branches. This feature is the fingerprint of the first-order transition $H_{FOT}$ in the high-temperature region. The bottom panel of Fig. 1 shows that in the AC loops this transition is detected with improved resolution: Paramagnetic peaks appear at the same fields where the $H_s$ jumps are measured. At lower fields the shielding capability is increased. On increasing frequency, the system enhances its shielding capability manifested as a $T'$ decrease. The field-location of the paramagnetic peak in AC loops, $H_{FOT}$, is frequency-independent.

Figure 2 (a) shows a set of AC magnetic data for applied fields up to 100 Oe. The paramagnetic peak, observed in $T'$ vs. $T$ measurements at $T_{FOT}$, shifts towards lower temperatures on increasing field. The peaks are sharp with an amplitude that slightly decreases on increasing field. Figure 2 (b) shows that at fields larger than 100 Oe shielding currents develop in the sample prior to the appearance of the paramagnetic peak. Nevertheless, the peak can be clearly detected in AC magnetic measurements up to 200 Oe.

The enthalpy of this first order transition is proportional to the height of the step in the magnetic induction at $T_{FOT}$. This magnitude can be obtained from the magnificity value at the transition field considering that $T' = B'/H_{ac}$ and according to Ref. 22

$$T' = 1 + \frac{2\Delta B}{\pi H_{ac}}$$  \hspace{1cm} (1)

for small ripple fields $H_{ac}$. Figure 3 (a) shows the $\Delta B$ evolution with $T_{FOT}/T_c$ obtained from applying Eq. (1) to $T'(T)$ data at different fields. We found a linear increase of $\Delta B$ up to temperatures $T_{FOT} \sim T_c$ within the error bars. These findings are in contrast to the seminal work of Ref. 2 reporting that for $FOT \geq 0.93T_c$, the jump in magnetic induction decreases dramatically down to 25% at $T_c$ (see Fig. 3 (a)). This result was interpreted within the framework of a crossover between an electromagnetic to Josephson coupling of pancake vortices on decreasing field (increasing $T_{FOT}$). This interpretation was proposed by the theoretical work of Ref. 13 that provided the functionality of $\Delta B(T)$ at the first-order transition.

A linear evolution of $\Delta B$ can be explained by considering that the inter-layer coupling is dominated by electromagnetic interactions between weakly bound pancake vortices undergoing large thermal fluctuations. Taking into account the dispersive electromagnetic line tension of vortices and the Lindemann criterion for vortex melting in this case

$$\Delta B = \frac{\mu k_B T_{FOT}}{\phi_0 d}$$  \hspace{1cm} (2)

where $k_B$ is the Boltzmann constant, $\phi_0$ the flux quantum, $d \approx 15.4 \mu m$ the CuO planes inter-layer distance and $\mu'$ a numerical constant. The latter constant depends on the Lindemann number for a given material and up to our knowledge there is no report of its quantitative value in the case of Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. The linear $\Delta B$ data obtained in our sample is reasonably well fitted with Eq. (2) for $\mu' = 0.6$.

The entropy-jump per vortex and per CuO layer entailed in the first-order transition can be obtained from $\Delta B$ data by means of the thermodynamic Clausius-Clapeyron relation

$$\Delta s = -\frac{\phi_0 d}{4\pi B_{FOT}} \frac{dH_{FOT}}{dT}.$$  \hspace{1cm} (3)

Figure 3 (b) shows that the $\Delta s$ data of our sample diverges close to $T_c$, as also reported in the previous work of Ref. 2. In order to explain the measured $T_{FOT}$ evolution of $\Delta s$ in terms of the nature of the inter-layer coupling that dominates in the system, Ref. 13 proposes that for electromagnetic interactions,
\[
\Delta s = \frac{\mu}{\pi (1 - (T_{FOT}/T_c))} d
\]

with \(\mu\) a material-dependent constant. Our \(\Delta s\) data is reasonably well fitted with the expression of Eq. 4 in the whole temperature range close to \(T_c\). Therefore, our results are in discrepancy with the data and interpretation provided in Ref. 2, where the divergent \(\Delta s\) was not satisfactorily fitted with Eq. 4. However this might be associated to the fact that in Ref. 2 \(\Delta B\) decreases close to \(T_c\). The difference between our data and those of Ref. 2 can have origin in our AC measurements being more sensitive to bulk currents than the DC measurements reported in Ref. 2, more affected by surface-barrier effects. However, inhomogeneities in the seminal sample can not be ruled out as possible sources of this discrepancy.

Another remarkable phenomenology in the high-temperature region of the phase diagram is that the irreversibility temperature, \(T_{IL}\), identified from the onset of the third-harmonic signal on cooling, develops at temperatures larger than those associated to the paramagnetic peak (see the low panel of Fig. 2(b)). This indicates the existence of a phase region with non-linear vortex dynamics at fields exceeding \(H_{FOT}\). A previous work reported a qualitatively similar phenomenology by means of magneto-optics techniques and proposed that bulk pinning still plays a role in the high-temperature liquid vortex phase. Here we study in detail the quantitative spanning of this non-linear vortex liquid as a function of frequency.

Figure 4 shows that as higher applied fields the shielding of the AC field starts at significantly higher temperatures than the occurrence of the first-order transition. The temperature of this phase increases with frequency and at high fields (300 Oe) can be as large as 20% of \(T_c\) for frequencies of the order of 1 kHz. This phenomenon might have origin from a residual effect of pinning, or Bean-Livingston barriers on the high-temperature liquid phase.

The vortex phase diagram of Fig. 4 also shows the connection between the low-temperature regime of the first-order transition, \(H_{SP}\), and the high-temperature regime \(H_{FOT}\) for \(0.4 \lesssim T/T_c \lesssim 0.5\). The \(H_{SP}\) transition field can be obtained from DC magnetic hysteresis loops as shown in Fig. 4(a) for three different temperatures. In these curves the local minimum (maximum) of the ascending (descending) branch becomes more evident on cooling. The loops occupy two field-quadrants and noticeably increase their width on cooling. This figure also indicates that the \(H_{SP}\) field is hard to detect from DC magnetic loops for temperatures larger than 33 K. This is due to a technical limitation of the DC technique that lacks resolution in order to measure bulk transitions when surface barriers dominate the magnetic properties.

We therefore use the AC hysteresis loop technique in order to track the second-peak transition in the intermediate temperature-regime. The AC transmittivity reflects the dimensionless normalized sustainable-current density, \(J = j(f)/H_{ac}\), from \(J = (1/\pi) \arccos(2T'/1)^{26}\). This formula was derived for an AC penetration in the Bean critical regime, an assumption that seems to be valid in view of the results presented in Ref. 15. A low-temperature AC loop at 35 K is shown in Fig. 4(b) depicting local minima in both, the ascending and descending branches. This minima can be ascribed to the second-peak transition \(H_{SP}\) since a minimum in \(T'\) corresponds to a maximum in the bulk \(J\). The \(T'\) signal evolves in a different manner for the high- and intermediate-temperature regimes. For temperatures larger than 0.66 \(T_c = 58\) K, the transmittivity presents paramagnetic peaks, developing at the flanks of the central depletion (see Fig. 2). For intermediate temperatures a sudden jump of \(T'\) is detected, as for instance in the AC loops measured at 40 and 45 K (see Fig. 4(b)). This step-like feature, manifested at a field \(H_{step}\), implies a frequency-independent drop of the magnetic hysteresis, consistent with the change in the width of the DC loops, and indicating that this transition is governed by the local value of \(B\) rather than \(H_{ac}\). The jump in \(T'\) is related to the sudden change in shielding currents with the sample becoming more transparent to the penetration of the AC ripple field at larger \(H\). Similarly, the height of the steps in \(T'\) decreases on increasing temperature.

The crossover temperature for the detection of the second-peak, or of the step-like feature, is time/frequency dependent. Figure 4 presents the evolution of the normalized sustainable-current density ex-
Vortex matter in the low-intermediate temperature regime. (a) DC loops: the transition field $H_{sp}$ is taken at the midpoint between the onset and the full development of the local minimum in the ascending branch. (b) Transmittivity AC loop measured with a ripple field of 0.9 Oe rms and 17 Hz parallel to $H$. The $H_{sp}$ field is obtained similarly as in DC loops (see arrow). The field location of the step-like feature, $H_{step}$, is taken at half the step height in the ascending field branch.

Figure 6. Normalized sustainable-current density $J$ as a function of applied field at 40 K. The frequency dependence of $J$ is obtained from AC magnetization loops measured with a ripple field of 0.9 Oe rms and frequencies ranging 1.5 to 985 Hz. A higher electric field in a transport $I(V)$ measurement. Since the $I(V)$ curves just below and above the first order transition cross — with the electric field in the high-field phase being larger than that in the low-field phase for the low-current density limit, and vice-versa for the high-current density limit —, varying the working point (by tuning the frequency) leads to either a step-like-behavior of the screening current (at low electric fields) or a peak-like curve (at high electric fields). The energy barriers for flux creep have $U(J)$ and $E(J)$ curves with different functionalities for fields larger or smaller than the transition one. On varying field close to the transition these curves cross, and the phase transition produces a discontinuous change on the electrodynamics of vortex matter. Detecting the transition with a high electric field (short measurement times) leads to an enhancement of shielding currents with increasing field, whereas with a low electric field (long times) a sudden decrease is observed.

IV. CONCLUSIONS

We revisited the vortex phase diagram in pristine Bi$_2$Sr$_2$CaCu$_2$O$_8$ vortex matter by applying AC and DC local magnetization techniques and reveal a new phenomenology regarding the thermodynamic properties and the nature of the first-order phase transition line. We show that in this case AC local-magnetic techniques probe the physics at shorter time-scales, having access to currents flowing in the bulk sample with improved sensitivity than DC measurements. As a consequence, we were able to track down the connection between the low $H_{sp}$ and high $H_{FOT}$ temperature regions of the first-order vortex phase transition in an intermediate temperature-regime, covering the whole temperature-
range of the transition. By combining simultaneous measurements of the first and third-harmonics of the magnetization we were also able to detect the existence of a non-linear vortex liquid phase at $T < T^c_{IL}$ spanning a vortex phase region that at 300 Oe can be as large as 20 K. The width of this non-linear liquid region increases on increasing frequency. From AC magnetic measurements of the first and third-harmonics of the magnetization we were also able to detect the existence of a non-linear vortex liquid phase at $T^c_{IL}$. By means of the thermodynamic Clausius-Clapeyron relation we estimated the entropy-jump per pancake vortex. We explain the temperature-evolution of the latent-heat of our sample considering solely the dominance of electromagnetic inter-layer coupling all along the $H^c_{FOT}$ transition. This is in clear contrast to the claim of Ref.\textsuperscript{2} that during the $H^c_{FOT}$ the dominant interlayer coupling changes nature on approaching $T^c$.

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