Paramagnetic ac susceptibility at the first-order vortex-lattice phase transition

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A sharp paramagnetic peak in the local ac susceptibility χ' at the first-order vortex-lattice phase transition in $Bi_2Sr_2CaCu_2O_8$ crystals is found. Observation of this peak establishes the thermodynamic nature of the transition and allows accurate evaluation of the equilibrium magnetization step height. The associated estimated entropy change Δs reaches values in excess of $6k_B$ per pancake vortex close to T_c. The local χ'' shows two independent dissipation peaks. The broader one is caused by the onset of irreversible magnetization, whereas the narrow one is due to hysteresis at the phase transition. [S0163-1829(96)50930-6]

Thermodynamic phase transitions of the vortex lattice in high-temperature superconductors (HTSC) have recently attracted significant scientific attention.[1–3] In particular the theoretical predictions of a possible first-order vortex-lattice melting transition[1,4,5] were intensively studied in $YBa_2Cu_3O_7$ (YBCO) crystals using resistive measurements.[6] Sharp transitions were also observed in mutual inductance measurements on $Bi_2Sr_2CaCu_2O_8$ (BSCCO) crystals[7,8] and in resistance measurements on nonuniformly irradiated BSCCO crystals.[9] Recently a thermodynamic confirmation of the first-order transition was obtained by revealing a step in magnetization in clean BSCCO (Refs. 10–14) and YBCO (Refs. 15,16) crystals using local[10,11] and global[12–16] dc measurements. Alternatively, the melting transition is commonly investigated using ac magnetization measurements.[2,17] In these studies the ac susceptibility dissipation peak χ'', accompanied by a *diamagnetic* dip in the in-phase susceptibility χ', is often interpreted as an indication of the melting transition and the associated premelting enhancement of the vortex-lattice pinning.[2,17–21] In contrast, in this paper we demonstrate that in BSCCO crystals the first-order vortex-lattice transition results in a sharp and very pronounced *paramagnetic* peak in χ' that was not reported previously. Furthermore, the small hysteresis associated with the first-order transition[6,10] results in an additional small sharp peak in χ''. This additional narrow peak, however, is not related to the regular and much broader χ'' peak that results from irreversible surface currents in BSCCO crystals. In addition, the thermodynamic interpretation of the dc magnetization step was recently questioned on the basis of torque magnetometry measurements in BSCCO.[22] The findings reported here clearly demonstrate the thermodynamic nature of the transition and confirm the dc magnetization results.

One of the unexpected features of the first-order vortex-lattice phase transition is the large entropy change at the transition in BSCCO crystals, as well as in YBCO crystals, reported by dc magnetization measurements.[10,14–16] This result currently has no adequate theoretical explanation. Our ac technique proves to be much more sensitive and allows an accurate evaluation of the magnetization step and entropy change very close to the critical temperature T_c where dc measurement is not reliable. The entropy change at the transition is found to attain values up to $6k_B$ per pancake vortex a factor of 3 larger than estimated previously. These extremely high values pose significant constraint and challenges for future theoretical developments.

Two high quality BSCCO crystals[23] (T_c=90 K) were studied. Crystal A was cut into a rectangular shape of $300\times240\times40$ μm^3 and was attached to an array of eleven 10×10 μm^2 Hall sensors fabricated photolithographically using GaAs/AlGaAs heterostructure.[10,24] Crystal B, $240\times110\times25$ μm^3, was studied using an array of 3×3 μm^2 sensors. Both dc and ac external magnetic fields, H_{dc} and H_{ac}, were applied parallel to the c axis of the crystals. The resulting position dependent local dc and ac fields, B_{dc} and B_{ac}, at the surface of the crystals were measured by the sensors as a function of the applied fields and temperature. The in- and out-of-phase fundamental frequency components B'_{ac} and B''_{ac} were measured simultaneously by lock-in technique.

Figure 1 shows the experimental local B'_{ac} in BSCCO crystal as a function of H_{dc} at three temperatures. The general behavior of B'_{ac} shows a gradual crossover from a com-

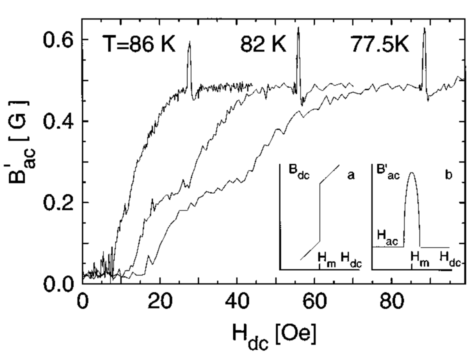

FIG. 1. The experimental B'_{ac} in BSCCO crystal B as a function of H_{dc} at 77.5, 82, and 86 K, H_{ac}=0.5 G rms, and f=37 Hz. The first-order phase transition is resolved as a sharp peak in B'_{ac} at $H_m(T)$. Insets: schematic behavior of the local B_{dc} (a) and the corresponding calculated B'_{ac} (b) as a function of H_{dc} in the vicinity of the phase transition.

pletely shielded diamagnetic Meissner state at low H_{dc} ($B'_{ac}=0$, $\chi'=-1/4\pi$) to an unscreened response at higher fields above the irreversibility line ($B'_{ac}=H_{ac}$, $\chi'=0$). The overall shape of the response is consistent with presence of geometrical barrier.[25] The astonishing feature of Fig. 1 is the sharp *paramagnetic* peak in the local susceptibility. The position of the peak shifts to higher fields with decreasing temperature following the temperature dependence of the first-order phase transition line derived from the dc measurements.[10] To the best of our knowledge this is a first report of such a unique paramagnetic response of χ' associated with vortex-lattice phase transition.

A first-order vortex-lattice phase transition results in a small magnetization step at the transition.[10–16,24,26–28] Inset *a* of Fig. 1 shows a schematic behavior of the local B_{dc} as a function of H_{dc} in the vicinity of the phase transition.[10,28] The field B_{dc} follows approximately the applied field (since the magnetization is small, $H_{dc}-B_{dc}\ll H_{dc}$) except at the transition H_m where a step of height ΔB in B_{dc} occurs. We now analyze the behavior of the ac response. At elevated temperatures the vortex-lattice phase transition in BSCCO occurs in the reversible magnetization region.[28] In this case the amplitude of the in-phase component of local ac field is given by $B'_{ac}=H_{ac}(dB_{dc}/dH_{dc})$. Therefore, away from the transition $B'_{ac}\simeq H_{ac}$ since $dB_{dc}/dH_{dc}\simeq 1$, whereas at the transition a sharp paramagnetic peak in B'_{ac} is present since dB_{dc}/dH_{dc} diverges. This is the origin of the observed B'_{ac} peak in Fig. 1. If the transition is smooth with characteristic width larger than the amplitude of the ac field H_{ac}, then the height of the peak is determined by dB_{dc}/dH_{dc} in the transition region. On the other hand, if the transition is sharp (as is the experimental case as shown below) with a width that is small compared to H_{ac}, then a straightforward Fourier analysis results in

$$B'_{ac}\simeq H_{ac}+\frac{2\Delta B}{\pi}\sqrt{1-\left(\frac{H_{dc}-H_m}{H_{ac}}\right)^2} \tag{1}$$

for $|H_{dc}-H_m|\leqslant H_{ac}$, where H_m is the value of the applied field H_{dc} at the transition. The calculated behavior of B'_{ac} is shown in inset b of Fig. 1. Note that in the peak region $B'_{ac}>H_{ac}$ and hence the response is *paramagnetic*. Moreover, since the height of the peak at $H_{dc}=H_m$ is independent of the amplitude of ac applied field, B'_{ac} can be significantly larger than H_{ac} at small ac fields.

Figure 1 clearly indicates that the ac technique results in a much more clear and decisive signal at the phase transition compared to the dc measurements.[10,11,28] We now analyze this χ' peak in more detail. Figure 2 shows B'_{ac} at the transition for various amplitudes of H_{ac} at T=80 K. From Eq. (1) we find that the height of the ac peak is given by $\Delta B\sqrt{2}/\pi$ independent of the amplitude of H_{ac}, whereas the full width at half maximum scales with H_{ac}, and is equal to $\sqrt{6}H_{ac}$ (the experimental B'_{ac} and H_{ac} are shown as rms rather than peak values which results in the $\sqrt{2}$ corrections). The data in Fig. 2 are consistent with both these predictions: the height of the peak is H_{ac} independent whereas the width grows linearly with H_{ac} and follows the predicted $\sqrt{6}$ very closely. This result indicates that the width of the phase transition is significantly narrower than the amplitude of the ac field, and that this method provides a very reliable measurement of ΔB. In contrast, if the phase transition would be broad, the width of the measured peak would be approximately independent of the amplitude of H_{ac} and the height of B'_{ac} peak, given by $H_{ac}(dB_{dc}/dH_{dc})|_{H_{dc}=H_m}$, would grow linearly with the amplitude of the ac field. In Ref. 22 it was suggested that the dc magnetization step is of artifactual origin related to the onset of irreversibility below the transition. We emphasize that onset of irreversibility results in a *diamagnetic* ac response. A sharp termination of irreversibility may cause an abrupt end of the diamagnetic signal but cannot turn it into a *paramagnetic* one. Moreover, irreversible magnetization results in an ac response that is usually significantly amplitude and frequency dependent, in contrast to our findings. The excellent agreement of our data with the calculated response clearly confirms the thermodynamic nature of the magnetization step.

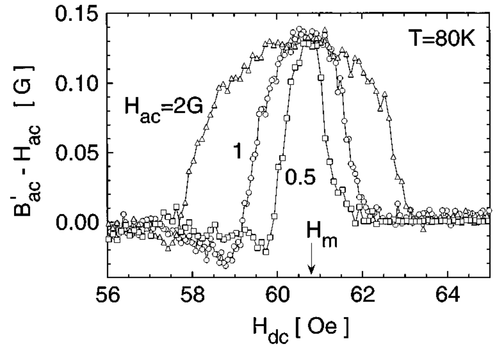

FIG. 2. The experimental $B'_{ac}-H_{ac}$ at the transition for various indicated amplitudes of H_{ac} rms ($T=80$ K, crystal A). The height of the peak is independent of amplitude of the ac field. The width of the peak scales as $\sqrt{6}H_{ac}$.

Another important feature of the vortex-lattice phase transition is described in Fig. 3. In a platelet crystal the field B_{dc} is not uniform even under equilibrium magnetization conditions and forms a dome-shaped profile across the sample.[29–31] Since B_{dc} is maximum in the center of the sample the melting transition occurs first in the central region and the liquid to solid interface moves gradually towards the

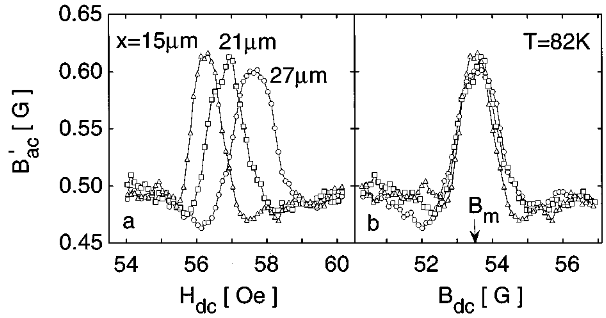

FIG. 3. (a) Local B'_{ac} peaks measured by three adjacent sensors 6 μm apart as a function of H_{dc} at $T=82$ K in crystal B (crystal center is at $x=0$). The value of the applied dc field at the phase transition $H_m(x)$ is position dependent due to dome-shaped field distribution $B_{dc}(x)$in a platelet crystal. (b) The same data plotted as a function of local B_{dc}. The first-order phase transition occurs at position independent thermodynamic B_m.

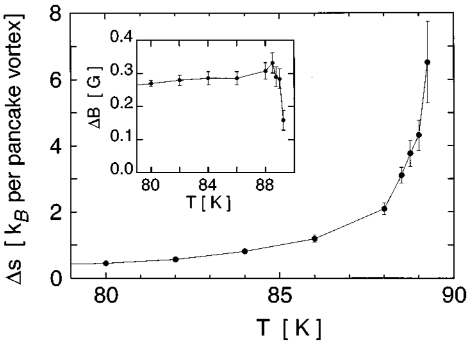

FIG. 4. The entropy change at the transition in the vicinity of T_c calculated per vortex per double CuO layer. Inset: temperature dependence of the dc induction jump at the phase transition ΔB calculated using Eq. (1).

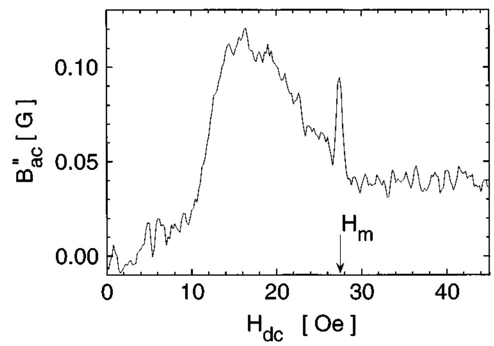

FIG. 5. The experimental B''_{ac} as a function of H_{dc} at $T=86$ K ($H_{\mathrm{ac}}=0.5$ G rms). The narrow dissipation peak corresponds to hysteretic first-order transition at H_m, whereas the broad one is due to the presence of irreversibility.

edges as H_{dc} is increased. This behavior is clearly presented in Fig. 3(a) showing the χ' peak measured by three adjacent sensors 6 μm apart. The phase transition occurs at about 1 Oe higher applied field at each of the shown locations starting from the central region and moving towards the edges. This behavior results in a much broader apparent transition width as measured by global magnetization.[13,14] Locally, however, the phase transition occurs at the same thermodynamic field $B_m(T)$ at all locations. This is demonstrated in Fig. 3(b) where the same data are plotted vs the local field B_{dc} measured by each of the sensors showing overlapping peaks. A similar picture is obtained when B'_{ac} is measured as a function of temperature. At $H_a=50$ Oe for example the three sensors of Fig. 3 show peaks that are progressively shifted by ~ 0.15 K to higher temperatures for sensors closer to the edge. This temperature shift is in agreement with the field shift shown in Fig. 3(a) taking into account the slope of $B_m(T)$ of about 6 G/K at $T\simeq 80$ K.[10] The consistency of the local response at various locations is an additional confirmation of the thermodynamic nature of the transition.

The above results demonstrate that this unique behavior of ac susceptibility can be used for accurate determination of ΔB and the entropy change at the transition ΔS. At intermediate temperatures our results confirm previously reported data;[10] however, the ac technique provides more precise quantitative data particularly in the vicinity of T_c, where dc measurements result in large error bars. Reliable information at high temperatures is of major importance since this temperature region displays the largest discrepancies with the existing melting[32–37] and decoupling[38–40] theories. Figure 4 shows the magnetization step ΔB and the entropy change per pancake vortex at the transition in the vicinity of T_c given by $\Delta s=-(d\phi_0/4\pi)(\Delta B/B_m)(dH_m/dT)$, where ϕ_0 is the flux quantum and $d\simeq 15$ Å is the interlayer spacing. The entropy change increases rapidly with temperature reaching values in excess of 6 k_B per vortex per double CuO layer, that are substantially higher than reported previously,[10] indicating a strongly first-order phase transition. Relatively high Δs values were recently reported also in YBCO crystals[15,16] although in this case $\Delta s(T_m)$ seems to drop rapidly in the vicinity of T_c in contrast to the apparently diverging behavior in BSCCO (Fig. 4).[10,14] Within the existing theoretical framework in the melting scenario Δs is predicted to drop monotonically with temperature $\Delta s(T)\approx(0.1d/\varepsilon)\sqrt{B_m(T)/\phi_0}$ where $\varepsilon\simeq 10^{-2}$ is the anisotropy parameter.[10,35] At 1 K below T_c, for example, $B_m\simeq 5$ G and the calculated $\Delta s\simeq 7\times 10^{-3}$ k_B. Our measured Δs is therefore about *three* orders of magnitude larger than expected. In the decoupling case Δs is predicted to be on the order of 0.5 k_B independent of temperature, so also in this scenario the measured Δs is an order of magnitude larger than estimated theoretically. Thus obviously our observation of diverging $\Delta s(T_m)$ close to T_c is incompatible with the existing theoretical predictions[10,26,27,32–40] and calls for further investigations.

Another interesting aspect of the first-order phase transition is hysteresis that may occur at the transition.[6,10,26,35] In the case of a small hysteresis, $\delta H<2H_{\mathrm{ac}}$, the analysis can be simplified by assuming two sharp transitions at $H_{\mathrm{dc}}=H_m\pm\delta H/2$ for the increasing and decreasing fields. The in-phase response in this case is modified only slightly compared with Eq. (1) (correction $\delta H/2$ is small),

$$B'_{\mathrm{ac}}\simeq H_{\mathrm{ac}}+\frac{\Delta B}{\pi}\left[\sqrt{1-\left(\frac{H_{\mathrm{dc}}-H_m+\delta H/2}{H_{\mathrm{ac}}}\right)^2}+\sqrt{1-\left(\frac{H_{\mathrm{dc}}-H_m-\delta H/2}{H_{\mathrm{ac}}}\right)^2}\right], \quad (2)$$

however, the hysteretic transition results in appearance of a sharp peak in the out-of-phase component

$$B''_{\mathrm{ac}}\simeq-\frac{\Delta B}{\pi}\frac{\delta H}{H_{\mathrm{ac}}} \quad (3)$$

at $|H_{\mathrm{dc}}-H_m|<H_{\mathrm{ac}}-\delta H/2$. Figure 5 shows the measured $-B''_{ac}$ as a function of H_{dc} at $T=86$ K. A sharp dissipation peak is observed at the phase transition. From Eq. (3) we evaluate the hysteresis width at the transition of $\delta H\simeq 0.26$ G. The hysteresis is usually ascribed to supercooling or superheating of the vortex lattice.[6,35] We find that δH depends on temperature and H_{ac}, and the microscopic origin of this hysteresis is not yet clear. The important conclusion from Fig. 5, however, is that there are *two independent* dissipation peaks. The broader peak is the usual dissipation peak observed near the irreversibility line, which in case of BSCCO at elevated temperatures is mainly due to geometrical and surface barriers and is not related to the vortex-lattice phase transition.[25] The new sharp peak in Fig. 5, on the other hand, occurs at $H_m(T)$ and is the result of the hysteretic first-order phase transition. We emphasize that this dissipation peak is not

related to the premelting ''peak-effect,''[20,21] since the latter is inevitably accompanied by a *diamagnetic* χ' dip in contrast to our *paramagnetic* peak. Note also that χ'' peak is position dependent like the χ' peak in Fig. 3, and therefore would not be readily detected in global measurements.

In conclusion, we have resolved sharp peaks in local χ' and χ'' at the first-order phase transition in BSCCO crystals. The paramagnetic χ' peak allows accurate evaluation of magnetization step at the transition ΔB. The associated entropy change Δs shows a diverging temperature dependence in the vicinity of T_c and reaches values significantly above 1 k_B per pancake vortex. The local χ'' shows two independent dissipation peaks. The usual broad peak occurs in the vicinity of irreversibility line, whereas the narrow one is due to hysteresis at the vortex-lattice phase transition.

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