Coexistence of superconductivity and antiferromagnetism in $HgBa_2Ca_4Cu_5O_y$: Multiharmonic susceptibility and vortex dynamics study

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(Received 10 December 2006; revised manuscript received 12 August 2007; published 27 December 2007)

We have studied the multiharmonic ac susceptibility response of $HgBa_2Ca_4Cu_5O_y$ (Hg:1245), a multilayered high-temperature superconductor (HTS) having two crystallographically inequivalent CuO_2 planes in a unit cell with very imbalanced carrier concentrations. Vortex melting lines are well described by the commonly accepted model of melting with moderate anisotropy factors of 40–50, depending on the doping level. The diamagnetic response with applied fields parallel to superconducting (a,b) planes also shows a quite robust supercurrent along the c axis. Our results are also discussed in connection with contradictory models and experiments regarding the coexistence of superconductivity and antiferromagnetism in HTSs.

DOI: 10.1103/PhysRevB.76.212508 PACS number(s): 74.72.Jt, 74.25.Ha, 74.25.Nf, 74.25.Qt

Multilayered superconducting cuprates are generally described by the formula $(CRL)Ca_{n-1}Cu_nO_{2n}$, where (CRL) is a charge reservoir layer such as $HgBa_2O_y$, $TlBa_2O_y$, $Tl_2Ba_2O_y$, $(Cu,C)Ba_2O_y$, $Ba_2(O,F)_y$, etc. $Ca_{n-1}Cu_nO_{2n}$ has an infinite-layer (IL) structure and n represents the number of CuO_2 planes between two CRLs, which supply holes to the above-mentioned CuO_2 planes. It was shown that some of the above-mentioned high-critical-temperature (T_c) superconductors have very interesting properties, mostly due to the fact that multilayered cuprates include two or more crystallographically inequivalent CuO_2 planes in a unit cell, outer planes (OPs) with pyramidal (five) oxygen coordination and inner planes (IPs) with square (four) oxygen coordination, usually with an inhomogeneous charge distribution between OPs and IPs. For example, Cu NMR studies[1] showed that in Hg- and (Cu,C)-based multilayered cuprates, the hole concentration in the inner planes is much smaller than the hole concentration in the outer planes, and the difference increases with both n and the total carrier concentration. Moreover, studies on the n dependence of T_c in $HgBa_2Ca_{n-1}Cu_nO_y$ [Hg:$12(n-1)n$] suggest that, for $n \geqslant 5$, superconductivity (SC) resides only in the outer planes, the inner planes having a too low hole concentration.[2] The occurrence of SC only in OPs is quite understandable, since it was proved[3] that superconductivity can exist in just one $(CaCuO_2)_2$ SC layer sandwiched between two $(Ba_{0.9}Nd_{0.1}CuO_{2+x})_5$ charge reservoirs blocks, so interlayer coupling is not necessary to obtain a SC state. Moreover, in $[(Ba_{0.9},Nd_{0.1})CuO_{2+x}]_5/[CaCuO_2]_2$ artificial superconducting superlattices[4] it was shown that the SC IL blocks are decoupled by the 2.1-nm-thick CRL block.

In order to investigate qualitatively the interlayer coupling between SC outer planes in $HgBa_2Ca_4Cu_5O_y$ (Hg:1245) and the influence of the IP block on vortex dynamics, we have studied the multiharmonic susceptibility and vortex melting lines of three Hg:1245 samples with different doping levels and, for comparison, of an optimum-doped Hg:1256 sample.

As many other multilayered cuprates, Hg:1245 can be grown only as polycrystalline samples using high-pressure synthesis, so direct probing of the c-axis supercurrent using single crystals or thin films is not possible. For this study we have prepared three types of Hg:1245: optimum doped with $T_c = 107$ K, slightly underdoped with $T_c = 100$ K, and underdoped with $T_c = 87$ K and, for comparison, an optimum doped ($T_c = 100$ K) Hg:1256.

Polycrystalline samples were prepared by the high-pressure synthesis technique as described in detail elsewhere.[5] For grain alignment, samples were ground, mixed with an epoxy resin in a 1:3 weight ratio, and kept for 10–12 h in a high magnetic field of 10–14 T, following the procedures from Ref. 6. Our samples consist of a collection of single crystals embedded into a cylindrical epoxy, all crystals having the c axis parallel to the symmetry axis of the cylinder, as revealed by the x-ray diffraction patterns, one example being shown in Fig. 1. The XRD pattern (in logarithmic scale) shows only the $(00l)$ lines of Hg:1245, a very small amount of Hg:1256 [again, just some $(00l)$ lines, probably as stacking faults], and an unidentified phase, marked with stars in Fig. 1. This unidentified phase does not have

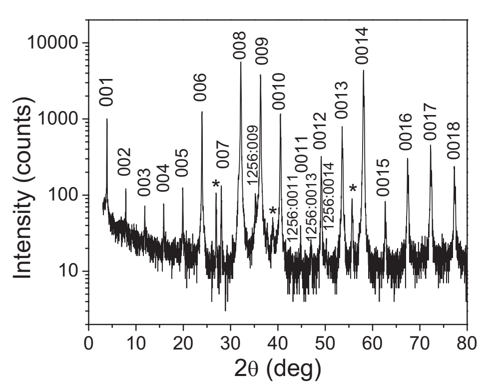

FIG. 1. Typical x-ray diffraction pattern of aligned Hg:1245. Apart from the $(00l)$ lines of Hg:1245, a small amount of Hg:1256 (stacking faults, $00l$ lines) and an unidentified nonsuperconducting compound are seen.

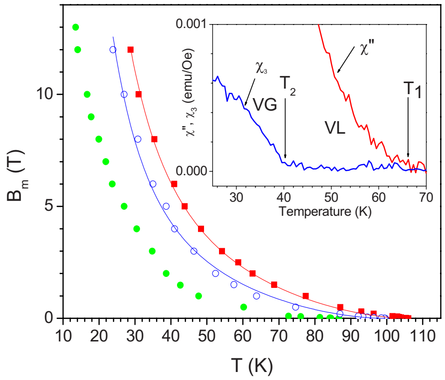

FIG. 2. (Color online) Vortex melting lines of the three Hg:1245 samples. Solid lines represent one-parameter fits with Eq. (1) in the two-fluid model. Inset: typical example of the out-of-phase and third-harmonics susceptibility response, zoomed in at low dissipation levels, for optimum doped Hg:1245 sample in a dc field of 6 T, ac field amplitude of 50 mOe, and ac field frequency of 10 kHz.

any superconducting properties; it appears also during the high-pressure synthesis of the nonsuperconducting infinite-layer compound $CaCuO_2$. Very small amounts of Hg:1223 and Hg:1234 (again as stacking faults), not seen in XRD patterns even in the logarithmic scale, were detected in the low dc field susceptibility response (in logarithmic scale), but since their vortex melting lines are known to us, our study was not influenced by these impurity phases.

For the vortex dynamics studies we have measured the multiharmonic ac susceptibility using a Quantum Design Physical Properties Measurement System Model 6000, in dc fields up to 14 T. Vortex melting lines that separate vortex glass (VG) phase from vortex liquid (VL) phase were determined from the onset of the third-harmonic susceptibility χ_3, our method[7,8] being based on the fact that in the VG phase, with low probing currents (i.e., low ac field amplitudes h_{ac}), the dissipation is strongly nonlinear, $E(J) \sim \exp[-(J_T/J)^{\mu}]$, while in the VL phase the dissipation is linear (Ohmic),[9] *and* on the fact that the third-harmonic susceptibility response is due to the nonlinear dissipation only,[10] while the fundamental out-of-phase susceptibility χ'' is due to both linear and nonlinear dissipation. An example of such a measurement, with both dc and ac fields perpendicular to SC planes is shown in the inset in Fig. 2, for Hg:1245 with $T_c=107$ K, in a dc field of 6 T, ac field amplitude of 50 mOe, and ac field frequency of 10 kHz. It can be clearly seen that the onset temperature of χ_3 (T_2, about 40 K) is much smaller than the onset temperature of χ'' (T_1, about 66 K), which means that, at 6 T, between 40 and 66 K there is a VL phase (Ohmic dissipation, since $\chi_3=0$ and $\chi''>0$) and for temperatures lower than 40 K there is a VG phase.

Similar $\chi_3(T)$ measurements performed at various dc fields, on the three Hg:1245 samples, allowed us to determine the vortex melting lines, shown in Fig. 2—namely, solid squares for the sample with $T_c=107$ K, open circles for the sample with $T_c=100$ K, and solid circles for the sample with $T_c=87$ K. Analysis of the melting transition in the framework of an anisotropic three-dimensional (3D) Ginzburg-Landau rescaling approach[11] gives the following temperature dependence of the melting field:

$$B_m(T) = \frac{C^2 c_L^4 \phi_0^5}{(k_B T)^2 \lambda_{ab}^4 \gamma (\cos^2 \alpha + \gamma^2 \sin^2 \alpha)^{1/2}}, \quad (1)$$

where C is a constant ($C \approx 1/4\pi^2$), c_L is the empirical Lindemann parameter (taken in the following to be 0.15), ϕ_0 is the magnetic flux quanta, λ_{ab} is the penetration depth along the superconducting (a,b) plane, γ is the anisotropy factor, and α is the angle between the magnetic field lines and the (a,b) plane. For our preferentially oriented samples and with our experimental setup, $\alpha=90°$. A (possible) very small mean angle of misorientation might introduce an additional experimental error in our analysis but, in our opinion, does not change qualitatively our findings, as we will show later on. A comprehensive analysis of theoretical models of melting lines and comparison with experimental data was published recently by Blatter and Geshkenbein.[12] Regarding the temperature dependence of the in-plane penetration depth, the "two-fluid" model gives $\lambda_{ab}(T)=\lambda_{ab}(0)[1-(T/T_c)^4]^{-1/2}$, the critical behavior of the 3D *XY* model gives $\lambda_{ab}(T) = \lambda_{ab}(0)(1-T/T_c)^{-1/3}$, and the mean-field model gives $\lambda_{ab}(T)=\lambda_{ab}(0)(1-T/T_c)^{-1/2}$. Apart from the heavily underdoped sample with $T_c=87$ K, the melting lines of the other samples Hg:1245 are very well described by Eq. (1) using the $\lambda_{ab}(T)$ given by the two-fluid model. The solid lines in Fig. 2 represent *one-parameter* fits with the above-mentioned model, the only free parameter being the anisotropy factor—namely, $\gamma=41$ for the optimum-doped Hg:1245 and $\gamma=48.3$ for the slightly underdoped Hg:1245, respectively. Rather good fits (not shown) were obtained also by using $\lambda_{ab}(T)$ from the critical behavior of 3D *XY* model, resulting, however, in slightly different values of γ (30%–40% smaller). The mean-field model gives a melting line that is too steep when compared with the experimental data. It was shown[12] that, by taking into account the suppression of the order parameter at high fields close to the upper critical field, the mean-field scaling describes very well the experimental data, but implies additional fitting parameters. The melting line of the heavily underdoped sample is not well described by the above-mentioned model (the one-parameter fit has significant deviations from the experimental points). It is quite likely that, in this case, the short-range Josephson coupling is rather weak, quite possibly comparable with (or smaller than) the weak, long-range, magnetic coupling. In fact, for this sample the melting line looks rather close to those resulting from the 2D vs 3D melting,[13] or to those calculated in Refs. 14 and 15 for weakly (magnetically) coupled layered superconductors.

Even if the resulting values of the anisotropy factor are subject to experimental errors, including a possible grain misalignment, and depend on the model chosen for the $\lambda_{ab}(T)$ dependence as well as on the experimental method, we believe that it is safe to consider that in our two Hg:1245 samples with higher T_c, anisotropy factors are lower than the usual values for $Bi_2Sr_2CaCu_2O_y$ (Bi:2212), which is well known to have a nonzero c-axis supercurrent. Also, the melting lines are steeper than the well-known melting line of Bi:2212.

Additional evidence of a nonzero $J_c^{(c)}$ in Hg:1245 may

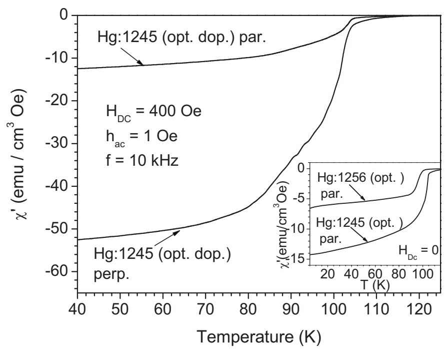

FIG. 3. In-phase susceptibility response of the optimum-doped Hg:1245 (T_c=107 K) in both parallel and perpendicular field configurations, for H_{dc}=400 Oe, h_{ac}=1 Oe, and frequency of 10 kHz. Inset: in-phase susceptibility response of the optimum-doped Hg:1245 (T_c=107 K) and Hg:1256 (T_c=100 K) in parallel field configurations, for zero dc field, h_{ac}=1 Oe, and frequency of 10 kHz.

result from the diamagnetic signal measured with both dc and ac fields parallel to the SC (a-b) planes, since in this configuration, the circulating currents responsible for the diamagnetic signal must contain segments of current along the c axis. The in-phase diamagnetic signal of the optimum-doped Hg:1245 (T_c=107 K) in both parallel and perpendicular field configurations is shown in Fig. 3, for H_{dc}=400 Oe, h_{ac} =1 Oe, and frequency of 10 kHz. We are aware of the fact that, in layered superconductors, this kind of measurements in parallel fields is quite tricky, since a small grain misalignment may induce quite high errors. However, if one assumes that the diamagnetic response in parallel field configuration comes only from grain misalignment and $J^{(ab)}$ circulating currents (i.e., $J_c^{(c)}=0$), from the values of the two signals at 40 K in the two configurations, the average angle of misalignment would be $\theta=\arctan(12.5/52)=\arctan(0.24)=13.5°$. In our opinion, such a high value of the average angle of misalignment is unrealistic since we have not seen, even in a logarithmic scale, non-(00l) lines in XRD pattern.

At this point, another question may arise: if the c-axis supercurrent $J_c^{(c)}>0$ could be associated with the single SC blocks without interlayer coupling within the inner CuO_2 planes—i.e., if there is a c-axis current between adjacent OPs separated by CRL, but not between adjacent OPs separated by the three IPs. To investigate this possibility, we have measured the susceptibility response in parallel fields of an optimum-doped (T_c=100 K) Hg:1256 sample. The comparison is shown in the inset of Fig. 3. Within the above-mentioned scenario, the supercurrent loops in parallel field configuration in the two samples have to be similar, and the only difference in the diamagnetic response should come from the slightly different number of such loops in grains with the same dimensions. More clearly, if assuming the same average thickness of SC grains in the two samples, the saturation diamagnetic response (χ_s') in parallel field configuration should scale as $1/c$, with c being the c-axis length of the respective unit cell. In our case [c(Hg:1245) =2.2168 nm, c(Hg:1256)=2.5303 nm] one should have χ_s'(Hg:1245)/χ_s'(Hg:1256)=1.14. From the inset in Fig. 3 it can be seen that χ_s'(Hg:1245,5 K)/χ_s'(Hg:1256,5 K) is about 2.17, too large for the above-mentioned scenario to hold. Based on these considerations, it is our belief that in Hg:1245 there is an interlayer coupling between SC OPs separated by the thin IPs layer and a c-axis supercurrent. Unlike our case, the absence of coupling in $[(Ba_{0.9},Nd_{0.1})CuO_{2+x}]_5/[CaCuO_2]_2$ superlattices[4] is due to the fact that the barrier separating two consecutive SC planes is too thick, about 2 nm, as compared with the thickness of the IP block in Hg:1245, which is 0.97 nm.

Our findings might also have some rather fundamental implications regarding the coexistence of superconductivity and antiferromagnetism. There is still no universally accepted theory of high-temperature superconductivity (HTS). Most models assume that doping creates holes in the valence band of an insulating, antiferromagnetic (AF) parent compound and that AF and SC are intimately related, one of the key issues being the coexistence, mixing[16,17] or nonmixing,[18] of SC and AF. Coexistence of SC and static spin-density-wave stripes was studied in the two-dimensional Hubbard model[19], and more recently, Mori and Maekawa[20] studied exactly the case of multilayered cuprates, calculated the Josephson coupling between the SC planes separated by an AF insulator, and showed that SC and AF can coexist. Until now there are several contradictory experimental results claiming either mixing,[21–24] or no mixing[25] of AF and SC, with important implications on the possible validity of microscopic theories. Lake *et al.* reported[21] that the AF correlations are induced in vortex cores and extend over the cores into the SC region in $La_{2-x}Sr_xCuO_4$ (LSCO) under magnetic field—that is, an AF proximity effect into the SC state. Later on, Bozovic *et al.*[25] used an advanced molecular beam epitaxy system to synthesize $La_{2-x}Sr_xCuO_4/La_2CuO_4$ (LSCO/LCO) heterostructures and measured their properties. They discovered that in one unit cell (1UC) LSCO/1UC LCO heterostructures there is no c-axis supercurrent (i.e., no supercurrent perpendicular to the SC planes) and concluded that AF and SC do not mix. On the other hand, μSR[22] (muon spin rotation) and Cu-NMR[23] studies revealed the coexistence of SC and AF in exactly the same material studied here, Hg:1245. It was found that for optimum-doped Hg:1245, the two outer planes undergo a SC transition at T_c=107–108 K, while the three inner planes do an AF transition below $T_N\approx 60$ K. With decreasing the doping level, T_c decreases, while T_N increases, and for extremely underdoped samples with T_c=72 K and $T_N\approx 290$ K, even in the OPs responsible for the SC transition, AF ordering was detected.[24] Based on a site-selective Cu-NMR study, Ref. 24 claims that there is even a uniform mixing of AF and SC on a single OP in the above-mentioned extremely underdoped Hg:1245. The latter type of Hg:1245 was unavailable for our studies, but the sample with T_c =107–108 K and $T_N\approx 60$ K is exactly our optimum-doped Hg:1245, whose crystal structure and properties of OPs and IPs are shown in Fig. 4.

So, if the interpretation of μSR and Cu-NMR measurements in Hg:1245 is correct, our results raise two more issues. The quite robust c-axis supercurrent in Hg:1245 means that supercurrent can tunnel through a thin (0.97 nm in our case) AF layer, unlike the case of Ref. 25 where the thickness of the AF La_2CuO_4 layer was larger than 2 nm. In a more

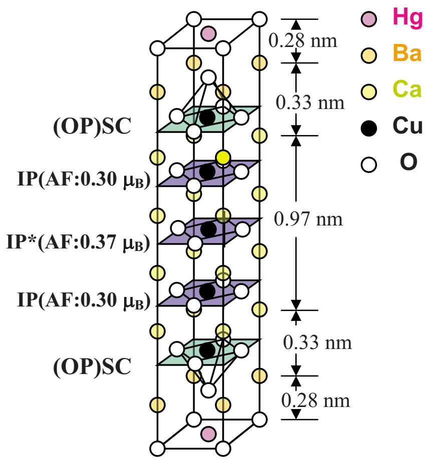

FIG. 4. (Color online) Crystal structure of Hg:1245 and illustration of the physical properties at each outer and inner plane, as of Refs. 23 and 24.

general picture, however, the thickness of the barrier is not the only factor that may lead to a zero c-axis supercurrent, since it was shown that a 20-nm-thick barrier made of underdoped LCO with $T_c=25$ K, at $T>T_c$ (i.e., in the normal-metallic state) does not block the c-axis supercurrent between LSCO SC electrodes (giant proximity effect).[26] So it is obvious that in both artificial superlattices and multilayered cuprates, the existence or nonexistence of a c-axis supercurrent is controlled by both the thickness and nature (AF insulator, metal, SC in normal state, etc.) of the barrier separating two SC layers. Recently, taking also into account the significant carrier concentration (even if not large enough for promoting a SC state) in the three IPs in Hg:1245, Mukuda *et al.*[27] suggested that the IPs are in fact in an antiferromagnetic metal state. The next important issue regards the vortex melting lines shown in Fig. 2, where it can be seen that there is no special feature at or around the Néel temperature T_N [which is about 60 K in the optimum-doped sample, a little higher (not measured) in the slightly underdoped sample and higher than T_c in the underdoped sample], so *AF ordering itself does not have a visible influence on the vortex melting lines*. This finding was quite a surprise for us, being in stark contrast with the anomalous melting line in overdoped (Cu,C)$Ba_2Ca_3Cu_4O_y$ that showed an upward kink at the temperature where the second superconducting gap opens significantly, as we recently reported in Ref. 8. We were expecting some anomalous behavior around T_N, but as can be clearly seen, the two samples not heavily underdoped do not have any special feature at T_N.

In conclusion, we have determined the vortex melting lines in three Hg:1245 samples having different carrier concentrations. Apart from the heavily underdoped sample, vortex melting lines are very well described by the commonly accepted anisotropic 3D melting model with moderate values of anisotropy factors (one order of magnitude smaller that in Bi-based HTSs). There is also a quite robust supercurrent along the c axis, as revealed from susceptibility measurements in parallel field configuration. Providing the validity of interpretation of μSR and Cu-NMR measurements suggesting that the three underdoped inner planes separating the two superconducting outer planes undergo an antiferromagnetic transition, our results also show the existence of a supercurrent tunneling through a (thin) AF layer and the fact that AF ordering itself does not affect vortex melting lines.

This work was supported by the Japanese Society for the Promotion of Science; by a Grant-in-Aid for Scientific Research on Priority Area "Invention of Anomalous Quantum Materials," Ministry of Education, Science, Sports and Culture of Japan; by the Romanian Ministry of Education and Research; by the European Commission through the Marie Curie Excellence Grant "NanoTechPinningHTS," by Japan Science and Technology Agency; and by AIST through germination research initiative "Basic Physics of Multi-band Superconductor" and through a grant for overriding priority research "High-Tc Frontier."

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