Vortex Dynamics in Hg-based Multi- and Super-Multi-Layered Cuprates

A Crisan$^{1,2}$, Y Tanaka$^3$, A Iyo$^3$, H Matsuhata$^3$, D D Shivagan$^3$, P M Shirage$^3$, T W Button$^1$, J S Abell$^1$, K Tokiwa$^4$ and T Watanabe$^4$

$^1$Department of Metallurgy and Materials, University of Birmingham, Edgbaston, Birmingham B15 2TT, U.K.

$^2$National Institute of Materials Physics, 105 bis Atomistilor Street, P.O. Box MG-7, Bucharest, 077125 Romania

$^3$National Institute of Advanced Industrial Science and Technology (AIST) Tsukuba Central 2, 1-1-1 Umezono, Tsukuba, Ibaraki 305-8568 Japan

$^4$Tokyo University of Science, Noda, Chiba 278-8510 Japan

E-mail: I.A.Crisan@bham.ac.uk

Abstract. Hg-based multi- and super-multi-layered cuprates HgBa$_2$Ca$_{n-1}$Cu$_n$O$_y$ (Hg:12($n-1)$) ($n\geq 4$) are composed of the charge reservoir layer (CRL) HgBa$_2$O$_x$, and of CuO$_2$ planes: outer planes (OP) with pyramidal oxygen coordination and inner planes (IP) with square oxygen coordination, with different charge distribution. This fact gives rise to very interesting phenomena, like coexistence of superconductivity (SC) in OPs with antiferromagnetism (AF) in IPs in Hg:1245. The dependence of $T_c$ on the number $n$ of CuO$_2$ planes is also interesting: for $n$ increasing from 4 to 5 and to 6, $T_c$ decrease from 125 to 108 and then to about 103 K; further increase in $n$ keeps $T_c$ constant at about 103 K. In Hg:1245 we show that the AF ordering at $T_N$ of 60 K has no influence on vortex dynamics, and there is a quite robust c-axis supercurrent. Vortex melting lines of various samples showed that Hg-based multi-layered cuprates ($n = 4, 5$) behaves in the usual way (increasing $n$ resulted in decrease of Josephson coupling (JC) and a less sharp melting line) and are well described by the usual anisotropic GL-based theory. Samples with $n \geq 6$ have all the same melting line, suggesting that, in this case, the short-range JC becomes much less important than the long-range magnetic coupling (MC). However, since the JC between two pancakes in OPs of adjacent unit cells (pancakes separated by the thin CRL) is large, we suggest that the vortex matter in Hg-based super-multi-layered cuprates is composed of magnetically-coupled pancake-vortex-molecules.

1. Introduction

Multi- ($n = 3$-$5$) and super-multi- ($n \geq 6$) layered HgBa$_2$Ca$_{n-1}$Cu$_n$O$_y$ (Hg:12($n-1$)) are composed of a charge reservoir layer (CRL) HgBa$_2$O$_x$, and of the infinite layer (IL) Ca$_{n-1}$Cu$_n$O$_{2n}$ with $n$ being the number of CuO$_2$ planes between two CRLs that supplies holes to the above-mentioned CuO$_2$ planes. They include crystallographically inequivalent CuO$_2$ planes in a unit cell, outer planes (OP) with
pyramidal (five) oxygen coordination and inner planes (IP) with square (four) oxygen coordination, usually with inhomogeneous charge distribution between OP and IP.

In Hg:1245 it was shown that the two optimally-doped OP’s undergo a superconducting (SC) transition at \( T_c = 108 \text{ K} \), whereas the three underdoped IP’s have an antiferromagnetic (AF) transition below \( T_N \approx 60 \text{ K} \) [1,2]. With decreasing the doping level, \( T_c \) decreases, while \( T_N \) increases, and, for extremely underdoped samples with \( T_c = 72 \text{ K} \) and \( T_N = 290 \text{ K} \), even in the OP’s responsible for the SC transition, AF ordering was detected [3]. Coexistence of superconductivity and antireffomagnetism is a key issue for microscopic theories of high-\( T_c \) superconductivity. Bozovic et al [4] used an advanced molecular beam epitaxy system to synthesize La\(_{2-x}\)Sr\(_x\)CuO\(_4\)/La\(_2\)CuO\(_4\) heterostructures and discovered that there is no \( c \)-axis supercurrent (i.e., no supercurrent perpendicular to the SC planes), and concluded that the AF nature of La\(_2\)CuO\(_4\) is responsible for this.

Another interesting property is the dependence of \( T_c \) on the number of CuO\(_2\) planes, \( n \). In Hg:12\((n-1)n \), a maximum \( T_c \) of about 135 K is obtained for \( n = 3 \), then \( T_c \) decreases to 125 K (\( n = 4 \)), to 108 K (\( n = 5 \)), and to 103 K (\( n = 6 \)). Quite astonishingly, for \( n \) higher than 6, Hg:12\((n-1)n \) retains a high and almost constant \( T_c \) of about 103 K, as revealed by dc magnetization and ac susceptibility measurements [5].

Here we present vortex dynamics studies of Hg:12\((n-1)n \). In order to investigate qualitatively the interlayer coupling between superconducting outer planes in Hg:1245, and the influence of AF ordering in the inner planes on vortex dynamics, we have studied the multi-harmonic susceptibility and vortex melting lines of three Hg:1245 samples with different doping levels, and, for comparison, of an optimum doped Hg:1256 sample. In the case of super-multi-layered Hg-based cuprates Hg:12\((n-1)n \) with \( n \geq 6 \), we have determined that vortex melting lines do not depend on the number of inner CuO\(_2\) planes, suggesting a magnetic coupling between pancake vortex molecules.

2. Experimental

Various samples were synthesized from HgO, CuO, Cu\(_2\)O, Ca\(_2\)CuO\(_4\) and the precursor Ba\(_2\)Ca\(_2\)Cu\(_3\)O\(_{7.5}\). Combining source materials, mixtures with nominal compositions HgBa\(_2\)Ca\(_n\).\(_3\)Cu\(_{n+2}\)O\(_{3n+2}\) were obtained, pressed into pellets, encapsulated in Au, and heated at 1000 – 1100 °C under a pressure of 3.5 GPa for 2h. As synthesized samples were ground, mixed with epoxy resin in a 1:3 weight ratio, and aligned in a magnetic field of 7T, for 10-12 h, at room temperature. Preparation is described in detail elsewhere [5].

Samples were characterized by X-ray diffraction (XRD) (for both powdered and grain-aligned samples), by energy dispersive X-ray analyzer attached to a scanning electron microscope (SEM-EDX) to check the cation ratio, and by transmission electron microscope. Superconducting properties were studied by four-probe electrical resistivity (for as-grown samples), by temperature-dependent DC susceptibility (MPMS Quantum Design) and by multi-harmonic AC susceptibility (PPMS Quantum Design).

Quite important for our study are the XRD patterns of grain-aligned samples. For \( n \) between 4 and 7, XRD patterns showed only (001) lines of the nominal phase, in some cases with a small amount (few %) of impurity (\( n \) different than the nominal one), as stacking faults, or a very small amount of CaCuO\(_2\) Infinite Layer (IL). Samples with nominal compositions Hg:1278 (\( n = 8 \)), Hg:12910 (\( n = 10 \)), and Hg:121314 (\( n = 14 \)), showed only (001) lines of a large number of phases with \( n \) between 5 and 16 [5]. Examples of such XRD patterns are shown in figure 1: (a) Hg:1245 showing only (001) lines of the nominal phase; (b) Hg:1256 showing (001) lines of the nominal phase and a very small amount of IL; and (c) Hg:1267 showing (001) of the nominal phase, a small amount of Hg:1278 (only (001) lines) and a very small amount of an unidentified phase, without any superconducting properties, this phase appearing also in IL samples.

Using TEM, we have seen in fact that, for \( n = 8 \), 10, and 14 samples, the grains consist of stacking faults, randomly distributed along the \( c \)-axis, of various Hg:12\((n-1)n \) layers with \( n \) between 5 and 16. Such an image is shown in figure 1(d) for the case of Hg:12910 sample.
Vortex melting lines, $B_m(T)$ or $T_m(B)$, which separates the vortex-glass (or solid) (VG) and vortex-liquid (VL) phases [6] can be determined by using the onset of third-harmonic susceptibility response, $\chi_3$ of bulk superconductors with preferentially-oriented crystallites, with very low ac field amplitudes [7,8]. In the VG state below $T_m(B)$, $E(J) \sim \exp[-(J/T_m)^\mu]$, while for $T>T_m(B)$ (in the VL state), $E(J) \sim J$ for sufficiently low current levels. At the same time, out-of-phase susceptibility, $\chi''$, is a measure of the total dissipation, while $\chi_3$ is a measure of the nonlinear dissipation only [9]. An example of such a measurement, with both DC and AC fields perpendicular to SC planes, is shown in the insert of figure 3, for an optimum-doped Hg:1245 sample with $T_c=107$ K. It can be clearly seen that the onset temperature of $\chi_3$, ($T_2$, about 40 K) is much smaller than the onset temperature of $\chi''$ ($T_1$, 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 DC fields allowed us to determine the vortex melting lines of all the analyzed samples.

3. Results and discussion

3.1. HgBa$_2$Ca$_4$Cu$_{7}$O$_{y}$ samples: coexistence of AF and SC reflected in vortex dynamics

For this subject we have studied 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. Crystal structure of optimum doped
Hg:1245 ($a = 0.385$ nm, $c = 2.21$ nm) showing the characteristics of the inequivalent CuO$_2$ layers is shown in figure 2 (note the thickness of the barrier composed of the three IP AF layers separating the two OP SC layers, of about 0.97 nm).

Figure 2. Crystal structure of Hg:1245 and illustration of physical properties at each outer and inner planes of optimum-doped sample [3].

Vortex melting lines determined from $\chi_3(T)$ measurements are shown in figure 3. The first important issue is 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 heavily 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$_2$Ca$_3$Cu$_4$O$_y$ that showed an upward kink at the temperature where the second superconducting gap opens significantly, as we recently reported [8]. We were expecting something similar around $T_N$, but, as can be clearly seen, the two samples in question do not have any special feature at $T_N$. Analysis of the melting transition in the framework of an anisotropic 3-dimensional (3D) Ginzburg-Landau rescaling approach [10] gives the following temperature dependence of the melting field:

$$B_m(T) = \frac{C^2 c_L^4 \Phi_0^5}{(k_B T)^2 \lambda_{ab}^2 \gamma^2 (\cos^2 \alpha + \gamma^2 \sin^2 \alpha)^{1/2}}$$

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), $\Phi_0$ is the magnetic flux quanta, $\lambda_{ab}$ is the penetration depth along the superconducting $(a,b)$-plane, $\gamma$ is the anisotropy factor, and $\alpha$ is the angle between the magnetic field lines and the $(a,b)$-plane. For our preferentially oriented samples and with our experimental set-up, $\alpha = 90^\circ$. 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 $\lambda_{ab}(T) = \lambda_{ab}(0)(1-T/T_c)^{1/2}$, and the mean-field model gives $\lambda_{ab}(T) = \lambda_{ab}(0)T/T_c$. Apart from the heavily underdoped sample with $T_c = 87$ K, the melting lines of the other Hg:1245 samples are very well described by equation (1) using the $\lambda_{ab}(T)$ given by the “two-fluid” model. Full lines in Fig. 3 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 (squares in figure 3), and \( \gamma = 48.3 \) for the slightly underdoped Hg:1245 (open circles in figure 3), respectively. The melting line of the heavily underdoped sample (full circles) is not well described by the above-mentioned model, looking rather close to those calculated in [11] for weakly (magnetically) coupled layered superconductors. Even if the resulted 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\(_{2}\)Sr\(_{2}\)CaCu\(_{2}\)O\(_{y}\) (Bi:2212), which is well known to have a non-zero c-axis supercurrent. Also, the melting lines are steeper than the well-known melting line of Bi:2212. In our opinion, vortex melting lines show that in Hg:1245 there is a c-axis supercurrent, and the presence of the 0.97 nm thick barrier composed of three AF IPs does not destroy superconductivity along the c-axis (c-axis coupling). Additional measurements in parallel fields (not shown) support this fact.

3.2. Vortex melting line of HgBa\(_{2}\)Ca\(_n\)Cu\(_{n-1}\)O\(_y\) \((n \geq 6)\): magnetically-coupled pancake vortex molecules.

In figure 4 are shown the vortex melting lines, \( B_m \) as function of reduced temperature, \( t = T/T_c(0) \), for multi- and super-multi-layered Hg cuprates: Hg:1234 single phase, Hg:1245 single phase (optimum doped one), Hg:1256 (almost) single phase, Hg:1267 (almost) single phase, “Hg:12910”-nominal and “Hg:121314”-nominal. It can be seen that all samples with \((n \geq 6)\) have practically the same melting line (which could not be fitted satisfactorily with the model previously discussed). Full lines are one-parameter (\( \gamma \)) fits with Eq.(1) of data for Hg:1234 and Hg:1245.

![Figure 4](image-url). Vortex melting lines Hg:1234, Hg:1245, Hg:1256, Hg:1267, Hg:”12910” and Hg:”121314”. Full lines are one-parameter fit with Eq.(1).

Figure 4. Vortex melting lines Hg:1234, Hg:1245, Hg:1256, Hg:1267, Hg:”12910”and Hg:”121314”. Full lines are one-parameter fit with Eq.(1).

In our opinion, the explanation for the coincidence of melting lines of Hg-12\((n-1)n\) \((n \geq 6)\) reside in the interplay between the Josephson coupling (JC), \( \Lambda_J \), and the magnetic coupling (MC), \( \Lambda_M \). At short distances, \( \Lambda_J \) is much stronger than \( \Lambda_M \). But, since JC is a short-range interaction, at large distances MC (long-range interaction) takes over as the dominant pancake-pancake interaction. As can be seen from figure 4, Hg:1234 has a very robust melting line and a quite small anisotropy factor \( \gamma = 17.5 \), which means a quite strong JC. Insertion of another CuO\(_2\) IP leading to Hg:1245 results in a significant shifting of the melting line towards lower temperatures, a larger \( \gamma \) of about 41, meaning a smaller JC. Now, it is known that the 3 IP’s in Hg:1245 have a very low carrier concentration, and they undergo an AF transition [1-3]. Another addition of an IP leading to Hg:1256 results in a further (but less significant) shift of the melting line, but, from now on, any more additions of IP’s in the unit cell do not affect the melting lines of super-multi-layered Hg-based cuprates. In our opinion, this
means that, in Hg:1245 there is still a significant JC, $\Lambda_{IP}^J$, between the pancake vortices in the two OP’s separated by the three IP’s, while in all other Hg-12($n$-1)$_n$ ($n \geq 6$), short-range $\Lambda_{IP}^J$ becomes much smaller than the long-range MC, $\Lambda_{IP}^m$. Being a long-range pancake-pancake interaction, $\Lambda_{IP}^m$ is not affected significantly by additions of extra IP’s in the unit cell. In fact, our experimental vortex melting lines of Hg:1256, Hg:1267, Hg:”12910” and Hg:”121314” resembles very much the theoretical (numerically calculated) melting lines of magnetically-coupled pancake vortices [11]. However, in super-multi-layered Hg-based cuprates, there are two types of pancake pairs: in OP’s separated by the thick ($n$-2) IP’s, for which $\Lambda_{IP}^J \ll \Lambda_{IP}^m$, and, respectively, in OP’s separated by the thin CRL, for which $\Lambda_{CRL}^{J} \gg \Lambda_{CRL}^{m}$. This latter assertion is obvious; otherwise, Hg:1234 for example, would have a very weak melting line. Therefore, the strongly (Josephson) coupled pancake pairs separated by CRL can be regarded as pancake molecules, which, in turn, are weakly (magnetically) coupled along the z-direction in the solid vortex phase, which can be called the Pancake Molecule Glass (or solid) (PMG). At higher temperatures, the thermal fluctuations overcome the magnetic coupling, and the PMG melts into a liquid (gas) phase, the Pancake Molecule Liquid.

Acknowledgements
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 the grant for overriding priority research “High-Tc Frontier”.

References
[1] Tokiwa K, Okumoto H, Imamura T, Mikusu S, Yuasa K, Higemoto W, Nishiyama K, Iyo A, Tanaka Y and Watanabe T 2003 Int. J. Mod. Phys. B 17 3540
[2] Kotegawa H, Tokunaga Y, Araki Y, Zheng G.-q., Kitaoka Y, Tokiwa K, Ito K, Watanabe T, Iyo A, Tanaka Y and Ihara H 2004 Phys. Rev. B 69 014501
[3] Mukuda H, Abe M, Araki Y, Kitaoka Y, Tokiwa K, Watanabe T, Iyo A, Kito H and Tanaka Y 2006 Phys. Rev. Lett. 96 087001
[4] Bozovic I, Logvenov G, Verhoeven M A J, Caputo P, Goldobin E and Geballe T H 2003 Nature 422 873
[5] Iyo A, Tanaka Y, Kodama Y, Kito H, Tokiwa K and Watanabe T 2006 Physica C 445-448 17
Iyo A, Tanaka Y, Kito H, Kodama Y, Matsuhata H, Tokiwa K and Watanabe T Physica C (in press)
Iyo A, Tanaka Y, Kito H, Kodama Y, Shirage P M, Shivagan D D, Matsuhata H, Tokiwa K and Watanabe T J. Phys. Soc. Japan (in press)
[6] Fisher D S, Fisher M P A and Huse D 1991 Phys. Rev. B 43 130
[7] Crisan A, Iyo A and Tanaka Y 2003 Appl. Phys. Lett. 83 506
[8] Crisan A, Tanaka Y, Iyo A, Cosereanu L, Tokiwa K and Watanabe T 2006 Phys. Rev. B 74 184517
[9] Fabbricatore P, Farinon S, Gemme G, Musenich R, Parodi R and Zhang B 1994 Phys. Rev. B 50 3189
[10] Blatter G, Geshkenbein V G and Larkin A I 1992 Phys. Rev. Lett. 68 875
[11] Fanghor H, Koshelev A E and Dodgson M J W 2003 Phys. Rev. B 67 174508