

**Anomalous vortex melting line in the two-component superconductor (Cu,C)Ba<sub>2</sub>Ca<sub>3</sub>Cu<sub>4</sub>O<sub>10+δ</sub>**A. Crisan,<sup>1,2</sup> Y. Tanaka,<sup>1,\*</sup> A. Iyo,<sup>1</sup> L. Cosereanu,<sup>3</sup> K. Tokiwa,<sup>4</sup> and T. Watanabe<sup>4</sup><sup>1</sup>National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba, Ibaraki 305-8568, Japan<sup>2</sup>National Institute for Materials Physics, P.O. Box MG-7, 077125 Bucharest, Romania<sup>3</sup>METRA, P.O. Box 51-16, 076550 Bucharest, Romania<sup>4</sup>Tokyo University of Science, Noda, Chiba 278-8510, Japan

(Received 21 July 2006; revised manuscript received 20 October 2006; published 14 November 2006)

Using the on-set of third-harmonic susceptibility we have measured vortex melting lines of high-pressure synthesized (Cu<sub>0.6</sub>C<sub>0.4</sub>)Ba<sub>2</sub>Ca<sub>3</sub>Cu<sub>4</sub>O<sub>y</sub> [(Cu,C):1234] and (Cu<sub>0.5</sub>C<sub>0.5</sub>)Ba<sub>2</sub>Ca<sub>2</sub>Cu<sub>3</sub>O<sub>y</sub> [(Cu,C):1223] with preferentially oriented crystallites and having various carrier concentrations. Vortex melting lines of all (Cu,C):1223 samples and of optimum-doped (Cu,C):1234 were very well described by the commonly accepted theory of melting lines within the two-fluid model. Overdoped (Cu,C):1234 proved to have an anomalous melting line, that was phenomenologically explained by the rather special gaps evolution, that is the significant opening of a second superconducting gap due to CuO<sub>2</sub> outer planes at a temperature lower than the critical one, where the first superconducting gap due to inner planes opens. The experimental melting lines of overdoped (Cu,C):1234 were modelled theoretically by assuming an empirical formula for the temperature dependence of anisotropy factor.

DOI: [10.1103/PhysRevB.74.184517](https://doi.org/10.1103/PhysRevB.74.184517)

PACS number(s): 74.25.Qt, 74.72.Jt, 74.25.Ha

**I. INTRODUCTION**

Recently there is an increasing interest in multilayered superconducting cuprates that are generally described by the formula (CRL) Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>2n</sub>, where (CRL) is a charge reservoir layer such as HgBa<sub>2</sub>O<sub>y</sub>, TlBa<sub>2</sub>O<sub>y</sub>, Tl<sub>2</sub>Ba<sub>2</sub>O<sub>y</sub>, (Cu,C)Ba<sub>2</sub>O<sub>y</sub>, Ba<sub>2</sub>(O,F)<sub>y</sub>, etc. Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>2n</sub> has an infinite layer structure and *n* represents the number of CuO<sub>2</sub> planes between two CRLs, which supplied holes to the above-mentioned CuO<sub>2</sub> planes. It was shown that some of the above-mentioned high-critical temperature (*T<sub>c</sub>*) superconductors have very interesting properties, mostly due to the fact that multilayered cuprates include two or more crystallographically inequivalent CuO<sub>2</sub> 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.

For example, Cu-NMR and  $\mu$ -SR studies revealed a coexistence of superconductivity (SC) and antiferromagnetism (AF) in five-layered compound HgBa<sub>2</sub>Ca<sub>4</sub>Cu<sub>5</sub>O<sub>y</sub> (Hg:1245), in which the two optimally doped OP's undergo a SC transition at *T<sub>c</sub>*=108 K, whereas the three underdoped IP's have an AF transition below *T<sub>N</sub>*~60 K.<sup>1,2</sup> More recently, in underdoped Hg:1245 it was discovered that AF ordering can uniformly coexist on a microscopic level (the same CuO<sub>2</sub> plane) with SC that takes place on OP's at *T<sub>c</sub>*=72 K.<sup>3</sup> Another interesting property of multilayered cuprates, that may bring important information to understand high-*T<sub>c</sub>* superconductivity, is the dependence of *T<sub>c</sub>* on the number of CuO<sub>2</sub> planes, *n*. Such experiments were performed on MBa<sub>2</sub>Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>y</sub>, with *M*=Hg, Tl, (Cu,C), on MSr<sub>2</sub>Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>y</sub>, with *M*=(Cu,Cr) and (Cu,V), and on Ba<sub>2</sub>Ca<sub>n-1</sub>Cu<sub>n</sub>O<sub>2n</sub>(O,F)<sub>2</sub> [F-02(*n*-1)*n*], for *n* up to 9,<sup>4-12</sup> and revealed that, in the case of *M*=Hg, Tl, (Cu,C), (Cu,V) as well as for F system, *T<sub>c</sub>* reaches a maximum for *n*=3 or 4, then decreases with increasing *n*, but still having high values

of around 100 K. In the F and Hg system, a high and almost constant *T<sub>c</sub>* was established for *n* between 5 and 9.<sup>12</sup> (Cu,Cr) system, however, shows a continuous decrease in *T<sub>c</sub>* with increasing *n*, dropping to zero for *n*=8. This fact shows that the reasons for *T<sub>c</sub>*(*n*) dependences is not yet fully understood. Other interesting aspects arise from the dependence of *T<sub>c</sub>* on sample doping. (Cu,C):1234 maintains a very high *T<sub>c</sub>* of about 117 K even in a heavily overdoped regime,<sup>13-15</sup> while (Cu,C):1245 show an unconventional variation of *T<sub>c</sub>* with doping level, i.e., *T<sub>c</sub>* decreases from 94.7 K (in the as-synthesized sample) to 93.3 K upon annealing in N<sub>2</sub> at 460 °C, and then increases to 99.0 K upon annealing in N<sub>2</sub> at 530 °C, reflecting the fact that the CuO<sub>2</sub> planes governing the bulk *T<sub>c</sub>* shift from the three IP's to the two OP's.<sup>16</sup> From viewpoint of band calculations or simple inspection on the electronic structure, all of the above-mentioned multilayered cuprates can be considered as multiband superconductors.<sup>17-21</sup>

Until now, there is little work on vortex dynamics in these multilayered cuprates. In particular, one of the most important aspects regarding vortex dynamics is the melting line for vortex matter, *B<sub>m</sub>*(*T*) or *T<sub>m</sub>*(*B*), which separates the vortex-glass (VG) and vortex liquid (VL) phases.<sup>22,23</sup> Recently, we developed a simple and straightforward technique for determining melting lines (and, consequently, anisotropy factors  $\gamma$ ) by using the third-harmonic susceptibility response of bulk superconductors with preferentially oriented crystallites, which proved to be very suitable especially for these polycrystalline samples grown by high-pressure synthesis.<sup>24</sup> In this paper we present measurements of the vortex melting line in overdoped (Cu,C):1234 that proved to behave anomalously, show the comparison with (Cu,C):1223 system, and discuss the above-mentioned anomaly taking into account the quite special gaps evolution in this two-component superconductor.

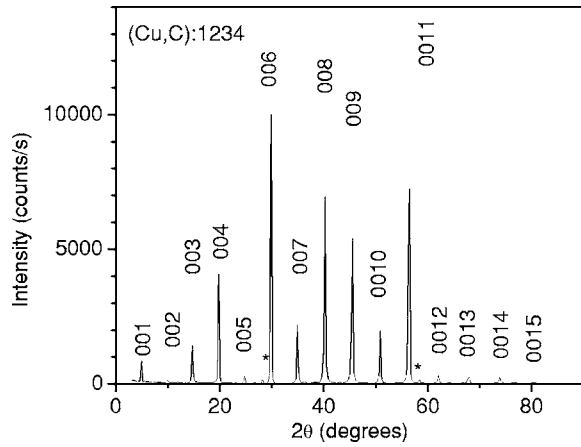


FIG. 1. X-ray diffraction pattern of over-doped (Cu,C):1234. The stars indicate a very small amount of infinite-layer compound  $\text{CaCuO}_2$ .

## II. EXPERIMENT

Sample preparation is presented in detail elsewhere.<sup>25–27</sup> Briefly, precursors having the nominal compositions  $\text{Ba}_2\text{Ca}_{1.6}\text{Cu}_{3.5}\text{O}_y\text{C}_x$  [for (Cu,C):1223] and  $\text{Ba}_2\text{Ca}_{2.7}\text{Cu}_{4.6}\text{O}_y\text{C}_x$  [for (Cu,C):1234] were prepared from mixtures of  $\text{BaCO}_3$ ,  $\text{CaCO}_3$ , and  $\text{CuO}$  powders at  $880^\circ\text{C}$  for 24 h in flowing  $\text{O}_2$  with one intermediate grinding. The residual carbon concentration  $x$  in both precursors was estimated to be about 0.1 by infrared absorption method. One mol of the respective precursor was mixed with 0.4 mol of  $\text{CaCO}_3$  and 0.52 mol of  $\text{AgO}$  (as an oxidizing agent) for (Cu,C):1223, and, respectively, with 0.3 mol of  $\text{CaCO}_3$  and 0.4 mol of  $\text{AgO}$  for (Cu,C):1234. The mixtures with nominal compositions of  $(\text{Cu}_{0.5}\text{C}_{0.5})\text{Ba}_2\text{Ca}_2\text{Cu}_3\text{O}_y$  and  $(\text{Cu}_{0.6}\text{C}_{0.4})\text{Ba}_2\text{Ca}_3\text{Cu}_4\text{O}_y$  were sealed in gold capsules and heated at  $960^\circ\text{C}$  for 4 h, and, respectively, at  $980^\circ\text{C}$  for 2 h under a pressure of 3.5 GPa by means of a cubic-anvil-type apparatus. The as-grown samples are highly over-doped and, for reducing the carrier concentration, some samples were ground into powders and post-annealed in flowing nitrogen gas in the temperature range of  $400^\circ\text{C}$ – $500^\circ\text{C}$  for 12 h. For grain alignment, samples were ground, mixed with an epoxy resin in 1:1 weight ratio, and kept for 10–12 h in a high magnetic field of 10–14 T. In Fig. 1, x-ray diffraction pattern of aligned (Cu,C):1234 is presented, showing only (001) peaks. A very small amount of impurity, namely the infinite-layer compound  $\text{CaCuO}_2$  was also detected (the very small XRD peaks marked with stars).

The principle of our method of determining vortex melting lines comes from the very basic properties of vortex matter.<sup>22,23</sup> In the VG state below  $T_m(B)$ , the electric field response to a current density  $J$  is strongly nonlinear, of the form  $E(J) \sim \exp[-(J_T/J)^\mu]$ , where  $J_T$  is a characteristic current density and  $\mu \leq 1$ . Exactly at  $T_m(B)$  the current voltage characteristic is a power-law, and, finally, for  $T > T_m(B)$  (in the VL state), one expects ohmic behavior  $E(J) \sim J$  for sufficiently low current levels. At the same time, it is well known that the out-of-phase susceptibility response of a superconductor,  $\chi''$ , is a measure of the total dissipation, linear

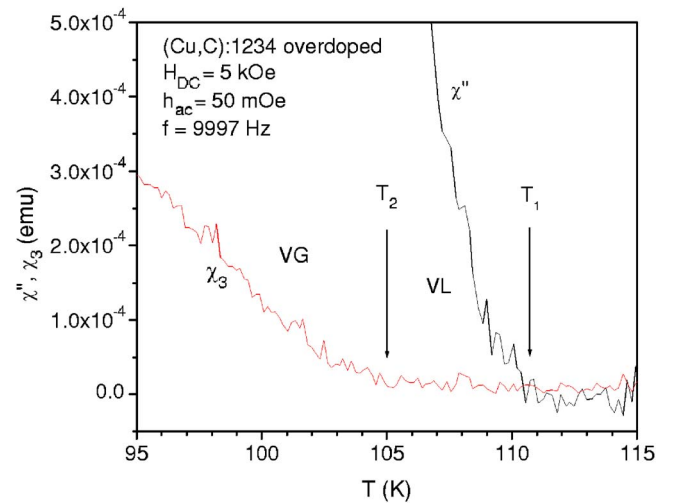


FIG. 2. (Color online) Temperature dependence of out-of-phase and third-harmonic susceptibility of over-doped (Cu,C):1234, in dc field of 5 kOe, ac field amplitude of 50 mOe, and ac field frequency of 10 kHz.

and nonlinear, while the third-harmonic susceptibility,  $\chi_3$ , is a measure of the nonlinear dissipation only.<sup>28</sup> Experimentally, we have probed the VG-VL melting transition by measuring the temperature dependence of both out-of-phase and third-harmonic susceptibilities, in dc fields  $\mu_0 H_{\text{DC}}$  up to 14 T, with very low ac field amplitudes  $h_{\text{AC}}$  of 30–50 mOe (ensuring in this way a low probing current density), using a physical properties measurement system (PPMS) Model 6000 (Quantum Design). An example of such measurements is shown in Fig. 2, for the over-doped (Cu,C):1234, in a dc field of 5 kOe, with ac field amplitude of 50 mOe and ac frequency of 10 kHz. It can be clearly seen that the on-set temperature of  $\chi''$ ,  $T_1$ , is higher than the onset temperature of  $\chi_3$ ,  $T_2$ . This means that, between  $T_1$  and  $T_2$ , there is dissipation ( $\chi'' > 0$ ), but it is a linear (Ohmic) one since  $\chi_3 = 0$ . Therefore, by using low current density (low  $h_{\text{AC}}$ ), we could probe the VL phase for  $T_2 < T < T_1$ , and the VG phase for temperatures below  $T_2$  (where  $\chi_3 > 0$ ). The onset temperature of  $\chi_3$  was taken as the melting temperature of the VG phase, at a given dc field.

## III. RESULTS AND DISCUSSION

A large number of the above-mentioned measurements were performed in various dc fields, on all five analyzed samples. The resulting melting lines of vortices are shown in Fig. 3(a), for (Cu,C):1223 [optimum doped ( $T_c = 121$  K), full circles; slightly over-doped ( $T_c = 115$  K), open circles; and over-doped ( $T_c = 76$  K), open squares], and in Fig. 3(b) for (Cu,C):1234 [optimum doped ( $T_c = 118$  K), triangles; and over-doped ( $T_c = 117$  K), full squares]. The lines in Fig. 3 will be addressed later on.

Analysis of the melting transition in the framework of an anisotropic three-dimensional (3D) Ginzburg-Landau rescaling approach<sup>29</sup> gives the following temperature dependence of the melting field:

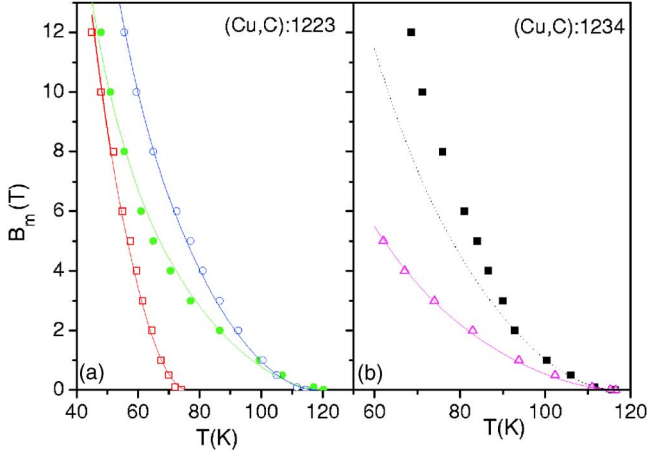


FIG. 3. (Color online) Experimentally determined vortex melting lines of (Cu,C):1223 (optimum doped, full circles; slightly overdoped, open circles; and overdoped, open squares), and of (Cu,C):1234 (optimum doped, triangles; overdoped, full squares). Full lines represent one-parameter fits with Eq. (1) using two-fluid model for the temperature dependence of the in-plane penetration depth.

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

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),  $\Phi_0$  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 setup,  $\alpha = 90^\circ$ . A comprehensive analysis of theoretical models of melting lines and comparison with experimental data was presented recently by Blatter and Geshkenbein.<sup>30</sup> 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 overdoped (Cu,C):1234, the melting lines of all the other samples are very well described by Eq. (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. 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  $\gamma$ . Mean-field model gives a melting line that is too steep when compared with the experimental data. It was shown<sup>30</sup> 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 values of anisotropy parameter resulting from the fits with Eq. (1) in the “two-fluid” model, taking  $\lambda_{ab}(0) = 140$  nm, for (Cu,C):1223 with different carrier concentrations are 24.8, 22.6, and 20.2, respectively, while for opti-

mum doped (Cu,C):1234,  $\gamma = 27.3$ . Apart from experimental errors, the absolute values of  $\gamma$  are subject to some uncertainty, for two reasons: (i) as previously mentioned, almost the same quality fits were obtained for  $\lambda_{ab}(T)$  dependence from the critical behavior of 3D XY model, but in this case the values of  $\gamma$  resulted to be about 30%–40% smaller, and (ii) the value of Lindemann parameter  $c_L$  is commonly accepted to be between 0.1 and 0.2, slightly depending on material,<sup>31</sup> and since  $c_L$  enters in Eq. (1) at fourth power (while  $\gamma$  enters at square), a small uncertainty in  $c_L$  will also be reflected in the absolute values of anisotropy parameter.

A close inspection of Fig. 3 shows that (Cu,C):1223 behave in a quite normal way, i.e., with increasing carrier concentration from optimum doped state towards overdoped states, melting lines become steeper but, in the same time, critical temperature decreases, while in the case of (Cu,C):1234, an overdoped state has a much steeper melting line than the optimum doped one, with  $T_c$  being practically the same. Also, the melting line of overdoped (Cu,C):1234 could not be fitted with a reasonable accuracy in any of the three models above mentioned. It can be clearly seen that, at a certain temperature, the experimental melting line deviates (has a very visible up-ward kink) from the trend at higher temperatures [dotted line in Fig. 3(b), actually a one-parameter fit in the two-fluid model of the experimental points at higher temperature], quite suddenly becoming steeper.

In our opinion, the explanation for this anomaly is the fact that overdoped (Cu,C):1234 is a rather special two-component superconductor with a large difference in the carrier concentrations between the two inequivalent layers, OP’s and IP’s, having a very weak interband interaction. A <sup>63</sup>Cu NMR study revealed that the temperature derivatives of the Knight shift due to the two inequivalent layers have the peaks situated at different temperatures.<sup>32</sup> Specific heat measurements showed that, apart from the usual sharp peak in the electronic specific heat at  $T_c$ , there is another smaller, quite broad peak at a smaller temperature.<sup>33,34</sup> Numerical simulations<sup>34</sup> performed in the framework of Suhl *et al.*,<sup>35</sup> reproduced quite well the specific heat experimental results for  $\Lambda_{22}/\Lambda_{11} \approx 0.75$  and  $\Lambda_{12}/\Lambda_{11} \approx \Lambda_{21}/\Lambda_{11} \approx 0.017$ , where  $\Lambda_{11}$  and  $\Lambda_{22}$  are intraband interactions in the two bands, while  $\Lambda_{12}$  and  $\Lambda_{21}$  are interband interactions. So, the parameter describing the “weakness” of interband interaction,  $\Lambda_{12}\Lambda_{21}/\Lambda_{11}\Lambda_{22}$  is, for overdoped (Cu,C):1234, about  $4 \times 10^{-4}$  indeed a very small value. For comparison, in MgB<sub>2</sub>, with the values of  $\Lambda_{ij}$  from Ref. 36, the same parameter is about  $4.6 \times 10^{-2}$ , that is two orders of magnitude higher. With these considerations, we can explain the anomalous melting line of overdoped (Cu,C):1234 by invoking a temperature evolution of the second superconducting gap  $\Delta_2$  similar to that described in Ref. 35 for two-band superconductors with very weak interband interaction:  $\Delta_2(T)$  is very small for temperatures close to  $T_c$ , and develops significantly (has a pronounced upward kink) at a certain temperature,  $T^*$ .

In the following, we propose a very simple phenomenological model to describe the anomalous vortex melting line of overdoped (Cu,C):1234. At high temperatures  $T > T^*$  (where the overdoped OP’s are in the almost gapless state), the anisotropy factor is  $\gamma_h$ . At lower temperatures  $T < T^*$ ,

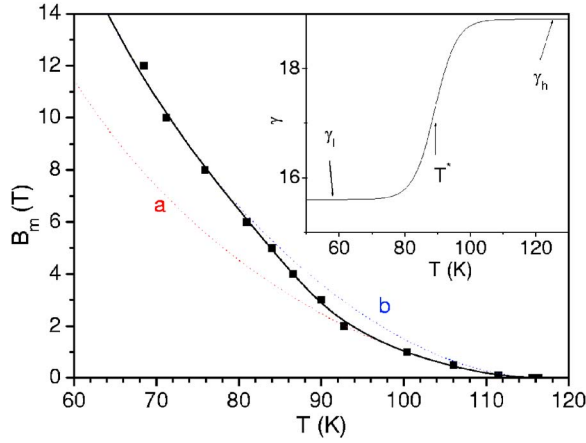


FIG. 4. (Color online) Experimentally determined vortex melting line of over-doped (Cu,C):1234 (full squares). Dot and full lines represent fits described in the text. Inset shows the proposed empirical temperature dependence of anisotropy factor, Eq. (2), with the parameters resulting from fits.

after the superconductivity in OP's become well established,  $c$ -axis coupling is higher resulting in a lower anisotropy factor  $\gamma_l$ . Since  $\Delta_2$  opens gradually, even if it increases strongly at  $T^*$ , the increase in  $c$ -axis coupling is expected to be a gradually one (even if rather steep). Therefore the use of a Heaviside-like step function for the temperature dependence of the anisotropy factor [ $\gamma(T) = \gamma_h$  for  $T > T^*$ ;  $\gamma(T) = \gamma_l$  for  $T < T^*$ ] would not be the right choice [indeed, our tentative fit of experimental melting line of (Cu,C):1234 with Eq. (1) in the two-fluid model for  $\lambda_{ab}(T)$  dependence, and with the above-mentioned  $\gamma(T)$  gave very poor results]. Instead, we introduce an empirical  $\gamma(T)$  dependence that looks like a broadened Heaviside-like step function, containing a hyperbolic tangent,

$$\gamma(T) = \frac{\gamma_h + \gamma_l}{2} + \frac{\gamma_h - \gamma_l}{2} \times \tanh \frac{T - T^*}{A}, \quad (2)$$

where parameter  $A$  reflects how sharp is the change in anisotropy, while  $T^*$  is the inflection point (symmetry point) of the tanh function and is related to the temperature at which  $\Delta_2$  opens significantly. As can be easily seen, at high temperatures  $\tanh[(T - T^*)/A] \rightarrow 1$  and  $\gamma(T) = \gamma_h$ , while at low temperatures,  $\tanh[(T - T^*)/A] \rightarrow -1$  and  $\gamma(T) = \gamma_l$ . The inset in Fig. 4 show the proposed empirical  $\gamma(T)$  dependence, with parameters  $\gamma_h$ ,  $\gamma_l$ ,  $T^*$ , and  $A$  resulted from the following analysis. At high temperatures, when  $\Delta_2$  is very small, over-doped (Cu):1234 has the anisotropy factor  $\gamma_h$ , and the dotted line (a) in Fig. 4 is a one parameter fit with Eq. (1) in the two-fluid model of the experimental points at high temperatures (between 92 and 117 K), from which resulted  $\gamma_h = 18.9$ . At low temperatures the anisotropy factor is  $\gamma_l$  and the dotted line (b) in Fig. 4 is a one-parameter fit, using the same model, of the experimental points at low temperatures (below 80 K), from which resulted  $\gamma_l = 15.6$ . Full line in Fig. 4 represent the *two-parameter* fit of the entire experimental melting line with Eq. (1), with  $\lambda_{ab}(T)$  dependence from the two-fluid model and  $\lambda(T)$  from Eq. (2) with the values of  $\gamma_h$

and  $\gamma_l$  resulted from the previous two fits of the high- and, respectively, low-temperature data. The resulting fitting parameters are  $T^* = 89.2$  and  $A = 6.55$ , and it can be clearly seen that our phenomenological model describes very well the anomalous vortex melting line of over-doped (Cu,C):1234.

We are not aware of any analytical formulation of vortex melting lines in two-component (two-gap) high-temperature superconductors. While several theoretical models for two band superconductors have been proposed, they are just for reproducing the temperature dependence of an upper critical field. Moreover these theories give qualitative different behavior of anisotropy factor. Gurevich<sup>37</sup> and Golubov and Koshelev<sup>38</sup> derived the quasiclassical Usadel equations for multiband SC, described the behavior of upper critical field, vortex core structures, and derived a model to replace the “classical” Ginzburg-Landau model. They commonly suggest the continuous change in either  $\gamma$  or “averaged” coherence length near  $T_c$ . They are unlike the  $\gamma(T)$  dependence we are proposing, that described very well our results. The above-mentioned theories were developed for describing the properties of MgB<sub>2</sub>, while the multilayered cuprate (Cu,C):1234 is very different: its two-component nature is due to the very different carrier concentration in the inequivalent OP's and IP's (in fact, one can tune this difference by selective over-doping and “engineer” the Fermi levels), and, unlike MgB<sub>2</sub>, interband interaction is indeed very small.

Moreover the two component superconductor has a principle tolerance for a nonsingular quantum phase dislocation relevant to an interband phase difference soliton (*i* soliton) theoretically.<sup>39–41</sup> It gives rise to a nonaxial vortex and a fractional flux<sup>42,43</sup> which are not taken into account in previous theories even though they are successful in MgB<sub>2</sub> physics. The nonaxial vortex and the fractional flux invoke too many (to be fully taken into account) plausible possibilities, like the crossover corresponding to a nonzero interband coupling counterpart of vortex sublattice melting demonstrated by a Monte Carlo simulation in liquid metallic hydrogen.<sup>44</sup> A two component superconductor was also modeled theoretically through a Josephson-coupled double layer XY model that showed the existence of three possible states of vortices associated with the relative phase of the layers.<sup>45</sup>

In conclusion, using third-harmonic susceptibility we have measured vortex melting lines of high-pressure synthesized (Cu,C):1223 and (Cu,C):1234 with preferentially oriented crystallites and having various carrier concentrations. Vortex melting lines of all (Cu,C):1223 samples and of optimum-doped (Cu,C):1234 were very well described (one-parameter fit) by the commonly accepted theory of Blatter, Geshkenbein, and Larkin within the two-fluid model. Over-doped (Cu,C):1234 proved to have an anomalous melting line, that was phenomenologically explained by the quite special temperature evolution of the second superconducting gap due to the very weak interband interaction.

#### ACKNOWLEDGMENTS

One of the authors (A.C.) gratefully acknowledges the Japanese Society for the Promotion of Science (JSPS). This work was partially supported by a Grant-in-Aid for Science

Research on Priority Area “Invention of Anomalous Quantum Materials,” Ministry of Education, Science, Sports and Culture of Japan; Romanian Ministry of Education and Re-

search; Japan Science and Technology Agency; and “Basic Physics of Multi-band Superconductor,” AIST germination research initiative (FY2002-04).

\*Corresponding author. Electronic address: y.tanaka@aist.go.jp

- <sup>1</sup>H. Kotegawa, Y. Tokunaga, K. Ishida, G.-q. Zheng, Y. Kitaoka, A. Iyo, Y. Tanaka, and H. Ihara, *Phys. Rev. B* **65**, 184504 (2002).
- <sup>2</sup>K. Tokiwa, H. Okumoto, T. Imamura, S. Mikusu, K. Yuasa, W. Higemoto, K. Nishiyama, A. Iyo, Y. Tanaka, and T. Watanabe, *Int. J. Mod. Phys. B* **17**, 3540 (2003).
- <sup>3</sup>H. Mukuda, M. Abe, Y. Araki, Y. Kitaoka, K. Tokiwa, T. Watanabe, A. Iyo, H. Kito, and Y. Tanaka, *Phys. Rev. Lett.* **96**, 087001 (2006).
- <sup>4</sup>H. Kusuhara, T. Kotani, H. Takei, and K. Tada, *Jpn. J. Appl. Phys., Part 2* **28**, L1772 (1989).
- <sup>5</sup>A. Iyo, Y. Aizawa, Y. Tanaka, M. Tokumoto, K. Tokiwa, T. Watanabe, and H. Ihara, *Physica C* **357-360**, 324 (2001).
- <sup>6</sup>H. Wakamatsu *et al.*, Proceedings of EUCAS 2005, Vienna, Austria.
- <sup>7</sup>Y. Kodama, M. Hirai, H. Kito, Y. Tanaka, A. Iyo, K. Tokiwa, and T. Watanabe (unpublished).
- <sup>8</sup>B. A. Scott, E. Y. Suard, C. C. Tsuei, D. B. Mitzi, T. R. McGuire, B.-H. Chen, and D. Walker, *Physica C* **230**, 239 (1994).
- <sup>9</sup>E. V. Antipov, A. M. Abakumov, and S. N. Putilin, *Semicond. Sci. Technol.* **15**, R31 (2002).
- <sup>10</sup>S. M. Loureiro, Y. Matsui, and E. Takayama-Muromachi, *Physica C* **302**, 244 (1998).
- <sup>11</sup>N. D. Zhigadlo, Y. Anan, T. Asaka, Y. Ishida, Y. Matsui, and E. Takayama-Muromachi, *Chem. Mater.* **11**, 2185 (1999).
- <sup>12</sup>A. Iyo, Y. Tanaka, Y. Kodama, H. Kito, K. Tokiwa, and T. Watanabe, *Physica C* (to be published); <http://dx.doi.org/10.1016/j.physc.2006.03.067>
- <sup>13</sup>H. Ihara, K. Tokiwa, H. Ozawa, M. Kirabayashi, M. Matsuhata, A. Negishi, and Y. S. Song, *Jpn. J. Appl. Phys., Part 2* **33**, L503 (1994).
- <sup>14</sup>O. Ogino, T. Watanabe, H. Tokiwa, A. Iyo, and H. Ihara, *Physica C* **258**, 384 (1996).
- <sup>15</sup>T. Watanabe, S. Miyashita, N. Ichioka, K. Tokiwa, K. Tanaka, A. Iyo, Y. Tanaka, and H. Ihara, *Physica B* **284-288**, 1075 (2000).
- <sup>16</sup>M. Hirai, Y. Kodama, H. Kito, Y. Tanaka, K. Tokiwa, T. Watanabe, and A. Iyo (unpublished).
- <sup>17</sup>C. O. Rodriguez, R. Weht, M. Weissmann, N. E. Christensen, and E. L. Peltzer y Blanca, *Physica C* **219**, 17 (1994).
- <sup>18</sup>C. O. Rodriguez, N. E. Christensen, and E. L. Peltzer y Blanca, *Physica C* **216**, 12 (1993).
- <sup>19</sup>N. Hamada and H. Ihara, *Physica B* **284-288**, 1073 (2000).
- <sup>20</sup>N. Hamada and H. Ihara, *Physica C* **357-360**, 108 (2001).
- <sup>21</sup>I. Hase, N. Hamada, and Y. Tanaka, *Physica C* **412**, 246 (2004).
- <sup>22</sup>M. P. A. Fisher, *Phys. Rev. Lett.* **62**, 1415 (1989).
- <sup>23</sup>D. S. Fisher, M. P. A. Fisher, and D. A. Huse, *Phys. Rev. B* **43**, 130 (1991).
- <sup>24</sup>A. Crisan, A. Iyo, and Y. Tanaka, *Appl. Phys. Lett.* **83**, 506 (2003).
- <sup>25</sup>A. Iyo, Y. Tanaka, N. Terada, M. Tokumoto, and H. Ihara, *Physica B* **284-288**, 867 (2000).
- <sup>26</sup>M. Hirai, A. Iyo, H. Kito, A. Crisan, K. Tokiwa, T. Watanabe, J. Arai, and Y. Tanaka, *Physica C* **388-389**, 427 (2003).
- <sup>27</sup>M. Hirai, A. Iyo, Y. Kodama, A. Sundaresan, J. Arai, and Y. Tanaka, *Semicond. Sci. Technol.* **17**, 423 (2004).
- <sup>28</sup>P. Fabbriatore, S. Farinon, G. Gemme, R. Musenich, R. Parodi, and B. Zhang, *Phys. Rev. B* **50**, 3189 (1994).
- <sup>29</sup>G. Blatter, V. B. Geshkenbein, and A. I. Larkin, *Phys. Rev. Lett.* **68**, 875 (1992).
- <sup>30</sup>G. Blatter and V. B. Geshkenbein, in *The Physics of Superconductors*, Vol. 1, Conventional and High-Tc Superconductors, edited by K. H. Bennemann and J. B. Ketterson (Springer, Berlin, 2003), pp. 725–936.
- <sup>31</sup>M. Tinkham, *Introduction to Superconductivity*, 2nd ed. (McGraw-Hill, New York, 1996), p. 337.
- <sup>32</sup>Y. Tokunaga, K. Ishida, Y. Kitaoka, K. Asayama, K. Tokiwa, A. Iyo, and H. Ihara, *Phys. Rev. B* **61**, 9707 (2000).
- <sup>33</sup>Y. Tanaka, A. Iyo, N. Shirakawa, M. Ariyama, M. Tokumoto, S. I. Ikeda, and H. Ihara, *J. Phys. Soc. Jpn.* **70**, 329 (2001).
- <sup>34</sup>Y. Tanaka, A. Iyo, N. Shirakawa, M. Ariyama, M. Tokumoto, S. I. Ikeda, and H. Ihara, *Physica C* **357**, 222 (2001b).
- <sup>35</sup>H. Suhl, B. T. Matthias, and L. R. Walker, *Phys. Rev. Lett.* **3**, 552 (1959).
- <sup>36</sup>A. A. Golubov, J. Kortus, O. V. Dolgov, O. Jepsen, Y. Kong, O. K. Andersen, B. J. Gibson, K. Ahn, and R. K. Kremer, *J. Phys.: Condens. Matter* **14**, 1353 (2002).
- <sup>37</sup>A. Gurevich, *Phys. Rev. B* **67**, 184515 (2003).
- <sup>38</sup>A. A. Golubov and A. E. Koshelev, *Phys. Rev. B* **68**, 104503 (2003).
- <sup>39</sup>Y. Tanaka, *J. Phys. Soc. Jpn.* **70**, 2844 (2001).
- <sup>40</sup>Y. Tanaka, *Phys. Rev. Lett.* **88**, 017002 (2002).
- <sup>41</sup>A. Gurevich and V. M. Vinokur, *Phys. Rev. Lett.* **90**, 047004 (2003).
- <sup>42</sup>E. Babaev, *Phys. Rev. Lett.* **89**, 067001 (2002).
- <sup>43</sup>A. De Col, V. B. Geshkenbein, and G. Blatter, *Phys. Rev. Lett.* **94**, 097001 (2005).
- <sup>44</sup>E. Smørgrav, J. Smiseth, E. Babaev, and A. Sudbø, *Phys. Rev. Lett.* **94**, 096401 (2005).
- <sup>45</sup>W. Zhang and H. A. Fertig, *Phys. Rev. B* **71**, 224514 (2005).