## Oscillatory Behavior of Vortex-Lattice Melting Transition Line in Mesoscopic $Bi_2Sr_2CaCu_2O_{8+\nu}$ Superconductors

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The vortex-lattice melting transition of a limited number of vortices confined in mesoscopic square superconductors was studied by *c*-axis resistance measurements using stacks of intrinsic Josephson junctions in Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+y</sub>. In contrast to the melting transition in bulk crystals, we have first found a clear oscillatory behavior in the field dependence of the melting temperature in small samples of 5–10  $\mu m$  square. The periods of the oscillations roughly obey the regularity of the matching conditions of square vortex lattices surrounded by a square boundary and the melting temperatures are enhanced around the vortex number of  $i^2$  (where *i* is an integer). The results suggest that a confinement effect by the square boundary stabilizes square lattice structures which are realized around  $i^2$  vortex number even in competition with the favorable Abrikosov triangular lattice in the bulk.

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A melting transition from a solid to a liquid induced by thermal fluctuation is a general phenomenon which can be observed in various matters. While it is a thermodynamic phase transition involving a large number of particles, how the melting is changed with decreasing the number of particles is an interesting issue, which has actually been studied in various atomic nanocrystals, e.g., metal [1] and semiconductor nanoparticles [2], due to not only the importance for practical application like a modification of their physical property but also fundamental interest in the melting process strongly influenced by boundary conditions. For this subject, the vortex matter of stringlike objects in superconductors could be a comparable system to be explored because of the advantage of an easily tunable vortex number by an external magnetic field.

The first-order melting transition of the vortex lattice has been investigated extensively since the discovery of high- $T_c$  cuprate superconductors, because the large thermal fluctuation and layered crystal structures are expected to realize novel new phases and transitions including melting [3]. The first thermodynamic evidence of melting transitions has been obtained experimentally for high-quality single crystals of Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+y</sub> (Bi2212)[4]. The nature of the transition in Bi2212 in a magnetic field normal to the CuO<sub>2</sub> plane has been understood as a phase transition from a Bragg glass to a vortex liquid, which is accompanied by decoupling of the pancake vortices due to the strongly layered structure [5].

On the other hand, the progress of microfabrication techniques has resulted in vortex states of mesoscopic superconductors attracting much attention since the latter half of the 1990 s, because the enhanced effect of confinement in a small area creates unique vortex states and configurations, such as giant vortices [6], symmetry-induced antivortices [7], and concentric vortex structures

(vortex shell) [8]. While most of this research has been performed using conventional superconductors to date, mesoscopic size high- $T_c$  superconductors can give us a good field in which to study the melting transition of a small number of vortices confined in a restricted area. Some studied related to this concept have already been conducted, in which the size effect of the vortex matter in Bi2212 single crystal microdisks was investigated for diameters as small as 5  $\mu$ m using local magnetometry with a micro-Hall sensor [9–11]. However, no influence on the vortex-lattice melting transition for the sample dimensions down to ~30  $\mu$ m has been reported [9,11].

One of the disadvantages of such magnetic measurements is that the response from the vortex system is less pronounced as the penetration depth increases, such as a situation near  $T_c$  in high- $T_c$  materials. An alternative method to detect the behavior of vortices in small crystals is to use intrinsic Josephson junctions (IJJs), which are the native atomic-scale Josephson junctions contained in the Bi2212 crystal structure [12]. We have successfully observed the melting transition through the transport properties of IJJs in a magnetic field [13]. Moreover, Kakeya et al. reported single vortex penetrations into stacks of IJJs with an in-plane area as small as  $2 \mu m^2$ [14]. Therefore, the use of IJJs as a probe may reveal the vortex states in mesoscopic Bi2212 with simultaneous observation of the individual vortex penetration and phase transition. In this Letter, we present a detailed study of the dependence of the *c*-axis resistance on the magnetic field at relatively higher temperatures, in which the oscillatory behavior of the melting transition is identified in square samples with in-plane sizes in the range of 5 to 10  $\mu$ m, indicating that the melting temperature is modified by a matching between lattice structures and the shape of the boundary.

High-quality single crystals of Bi2212 were grown by the traveling-solvent floating-zone method [15]. Stacks of IJJs were fabricated from these single crystals based on the double-side etching technique [16,17]. A scanning ion microscope (SIM) image of a fabricated sample is shown in the inset of Fig. 1. Several samples were prepared with shapes that were almost square (lengths from 5 to 10  $\mu$ m) to study the influence of the in-plane size of the IJJ stacks on the vortex state. The number of IJJs in the stacks is not well controlled, but varies from 110 to 150 junctions among samples, as estimated from room temperature resistance measurements. Transport measurements were performed by the four-probe technique, using a current source (Keithley 6430) and a nanovoltmeter (Keithley 2182) in a chamber with a cryocooler. The electric resistance R is [V(+I) - V(-I)]/2I with an applied dc current I. The magnetic field was applied parallel to the c axis of Bi2212.

The melting transition of the vortex lattice can be detected by measurements of the *c*-axis resistance  $R_c$  and/or the critical current  $I_c$  of the IJJ stacks due to an abrupt change of the interlayer coupling that reflects the alignment of pancake vortices [13]. Figure 1 shows the magnetic field dependence of  $R_c$  at 80.0 K in  $5 \times 5 \ \mu m^2$  sample. Although the appearance of  $R_c$  changes with the applied current due to the strong nonlinearity of the *I-V* characteristics, a step corresponding to the melting transition emerges in almost the same field, as indicated by the arrows, which suggests the intrinsic nature of the step, i.e., the melting transition of the vortex lattice (see Supplemental Material for the current dependence of the melting transition and the detection by  $I_c$  measurement [17]). Moreover, in the measurements of  $R_c$ , periodic

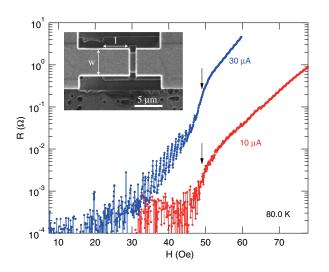


FIG. 1 (color online). Magnetic field dependence of *c*-axis resistance with different applied currents in  $5 \times 5 \ \mu m^2$  IJJs. Although there is a large current dependence, the melting transitions are observed as steplike enhancements of the resistance in both cases indicated by arrows. The inset shows a top view of  $5 \times 5 \ \mu m^2$  *z*-shaped IJJs by a SIM.

spikes (or oscillations at higher fields) due to the individual vortex penetrations into the stack are observed with an appropriate selection of the applied current. Hence, the detailed field dependence measurements of  $R_c$  reveal the vortex phase diagram in small Bi2212 crystals.

To visualize the single vortex penetrations and melting transition line,  $d \log(R_c + \delta R)/dT$  is mapped on the H-T plane, where they are highlighted due to the steep change of  $R_c$ . Here,  $\delta R$  is a small constant to suppress noise in the image in the region of  $R_c \sim 0$ . In Fig. 2, the melting transition line corresponds to the bright oscillating part from the top left to the bottom right, which is clearer under high fields and low temperatures, and gradually becomes obscure toward  $T_c$ , probably due to a decrease in the number of vortices. Note that another upper straight line that merges with the melting line near  $T_c$  is not an intrinsic phenomenon, but it is related to the critical current of the IJJs. When the applied current exceeds  $I_c$  of the 0th branch in a field during the H scan,  $R_c$  rapidly increases above this field because the IJJs transit to a resistive state. Thus, the position of this extrinsic line is dependent on the applied

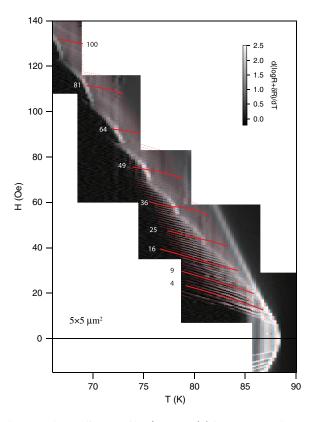


FIG. 2 (color online).  $d \log(R_c + \delta R)/dT$  map on the *H*-*T* plane of  $5 \times 5 \ \mu m^2$  IJJs.  $\delta R$  is set to 1 m $\Omega$  to improve the appearance of the image. The melting transition line and the individual vortex penetrations can be recognized as a white oscillating line and as fine white slash lines, respectively. The red dotted lines are used as a guide to count the number of vortices. The  $i^2$ th lines of the individual penetrations are emphasized by the red lines.

current. Another feature is the many fine slash lines numbered in Fig. 2, which are significant at low fields and correspond to the individual vortex penetrations. The lowest slash line indicates the location of the first vortex penetration into the stack. This phenomenon was first reported by Kakeya et al. for a small in-plane area of IJJs less than  $2 \mu m^2$  [14]. In the present experiments, this could be observed in all samples measured, even in a large sample with an in-plane area of 100  $\mu$ m<sup>2</sup>. The field intervals of the slash lines  $(\Delta H_p)$ , which correspond to the field required to add one more vortex, are almost constant in higher fields, while it is largest between the first and second lowest slash lines and there is a hysteresis dependent on the sweep direction of the field. Kakeya et al. [14] reported that the change of  $\Delta H_p$  is attributed to shrinkage of the effective in-plane area for vortices by the screening current. In the present samples, the higher-field value of  $\Delta H_p$  is still slightly larger than the expected value of  $\phi_0/wl$ , which is the magnetic flux density to place one flux quantum  $\phi_0$  in the in-plane area, and it implies the influence of the circulating current or damage at the sample edges during fabrication.

Figure 3 shows the temperature dependence of the melting transition fields and the first penetration fields  $H_{p1}$ , which were extracted from the  $d \log R_c/dT$  map in Fig. 2. To evaluate the periods of oscillatory behavior of the melting transition line, the monotonic dependence of  $H_m$  on the temperature is subtracted using a fitting curve by  $H_m = H_0(T_c/T - 1)$  (broken line in Fig. 3) from the decoupling theory of vortex strings that consist of pancake

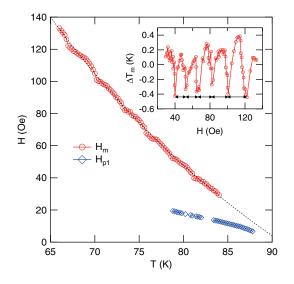


FIG. 3 (color online). Melting transition field (red open circles) and first penetration field (blue open diamonds) as a function of temperature, extracted from the map of Fig. 2 with a fitting curve by the decoupling theory. The inset shows the difference from the fitting curve of the melting temperature ( $\Delta T_m$ ) as a function of the magnetic field. The period of oscillatory behavior for the melting transition is extracted from this plot, as indicated by the arrows.

vortices [22]. The difference  $(\Delta T_m)$  from the fitting curve is plotted in the inset of Fig. 3, which shows a feature of broad peaks and sharp dips. The period of the oscillations is taken from the intervals of the dips rather than the peaks, due to the lesser ambiguity of the dip positions.

The oscillation behavior of a phase transition line in a vortex system is known to be related to the Josephson vortex phases in oxygen-deficient  $YBa_2Cu_3O_{7-\delta}$  single crystals [23,24]. The oscillations of the transition line of smectic-to-liquid [23] or slush-to-liquid [24] phases were interpreted by the commensurability between the periodicity of the CuO<sub>2</sub> layers and configurations of the Josephson vortices. In the present work, a matching effect between the confined pancake-vortex lattice and the surrounding sample boundaries may be relevant to the oscillatory behavior of the melting transition. Therefore, it is important to determine the number of vortices present in the in-plane area at a certain melting transition field to estimate the vortex configuration. This can be realized by careful counting of the number of slash lines as marked in Fig. 2. As a result, the red solid lines in Fig. 2 that indicate the  $i^{2}$ th lines (where *i* is an integer) almost coincide with the local maximums of the oscillating part of  $T_m$  defined in the inset of Fig. 3. The robustness of a vortex lattice against thermal fluctuation, which is the factor to determine the melting temperature, would be reinforced when the vortex lattice matches up with the boundary shape; therefore, the observed enhancement of  $T_m$  at approximately  $i^2$ vortices suggests that the expected vortex configuration, which causes the oscillations of the melting transition, is the square lattice structure.

To confirm the scenario of the matching effect by the square lattice structure, the sample-size dependence of the melting transition line was investigated for three other samples [17]. All of the samples exhibit sharp jumps of resistance accompanied with the melting transition and the oscillatory behavior of the transition line. Although the individual vortex penetrations are also observed for these samples, the disappearance of the slash lines at higher fields, especially in larger samples, makes it difficult to count the number of vortices. Instead, the period of the  $\Delta T_m$  oscillations (see the inset of Fig. 3 for the definition) was compared. The inset of Fig. 4 shows that there is a tendency for the period to increase with the magnetic field and with a reduction in the sample size. To understand this feature, the data for the expected number of vortices Nare replotted in Fig. 4 instead of the magnetic field H using the conversion  $N = H/\Delta H_p$ . All of the results for different samples seem to coincide on a single curve. If the formation of a square lattice stabilizes the system by the matching effect, then the expected matching numbers of vortices are 4, 9, 16, ...,  $i^2$  and the interval that corresponds to the period is expressed by  $(i + 1)^2 - i^2$ . Hence, in the unit of the number of vortices, the period of the oscillations is expected to be  $2\sqrt{N_{\text{total}}} + 1$  as a function of the total

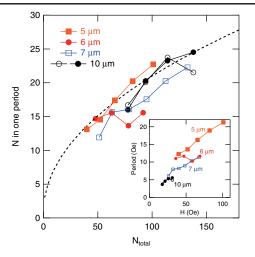


FIG. 4 (color online). Change of vortex numbers in one period of oscillations as a function of the total vortex number for four different sized samples. The vortex numbers were converted from the magnetic field dependence of the oscillation periods shown in the inset by dividing both axes by the period for the individual vortex penetrations ( $\Delta H_p$ ). With respect to the 10  $\mu$ m IJJs, measurements were performed twice (solid and open circles) to check the reproducibility. In the inset, the magnetic field of the horizontal axis is taken from the smaller dip field of both dips that define one period in the  $\Delta T_m$  oscillations.

number of vortices  $N_{\text{total}} = i^2$ , which is indicated as a dashed line in Fig. 4. There is reasonable coincidence with the experimental results, which supports the matching effect between the square lattices and the square boundary.

The filling rule of vortices in superconducting squares has been studied by Zhao et al. for micrometer-sized Nb thin films by comparison of direct observation by the Bitter decoration technique with simulations [25]. They found the formation of shells similar to disks and a periodic filling of the outermost and internal shells. The rule is that with increasing  $N_{\text{total}}$ , the outermost shell is filled until the number of vortices in it becomes 4*i* (where *i* is an integer), i.e., commensurate with the square shape. The vortices then fill the internal shells until the number becomes sufficiently large enough to create the outermost shell with 4(i + 1)vortices. In this process, a square lattice is found at small  $N_{\text{total}}$ , i.e., 4, 9, 16, although for sufficiently large  $N_{\text{total}}$  the vortices do not form clear square lattices even with commensurate numbers in the simulation, where the square lattice is distorted because the vortices tend to recover the triangular structure far from the boundary. Hence, it is difficult to expect a perfect square lattice with a large number of vortices, i.e.,  $N_{\text{total}} = 100$ , at which the oscillations of the melting transition still occur. A configuration of a deformed square lattice at large  $i^2$  may be relevant to the oscillations, or the periodic filling of the outermost and internal shells is an alternative explanation of the oscillatory behavior. This remaining issue of the relation between the melting temperatures and vortex configurations would be solved by a direct visualization of the confined vortices and/or a theoretical study of the melting transition of vortex crystals in mesoscopic high- $T_c$  superconductors.

In summary, we conducted a detailed study on the vortex-lattice melting transition of pancake vortices confined in mesoscopic square Bi2212 single crystals by out-of-plane resistance measurements of the IJJs stacks. We first found that the influence of the vortex confinement modulates the melting transition temperature, which results in the oscillatory behavior of the phase transition line in the H-T phase diagram. The size dependence and identification of the number of vortices using the individual vortex penetrations suggest that the melting temperature is enhanced when the configuration of vortices becomes a (deformed) square lattice.

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