Observation of Structures of Chain Vortices Inside Anisotropic High-T_c Superconductors

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In order to elucidate the formation mechanism of unconventional arrangements of vortices in high-T_c superconducting thin films at an inclined magnetic field to the layer plane, we investigated the structures of vortex lines inside the films by Lorentz microscopy using our 1-MV field-emission electron microscope. Our observation results concluded that vortex lines are tilted to form linear chains in $YBaCu_3O_{7,8}$. Vortex lines in the chain-lattice state in $Bi_2Sr_2CaCu_2O_{8+\delta}$, on the other hand, are all perpendicular to the layer plane, and therefore only vortices lined up along Josephson vortices form chains.

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When a magnetic field is applied to a type-II superconductor, the field penetrates it in the form of flux lines, or magnetic vortices, which usually form a triangular lattice. This is the case even for anisotropic superconductors as long as the magnetic field is directed along the anisotropy *c* axis. When the magnetic field is greatly tilted away from the *c* axis, however, the Bitter images show that the vortices no longer form a triangular lattice, but instead form arrays of linear chains along the direction of the field tilting for $YBaCu_3O_{7,8}$ (YBCO) [1] and alternating domains of chains and triangular lattices for $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) [2–4]. While the chain state in YBCO can be explained by the tilting of vortex lines within the framework of the anisotropic London theory [5–7], consistent with the experimental data [1], the chain-lattice state in Bi-2212 has long been an object of discussion. It has been attributed to two sets of vortex lines having different orientations [3,8–12], one set forming chains and the other set triangular lattices. Koshelev [12], for example, proposed that vortices that perpendicularly intersect Josephson vortices form chains and the rest of the vortices form triangular lattices. Recent investigations [4] report detailed observations of "pancake" vortices at a tilted magnetic field. No direct evidence for such mechanisms, however, was given experimentally due to the lack of methods to observe vortex lines inside superconductors.

Lorentz microscopy with our newly developed 1-MV electron microscope [13] has made it possible to distinguish between two vortex lines that are oriented in different directions [14], and was used for the present experiments determining the tilting of vortex lines. The schematic of our experiments is illustrated in Fig. 1(a). Film samples, 300–400 nm thick, of single-crystalline YBCO ($T_c = 92$ K) and Bi-2212 ($T_c = 85$ K) were prepared by thinning a region 30 μm × 100 μm, of a YBCO single crystal with a focused ion beam machine (Hitachi FB-2000) and by cleaving a Bi-2212 single crystal, respectively. These samples, the surfaces of which were parallel to the layer plane, were tilted around the y' axis and an electron beam was incident onto them from above. A magnetic field of 0–10 mT was applied obliquely to the surface of the samples at incidence angles (θ) of 70°–90°, and vortices in arrangements reflecting the anisotropic layered structure of the materials were observed as Lorentz micrographs.

Typical examples of Lorentz micrograph and Bitter pattern of the chain-lattice state in Bi-2212 are shown in Figs. 1(b) and 1(c), respectively. In both pictures, one can see two chains of vortices in the vertical direction and more or less triangular lattices in between. In the case of YBCO, however, only chains were observed to be produced; there were no lattices [1]. These apparently similar pictures in Figs. 1(a) and 1(c) contain different information about vortices. While the Bitter pattern indicates fine ferromagnetic particles gathered at the locations of the vortex magnetic fields on the surface of the film, the Lorentz micrograph is formed by the electron phase shifts caused by vortex magnetic fields mainly inside the film due to the Aharonov-Bohm effect [15]. In the case of vortex lines that are perpendicular to the film plane, the two images

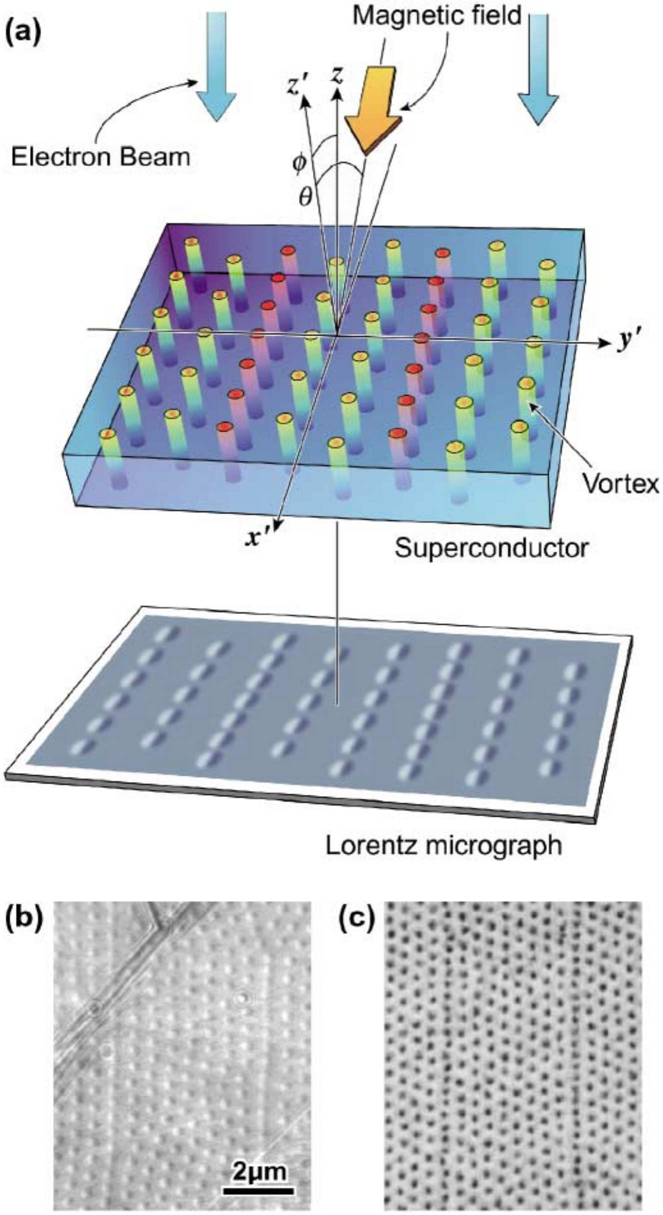

FIG. 1 (color). Observation of magnetic vortices in high-T_c superconductors at a tilted magnetic field. (a) Lorentz microscopy of vortices in a superconducting thin film. (b) Lorentz micrograph of vortices in Bi-2212 film. (c) Bitter pattern of vortices in Bi-2212. Similar chain-lattice patterns are observed in both (b) and (c). However, these two images are different in that Lorentz micrograph [14] is formed by the electron phase shifts caused mainly due to the vortex magnetic field inside the film and that Bitter pattern [1] is formed by the fine iron particles gathered at the locations where the vortex magnetic field appeared just outside the superconductor surface.

look very similar. The Lorentz images of vortex lines that are tilted considerably, however, become quite different from the Bitter images, i.e., elongated and weak in contrast [16], as demonstrated using vortex lines trapped along tilted columnar defects [14].

We first observed YBCO films using Lorentz microscopy to investigate whether vortex lines actually tilted to form chains as predicted [1] when the magnetic field was tilted away from the c axis. Examples of the Lorentz micrographs obtained are shown in Fig. 2. The vortex images look circular when θ is less than 75° under the defocusing condition of $\Delta f = 300$ mm [Fig. 2(a)]. When θ increases above 80°, the vortices begin to form chains in the vertical direction in Figs. 2(b), 2(c), and 2(d), though the vortex chains are not so straight due to the pinning. At the same time, the vortex images become gradually elongated in the direction of the chains. The gradual image elongation with an increase in θ provides direct evidence that vortex lines tilt in the direction of the chains as illustrated below in Fig. 4(e). This confirms our previous understanding that the chain structure is formed by the attractive interaction between tilted vortex lines in YBCO. The tilt angles of these vortex lines cannot be quantitatively measured from these images without careful comparison between the observed images and the simulated images under various defocusing distances (Δf), and this work is now in progress.

The chain-lattice state in Bi-2212 cannot be explained by the same mechanism, i.e., the tilting of vortex lines [12]. We therefore investigated how the chain-lattice state is formed in Bi-2212 by observing the three-dimensional arrangement of vortex lines inside the superconductor by Lorentz microscopy. An example of a Lorentz micrograph of the chain-lattice state at a magnetic field tilted by 85° is shown in Fig. 3. All the images of chain and lattice vortices are the same and are circular. If vortex lines are tilted greatly just as the magnetic field, their images must be elongated and weak in contrast as we reported in Ref. [14]. We can thus conclude that neither chain vortices nor lattice vortices in Fig. 3 are tilted, but these two kinds of vortices are both perpendicular to the layer plane. We cannot exclude the small-angle tilting of the vortex lines, however,

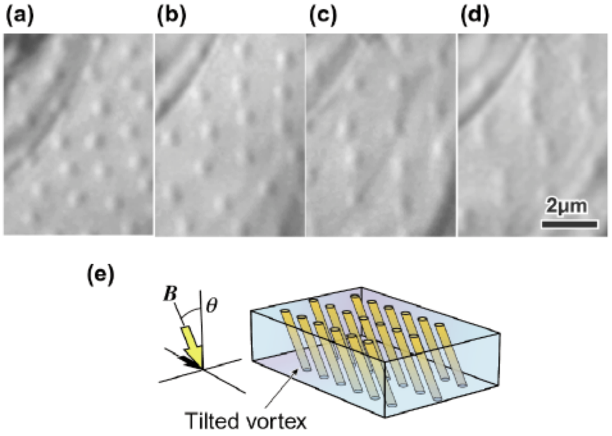

FIG. 2 (color). Lorentz micrographs of vortices in YBCO film sample at tilted magnetic fields ($T = 30$ K, $B_{z'} = 0.3$ mT); (a) $\theta = 75°$; (b) $\theta = 82°$; (c) $\theta = 83°$; (d) $\theta = 84°$. (e) Schematic of tilted vortex lines. When the tilt angle becomes larger than 80°, the vortex images begin to become elongated, and at the same time begin to form arrays of linear chains. This implies that chain vortices in YBCO are produced by some attractive force between tilted vortex lines.

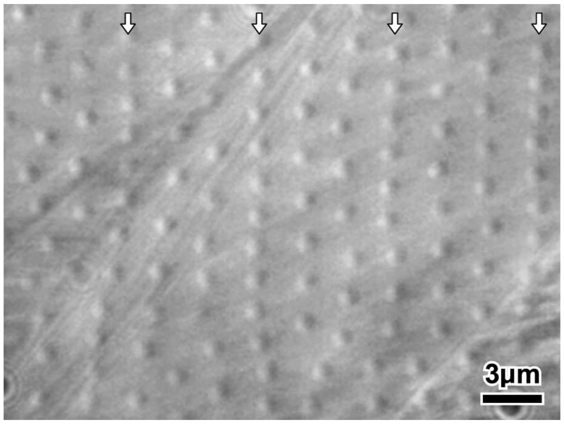

FIG. 3. Lorentz micrographs of vortices in Bi-2212. The chain-lattice structure is formed at a magnetic field tilted by $T = 50$ K, $B = 5$ mT. The chains are indicated by white arrows. If vortex lines are tilted at an angle comparable to the tilt angle of the magnetic field, 85°, the vortex images must be elongated and weak in contrast under this defocusing condition. Since the images of chain vortices as well as of lattices vortices are not elongated but circular, the vortex lines are not tilted but perpendicular to the layer plane.

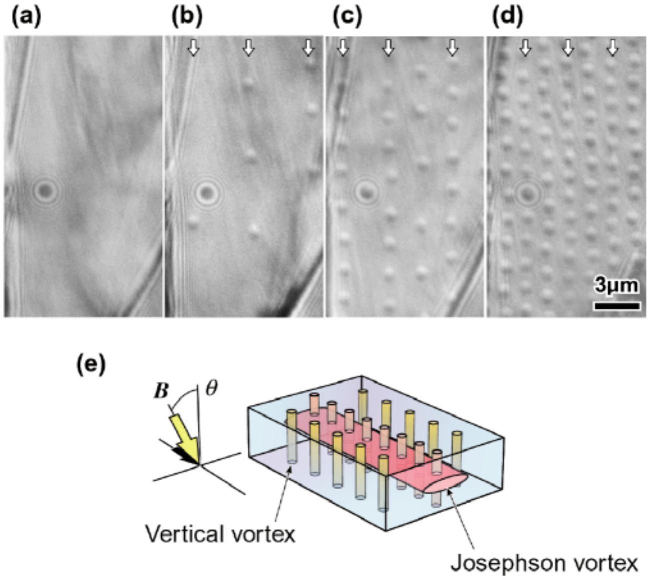

FIG. 4 (color). A series of Lorentz micrographs of vortices in a field-cooled Bi-2212 film sample when a magnetic field $B_{z'}$ perpendicular to the layer plane begins to be applied and increases at a fixed in-plane magnetic field $B_{x'}$ of 5 mT at $T = 50$ K. (a) $B_{z'} = 0$. (b) $B_{z'} = 0.02$ mT. (c) $B_{z'} = 0.1$ mT. (d) $B_{z'} = 0.17$ mT. (e) Schematic of the vortex lines consisting of vertical vortices and Josephson ones. When a magnetic field is applied parallel to the layer plane as in (a), no vertical vortices are produced, and therefore no vortex images can be seen, though Josephson vortices parallel to the layer plane should exist but cannot be observed by Lorentz microscopy due to the wide distribution of the vortex magnetic field. When the vertical magnetic field increases, vertical vortices at first begin to appear along straight lines indicated by white arrows as in (b), which are considered to be determined by Josephson vortices. Since vortices are arranged along straight lines even at large intervals between vortices and therefore no interaction between them seems to take place, we can find no other reason for the production of chain vortices than the assumption that vertical vortices crossing Josephson vortices form chains as illustrated in (e). Above $B_{z'} = 0.1$ mT, vertical vortices appear also between chain vortices as shown in (c) and (d).

if the tilt angle was much less than that of the magnetic field, 85°, since our method cannot detect the effect of such slight tilting.

We therefore carried out experiments to investigate whether or not the chain-lattice state was formed using the samples with vertical columnar defects much denser than vortices. The defect density corresponded to the vortex density of 20 mT. Since our previous experiments [14] assures us that vortex lines are trapped along tilted columnar defects regardless of the direction of the applied magnetic field as far as sample temperature T is above 20 K, the experiments were performed at $T = 50$ K, $B = 9$ mT, and $\theta = 85°$. Even under conditions where almost all the vortex lines were trapped along vertical columnar defects which were therefore perpendicular to the layer plane, we were able to observe the chain-lattice state, though the chain vortices were not arranged in such straight lines as those in samples free of columnar defects as shown in Figs. 1(b) and 1(c), but were zigzag a little bit due to the random distribution of columnar defects. This result provides evidence that the chain-lattice state can be formed even with nontilted, vertical vortices. If we accept that all the vortices are equally vertical, we can find no reason for specific vertical vortices to form chains. Therefore, we need some mechanism for specific vortices to form chains, such as the perpendicular crossing of these specific vortices with Josephson vortices. To confirm this possibility, we attempted to directly detect the Josephson vortices by Lorentz microscopy. The magnetic flux distribution of a Josephson vortex, however, was too thick, say, 50 μm wide in the layer plane to detect directly with our method. [See Fig. 4(e).]

Therefore, instead of directly observing Josephson vortices we carried out experiments to obtain evidence for the existence of Josephson vortices as follows. In the first experiment, a magnetic field $B_{x'}$ was applied parallel to the layer plane, and then a magnetic field perpendicular to the layer plane $B_{z'}$ was applied and gradually increased. A series of resultant Lorentz micrographs during this process are shown in Fig. 4. At $B_{x'} = 5$ mT and $B_{z'} = 0$, no vortex images can be seen [Fig. 4(a)], since there are no vertical vortices, but only Josephson vortices if any. When $B_{z'}$ increased from zero, images of vertical vortices began to appear. The vortices were not randomly distributed, but arranged along horizontal straight lines as shown in Fig. 4(b) as if they favored falling into lines of straight ditches (indicated by white arrows) though these are not visible.

These experimental results showing that vortices are located along unseen straight lines, even when vortices are so sparse that the attractive interaction between vortices may not occur, cannot be explained without assuming the existence of some kind of ditches for vortices. When $B_{z'}$

further increases, the vertical vortices began to appear also between the chains, thus forming the chain-lattice state [Figs. 4(c) and 4(d)]. These results lead to the conclusion that these ditches must consist of the Josephson vortices [see Fig. 4(e)] that should be produced when a magnetic field $B_{x'}$ is applied parallel to the layer plane. These Josephson vortices were once thought to have no interaction with vertical vortices but were shown by Koshelev [12] to act as potential wells (ditches) for vertical vortices. One may notice in Figs. 4(b), 4(c), and 4(d) that vortex chains are not always fixed in their locations as well as in their intervals. The locations of Josephson vortices can be changed because each Lorentz micrograph was photographed every time under new field-cooled conditions: the sample temperature was first increased above T_c, then a magnetic field was applied, and finally the sample was cooled in the presence of an applied magnetic field. The spacing of the ditches indicated by white arrows in Fig. 4 is 3–5 μm. This value is shorter than the estimated spacing 8 μm if gamma is assumed to be 200 [12]. This discrepancy may be due to the small thickness 300 nm of the film comparable to the magnetic vortex size.

We then carried out an observation of dynamics to see how vertical vortices begin to penetrate the film when $B_{z'}$ is increased from zero at a fixed $B_{x'}$ of 5 mT. Josephson vortices must have already been produced at $B_{z'} = 0$. When we gradually increased $B_{z'}$, vortex images began to enter our field of view one after another from one side along Josephson vortices which were fixed in their locations in this case, and then also between them. These static and dynamic behaviors of vortices confirm Koshelev's model [12] that vertical vortices can be stably located at Josephson vortices, thus forming chains even if they are extremely sparse, and can easily move along the directions of the Josephson vortices. Our previous finding [17] of the oscillation of chain vortices along the chain direction at much lower temperature than T_c also supports this conclusion.

The mechanisms that produce unconventional vortex arrangements at a tilted magnetic field reflecting the anisotropic layered structure of high-T_c superconductors were thus elucidated by direct observation of vortex lines inside the superconductors using our 1-MV holography electron microscope. While the chain structure in YBCO is formed by the attractive force between tilted vortex lines, the chain-lattice state in Bi-2212 is formed by the interaction of chain vortices with Josephson vortices as predicted by Koshelev [12].

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