

## Flux pinning in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin films with ordered arrays of columnar defects

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Ordered arrays of columnar defects have been fabricated by direct electron-beam writing on  $c_{\perp}$ -oriented  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) thin films. Serving as strong pinning sites, columnar defects cause a 70% enhancement of critical current density  $J_c$  at  $T=77$  K in magnetic fields parallel to the  $c$  axis. The matching effect of the pinning force density has been observed, due to commensurability between flux lines and arrays of columnar defects. The pinning strength of columnar defects has been compared with that of point defects, and found to be greater than the latter in YBCO films. These results suggest that this way of pinning center fabrication could be a promising tool in studies of the pinning mechanism in high-temperature superconductors. [S0163-1829(96)50142-6]

Since the discovery of high-temperature superconductors (HTS's), the pinning mechanism in HTS's has stimulated much scientific and technological interest. For practical purposes, various kinds of irradiation have been used to introduce artificial pinning defects to increase the critical current density  $J_c$  in magnetic fields. It is found that columnar defects created by heavy-ion irradiation greatly enhance  $J_c$  in single crystals,<sup>1,2</sup> in contrast to point defects created by electron or neutron irradiation.<sup>3,4</sup> It is also well known that  $J_c$  of as-prepared  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) thin films is known to be even higher than that of irradiated single crystals, presumably due to the intrinsic strong pinning sites. Artificial columnar defects increase  $J_c$  only moderately,<sup>5,6</sup> and occasionally a negative result is reported.<sup>7</sup> On the scientific side, vortex phases associated with different types of defects have been proposed and identified recently.<sup>8</sup> Though still with some controversy,<sup>9</sup> it is generally suggested that a transition from a vortex liquid to a vortex glass is expected when the pinning is dominated by point defects, while a transition to a Bose-glass phase is expected in the presence of columnar defects.

In spite of all the efforts to meet these technological and scientific challenges in cuprates, the subject of the ordered arrays of columnar pinning centers, and the related effects of commensurability are much less studied. In conventional superconductors, the magnetic matching effect has been observed, usually with ordered artificial defects<sup>10-13</sup> or with layered structures.<sup>14,15</sup> Flux pinning by an array of columnar defects in Josephson Junctions has also been studied.<sup>16</sup> In YBCO, the matching effects of vortex lines with the twin boundary in single crystals<sup>17</sup> or due to thin film thickness<sup>18</sup> have been observed. However, to the best of our knowledge,

there is still no report on the case of ordered arrays of columnar defects in YBCO or any other cuprate. One of the reasons could be the difficulty in fabricating ordered arrays of effective artificial pinning centers by standard lithographic techniques, since the size of these artificial defects should not be much larger than the coherence length  $\xi$ . In this paper, we report a way of fabricating pinning centers. By direct electron beam writing, we have fabricated square lattice pinning centers in YBCO films. We show that these pinning centers lead to an enhanced critical current density  $J_c$  in applied magnetic fields. When the magnetic flux lattice is commensurable to the pinning center lattice, the matching effect is observed. These artificial defects are columnlike, and a comparison of their pinning strength in YBCO films with that of point defects is briefly discussed.

The  $c_{\perp}$ -oriented YBCO films we used were prepared on  $\text{LaAlO}_3$  using the  $\text{BaF}_2$  (Mankiewich) process. The oxygen partial pressure was 300 mtorr and annealing temperature in wet oxygen was 775 °C. Details of preparation were described in Ref. 19. Films with thickness  $d=50$  and 90 nm were used. As-prepared films had critical temperature  $T_c > 90$  K, and resistivity  $\rho=240\sim 300$   $\mu\Omega$  cm at room temperature. Films were patterned into  $2\sim 4$   $\mu\text{m}$  wide and 4  $\mu\text{m}$  long microbridges.  $T_c$  of the bridges was the same as that of as-prepared films, and all bridges had  $J_c=4\sim 5\times 10^6$  A/cm<sup>2</sup> at 77 K determined by the four-probe measurement and 1  $\mu\text{V}$  voltage criterion for detection of the critical current.

To create artificial pinning defects by direct electron beam writing, we used a Phillips CM-12 scanning transmission electron microscope (STEM) with 120 keV high tension. The underlying mechanism of damage is  $T_c$  suppression due to in-plane oxygen displacements by electron irradiation, and

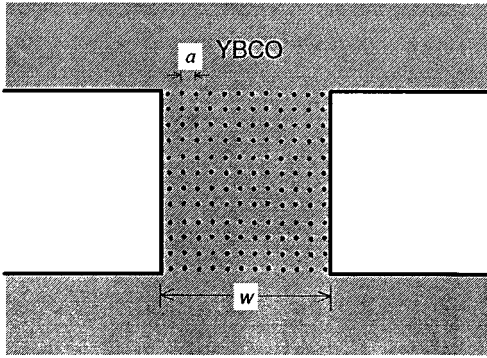


FIG. 1. A modified microbridge is illustrated. The dot matrix represents the damaged regions due to the highly focused electron beam. Lattice constant  $a$  is 90 nm and the size of dots is 10–20 nm. The bridges are 4  $\mu\text{m}$  long and the width  $W$  is 2–4  $\mu\text{m}$ .

has been carefully studied in Refs. 20 and 21. The square lattice of defects was fabricated in a way similar to that of preparation of Josephson junctions in Refs. 22 and 23. The damage was produced by stopping a focused electron beam (about 1.5 nm in diameter) at discrete points on the microbridge for certain dwell time, and created a square dot matrix with the help of computer programming control. Figure 1 illustrates the square lattice of artificial pinning centers on a microbridge. The lattice constant  $a$  is 90 nm. The current used was 80 pA and the dwell time for each spot was one second. The nominal spot size of the focused beam was 1.5 nm, but the real size of the artificial defects was determined by the beam spreading inside the films. A method similar to that proposed in Ref. 24 was used to estimate the size of the defects experimentally. The damaged area was found to be in the range of 10–20 nm in this way, which is consistent with the results of Monte-Carlo simulations.<sup>25</sup> Assuming a Gaussian distribution for the damaged area profile, the highly damaged region (where the pinning is strong) could be even smaller. Therefore, the geometric shape of these artificial pinning centers is columnar for films with  $d=50$  and 90 nm.

Ordered arrays of columnar defects were fabricated on two microbridges with thickness  $d=50$  and 90 nm.  $T_c$  of the bridges remained the same as expected when the separation between columnar defects was larger than the size of defects themselves. Before and after beam writing,  $J_c$  was measured at various temperatures  $T$  from 15 K to  $T_c$  and magnetic field up to 5 T parallel to the  $c$  axis. The lower limit of the temperature range was due to large  $J_c$  of the bridges at low temperatures. The effects of these columnar defects on two bridges were qualitatively the same. Figure 2(a) shows the temperature dependence of  $J_c$  at external magnetic fields  $H=0$  and 1.38 T for the 50 nm thick sample. In spite of a small decrease in  $J_c$  through most of the temperature range at  $H=0$ , the enhancement of  $J_c$  is obvious at  $H=1.38$  T. At zero external magnetic field,  $J_c$  at 77 K decreases by 15%. This 15% decrease can be well explained by the expected decrease of superconductive cross section due to columnar defects with diameter  $D \approx 10$  nm and separation  $a=90$  nm. For our samples,  $J_c \sim (1-t)^{3/2}$  both before and after electron beam (e-beam) writing, as shown in Fig. 2(b). This  $(1-t)^{3/2}$  dependence has been considered as the signature of columnar defect pinning.<sup>7,10,13</sup> The deviation near  $T_c$  is expected when  $\xi$  is eventually larger than  $D$ <sup>13</sup>. The same temperature depen-

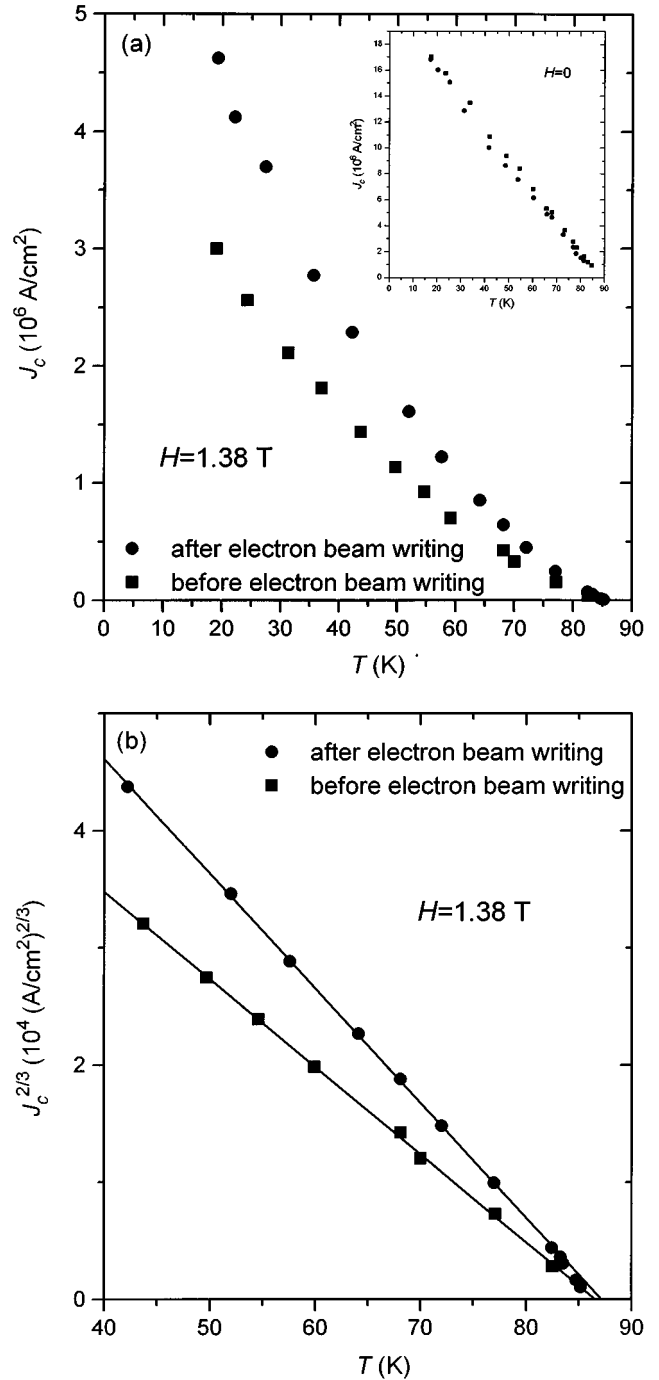


FIG. 2. The effects of the artificial columnar defects on  $J_c$  of the microbridge with  $d=50$  nm. (a)  $J_c$  vs  $T$  at  $H=1.38$  T. Inset:  $J_c$  vs  $T$  at  $H=0$ . (b)  $J_c^{2/3}$  vs  $T$  at  $H=1.38$  T.

dence of  $J_c$  both before and after the induction of artificial pinning centers not only highlights the pinning effects of the artificial columnar defects, but also implies that the mechanism of the intrinsic pinning in YBCO films be very similar to that of columnar defects.

The magnetic field dependence of  $J_c$  at  $T=77$  K is shown in Fig. 3(a) for the bridge before and after e-beam writing. An increase of 60–70% in  $J_c$  has been observed at high magnetic fields, which is comparable to the best results in the literature.<sup>5,6</sup>

The columnar defects form a square lattice. It has been

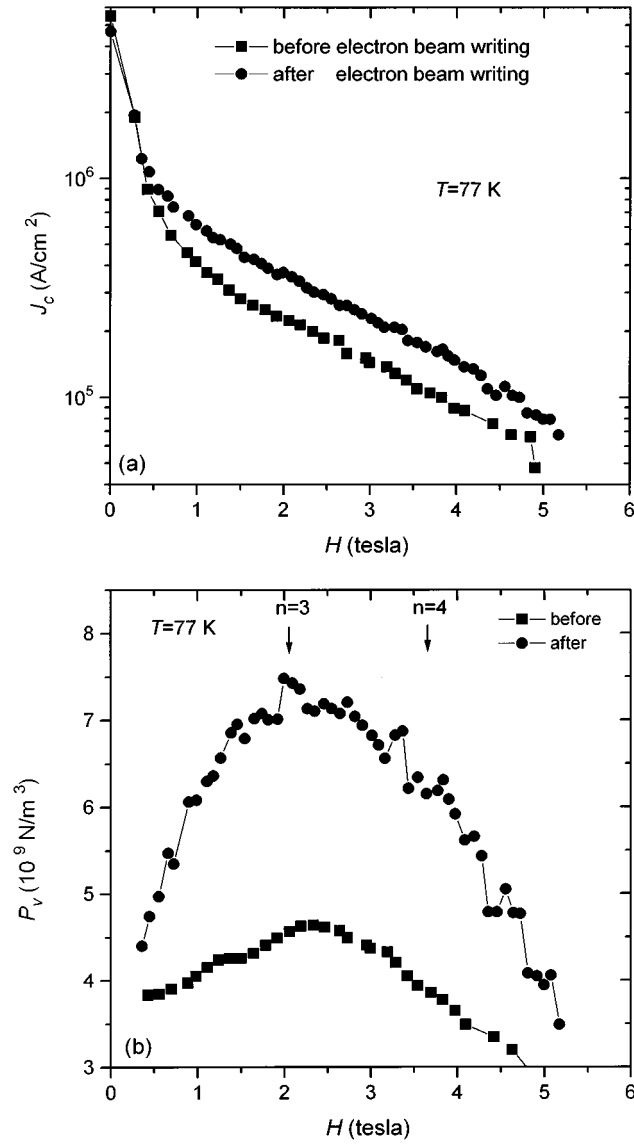


FIG. 3. (a)  $J_c$  vs  $H$  at  $T=77$  K. (b) Pinning force density  $P_v = J_c \times B$  vs  $H$ . The arrows mark the magnetic fields at which  $a = nl$  (see text). The sample is the same one as in Fig. 2.

proven that the imposed artificial pinning potential could be much larger than the difference between elastic energies for a square and a triangular flux line lattice (FLL) in superconducting films.<sup>26</sup> Therefore, for a square lattice of pinning centers with lattice constant  $a$ , a ‘‘maximum’’ pinning force density  $P_v$  can happen at certain magnetic fields when FLL matches the artificial columnar defect lattice (i.e.,  $a = nl$ , where  $n$  is an integer,  $l^2 = \phi_0/B$ ,  $B = \mu_0 H$ , and  $\phi_0$  is a quantum of flux). In this case FLL is not distorted, and has a minimum elastic energy. The matching effect has been reported in conventional superconductors.<sup>10–15</sup> Figure 3(b) shows such ‘‘peaks’’ of  $P_v = J_c \times B$  for the bridge with  $d = 50$  nm. Furthermore,  $P_v(H)$  of the 90 nm thick bridge is shown in Fig. 4. Although  $P_v$  is rather different at different temperatures, the positions of the corresponding peaks show only marginal (if any) temperature dependence. Most peaks corresponded to certain  $n$  of  $a = nl$  as marked in Figs. 3 and 4. A few unidentified peaks could be due to the multiple

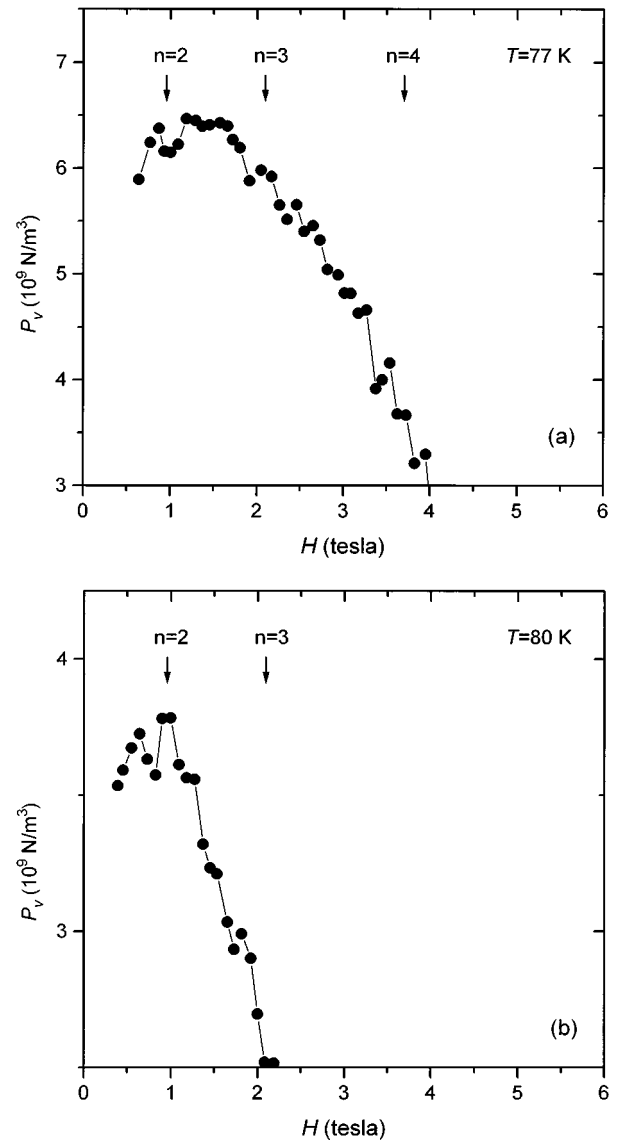


FIG. 4.  $P_v$  vs  $H$  at (a)  $T=77$  K and (b)  $T=80$  K for the microbridge with  $d=90$  nm. The arrows mark the magnetic fields at which  $a = nl$  (see text).

quantum trapping,<sup>10,26</sup> which occurs when the diameter of the columnar defect is larger than  $\xi_{ab}$ . For examples, the first peak in Fig. 4(b) might correspond to  $H_3 \equiv 3\phi_0/a^2$  in Ref. 26. At  $T=68$  K,  $P_v$  does not show clear peaks as at  $T=77$  and 80 K, presumably because the energy difference due to FLL distortion is relatively weak compared with the columnar defect pinning energy at lower temperatures.

The unit length pinning force of a columnar defect can be written as  $K/d \approx (2\pi\phi_0\mu_0)^{1/2} H_{c2}^{3/2}/8\kappa^2$ , where  $K$  is the interaction force between FL and the columnar defect,  $H_{c2}$  is the upper critical field, and  $\kappa$  is the Ginsburg-Landau parameter. Note that this expression is consistent with the temperature dependence of  $J_c$  in Fig. 2. Assuming a direct sum of pinning force, the pinning force density of the artificial columnar defects is  $\Delta P_v = (K/d) \times (1/a) \times (1-b)$ , where  $b \equiv H/H_{c2}$ . The factor  $(1-b)$  is included to account for the overall gap suppression by the magnetic field. It is reasonable to assume that the difference of the critical current den-

sity  $\Delta J_c$  before and after e-beam writing is due to artificial pinning. Therefore,  $\Delta J_c$  can be estimated from  $\Delta P_v = \Delta J_c \times B$ . At 77 K, the measured  $\Delta J_c = 2.3$  and  $0.9 \times 10^5$  A/cm<sup>2</sup> for  $H = 1$  and 3 T, respectively; the calculated  $\Delta J_c = 3.7$  and  $1.0 \times 10^5$  A/cm<sup>2</sup>. Here  $H_{c2} = 10$  T has been taken.<sup>27</sup> For a scaling law, a  $b^\alpha$  term, where  $\alpha \leq 1$ , should be further introduced to describe the general behavior of  $P_v(H)$ . Since this  $b^\alpha$  term is neglected and the direct sum of pinning force is a simple model which usually gives a larger value of  $P_v$ , we consider the estimates of  $\Delta J_c$  satisfactory.

To compare pinning of columnar defects with that of point defects, we also performed uniform electron irradiation on bridges with  $d = 50$  nm. The fluence was particularly chosen to be about the ‘‘average fluence’’ of the dot matrix writing. It was found that the increase in  $J_c(H)$  was only marginal. The volume density of the artificial point defects, which depress superconductivity, can be estimated as in Refs. 20 and 21. The measured  $\Delta J_c(H)$  at 77 K is at least

four times smaller than the estimates from the direct sum of the point defect pinning, in contrast to that in the case of columnar defects. Detailed comparisons between columnar and point defects will be reported elsewhere.

In conclusion, we have presented the studies of a way of flux pinning in YBCO thin films. Ordered arrays of columnar defects have been fabricated by direct electron beam writing. The artificial defects results in an obvious increase in  $J_c(H)$ . The peaks of pinning force density at particular magnetic fields have been observed and can be explained by the matching effect. These results suggest that this way of pinning pattern fabrication could serve as a promising tool in studies of flux pinning mechanism and vortex dynamics in high-temperature superconductors.

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