Vortex Structure in $YBa_2Cu_3O_7$ and Evidence for Intrinsic Pinning

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Using the high-resolution Bitter-pattern technique, we have studied the vortex structure in crystals of the high-T_c superconductor, $YBa_2Cu_3O_7$, for fields parallel to the c axis. We describe the vortex pinning by twin boundaries and show that a vortex lattice develops in twin-free regions. But the results also show directly that vortices are pinned at low temperatures even in crystals which are essentially defect free. We believe this pinning must be intrinsic to the new superconductors.

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In a magnetic field, type-II superconductors exist in a mixed state of flux bearing vortices in a superconducting matrix. The vortices and their interactions with material defects or pinning centers determine many of the properties studied and exploited in superconductors. In particular, zero resistance at high fields usually requires both a rigid vortex lattice and vortex pinning. Gammel *et al.*[1] used the Bitter-pattern technique, which produces images of the vortices, to show that the new superconductor $YBa_2Cu_3O_7$ has a mixed state of vortices with magnetic-flux quantum $\Phi_0 = hc/2e$, i.e., just as in conventional type-II superconductors. Unexpectedly, however, they observed no ordered vortex lattice. This could, in principle, be evidence of a high-temperature vortex liquid.[2,3] It is also unusual and probably related that some macroscopic experiments indicate that the vortices are mobile above and immobile below a reasonably well-defined "irreversibility" temperature $T^*(H)$.[4,5] However, more or less conventional "flux creep" models explain some recent experiments.[5-7] The low-temperature immobility, speaking generally or in the context of the flux creep model, implies vortex pinning by material defects even though the materials studied were crystals. In fact, a candidate defect does exist for $YBa_2Cu_3O_7$. The "crystals" are usually composed of macroscopic domains of crystal twins, mirror reflected versions of the same crystal orientation. These defects have been observed to pin vortices[8] and have been studied in recent theoretical work.[9] Our vortex images provide information on twin boundary pinning and demonstrate that a vortex lattice can occur in twin-free crystals. But they also prove the existence of another kind of pinning, apparently not related to a known crystal defect.

The high-resolution Bitter-pattern technique[1,10,11] employs fine ferromagnetic particles to "decorate" the surface of a superconductor. The resulting image of the vortices is then observed, usually in an electron microscope. We have studied platelike crystal samples of $YBa_2Cu_3O_7$. A typical crystal is 15 μm thick (c axis) and 1 mm across (a-b plane) and is twinned but has apparently no other such common, gross defects.[12,13] We will allude also to decorations of $Bi_2Sr_2CaCu_2O_8$.[14] In most of the experiments, the samples are cooled rapidly to 4.2 K in a magnetic field perpendicular to the a-b plane so that flux is "trapped." In discussing the observations it will be useful to consider that pinning may arise from a distribution of defects at positions r_i and with pinning strengths u_i. Elemental vortex interactions should produce an ordered (archetypally hexagonal) lattice which becomes increasingly rigid for higher local inductions $B_{\text{local}} = (\frac{2}{3})^{1/2}\Phi_0/a_\phi^2$, where a_ϕ is the lattice spacing. Pinning obstructs flux motion and produces vortex arrangements uncharacteristic of static equilibrium. For example, if r_i and r_{i+1} denote adjacent pins, then the vortex lattice will be obscured if $s \equiv \langle r_i - r_{i+1} \rangle \simeq a_\phi$ but only local variations in flux density and its gradient will be observed for large s.

We first describe some peripheral results. In highly twinned crystals, as in Ref. 1, one observes patterns with no long-range order in the vortex lattice. Experiments in which a field is applied at low temperatures produce flux penetration only a short distance into the superconductor with implied field gradients of order 0.1 T/μm (vortices are not resolved). This value corresponds[15] to a critical current density approaching 10^7 A/cm^2. The gradient is enormous compared to what follows and indicates strong pinning. The experiments also show that sample edges are superconducting. We had observed long-range order for trapped flux in $Bi_2Sr_2CaCu_2O_8$ crystals where there is no twinning. Therefore we selected $YBa_2Cu_3O_7$ crystals which showed untwinned areas (or unresolvable twins) optically.

Figure 1(a) shows a pattern obtained near a small twin-free region of a sample cooled at 20 G. Figures 1(b) and 1(c) show decorations of a crystal with a large twin-free region in successive experiments at 40 and 80 G. In these last two experiments the field was removed about 10 min before decoration. In Fig. 1(a) one sees the effect of twin boundaries but also, in the central region, the beginnings of vortex ordering because this small region is twin free. The observation is almost exactly that of Osip'yan and co-workers.[8] The degree of order is not, however, significantly greater than observed by Gammel *et al.* at such low fields. One expects that

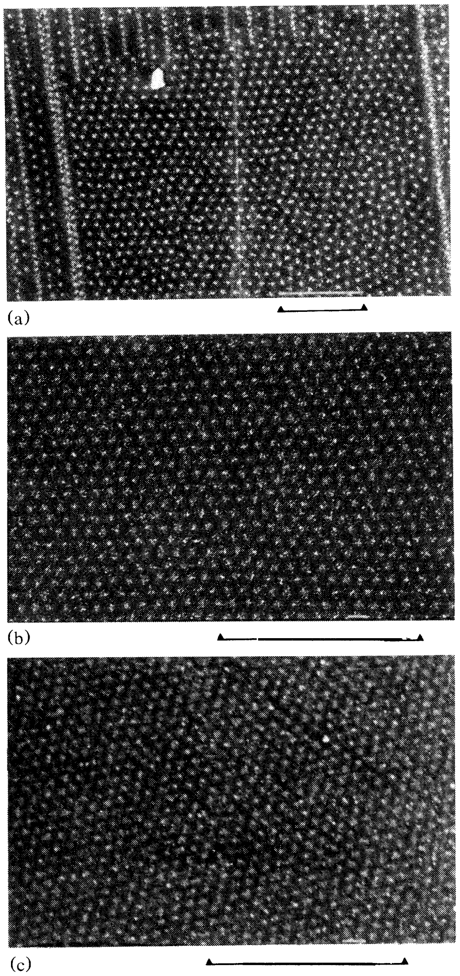

FIG. 1. The vortex pattern in twin-free regions of $YBa_2Cu_3O_7$ crystals cooled in fields of 20, 40, and 80 G to produce local fields of about 15, 35, and 65 G in the areas shown. The markers are 10 μm.

order increases with B_{local}, a test that failed in Gammel *et al.* Here order increases with field as indicated, somewhat inadequately, in Figs. 1(b) and 1(c). The ordered lattice extends far beyond the areas shown and is greater for the higher field as in conventional superconductors.

Figure 2 shows a more densely twinned area of the sample cooled at 20 G. Vertical strings of vortices delineate the twin boundaries. The coincidence of the vortex strings with twin boundaries all over the sample was verified by polarized light microscopy. The overall correspondence leads us to conclude that the "twin-free" regions are just that, and not just regions where twins are unresolved optically. Also, there is evidence that the vortices are pinned in, not against, the boundaries, i.e., the boundaries are potential minima. For example, vortex densities greater than the applied field occur at some boundaries and vortices are found at locations where local vortex deficits imply they would not be held unless they were at a minimum. Such a case will be shown below. The vortex strings are an accident of the approximate commensuration of the vortex spacing and the twin boundary spacing in this example: $s \simeq a_\phi$. It should be clear that a somewhat smaller twin boundary spacing would produce a disordered array without such clearly expressed lines. We believe this was the case for Gammel *et al.*

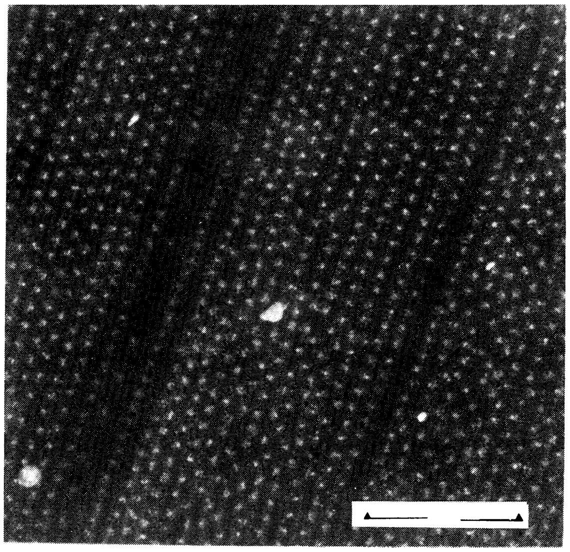

FIG. 2. The vortex pattern in a more highly twinned region for the sample cooled at 20 G. The marker is 10 μm.

Figure 3 and 4 show the vortex pattern near parts of the edge of the sample cooled at 40 G. Here flux gradients are largest. There is a vortex-free belt near the sample edge which is remarkably well defined except where twin boundaries (Fig. 4) have held vortices in the

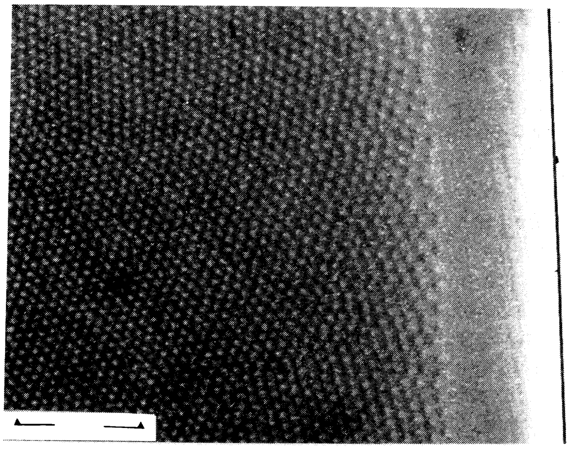

FIG. 3. The vortex pattern near the edge (black line) of the sample cooled at 40 G and decorated after the field was removed at 4.2 K. The marker is 10 μm.

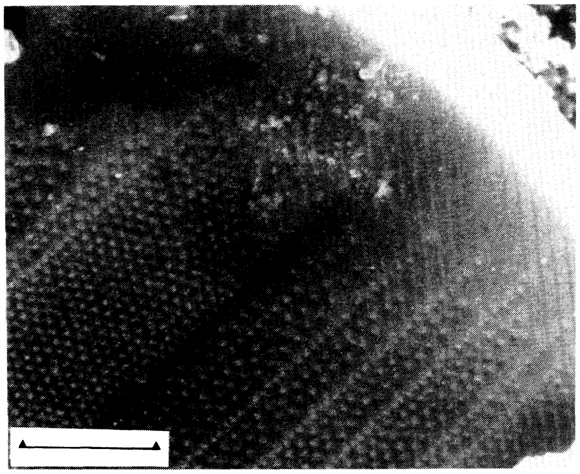

FIG. 4. The same as Fig. 3 but in an area where a few twin boundaries extend to the sample rim. The marker is 10 μm.

zero-field belt. From the edge of this belt, the flux increases gradually as one approaches the sample center where the higher and more nearly uniform fields of Fig. 1 occur. In contrast to the twinned regions and to the analogous patterns observed in conventional hard superconductors, there are no abrupt changes in flux or flux gradient in the twin-free region. The pattern reflects the equilibrium reached with the diamagnetic field of the flat plate in the field in which it was cooled, as if there were no pinning. The most singular aspect is the belt which has been observed previously only in type-I superconductor (small pinning) plates in the intermediate state.[16] The belt and the flux gradient represent the response of the vortices to the applied field combined with the diamagnetic field from the currents generated at the sample rim when some of the sample flux was expelled. Minimization of the external field energy for the thin sample provides a force driving vortices toward the sample center. The flux gradient reflects the opposing force originating in the repulsion between vortices. A flux-free belt of such large size can only be obtained for a thin plate; it may be varied by changing the applied field and hence the magnetic pressure on vortices.[16] The overall pattern is remarkable, first, in that there is so little evidence of pinning as the pattern formed in cooling and, second, that there has been no response to the changed magnetic environment following the field removal. In particular, although the rim currents have been reduced, in fact reversed, the vortices near the edge of the belt have not responded. Each is unambiguously attracted to the rim yet none has moved. Hence they are pinned individually.

The pattern itself reflects no pinning, but at 4.2 K there are many pins which are not associated with twinning or any comparably known and visible defect. These "invisible" pins apparently become active at some temperature below that at which the vortex pattern was established. To "freeze" the pattern without disturbing it the pins must be very dense. Then they cause only unresolvable changes in vortex position. Applying this requirement, $s \ll a_\phi$, to the densest flux regions produces the best bound on the pin spacing s. We estimate $s < 100$ nm since larger spacings would produce a noticeable disordering of the lattice just as occurs with the twin boundary pinning. We believe that this bound is conservative in the sense that any fluctuations in spacing or density on scale $\approx a_\phi$ would be detected: The pins must also be very uniformly distributed. Furthermore, at the temperature near which the freezing occurred, the same conditions must have applied. If there were a wide distribution of u_i, the effective pin site spacing would vary with temperature. The first to appear in cooling would distort the lattice. Therefore we infer that the pins must be very uniform energetically so that at no time was the effective pin density $\simeq a_\phi$, i.e., the pins appear rather abruptly in cooling. It is natural to associate the freezing temperature with the irreversibility temperature $T^*(H)$. Then the observations may also be relevant to $Bi_2Sr_2CaCu_2O_8$ and the unusual results of the macroscopic experiments may involve the nature of the invisible pins.

Subject to the discovery of a new, regularly occurring defect in these already well-studied materials, our observations suggest a pinning mechanism intrinsic to the material in some sense. Because the coherence length is very small, possibly only 1 nm, the vortices may interact with the crystal lattice or with oxygen vacancies, with point defects occurring at or near molecular densities.[17] Alternatively, a self-pinning mechanism is also conceivable. That is, the vortex and its fields may represent a disturbance sufficient to locally alter the electronic properties of the material or the superfluid, thereby providing its own potential minimum.[18] Such mechanisms, however vaguely defined here, would naturally satisfy the requirements for a dense, uniform pinning source in even the best crystals.

In conclusion, we have established that a well-ordered vortex lattice can develop in $YBa_2Cu_3O_7$ and have shown how the presence of twin domains inhibited the observation of lattices in some earlier experiments. The twin boundaries are energy minima for the vortices and dominate pinning at some temperature above the decoration temperature, 4.2 K. The earlier disordered arrangements have been cited as possible evidence for a high-temperature vortex liquid, but we have shown that twin pinning is sufficient to produce the disorder seen in those experiments. On the other hand, the observance of a lattice does not rule out the existence of such a state since, needless to say, liquids commonly freeze into crystalline solids. The specific question is thus left moot by our experiments, but we do infer a rather high mobility at high temperatures. The low mobility at low temperatures is observed directly as a restraint on individual vortices. The source of this pinning is undetermined but must be very dense [$>1/(100\ \mathrm{nm})^2$], uniformly distributed spatially, and of a well-defined energy. Since the invisible pins are not effective in pinning at high temperatures, resistance-free conduction may require some modification of them once they are understood. Alternatively, since the twins seem to be more conventional defects and are effective at a higher temperature, increasing their density should provide larger critical currents, at least in $YBa_2Cu_3O_7$.

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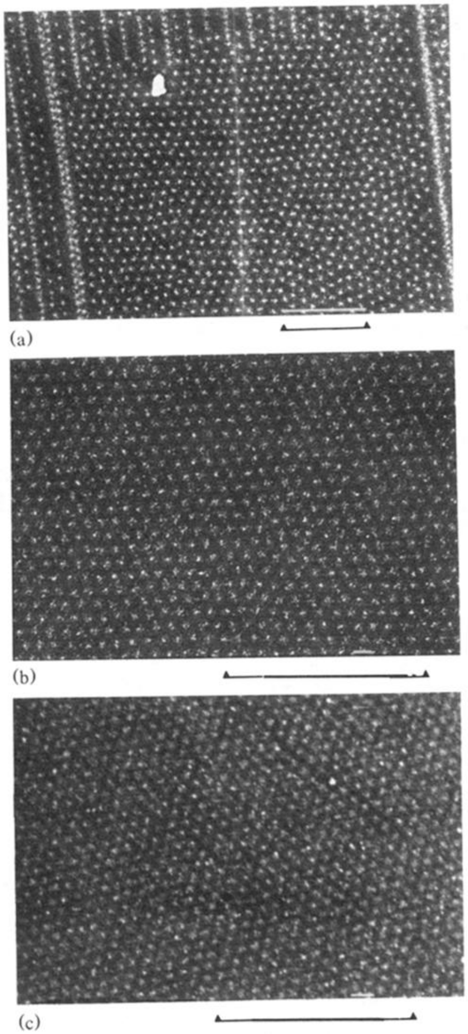

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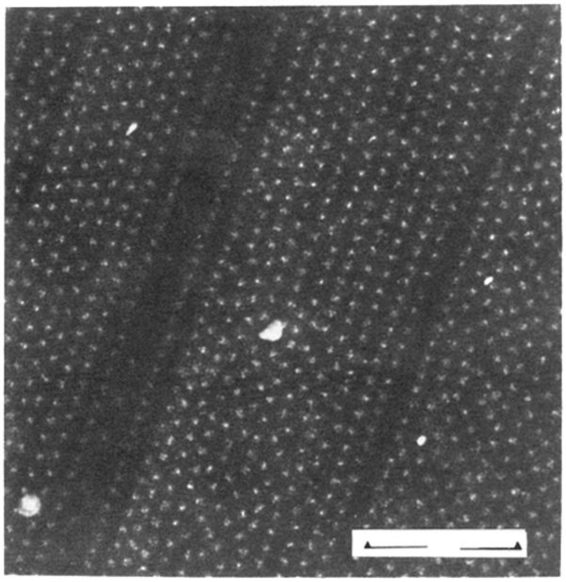

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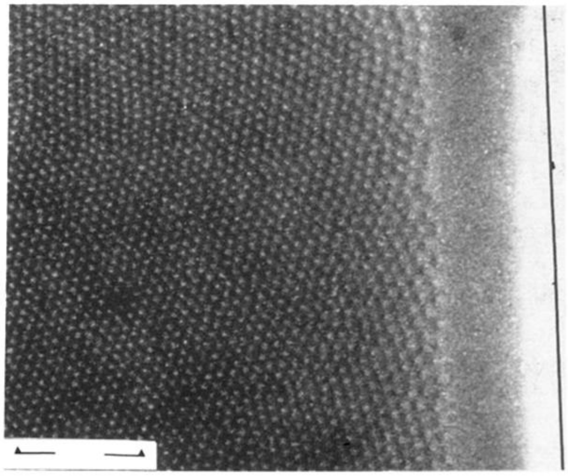

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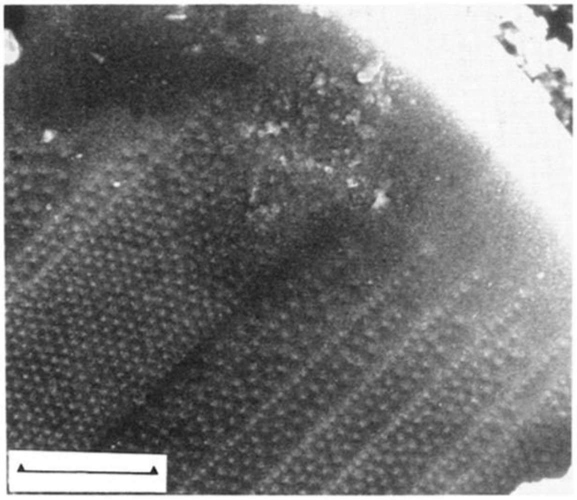

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