

## Neutron-irradiation effects on the $V$ - $I$ characteristics of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ twinned crystals: Linking transport results in a variety of copper oxide superconductors

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We have investigated the depinning process of magnetic flux vortices in twinned crystals of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  in intermediate applied magnetic fields parallel to the crystallographic  $c$  axis through precise resistivity measurements before and after fast-neutron irradiation. Of particular interest for each field is the power-law dependence of the electric field on current density,  $E \propto J^n$ , at a well defined temperature which separates a nonlinear region below from a more linear, dissipative region above. We find that  $n$  shows a striking dependence on the estimated value of the Larkin-Ovchinnikov length. The relation is not only manifest before and after irradiation but also for a variety of samples, including films, crystals, and at least two copper oxides other than  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ . [S0163-1829(97)50218-9]

Of fundamental interest to the study of high-temperature superconductors (HTS) is the character of fluxon depinning in an applied current. The theoretical  $H$ - $T$  diagram for the copper oxides has been divided into a diverse array of special cases concerning the interplay of pinning, thermal activation, collective effects, entanglement, etc., of magnetic flux lines.<sup>1</sup>

Many experimental techniques have been used to test these models. In particular, four-terminal dc  $V$ - $I$  measurements have been made for a wide range of chemical compositions and sample types. The vortex glass (VG) theory<sup>2</sup> has enjoyed success in studies of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) films,<sup>3</sup> except at very high ( $B > 5$  T as in Ref. 4) and very low fields ( $B < 50$  mT as in Ref. 5).

While a clear understanding of vortex behavior in clean untwinned YBCO crystals is emerging,<sup>6</sup> the discussion of resistivity measurements in twinned and/or defect-ridden YBCO crystals lacks consensus.<sup>7-19</sup> Outlooks include those who claim that the model of a second-order transition is quite relevant in crystals given resistive measurements of sufficient signal resolution<sup>16</sup> and those who claim that no finite-temperature glass transition can be realized in three dimensions when the effects of screening are considered.<sup>19</sup> The Bose-glass (BG) model<sup>9</sup> is expected to be relevant in the presence of correlated disorder and has been supported experimentally.<sup>14</sup>

We present four-terminal resistivity versus current-density measurements of twinned YBCO crystals including the effects of fast-neutron irradiation, and an interpretation of present and previous results for a variety of materials. We use heavily twinned crystals to suppress the abrupt lattice melting that one expects in clean, untwinned crystals.<sup>6</sup> Previous irradiation studies of resistivity in single crystals have emphasized proton damage,<sup>9,11</sup> columnar defects,<sup>12,14</sup> or electron damage,<sup>15</sup> but fast-neutron flux creates the most isotropic and most pointlike damage of all irradiations employed to date. The damage and pinning enhancement resulting from such irradiation of YBCO crystals have been well documented.<sup>20,21</sup>

The YBCO crystals were synthesized by the self-flux method in Au crucibles at Argonne National Laboratory. The  $T_c$  values for samples 1 and 2 are, respectively, 90.7 K and 92.6 K at ambient field with transition widths much less than 0.5 K. The fast-neutron irradiation, at a flux of  $1.5 \times 10^{17}$  n/cm<sup>2</sup> was carried out at the Missouri University Research Reactor (MURR).

The experimental procedure for taking and analyzing dc  $\rho$ - $J$  data has been described previously.<sup>5</sup> For each sample and each field, resistivity-current-density isotherms were taken around  $T_p$ . This temperature is defined by a power-law dependence of  $E$  (hence  $\rho$ ) on  $J$  and the separation of two apparently distinct types of behavior. Below  $T_p$ , isotherms display increasing negative concavity and very little spread with changing temperature, while those above  $T_p$  show both positive concavity on a  $\log \rho$ - $\log J$  plot and a marked increase in spacing with changing temperature. For sample 2, data were collected before and after fast-neutron irradiation (Fig. 1).

We examine the power-law slope— $n$  taken from  $E \propto J^n$ —of the  $T_p$  isotherm at each field. We determine a  $T_p$ , as shown in Fig. 1, by choosing the best fit to power-law dependence of an isotherm that separates the closely bunched, nonlinear isotherms below from the less crowded isotherms of positive concavity above. It is important to note that the resistivity resolution of the experiment can influence the determination of  $n$ . When compared to the film data, the resistivity floor of the crystal measurements is higher by three orders of magnitude. This is primarily due to geometrical factors. We have tried cutting and reanalyzing data sets for films and crystals at higher  $\rho$  levels. This does not always appreciably change the reported  $n$ , but when it does, it necessarily *decreases* the value. For instance, cutting the film data for 2.5 T at the crystal resistivity floor gives  $n=2.5$  instead of 3.2. Similarly, if one extended the resolution of the crystal measurements, we would expect, if anything, higher values of  $n$  than those we report here.

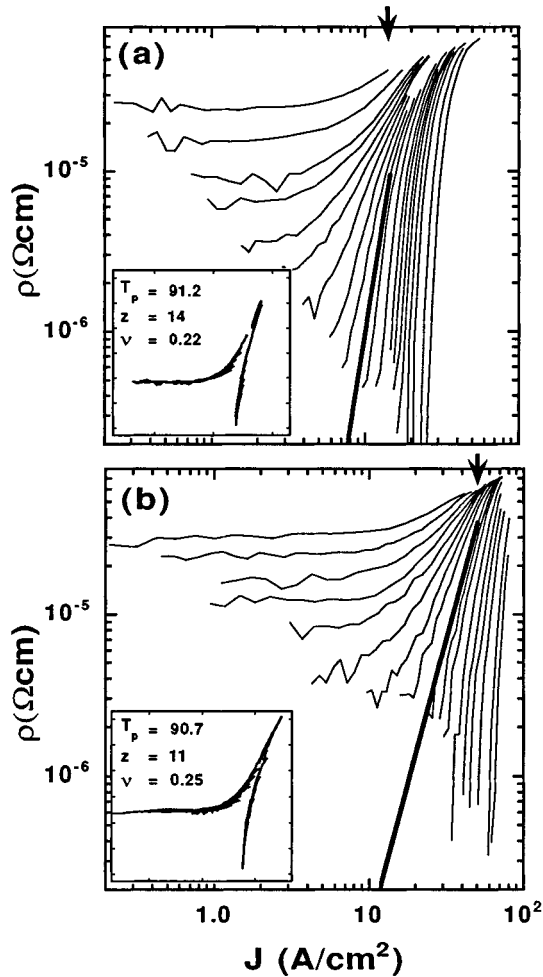


FIG. 1.  $\rho$ - $J$  isotherms for an YBCO crystal with 0.5 T parallel to the  $c$  axis (a) before and (b) after fast-neutron irradiation. The heavy power-law lines highlight the fit at  $T_p$  in both cases, while the arrows denote  $J_d$ . Temperatures range from 89.5 to 92 K for (a) 89 to 91.8 K for (b), with an average spacing of 100 mK and temperature stability of 5 mK each. In both cases, however, the spacing below  $T_p$  is about 200 mK; the isotherms are very closely spaced in this region, and some data are therefore excluded for clarity.

The other key to this analysis is the Larkin-Ovchinnikov length,  $L_p$ . Larkin and Ovchinnikov first predicted that the order of the Abrikosov vortex lattice would be disrupted by pinning centers in a nonideal sample.<sup>22</sup> The length  $L_p$  describes the average size of pinned vortex lattice domains that retain an Abrikosov-like lattice structure. One expects this length to be larger in crystals than in more disordered samples, such as thin films. We calculate an estimate of this length in the fashion used by Dekker *et al.*,<sup>23</sup> namely,

$$L_p = \left( \frac{kT_p}{\gamma\phi_0 J_d} \right)^{1/2}. \quad (1)$$

Here,  $\gamma$  is the mass anisotropy of the material defined by  $\gamma = (m_{ab}/m_c)^{1/2}$  (1/5 for YBCO, 1/20 for  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ , and 1/50 for  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$ ),  $\phi_0$  is the flux quantum, and  $J_d$  is the current at which the  $T_p$  isotherm starts to deviate from power-law behavior (see Fig. 1). Using Eq. (1) we obtain reasonable numerical values for our films (from 40–50

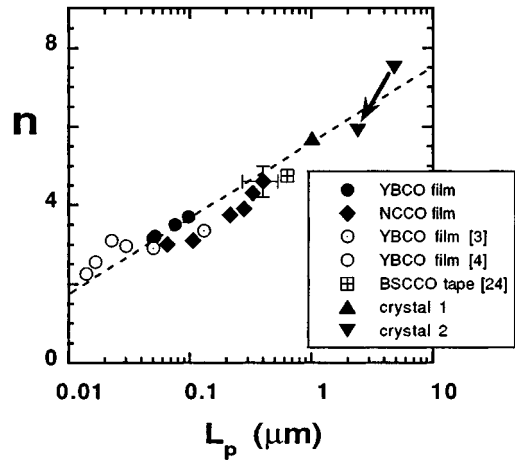


FIG. 2. Results for a variety of samples and fields ( $\parallel c$ ). Solid symbols denote data taken by the authors, including previously published thin-film work, while open symbols denote data from the literature. Multiple points for a given sample show the increase of  $n$  and  $L_p$  in decreasing applied fields. The crystal points each correspond to 0.5 T and the arrow highlights the effect of irradiation. Uncertainties are typically no larger than those shown for the NCCO data point.

nm at high fields to 100 nm or more for low fields), for twinned crystals (1–5  $\mu\text{m}$  for our two twinned YBCO crystals), and for the  $\text{Bi}_2\text{Sr}_2\text{Ca}_2\text{Cu}_3\text{O}_{10}$  tape (0.7  $\mu\text{m}$  extracted from Ref. 24).

An interesting picture emerges when we plot  $n$  versus  $L_p$  as in Fig. 2. We attribute the dependence of  $n$  on  $L_p$  to the sensitivity of depinning current distributions to the density of vortex lattice dislocations. Each point corresponds to a given sample and a specific applied magnetic field parallel to the  $c$  axis. In this view, we can easily compare the results for the YBCO crystals to those of our thin films of YBCO,<sup>5</sup> and  $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4-\delta}$ .<sup>25</sup> In addition, we have included data from other researchers.<sup>3,4,24</sup> The small uncertainties in Fig. 2 are due mostly to the inherent uncertainty in choosing  $T_p$  (which affects both  $n$  and  $L_p$ ). Though a wealth of resistivity data exist for high-temperature superconductors, many of the published transport measurements were not suitable for this analysis for one or more of the following reasons: they did not include data at high  $J$  (flux flow regime) that help one determine  $J_d$ ; the isotherms had spacings of more than 1 K and/or overly coarse current resolution, such that an accurate  $n$  value could not be estimated; the published data were too noisy to yield reliable values for  $n$  and/or  $J_d$ . Even given these criteria, the data presented are not meant to be exhaustive. We note that crystals with columnar pins have also shown a decreasing  $n$  value for increasing applied fields.<sup>12</sup>

Multiple points appear for a given sample because the calculated estimate for  $L_p$  always increases for decreasing applied fields (e.g., our YBCO film data points range from 5 T to 15 mT) mainly due to a marked decrease in  $J_d$  (and, to a lesser extent, an increasing  $T_p$ ) for decreasing fields. Though at a constant temperature an augmented pinning volume is predicted for an increasing field,<sup>1</sup> we emphasize that the points for a given sample in Fig. 2 are not isothermal. In fact, the field dependence we observe at  $T_p$  matches the arguments of Worthington *et al.* who noted both a decrease in

thermal smearing at lower values of temperature (corresponding here to higher values of applied field) and an increase in pinning effectiveness with the decreasing lattice constant,  $a_0$ .<sup>9</sup>

It is tempting to interpret  $L_p$  as an inverse measure of pinning site density and, hence, as a direct measure of sample purity, but Eq. (1) is derived by comparing available thermal energy to the energy required to nucleate a flux loop. At  $T_p$ , the thermally nucleated fluctuations are as large as the regions of Abrikosov-like order and the estimate for the Larkin-Ovchinnikov length in Eq. (1) follows. Since it is extracted from evidence of vortex depinning, it cannot reliably probe length scales lower than  $a_0$  for any given sample and field. The interpretation of  $L_p$  should be limited to, at best, a rough estimate for the effect of disorder on a system of flux lines.

In addition, Fig. 2 cannot be easily divided into single-vortex, small-bundle, and large-bundle pinning regimes for which different behaviors are predicted (Blatter *et al.*).<sup>1</sup> The ratio of  $L_p$  to  $a_0$  is generally much higher for crystals than films, which is evident from Fig. 2 since the data for each sample include a point for either 0.5 T or 1.0 T so that an intersample comparison can be made with a constant  $a_0$ . However, for a given sample we see  $L_p$  decrease and, simultaneously,  $L_p/a_0$  increase with increasing field. Hence, larger values of  $L_p$  do not necessarily correspond to greater numbers of vortices in the pinning volume.

The striking dependence of  $n$  on  $L_p$  suggests a continuous link between the heretofore distinct interpretations of thin-film data and crystal data. In addition, it has ramifications for theories which purport a universality class that depends on  $n$ . The power-law exponent is directly related to the VG dynamic scaling exponent  $z$ ; assuming a three-dimensional system,  $n = (z + 1)/2$  and since the VG theory predicts universal  $z$  values in the range of 4–7,<sup>2</sup> the results presented in Fig. 2 would seem to confine the theory to a small range of  $L_p$  values (20 to 200 nm). Though the BG model is expected to be more relevant to the data of twinned crystals, similar confinement follows as it assumes  $n = (z' + 1)/3$  with  $z'$  values proposed in the range of 6–8.<sup>9</sup> Several different, not necessarily mutually exclusive, explanations of the  $n$ - $L_p$  relation deserve attention.

Some percolative models, particularly that of Yamafuji and Kiss (YK), are based on distribution functions of pinned fluxoid clusters.<sup>17,18</sup> Of the many simulations of vortex depinning that have appeared in the literature,<sup>1</sup> our data seem most consistent with the picture of variable pinning distributions used by both YK and by Wördenweber.<sup>26</sup> Wördenweber convincingly simulated  $V$ - $I$  traces from Gaussian distributions of depinning currents (hence Gaussian distributions of pinning forces). This approach has enjoyed wider acceptance in low- $T_c$  literature than in high- $T_c$  literature. In particular, Warnes and Larbalestier treated a superconducting sample as a series of pinned vortex filaments, each with its own depinning current, such that the sample could be suitably described by a current distribution function.<sup>27</sup> They showed that, in principle, depinning current distributions followed from clean  $V$ - $I$  data via

$$Af(I) = \frac{d^2V}{dI^2}, \quad (2)$$

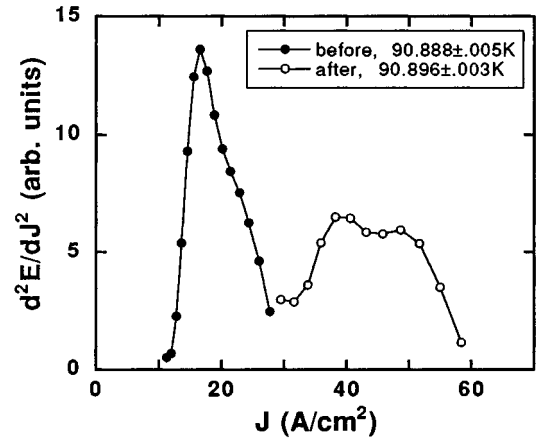


FIG. 3. Second derivatives of isothermal data approximating the form of  $f(J)$  for crystal 2 before and after fast-neutron irradiation. Both data sets correspond to 0.5 T applied  $\parallel c$ .

where the factor  $A$  incorporates details of the flux flow process. Based on this, we examine  $d^2E/dJ^2$ ; this approximation produces reasonable distributions (Fig. 3). Though one might expect the pinning sites introduced by fast neutrons to show a much different  $f(I)$  than the native pinning sites, we note that irradiation-induced and native pinning sites have been found to display similar pinning strengths.<sup>28</sup>

Using the VG scaling approach, it is possible to collapse the data for a given crystal and field onto two curves (see insets, Fig. 1) as we did in our thin film efforts.<sup>5,25</sup> However, as we noted there, the critical exponents are not universal and they do not match values predicted by the VG theory. Whereas the VG model is considered more applicable to films than crystals, the BG model depends on correlated disorder and we assume that it is relevant for twinned crystals,<sup>9</sup> especially for data collected prior to fast-neutron irradiation. However, the derived exponents do not fall near the BG predicted values for  $z'$  and  $\nu'$ . The troubling lack of a universality class suggests the exploration of interpretations of dc  $V$ - $I$  data that do not involve a phase transition, in agreement with the conclusions of Legris *et al.*<sup>12</sup> who measured  $V$ - $I$  data in YBCO crystals with columnar defects. YK simulate scaling collapses from a percolative standpoint.<sup>17</sup> Though they predict field-dependent and disorder-dependent critical exponents, they have not yet done so in a quantitative fashion.

It appears that one could pursue a phenomenological modification of the scaling relations, taking into account the role of  $L_p$ , so that a new set of critical exponents might emerge showing true universality. This is especially tempting when one considers the qualitative similarity of much of the resistivity data. Using the VG construction as a starting point and focusing only on the YBCO data in Fig. 2, the best fit of the data yields

$$z = a_1 + a_2 \ln(L_p). \quad (3)$$

This can be recast in terms of  $J_d$  since it is the variation in  $J_d$  in Eq. (1) that most strongly affects  $L_p$  over the range we have explored. Similarly, one could present the static scaling exponent,  $\nu$ , as a function of  $L_p$  or  $J_d$ , and hence by accounting for the effect with an extra parameter, we could

define a  $z^*$  and  $\nu^*$  which would remain constant. However, the introduction of an extra parameter does not produce a universal set of scaling functions  $F_{\pm}$ . The shapes of  $F_{\pm}$  depend directly on the original  $z$  and  $\nu$ . As a major justification for critical scaling is finding universal functions, there is little merit in this modification.

To make a more meaningful effort to modify the scaling analysis, one might incorporate the new length scale at a more fundamental level. In the language of the renormalization group,  $L_p$  appears to be a relevant perturbation to the system since it forces the flow under renormalization to a new, distinct, fixed point. It is beyond the scope of this effort to modify the scaling approach, but such a modification should be quite fundamental. The universal power-law dependence of  $E$  on  $J$  at the critical isotherm is a crucial part of critical scaling hypotheses, since it is precisely this that assures the cancellation of the diverging glass correlation length at  $T_p$ .

In summary, the dependence of  $n$  on  $L_p$  casts doubt on the relevance of  $V$ - $I$  measurements in exploring second-order vortex phase transitions in any high- $T_c$  system. Our work also demonstrates that the depinning process in crystals of YBCO is related in a continuous manner to that in both thin films and other copper-oxide superconductors. If the  $f(I)$  outlook is appropriate to the materials included, the results point to a broadening of depinning current distributions with an increase of the average pinning site density.

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