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Flux-pinning mechanisms in thin films of $YBa_2Cu_3O_x$

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We consider data from several experiments taken at very small induction in order to discern the nature of the pinning interaction in some thin-film samples of $YBa_2Cu_3O_x$ with very large critical current densities. Analysis of typical pinning energies and critical current densities indicate that the pinning is due to a large density of point defects. We propose a simple model of pinning by point defects in the CuO_2 planes that predicts a spacing between defects of 53 Å. This large defect density may help to explain other properties of these films.

Critical current densities typically observed in thin films of $YBa_2Cu_3O_x$ are very high, much higher than in bulk samples and somewhat higher even than in single crystals. It is clearly of interest to understand the origin of these high critical current densities. More specifically, it is of interest to establish the origin of the flux pinning in these thin films. Most studies of flux pinning in the high-temperature superconductors have focused on the high-field regime where the bare vortex-defect interaction is complicated by the interactions between vortices. Recent experiments on thin films at very low magnetic induction are yielding critical current densities and estimates of the pinning energies that presumably reflect the bare vortex-defect interaction. The interpretation of these data is considerably simpler than the high-field data and can reveal important facts about the flux-pinning processes active in these materials. In this Rapid Communication we analyze these data using simple models. We find on general grounds that the data imply a very large density of pinning sites, beyond the density of the extended defects seen in typical transmission electron micrographs.[1] We conclude that a very high density of still unidentified local defects must be present in these high current-density materials.

The experiments that we consider in our analysis include transport critical current measurements of thin lines,[2] magnetization hysteresis,[3] and low-frequency noise measurements.[4] These measurements have been performed on nominally identical thin-film specimens of $YBa_2Cu_3O_x$ prepared[3] *in situ* by single-target sputtering. These films are from 2000 to 4000 Å thick, completely c axis textured, and extremely smooth. Since the vortices are principally in the c-axis direction in these experiments, we need to consider only pinning in the a-b plane. We focus on the data at low temperatures in order to remove the complications associated with large thermal fluctuations. Typical critical current densities J_c as measured by transport or magnetization are about 3×10^7 A/cm^2. Estimates of the pinning energy U_0 range from 50 to 200 meV according to the noise and transport experiments.

A pinned vortex has a lower free energy than a free vortex in a superconductor. This difference can be characterized by an energy per unit length ε so that a pinned vortex segment of length l_p has a smaller energy of magnitude $\Delta E=\varepsilon l_p$. The amount of force F_p necessary to depin the vortex segment is related to the pinning energy by the characteristic distance δ that a vortex must be displaced from the pinning site for the vortex-defect interaction to $F_p\approx\Delta E/\delta=\varepsilon l_p/\delta$. In absence of collective effects, J_c is obtained by equating the Lorentz force and the pinning force. If a vortex segment of length l is pinned over a portion of its length l_p ($l_p<1$), this yields $J_c\Phi_0 l/c=\varepsilon l_p/\delta$, where Φ_0 is the flux quantum and c the speed of light. Given J_c, ε, and δ, one can solve for l_p/l, the fraction of the vortex that is pinned,

$$\frac{l_p}{l}=\frac{\delta J_c\Phi_0}{\varepsilon c}\,. \tag{1}$$

We have considered two basic pinning mechanisms, core pinning and magnetic[5] pinning, and concluded that core interactions are most likely responsible for the pinning in these thin films. To examine core pinning we will assume that the core of the vortex sits in a defect that is "normal" in the sense that the order parameter is completely depressed in the vicinity of the defect. The energy gain per unit length ε_c of a vortex in such a defect is just the condensation energy in the core region;

$$\varepsilon_c=\frac{H_c^2}{8\pi}\pi\xi_{ab}^2=\frac{\Phi_0^2}{64\pi^2\lambda_{ab}^2}\,, \tag{2}$$

where ξ_{ab} and λ_{ab} are the coherence length and penetration depth in the a-b plane. If we assume a very sharp discontinuity in the order parameter then we realize the minimum value of $\delta\approx\xi_{ab}$ and the maximum value of J_c in Eq. (1). Substituting Eq. (2) into (1) with $\xi_{ab}=15$ Å, $\lambda_{ab}=1400$ Å, and the typical J_c yields $l_p/l=0.27$. This result is remarkable in that we have assumed the strongest pinning forces possible and arrived at the conclusion that the vortex must be pinned along $\frac{1}{4}$ of its length. For the local defect model that we present below this implies that the vortex is pinned about every fourth unit cell along the c axis, i.e., about every 47 Å. Any reduction in the amount that the order parameter is depressed or increase in the characteristic pinning length δ will only increase l_p/l. These simple considerations lead to the striking conclusion that the vortices in these $YBa_2Cu_3O_x$ thin films are extremely well pinned. At this point we still do not suppose that the pertinent defects are actually local in nature, only that the pinning is very frequent along a vortex line.

The first hint that the pinning defects are local in nature can be gleaned by considering the magnitude of the pinning energies determined from experiment. When considering an isolated vortex the relevant question to ask is what is the length of vortex-line d that actually depins (due to thermal activation, for example). The associated pinning energy would then be $U_0 \approx (l_p/l)\varepsilon_c d$. To determine the value of d consider the difference in energy of the two vortex configurations shown in Fig. 1. In Fig. 1(a) we show a straight vortex line intersecting a line of pinning sites that, at this point, we suppose could arise from a single extended defect or many local defects. In Fig. 1(b), we show a distortion of the same vortex line such that the line is pulled from its pinning sites by a characteristic distance Δx over a length d. We choose the distortion to have the geometry shown in Fig. 1(b) for computational simplicity. Also, since we shall assume $d \gg \Delta x$, this distortion minimizes the "kinks" in the vortex line. A severely distorted line with large kinks or with $d \approx \Delta x$ would not permit the simple analysis that we present below. The distorted line has larger energy for two reasons: (1) the vortex is pulled away from the pinning sites over a length d, and (2) the vortex line is longer, which gives an increased energy owing to the line tension. We call these two contributions ΔE_p and ΔE_t, respectively. We estimate

$$\Delta E_p \approx d\left(\frac{l_p}{l}\varepsilon_c\right). \tag{3}$$

ΔE_t is just the difference in line energy for the two configurations,

$$\Delta E_t \approx \int_{-d/2}^{d/2} dz \left(1+\left|\frac{dx}{dz}\right|^2\right)^{1/2} \tau\left(\frac{dx}{dz}\right) - \int_{-d/2}^{d/2} dz\, \tau\left(\frac{dx}{dz}=0\right).$$

In this expression we allow the possibility of an anisotropic mass because the line tension τ is a function of the direction of the vortex line (given by dx/dz, z axis$\parallel c$ axis).[6] If we assume that $|dx/dz| = \text{const} = 2\Delta x/d \ll 1$ we find

$$\Delta E_t \approx \left(\frac{\Phi_0}{4\pi\lambda_{ab}}\right)^2 \ln(\kappa)\frac{m_{ab}}{m_c}\frac{2\Delta x^2}{d}, \tag{4}$$

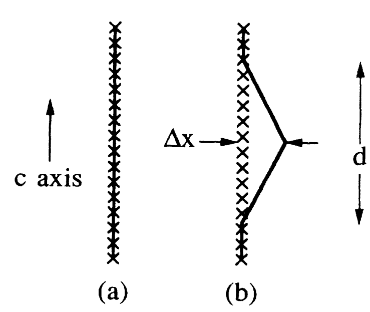

FIG. 1. (a) A vortex line in the c-axis direction is pinned frequently along its length. The pinning points are marked as ×'s. (b) The same vortex that is distorted from the pinning points a width Δx over a length d.

where m_{ab} and m_c are the mass values in the a-b plane and along the c-axis direction, respectively. The total energy of the distortion $\Delta E_p + \Delta E_t$ is given by

$$U = \frac{\Phi_0^2}{8\pi^2\lambda_{ab}}\left[\frac{1}{8}\frac{l_p}{l}\frac{d}{\lambda_{ab}} + \ln(\kappa)\frac{m_{ab}}{m_c}\left(\frac{\Delta x}{\lambda_{ab}}\right)^2\frac{\lambda_{ab}}{d}\right],$$

where we have used Eqs. (2)-(4). Notice that the portion of this energy associated with pinning increases with d while the portion associated with the line tension decreases. At the optimal value of d this function has a minimum value U_0, which we interpret as the characteristic pinning energy of the system. Minimization with respect to d yields

$$d = \Delta x\left(8\ln(\kappa)\frac{m_{ab}}{m_c}\frac{l}{l_p}\right)^{1/2}$$

and

$$U_0 = \left(\frac{\Phi_0}{4\pi\lambda_{ab}}\right)^2 \Delta x\left(2\ln(\kappa)\frac{m_{ab}}{m_c}\frac{l_p}{l}\right)^{1/2} = 2\frac{l_p}{l}\varepsilon_c d. \tag{5}$$

The evaluation of these expressions requires a specification of the size of the lateral distortion Δx. For now we invert the problem and use Eq. (5) to solve for Δx, which in order to achieve permanent depinning should be of order the distance between pinning sites. Generally Δx and U_0 may depend on the size of the applied current for the experiment of interest. We neglect such corrections, since it is our goal only to establish rough estimates of the relevant quantities. Assuming that $l_p/l = \frac{1}{4}$, $m_{ab}/m_c = \frac{1}{25}$, $\lambda_{ab} = 1400$ Å, $\xi_{ab} = 15$ Å, $\kappa = \lambda_{ab}/\xi_{ab}$, and a typical value $U_0 = 100$ meV, we find that $\Delta x = 38$ Å and $d = 92$ Å. We point out that this Δx is much smaller than the typical 500-Å spacing of the extended defects observed in TEM and that Δx is roughly the average separation between pinning sites suggested above. If, in fact, the relevant Δx were the spacing between the extended defects, then the pinning energy of Eq. (5) would be about 1.3 eV, significantly higher than the experimental values. These observations require that the pinning must be due to a large density of local defects.

The remaining issue is what local defect density is actually necessary to allow such frequent pinning of a vortex line and is this density consistent with the defect spacing implied by the analysis of the pinning energy. As a model we consider pinning by small regions of reduced-order parameter in the strongly conducting CuO_2 plane pairs. These local defects are presumably caused by some crystalline imperfection in the vicinity of the planes. We assume that at each pinning site a vortex gains the condensation energy of a length of vortex equal to the lattice spacing: $\Delta E_p(\text{per pin}) = c\varepsilon_c$, where c is now the lattice spacing. This is equivalent to saying that all the condensation energy resides in the planes. Consider an isolated vortex, nominally in the c-axis direction, that "wanders" through the crystal in order to pick up pinning sites. The vortex line will decrease its free energy when it picks up a pinning site, but it will increase its free energy by wandering to pick up those sites. The competition between these two effects will determine how frequently a vortex line is pinned. In Fig. 2 we present a vortex line pinned by local defects at the points marked as ×'s. The vortex has a characteristic distortion width s and length t between pinning sites. The increase in the free energy associated with this distortion is just the quantity ΔE_t of Eq. (4) if we make the replacement $\Delta x \to s$ and $d \to 2t$. The net free-energy gain of the vortex segment of length t is just $\Delta E_p - \Delta E_t$, which we can write as [using Eqs. (2) and (4)]

$$\Delta E_p - \Delta E_t = c\varepsilon_c - 2\ln(\kappa)\frac{m_{ab}}{m_c}\frac{s^2}{t}\varepsilon_c = t\left(\frac{l_p}{l}\varepsilon_c\right). \quad (6)$$

The last equality enforces the frequent pinning necessary to explain J_c [Eq. (1)]. Since the vortex lines will take maximum advantage of the available defects to lower their free energy, we want to find the minimum possible defect density which will still satisfy Eq. (6). Given Fig. (2), this implies that the defect density is approximately $n_v = 1/\pi s^2 t$. To find the minimum density we eliminate s^2 in the expression for n_v using Eq. (6) and minimize n_v with respect to t. The result is

$$n_v = \ln(\kappa)\frac{m_{ab}}{m_c}\left(\frac{l_p}{l}\right)^2 \frac{27}{2\pi c^3}.$$

For the parameters mentioned above we find $n_v \approx 3\times 10^{19}$ cm^{-3}, which corresponds to a defect every 188 unit cells. Also, $t = \frac{2}{3}(l/l_p)c = 31$ Å and $s = (\pi t n_v)^{-1/2} = 18$ Å. This value for t is a refinement of our previous estimate of the frequency of pinning sites that accounts for the competition between pinning and line energies. Notice that the mass anisotropy plays a key role in reducing the necessary defect density by allowing the vortex to bend more easily. The density of defects in a single layer is then $n_p = cn_v \approx 3.5\times 10^{12}$ cm^{-2}. The spacing between the defects in a single plane is $\Delta x \approx (n_p)^{-1/2} = 53$ Å. Given the simplicity of the models employed, this result agrees remarkably well with the value $\Delta x = 38$ Å that we derived in our analysis of the pinning energies.

Before considering the effects of these local defects further, we pause to consider the approximations made in the above analyses. We have assumed in the pinning-energy analysis that the flux line is actually uniformly pinned along its length (but with an energy reduced by a fraction l_p/l). Since the pinning sites are presumably randomly distributed, the above analysis is only valid if d is much

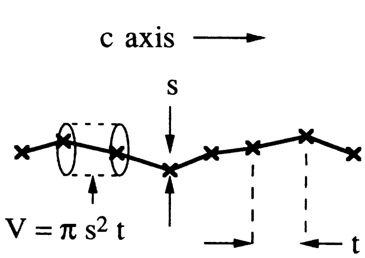

FIG. 2. A vortex line in the c-axis direction wanders through a superconductor picking up pinning sites. The characteristic distance between pinning sites is t and the characteristic lateral displacement of the vortex line is s. There is approximately one pinning site in a volume $V = \pi s^2 t$, as indicated by the cylinder.

larger than the distance between pinning sites t. In this case $t/d = \frac{1}{3}$, which is fair. We also assumed in Eq. (4) that $dx/dz \approx 2\Delta x/d \ll 1$. In this case $2\Delta x/d = 0.8$ so that the approximation is poor. Similarly, $s/t = 0.56$. The marginality of these approximations is acceptable, however, given the crudity of the models and as our main goal is to provide an overall framework for understanding the pinning interaction.

The analysis above indicates a very high density of local defects. A large defect density would have significant impact on other properties of the superconductor. At each defect, since it is a pinning site, the order parameter must be suppressed out to a distance $\xi_{ab} = 15$ Å around it. If the spacing between defects in the plane is only 53 Å, then a very substantial fraction of the plane is effectively normal. As the temperature increases this effect is exacerbated by the increase in the size of the coherence length. Though we have not observed such small defects directly, there is considerable indirect evidence for their existence.[7] For example, these films typically have transition temperatures of 80 to 86 K and c-axis lattice parameters of 11.72 to 11.8 Å. Further, these differences are robust against annealing in oxygen at moderate temperatures. In ceramics, crystals, and even postannealed thin films such treatment readily changes the oxygen content on the CuO chain layers. These observations may be a consequence of a large density of very small defects. We can estimate the effect on the T_c of the material by modeling the interface between the defect and the strongly superconducting material as a normal-superconducting interface.[8] The resulting depression in T_c is $\Delta T_c/T_c \approx [\pi\xi_{ab}(0)/(2\Delta x)]^2 \approx 0.21$. Considering the crudeness of the approximation, this value agrees fairly well with the observed values of about 0.1. Also, the penetration depth will be enhanced because the volume of the superconducting condensate is reduced. A rough estimate of this effect yields an enhanced low-temperature penetration depth $\lambda'_{ab} \approx \lambda_{ab} \times [\Delta x^2/(\Delta x^2 - \pi\xi_{ab}^2)]^{1/2} = 1600$ Å for $\Delta x = 53$ Å and $\lambda'_{ab} = 2000$ Å for $\Delta x = 38$ Å. These values somewhat underestimate the measured values of these films of 2500 Å, but seem reasonable given our simplistic model of the penetration-depth enhancement.[9] Hence the high local defect density that we propose is consistent with other properties of these thin films.

A similar analysis of the bismuth- and thallium-based superconductors is hampered by a lack of data. Nonetheless it is possible that the larger anisotropies will introduce some new considerations. If, for example, we leave all the parameters unchanged except the mass ratio in Eq. (5) we find that $U_0 = 16$ meV and $d = 15$ Å for $m_{ab}/m_c = 1000$. In this case the approximations made in the analysis are not at all good. Since $d \approx c$, an appropriate model would have to address carefully the actual layered nature of the superconductivity. Also, a similar analysis of $YBa_2Cu_3O_x$ single-crystal samples may yield important clues as to the nature of the defects in these materials.

In conclusion, we have examined the results of several experiments at very small induction in order to study the pinning mechanism in some thin-film specimens of Y-$Ba_2Cu_3O_x$. We have shown that the large critical currents that we obtain in these materials imply that the vortices are pinned very frequently along their length by core interactions. The pinning energies observed suggest that the pinning is due to a large density of local defects. A model based on pinning by local defects on the CuO_2 planes yields a spacing that agrees well with the analysis of the pinning energies. This large defect density may explain other physical properties of the cuprate superconductors.

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[5]Magnetic pinning is the result of changes in the field and current distribution outside the vortex core when a vortex sits on a defect. Since magnetic pinning occurs on a length scale λ_{ab}, the only evident sources of magnetic pinning in these materials are the many extended defects observed by TEM such as twin boundaries, grain boundaries, and stacking faults. Simple considerations reveal magnetic pinning energies ε_m that are at best comparable to ε_c. Equation (1) then forces us to conclude that $\delta \approx \xi_{ab}$, which is inconsistent with the pinning extending over a length scale λ_{ab}. Thus the strong pinning observed in these materials is likely not magnetic in origin.

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