Flux pinning and critical currents at low-angle grain boundaries in high-temperature superconductors

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Calculations of the pinning potential at low-angle grain boundaries in high-temperature superconductors are presented which fully incorporate the periodic nature of the low-angle boundary. A nonlocal kernel provides a smooth transition from an Abrikosov vortex far from the boundary to a Josephson vortex near the dislocations. We examine the angular dependence of critical current in the two idealized limits of pure strain and pure band bending. Recent data appear limited by band bending with significant potential for improvement through doping. © *2002 American Institute of Physics.* [DOI: 10.1063/1.1502907]

The seminal experiments of Dimos *et al.*[1] and other workers[2–5] revealed the remarkable sensitivity of the critical current in the high-temperature superconductors to the presence of a single grain boundary. Over the range of misorientation angles from $\sim 5°$ to 45° the critical current drops approximately exponentially by about 3 orders of magnitude. Above about $4°$ evidence of weak coupling is found,[6] indicative that the boundary is acting as a Josephson junction.[7] More recently it has been shown that doping can have a significant effect on the grain boundary critical current and a band-bending model has been proposed to account for the effect.[8–10]

In the regime above 10° misorientation the exponential drop has been successfully modeled as transmission through a continuous, long Josephson junction with a width determined through a structural unit description of grain boundary atomic structure.[11] Pinning models incorporating a nonlocal kernel have been developed for this regime,[12–14] and the effect of inhomogeneities have been investigated in a linear approximation.[15] At low angles, however, the situation is more complex as the boundary comprises an array of discrete dislocation cores and the vortex cannot be assumed to remain in the Josephson junction. The boundary plane alternates between ''bad'' (Josephson-like) regions at the dislocation cores and ''good'' regions in between. A Ginsburg–Landau formulation is appropriate for conditions when the coherence length is *large* compared to the dislocation spacing along the boundary, and the critical current is determined by the fraction of ''good'' grain boundary.[16] The situation is quite different if the dislocation spacing exceeds the coherence length which occurs for misorientations below $4°$, the regime important for coated conductor technologies.[17,18] Here *flux pinning* will determine the critical current, and the inhomogeneous nature of the boundary cannot be ignored. In this letter we present a generalization of previous treatments in which the vortex changes smoothly from an Abrikosov-like vortex away from the cores to a Josephson-like vortex in the vicinity of the cores. By including both the nonlinearity of the kernel and the finite extent of the Josephson junction we allow the vortex to explore the good regions of the boundary plane. Thus, we incorporate the minimum new physics necessary to give a physically realistic description of a low angle grain boundary.

Initial studies assumed the bad regions had a radius determined by strain[19] which destroys the delicate Cu $3d$–O $2p$ hybridization.[20] Previous studies[13,21] showed $D_B(\theta)/D(\theta) = 1.168\sqrt{\theta}$.[22] With increasing misorientation θ, the width of the nonsuperconducting zone $D_B(\theta)$ decreases, as does the dislocation spacing $D(\theta) = b/2\sin(\theta/2)$, where b is the Burgers vector.

There is increasing evidence for strong band-bending effects in perovskites and related materials due to an intrinsic nonstoichiometry at the dislocation cores.[23,24] Each dislocation core is surrounded by a hole-depletion zone such that the total number of depleted holes equals its charge. In a low angle boundary, and we therefore assume D_B to be independent of θ. We calculate pinning barriers for two idealized limiting cases, first, with D_B given by Eq. (1) assuming no band bending, and second with D_B to be $D_B \ll D(\theta)$, determined only by band bending. In reality both effects will occur simultaneously but we separate them here to demonstrate their very different angular dependence.

To calculate the pinning potential we use the approach of Agassi *et al.*[25] For a coordinate system $\{x,y\}$ perpendicular and parallel to the boundary [Fig. 1(a)], the critical current density *across* the grain boundary $j_x(y)$ involve the condensate gauge-invariant phase discontinuity $\chi(y)$ [Fig. 1(b)]. In a good passage the current–phase relation is as in the bulk. For a point x_M within the boundary width d_{GB}, as

$$j_x^{\mathrm{good}}(y) = -\frac{c}{4\pi\lambda^2}\left\{ A_x(x_M,y) + \frac{\Phi_0}{2\pi}\left.\frac{\partial\phi}{\partial x}\right|_{x=x_M}\right\}, \qquad (1)$$

where A_x is the electromagnetic potential, $\Phi_0 = \pi\hbar c/|e|$ is the flux quantum, λ is the penetration depth and the sign

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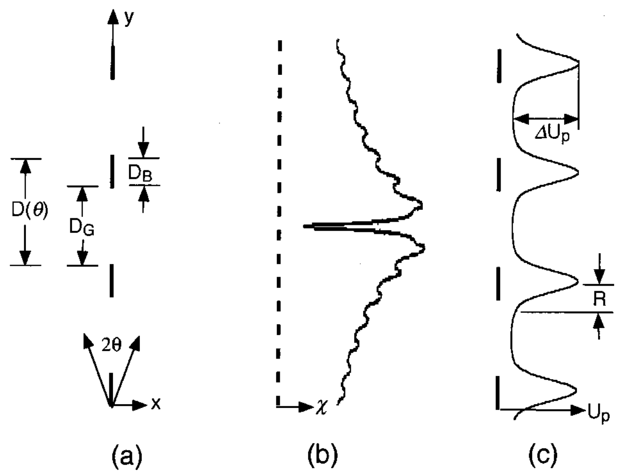

FIG. 1. Schematics showing (a) a two-dimensional model for a low angle grain boundary with angle 2θ between equivalent crystallographic directions in the two grains; (b) the gauge-invariant phase discontinuity $\chi(y)$ for a vortex located in the center of a good passage in a 3° grain boundary [note change of y scale with respect to (a)]; (c) the variation in pinning potential U_P parallel to the boundary plane which determines the critical current across the boundary. The pinning barrier ΔU_P rises over a distance R.

presumes for electron superconductivity. We make the approximation $\partial\phi/\partial x \cong [\phi(x=d_{\rm GB},y)-\phi(x=0,y)]/d_{\rm GB} \cong \Delta\phi/d_{\rm GB}$, and relate $\Delta\phi$ to $\chi(y)$ in the standard way:

$$\chi(y)=\phi(d_{\rm GB},y)-\phi(0,y)+\frac{2|e|}{\hbar c}\int_0^{d_{\rm GB}} dx A_x(x,y)$$

$$\approx \Delta\phi+\frac{2|e|}{\hbar c}d_{\rm GB}A_x(x_M,y). \tag{2}$$

Inserting $\Delta\phi$ from Eq. (3) into Eq. (1) gives a convenient linear relation for the critical current density, $j_x^{\rm good}(y)=-c\Phi_0/(8\pi^2\lambda^2 d_{\rm GB})=-j_G\chi(y)$, in which we identify j_G as the critical current of the grains.

In the bad regions we have $j_x^{\rm bad}(y)=-j_{JJ}\sin[\chi(y)]$ where j_{JJ} is a Josephson critical current. The grain boundary comprises a periodic array of these two domains, with critical current $j_x(y)=-Q_p(y)j_G\chi(y)+[1-Q_p(y)]j_{JJ}\sin[\chi(y)]$ where $Q_p(y)$ is a Kronig–Penney form factor taking the values 1 or 0 for good or bad regions, respectively. We then combine this with Ampere's law and the London equations and arrive at the key equation for the phase discontinuity χ:

$$\frac{\lambda_{JJ}^{2\,\rm good}}{\pi\lambda}\int_{-\infty}^{\infty}dy'\frac{\partial^2\chi(y')}{\partial^2 y'}K_0\left(\frac{|y-y'|}{\lambda}\right)$$

$$=-\frac{j_x(y+y_0)}{j_G}\sin[\chi(y)]$$

$$-\frac{2\lambda_{JJ}^{2\,\rm good}}{\lambda^2}\frac{y}{\sqrt{x_0^2+y^2}}K_1\left[\frac{1}{\lambda}\sqrt{x_0^2+y^2}\right], \tag{3}$$

where $\lambda_{JJ}^{2\,\rm good}=c\Phi_0/(16\pi^2\lambda j_G)$ is defined in analogy to the Josephson penetration depth. The solution reflects the periodicity of $j_x(y)$ [see Fig. 1(c)], and x_0 and y_0 are the location of the probe vortex. Inserting the solution into the free energy functional gives the pinning potential.[25] The present results are qualitatively different from a superconductor superlattice model reported previously.[21] The present model is fully two dimensional [see Fig. 1(a)] which allows unconstrained redistribution of the shielding currents. A plot of the periodic pinning potential per unit vortex length U_P is shown schematically in Fig. 1(c).

The full solution of Eq. (3) is computationally intense. However, since $j_{JJ}\ll j_G$ we take $j_{JJ}=0.01j_G$ in all cases. Second, the critical current across the boundary is proportional to the maximum gradient of U_P in the y direction,[26] which we approximate as $\Delta U_P/R$. The functional form of $U_P(y)$, is no longer required, just the pinning barrier ΔU_p [see Fig. 1(d)]. Assuming R scales with the size of the bad zone D_B, then R can be normalized out by taking critical current ratios for two different angles θ and θ_o[27]

$$\frac{j_c(\theta)}{j_c(\theta_o)}\approx\frac{\Delta U_P(\theta)}{\Delta U_P(\theta_o)}\frac{R(\theta_o)}{R(\theta)}. \tag{4}$$

Choosing θ_o sufficiently small, the critical current becomes relative to that in the *grains* j_G, exactly as measured experimentally.

The critical current across the boundary is now given by

$$\frac{j_c(\theta)}{j_G}\approx\frac{\Delta U_P(\theta)}{\Delta U_P(\theta_o)}\frac{D(\theta_o)}{D(\theta)}\quad\text{for the strain model}$$

$$or\,\frac{\Delta U_P(\theta)}{\Delta U_P(\theta_o)}\quad\text{for the band bending model,}$$

giving a robust prediction for the angular dependence of critical current for the two models.

We compare the predictions to available bicrystal data at 5 K, that of Verebelyi *et al.*[28] and Dimos *et al.*[1] One complication concerns the normalization to a low angle boundary. Verebelyi *et al.*, pointed out that grain boundaries of 1.8° or less are *indistinguishable* from the grains due to the presence of twin domains in YBCO; the lattice rotates by 1.8° across each twin boundary, so that intersections of twin domains would be expected to lead to 1.8° grain boundaries. Therefore, all grains contain 1.8° grain boundaries and *no smaller angle can be measured experimentally*.[29]

We therefore normalize our theoretical predictions to an angle of $\theta_o=1.8°$, and use the value $j_G=6.2\times10^7\ \mathrm{A\,cm^{-2}}$ extrapolated from Verebelyi *et al.*, which is close to the depairing limit. Figure 2 compares the predicted angular dependence of j_C with bad zones appropriate to the band bending and strain criteria. In the band-bending case, the constant width of the bad zone leads to a very rapid drop in critical current with angle. In effect, the current is pinched off as the dislocation spacing D reduces towards the size of the bad zone D_B. The choice of $D_B=3$ nm matches very well the data of Verebelyi *et al.* Above 6° D_G falls below zero, marking the transition to the tunneling regime, which is outside the scope of the present model.

In the strain model (dashed line, using identical parameters) the critical current is not pinched off so rapidly since the diameter of the bad zone reduces with increasing angle. It does not match the data of Verebeleyi *et al.* so well, but *parallels* the data of Dimos *et al.*[30] This may explain the very different angular dependence of the two sets of data. Using lower values of j_G, the fit to the Dimos data is even worse. A possible explanation is that their crystals may have had a larger angular spread within the grains. This would

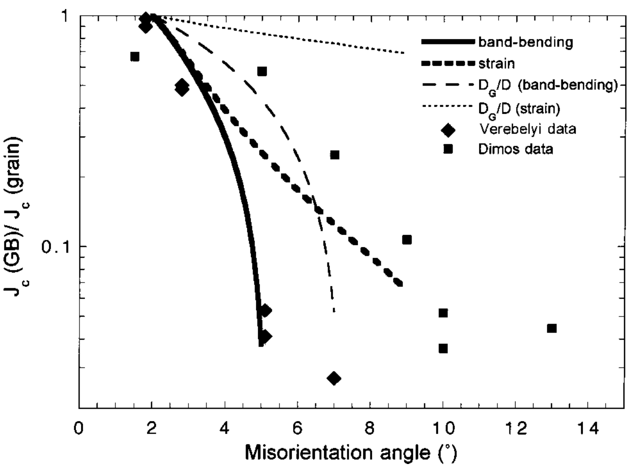

FIG. 2. Angular dependence of grain boundary critical current normalized to the grains comparing theoretical predictions for the band-bending (solid line) and strain (dashed line) mechanisms to the experimental results of Ref. 29 and Ref. 1. Also shown are D_G/D for the band bending and strain models, the geometric reduction in the good fraction of the boundary plane.

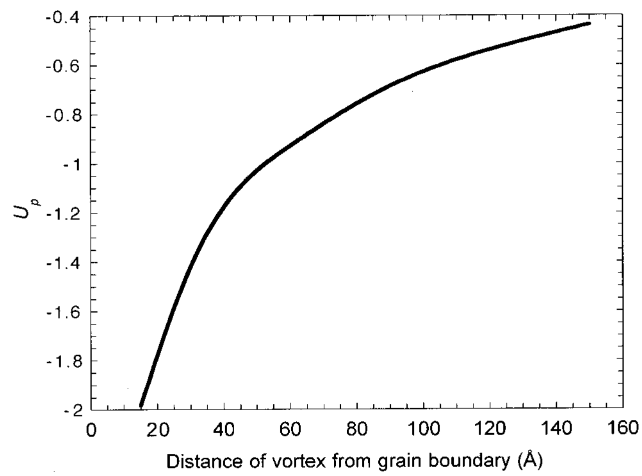

FIG. 3. Variation in pinning potential normal to the boundary plane which controls the critical current parallel to the boundary, calculated in the band bending model for a misorientation angles of 3° in units of $(\Phi_0/4\pi\lambda^2)^2$ per vortex unit length. The slope indicates a critical current parallel to the boundary of 1.7×10^8 A cm^{-2}, close to the depairing limit.

shift their experimental points to the right and explain their lower values of j_G. Note that in both models the critical current falls much *faster* than the geometric reduction in size of the good region D_G (shown as narrow lines in Fig. 2).[19]

At any given misorientation, the difference between the band bending and strain calculations indicates the potential for improving j_c through doping to a flat band condition. At angles of 5°–10° where the current is pinched off by band bending but not by strain, an *order of magnitude improvement* can be expected.

Finally we turn to the critical current *parallel* to the boundary, which depends on the cross section of the pinning potential surface *normal* to the boundary plane. Calculated values of the pinning potential as a function of position from the boundary plane are shown in Fig. 3. These are practically independent of misorientation angle and close to the depairing limit, as measured experimentally for a twin boundary.[31]

In conclusion, flux pinning calculations for the critical current density across and parallel to a low angle grain boundary are in reasonable agreement with experiment. Results suggest that band bending is a serious practical limitation at boundaries of 5°–10°, with the possibility of an order of magnitude improvement in critical current if doping can achieve a flat band condition.

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