Analysis and prediction of the critical current density across [001]-tilt $YBa_2Cu_3O_{7-\delta}$ grain boundaries of arbitrary misorientation angles

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(Received 20 July 1995)

A qualitative analysis of the expected dilatation strain field in the vicinity of an array of grain-boundary (GB) dislocations is presented. The analysis provides a basis for the prediction of the critical current densities (j_c) across low-angle $YBa_2Cu_3O_{7-\delta}$ (YBCO) GB's as a function of their energy. The introduction of the GB energy allows the extension of the analysis to high-angle GB's using established models which predict the GB energy as a function of misorientation angle. The results are compared to published data for j_c across [001]-tilt YBCO GB's for the full range of misorientations, showing a good fit. Since the GB energy is directly related to the GB structure, the analysis may allow a generalization of the scaling behavior of j_c with the GB energy.

I. INTRODUCTION

Measurements on $YBa_2Cu_3O_{7-\delta}$ (YBCO) thin films grown on bicrystal substrates have shown that the critical current density (j_c) across an individual YBCO grain boundary (GB) decreases with the misorientation angle between adjacent grains.[1–4] Highly disordered material around the dislocation cores was initially suggested to exhibit depressed superconducting properties thus reducing the effective superconduction area of the GB's.[1,2] Elliptical regions with a relatively large component of the strain field in a direction perpendicular to the GB plane were later assumed to have depressed superconducting properties.[5] The models described the behavior of low-angle GB's based on the variation in dislocation density as a function of misorientation angle. In thin films, additional strain may be induced in the boundary region from the interaction between the film and its substrate. Residual strain remaining from mismatches in lattice parameters and thermal-expansion coefficients would then add to the strain introduced by the dislocations.[6,7]

Several other investigations[8–10] have given additional information on the transport mechanisms and the microstructure of GB junctions. However, there is no conclusive evidence concerning the nature of boundary barrier. Gao *et al.*[9] studied the dislocation cores of [001]-tilt low-angle GB's in polycrystalline metallorganic chemical-vapor-deposited YBCO films by high-resolution electron microscopy (HREM) and image simulations. They found that low-angle GB's consist of a wall of discrete edge dislocations separated by relatively perfect lattice matching regions. From a comparison of HREM images with simulated images, the dislocation cores appeared to be Cu rich with a core radius of about 1 nm. It was inferred that the transition from strong coupling to weak coupling observed near misorientation angles of 11° by Dimos, Chaudhari, and Mannhart[2] was due to overlap of Cu-rich cores, which would be insulating in character. Using a self-consistent method, Winkler *et al.*[8] determined the barrier/boundary of a 32°[001]-tilt YBCO GB to be mostly dielectric with a (physical) thickness of a few nanometers. Ivanov *et al.*[10] studied the transport properties through a 4°[001]-tilt GB weak link as a function of temperature and found that the properties may be explained in terms of an S-S'-S weak-link model (where the barrier S' has a lower superconducting transition temperature than the electrodes S). Thus, a depression of the superconducting properties is also likely to occur at the stressed regions surrounding GB dislocations and to increase with increasing stress.

All components of the stress field in the GB's need to be considered when a full understanding of the effects of strain on the superconducting properties is to be obtained. The dilation of the lattice surrounding the GB dislocations includes all stress components and can be assumed to induce variations of the properties. Li[11] has calculated the form of the dilatation strain field for a finite wall of edge dislocations in an isotropic cubic material (see schematic in Fig. 1). The effects of the tensile stresses dominate the volume changes produced in the lattice by edge dislocations.[12] However, both

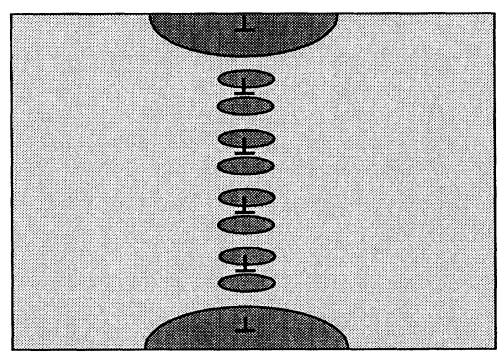

FIG. 1. Schematic of the dilatational strain field for a finite wall of edge dislocations. At large distances from the wall, the dilatational field approaches that of a single dislocation with Burger's vector Nb where b is the Burger's vector of an individual dislocation and N is the total number of dislocations building up the wall. At small distances from the wall, the dilatation of the individual dislocations introduces small loops. The size of these loops in the direction perpendicular to the wall is of the order of the dislocation spacing in the wall.

edge and screw dislocations introduce dilatation in the lattice when second-order effects are considered. Lattice dilatation in high-angle GB's have been observed in a variety of materials using HREM.[13,14] The volume expansions found in GB's of metallic materials could be directly related to the electrical resistivity of the GB and to the GB energy.[15,16]

A number of GB models allow the determination of the relative GB energy.[17,18] Read and Shockley[19] developed a model based on continuum elasticity theory. The GB energy was divided into two types of energy corresponding to the two modes by which the lattice accommodates the lattice misfit at the GB. One type represents the energy of the atomic disorder at the dislocation cores where Hooke's law is no longer valid. The second type corresponds to the energy of the lattice deformation which extends over distances that are comparable to the spacing between the dislocations. The model shows a good agreement with experimental data of GB energy as a function of misorientation angle between adjacent grains for low-angle GB's up to about 20–25°. Another model presented by van der Merwe[20] extended the model of Peierls and Nabarro,[17] which described the strain energy of a single dislocation, to an array of dislocations in a GB. Even though the model assumed an unrealistic sinusoidal force between the two grains (half-crystals), it extended the range of misorientation angles for which a good agreement with experimental data could be obtained. The energy, $E(\theta)$, of a tilt boundary with a misorientation angle θ is given by[20]

$$E(\theta)=[\mu_0 a/2\pi^2(1-2\nu)](1+\gamma-(1+\gamma^2)^{1/2} - \gamma\ln\{2\gamma[(1+\gamma^2)^{1/2}-\gamma]\}) \quad (1)$$

with

$$\gamma=[\pi\mu(1-2\nu)/\mu_0(1-\nu)]\tan(\theta/2)\sec(\theta/2), \quad (2)$$

whee a is the lattice parameter, μ is the shear modulus, μ_0 is a constant of the order of μ, and ν is the Poisson's ratio.

For crystals with cubic symmetry, the tilt GB's with the [100] (or [001]) rotation axis have fourfold symmetry. Therefore, in this case the GB energy should be symmetric around $\theta=\pi/4$. This was obtained by redefining the GB energy, based on expression (1), as

$$E_s(\theta)=E(\theta)+E(\pi/2-\theta)-E(\pi/2). \quad (3)$$

The symmetrical expression (3) is then comparable to the expression of Shockley and Read for tilt misorientation angles between 0 and 21° (see Fig. 7 of Ref. 20).

For large-angle GB's, arrays of secondary GB dislocations are expected to accommodate the angular deviation from special misorientations with a high density of coincidence site lattice (CSL) points or low Σ values (Σ is the unit-cell volume of the CSL divided by the volume of each crystal lattice point).[21,22] The secondary GB dislocations add to the primary lattice dislocation content comprising the special boundary. Therefore, cusps in the GB energy are expected for relatively low Σ boundaries. CSL misorientations around the [001] rotation axis for nearly tetragonal YBCO have been calculated by Singh, Chandrasekhar, and King.[23] For an axial ratio a^2:b^2:c^2::1:1:9, they obtained $\Sigma1$, $\Sigma5$, $\Sigma13$, $\Sigma17$, $\Sigma25$, $\Sigma29$, $\Sigma37$, and $\Sigma41$ at 90°, 36.87°, 22.62°, 28.072°, 16.26°, 43.603°, 18.925°, and 12.68°, respectively. Zhu, Corcoran, and Suenaga[24] have considered both types of dislocations, i.e., primary and secondary, in their calculation of YBCO GB energies. In their calculation the deviation from a CSL orientation can be accommodated by secondary GB dislocations as well as the oxygen content (c/a ratio) at the boundary. They compared the elastic strain energy of GB's with misorientation angles between 44° and 50°, which are in the neighborhood of two ideal tilt boundaries. Differences in core energies were neglected, since these ideal tilt boundaries have similar densities of coincidence sites per unit cell (i.e., similar Σ values).

In this paper we explore the possibility of relating j_c across an individual YBCO GB to the inverse of its energy for the entire range of [001]-tilt misorientation angles. A qualitative analysis of the expected strain field in the vicinity of individual dislocations in an array of dislocations is presented. The analysis gives a simple physical picture of how the transmission properties of the boundary could be determined by GB dislocations, their cores and associated strain fields. The analysis also provides a basis for the relation of j_c across a low-angle GB to its energy. The introduction of the GB energy allows the extension of the analysis to high-angle GB's using established theories which predict the GB energy as a function of misorientation angle. The results are compared to published data for j_c across [001]-tilt YBCO GB's for the full range of misorientation angles.

II. ASSUMPTIONS OF THE MODEL

We use the expression of van der Merwe for the GB energy of a tilt boundary as a function of the misorientation angle. Van der Merwe's expression displays the main features expected for a first approximation of the GB energy curve as a function of misorientation angle θ, e.g., a linear increase with θ for low-angle GB's and an apparent saturation for high-angle GB's. The expression obtained by Read and Shockley, $E(\theta)=E_0\theta\ln(\theta_0/\theta)$, could alternatively be used, but, this approximation breaks down when $\theta>\theta_0/e$. In a first approximation, no fine structure in the GB energy for high-angle GB's (i.e., cusps for special low-energy misorientations) is considered. The available j_c data for YBCO bicrystal films does not display clear peak structures. These are probably smeared out by the scatter in the data and by the wave nature of the YBCO bicrystal film boundaries.[25–28] However, local maxima in j_c for special misorientation angles could be accounted for by incorporating the cusps in the GB energy.

We restrict our analysis to pure symmetrical [001]-tilt boundaries. We consider this type of GB as a wall of identical, parallel and equidistant edge dislocations (see schematics in Figs. 1 and 2). A formal treatment of elasticity problems for YBCO, which is an orthorhombic material, requires taking into account its anisotropic elastic properties.[29] However, the symmetry of the GB's considered here, disregarding surface-image effects, reduces the determination of the stress distribution around the GB dislocations to a problem of plane strain in the YBCO basal plane. The basal plane is supposed to be nearly isotropic since the elastic properties along the a and b directions do not differ very much and since the properties along both directions get sort of averaged in the basal

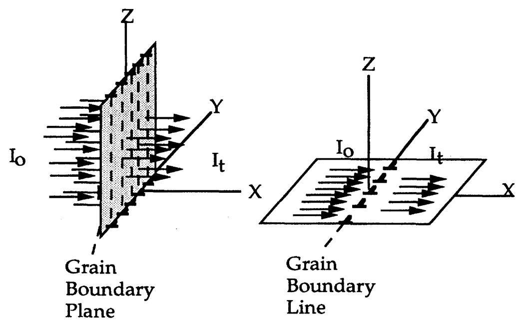

FIG. 2. Schematic representation of a pure symmetrical tilt boundary considered as a wall of identical and equally spaced edge dislocations. A beam containing I_0 paired electrons coming from the left side impinges perpendicularly to the GB. The number of transmitted pairs per unit area I_t (not reflected or removed from the forward supercurrent) are responsible for the supercurrent that is measured across the boundary.

plane due to the presence of {110} twins. Hence, the plane strain is expected to be qualitatively similar to the case of a similarly oriented cubic isotropic material. The main features of the stress distribution around the GB dislocations for the cubic case provide the necessary insight for our qualitative analysis.

Our model assumes that regions with a local dilatation (or contraction) of the YBCO unit cell above (or below) a critical value have depressed superconducting properties. Therefore, the supercurrents are essentially confined to *channels* along the lines of zero dilatation. Then, for low misorientation angles where GB dislocations are well separated, the GB's will behave as parallel arrays of superconducting bridges with dimensions (length in the direction of current transport and width of the cross section) of the order of the dislocation spacing. A configuration for high-T_c bicrystal GB's, consisting of a parallel arrays of Dayem bridges of width comparable to the coherence length and separated by normal regions, has previously been suggested by Sarnelli, Chaudhari, and Lacey,[30] based on measurements of the residual critical current across GB's in the presence of large external magnetic fields.

The width of the channels decreases as the dislocations get closer, i.e., as the dislocation density ρ increases. The major section of the channels passes between the dislocations, but a fraction of the channels would encounter the dislocation cores (see Fig. 1). We also assume that the fraction of the supercurrent that approaches the dislocation cores becomes further reduced from an interaction between the pairs and the dislocation cores, but that it can partially tunnel through this region. It is in fact such a contribution that will determine the j_c as the dislocations begin to overlap with increasing misorientation angle since the high-angle GB's essentially consist of a uniform layer of misfit.

III. MATHEMATICAL EXPRESSIONS

A unit area of a pure-tilt GB consisting of a distribution of dislocations with density ρ (ρ increases with increasing misorientation angle) is schematically shown in Fig. 2. I_0 paired electrons coming from the left side impinge perpendicularly to the GB (i.e., the wall of edge dislocations). From the initial I_0 paired electrons, the regions under a critical dilatation reduce the value to I_0'. I_0' is assumed to be proportional to the width of the channels, and these are assumed to be approximately inversely proportional to the density of dislocations ρ. Then

$$I_0'=I_0\alpha/\rho, \tag{4}$$

where α is a constant of proportionality. Effectively I_0' paired electrons have the opportunity to cross the dislocation wall. The probability that a coupled pair becomes reflected (or removed from the forward supercurrent) when interacting with a dislocation or its core is specified by a microscopic cross section $d\sigma$. The total cross-section area associated with all the dislocations is given by

$$\sigma=\rho d\sigma. \tag{5}$$

The number of pairs impinging at the dislocation wall that get reflected or removed from the forward supercurrent is

$$I_d=I_0'\rho d\sigma. \tag{6}$$

The number of transmitted pairs per unit area, and therefore responsible for the supercurrent that is measured across the GB is

$$I_t=I_0'-I_d=I_0'-I_0'\rho d\sigma=I_0'(1-\rho d\sigma), \tag{7}$$

$$I_t=I_0(\alpha/\rho)(1-\rho d\sigma). \tag{8}$$

A convenient way to estimate the density of dislocations is as follows. The total energy $E(\theta)$ (per unit area) of a GB is equal to the energy contributions from all the dislocations. The total number of dislocations in a unit area is given by ρ. If each dislocation contributes with equal average energy E_0 to the total GB energy, then

$$E(\theta)=E_0\rho \tag{9}$$

from which

$$\rho=E(\theta)/E_0. \tag{10}$$

Replacing (10) in (8), we obtain $[E(\theta)>0]$

$$I_t=I_0[\alpha E_0/E(\theta)][1-E(\theta)d\sigma/E_0]. \tag{11}$$

We rename $E_0/d\sigma=E_0'$ and $\alpha d\sigma=\alpha'$ so that the ratio of j_c across the GB, $j_c^{\mathrm{GB}}(\theta)$, to the intragrain j_c, $j_c(0)$, is finally given by

$$j_c^{\mathrm{GB}}(\theta)/j_c(0)=I_t/I_0=\alpha'[E_0'-E(\theta)]/E(\theta). \tag{12}$$

The ratio $E_0/d\sigma(=E_0')$ is related to the dislocation cores and to the nature of their interaction with the electron pairs. In the case of YBCO thin films, α' will be sensitive to the different substrate materials since the residual strain from the substrate/film mismatch influences the width of the channels around the lines with zero dilatation. Similar or even more complex effects can be expected from the degree of oxygenation in the YBCO material. In our model, α' and E_0' are taken as fitting parameters to be determined from experimental measurements. This intuitive picture holds as long as the

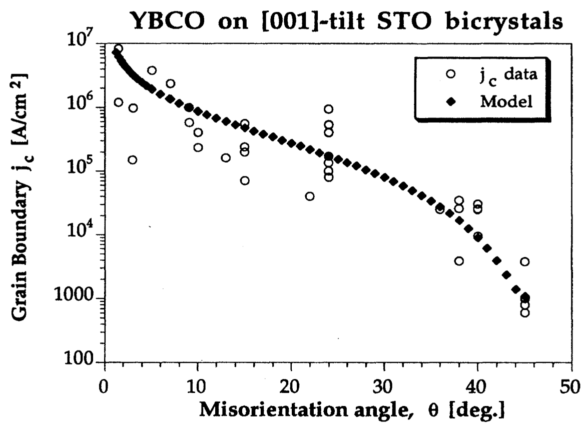

FIG. 3. Misorientation dependence of j_c predicted by our model, plotted together with j_c data across GB's of YBCO films on $SrTiO_3$ (STO) bicrystal substrates extracted form the work by Gross (Ref. 4). A good fit was obtained with $I_0\alpha' = 1.243\times 10^6$ A/cm^2 and $E_0' = 1.542$ J/m^2.

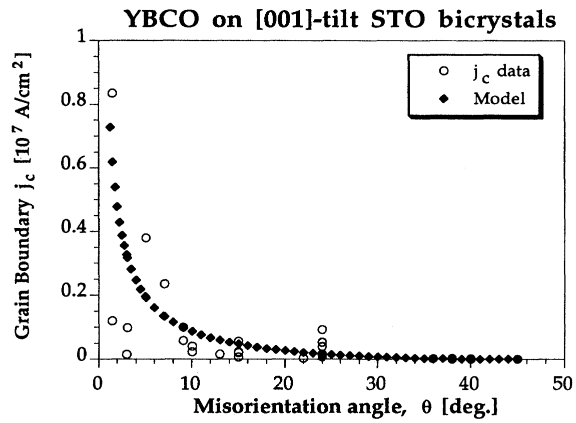

FIG. 4. Plot of the data from Fig. 3, displayed in a linear scale.

microscopic cross sections ($d\sigma$) surrounding the dislocations, as well as the regions with depressed superconducting properties do not overlap. However since relation (12) is expressed only in terms of the GB energy, we investigated whether it can also predict the behavior for high-angle [001]-tilt GB's, assuming that the model of van der Merwe describes the relative GB energy well in a first approximation. This is analogous to the extrapolation of the expressions for the GB energy themselves, since they are built for low-angle GB's consisting of individual dislocations, nevertheless they seem to predict the energy for angles where the concept of individual dislocations is no longer valid.

IV. RESULTS AND DISCUSSION

We remind the reader that under our assumption of a nearly isotropic YBCO basal plane we should use expression (3), symmetrical around $\pi/4$, for the case of [001]-tilt YBCO GB's. We evaluated expression (3) using $a=3.9$ Å, $\mu_0=\mu=59$ GPa, and $\nu=0.281$ for YBCO,[29] and we replaced the obtained values in expression (12) using $I_0\alpha' = 1.243\times 10^6$ A/cm^2 and $E_0' = 1.542$ J/m^2. The results are plotted in Fig. 3 (using a logarithmic scale) together with j_c data across GB's of YBCO films on $SrTiO_3$ (STO) bicrystal substrates replotted from the work by Gross.[4] Similar results can be obtained with the expression of Read and Shockley, $E(\theta)=E_0\theta\ \ln(\theta_0/\theta)$, however the approximation breaks down when $\theta > \theta_0/e(\approx 39°/e)$.

The curve obtained with our model qualitatively improves the fitting of the data as compared to the nearly exponential dependence (with negative exponent) claimed in the aforementioned work.[4] We obtain a curve that runs somewhat similar to the exponential dependence for high-angle GB's, but modulated so that it shows more tendency to approach regions with a high density of data points. Moreover, an exponential dependence of j_c as a function of the misorientation angle suggests the presence of a barrier layer at the boundary which increases in thickness as a function of misorientation angle. However, such interpretation would disregard and contradict the actual changes in the GB microstructure as a function of misorientation angle. Figure 4 shows the same data from Fig. 3 plotted using a linear scale instead. The model shows the main features experimentally observed for this dependence, i.e., an approximately linear decrease of j_c^{GB} for low misorientation angles and an apparent saturation for large misorientation angles, although the slope of the linear portion appears slightly steeper than the data from Dimos *et al.*[1,2] However, this steeper slope may indirectly contribute to the understanding of why the j_c data shows a large scatter in the low-angle region. From the previous discussions, it is apparent that the changes in microstructure and nature of the barrier layer are more complex in the low-angle region than in the high-angle region. In the low-angle region, the critical current across the GB would be due to combined contributions of current transport along nearly undistorted well lattice-matched regions and to tunneling through stressed regions surrounding GB dislocations and through the dislocation cores. To consider that the radii of the dislocation cores remain nearly constant for different misorientation angles is probably a good assumption, consistent with previous HREM investigations.[9] However, as the misorientation angle increases the magnitude of the stress field increases and its spatial extent becomes concentrated perpendicular to the boundary plane. It is therefore somewhat misleading to attribute the linear dependence of j_c as function of the misorientation angle (in the low-angle region) only to either cores or stress fields of constant radii, that increase linearly in number with the misorientation angle.

We reexamine expression (11) [which is the same as (12) with only different nomenclatures for the fitting parameters], in the two regions of low-angle GB's and high-angle GB's separately. For low-angle GB's, $E(\theta)$ varies as θ and the term $E(\theta)d\sigma/E_0$ is small compared to 1. Therefore expression (11) can be approximated to

$$I_t \approx I_0[\alpha E_0/E(\theta)]. \tag{13}$$

This will vary inversely proportional to the misorientation angle as expected for low-angle GB's. For high-angle GB's $E(\theta)$ varies much more slowly and it can be interpreted to be concentrated in a fairly uniform layer of misfit at the GB, with an associated concentrated dilatation field. In this case (11) can be seen as an overall reduced current due to tunneling across the boundary layer. Both $E(\theta)$ in the denominator and in the term $[1-E(\theta)d\sigma/E_0]$ would account for the amount of reduced current. In fact, it appears as if the cross section $d\sigma$ would be giving an average value of the effect of the dislocations, their cores and the concentrated dilatation regions.

In the intermediate range of GB misorientations (between abut 10–20°), the GB changes from a situation in which the misfit (and the dilatation) is concentrated in loops along the glide planes of individual dislocations, to one in which the regions of misfit lie perpendicular to the glide plane. It is then likely that as the misorientation angle increases from low angle to intermediate angles, our expression is underestimating some current concentration in the superconducting regions between dislocations and the increasing contribution of tunneling across the dilated regions at the boundary. The assumption of a constant $d\sigma$ is less suitable here too. It should also be remembered that the "real" curve of the GB energy as a function of the misorientation angle could have finer structure in the large-angle range, with cusps for low Σ boundaries which have not been contemplated with the expression of van der Merwe. The fine structure in the energy would then give local peaks in the j_c data which may be confused with the scatter (see for example the locally high experiment j_c data for $\theta=24°$ in Fig. 3, which is close to a $\Sigma 13$ boundary). The possibility of maxima in j_c across GB's (Refs. 31 and 32) with low-energy misorientations is consistent with the present analysis. They would be accounted for within the frame of a slightly refined analysis incorporating cusps (or local minima) in the GB energy.

The j_c across GB's of YBCO films on yttria-stabilized-ZrO_2 bicrystal substrates and its deviation from the YBCO/STO case can also be understood within the frame of this model.[26] According to the analysis, the extent of the weak-link region is concentrated at the boundary for high-angle GB's. This agrees well with a recent self-consistent determination of the thickness of the weak-link region.[8] Additional features of the behavior of j_c across GB's can also be explained with this analysis. Different local structures of the boundary plane are contained in a boundary length of a few microns,[25–28] typical width for GB junctions. The different structures would give rise to local variations in the GB energy. This variation would translate into an inhomogeneous distribution of j_c as frequently observed from measurements of j_c as a function of applied magnetic field.[3,33,34]

V. CONCLUSIONS

In summary, an analysis and a prediction for a scaling behavior of j_c as a function of the GB energy has been presented. The analysis assumes that regions of suppressed superconducting properties are produced where the dilatation stress field at the GB distorts the YBCO unit cells beyond a critical value. A qualitative analysis of the expected dilatation strain field in the vicinity of an array of dislocations is presented. The analysis provides a basis for the prediction of the j_c across a low-angle YBCO GB as a function of its energy. The introduction of the GB energy allows the extension of the model to high-angle GB's, assuming that the model of van der Merwe describes well the YBCO GB energy as a function of misorientation angle. The results are compared to published data for j_c across [001]-tilt YBCO GB's for a full range of misorientations, showing a good fit. Even more important, the analysis is qualitatively based on the expected changes of the GB microstructure, i.e., GB energy, as a function of the misorientation angle. Thus, the analysis establishes a connection between the GB structure and the superconducting transport properties.

ACKNOWLEDGMENTS

Discussions with Professor Anders Thölén, Professor Tord Claeson, Dr. Vitaly Shumeiko, Dr. Zdravko Ivanov, and Dr. Dag Winkler were very fruitful. Financial support from the Swedish Natural Science Research Council (NFR) and from the Swedish National Board for Industrial and Technical Development (NUTEK) is gratefully acknowledged.

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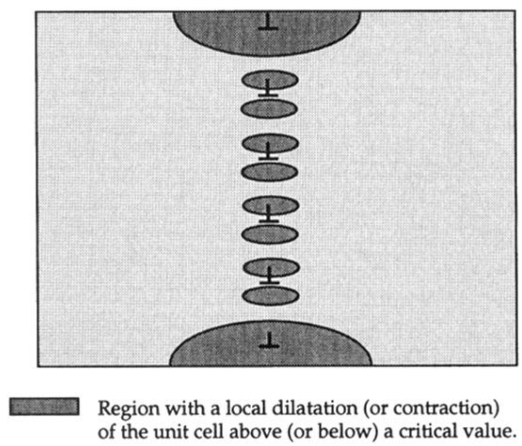

FIG. 1. Schematic of the dilatational strain field for a finite wall of edge dislocations. At large distances from the wall, the dilatational field approaches that of a single dislocation with Burger's vector Nb where b is the Burger's vector of an individual dislocation and N is the total number of dislocations building up the wall. At small distances from the wall, the dilatation of the individual dislocations introduces small loops. The size of these loops in the direction perpendicular to the wall is of the order of the dislocation spacing in the wall.

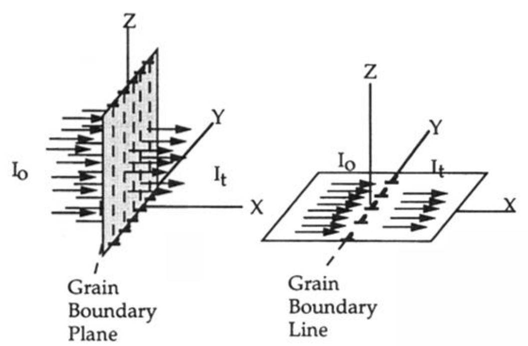

FIG. 2. Schematic representation of a pure symmetrical tilt boundary considered as a wall of identical and equally spaced edge dislocations. A beam containing I_0 paired electrons coming from the left side impinges perpendicularly to the GB. The number of transmitted pairs per unit area I_t (not reflected or removed from the forward supercurrent) are responsible for the supercurrent that is measured across the boundary.