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Dilatation stresses and transport properties of grain boundaries in high-T_C superconductors

I.A. Ovid'ko *

Institute of Problems of Mechanical Engineering, Russian Academy of Sciences, Bolshoj 61, Vas.Ostrov, St.Petersburg 199178, Russia

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Abstract

A theoretical model is suggested which describes the effect of grain-boundary-induced dilatation stresses on the high-T_C superconducting properties of grain boundaries in cuprates. In the framework of the model, the dilatation stresses cause weak deviations from bulk stoichiometry within grain boundary cores and give rise to electric-charge inhomogeneities within and near grain boundary cores. The regions with "non-ideal" stoichiometry and electric charge density suppress high-T_C superconductivity, in which case the critical current density J_c across grain boundaries essentially decreases compared to the bulk. The J_c enhancement in polycrystalline cuprates due to doping is discussed in the framework of the model. The effect of grain boundary dislocations on stoichiometric inhomogeneities in vicinities of high-angle boundaries is theoretically analyzed. The specific structural and behavioral features of high-quality c-axis twist boundaries in BiSrCaCuO superconductors are discussed. Results of the suggested model are compared with experimental data reported in scientific literature. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

Polycrystalline high-transition-temperature (T_C) superconductors are characterized by the transport critical-current density (J_c) whose values are essentially lower (commonly by orders) than those of their single crystalline counterparts, e.g. [1–6]. This property of high-T_C superconductors plaques their high-current applications that are potentially capable of being revolutionary in electrical engineering. The undesired, from an applications viewpoint, low J_c values in polycrystalline superconducting materials are related to the complicated suppressing effect of grain boundaries (GBs) on high-T_C superconductivity [5–7]. At present, though there are many experimentally documented facts concerning the effect of GBs on the transport critical-current density in high-T_C superconductors [5–7], its physical mechanism(s) is (are) under discussion [5–21].

We think that spatially inhomogeneous dilatation stress fields associated with GB cores induce local deviations from bulk stoichiometry within boundary cores and electric-charge inhomogeneities within and near boundary cores. The stoichiometric and electric-charge inhomogeneities are capable of playing an important role in suppression of the superconducting properties of polycrystalline high-T_C cuprates. This idea is consistent with experiment-based representations of the influence of defects on chemical composition inhomogeneities in polyatomic solids [22] and is in, at least, qualitative agreement with the following experimental facts related to polycrystalline high-T_C superconductors:

1. Existence of deviations from bulk stoichiometry near GBs in samples fabricated at highly non-equilibrium conditions [5,18,23].
2. Existence of hole depletion zones in vicinities of GBs [7,24,25].
3. Dramatic distinction between the properties of low- and high-angle GBs [1–6].
4. Existence of variations of the superconducting properties along GB planes [5,26–30].
5. Doping-induced enhancement of J_c in Ca-doped YBaCuO superconductors [16].

The main aim of this paper is to elaborate a theoretical model which describes the effect of GB-induced

* Tel.: + 7-812-3214764; fax: + 7-812-3214771.
E-mail address: ovidko@def.ipme.ru (I.A. Ovid'ko).

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dilatation stresses on critical-current density across GBs in high-T_C superconductors, taking into consideration the experimentally documented facts 1–5.

2. Model of the effect of grain boundaries on high-T_C superconductivity: key points

The generic feature of all cuprate superconductors is their polyatomic structure [5,31]. The generic feature of all GBs in cuprates (as well in other solids) is their role as sources of dilatation stress fields [32]. Since atoms of different types exhibit different behaviors in response to dilatation stresses [22], dilatation stresses associated with GB cores induce local violations of ideal (optimal for high-T_C superconductivity) stoichiometry within GB cores in polyatomic cuprates. This is a generic feature of all polycrystalline high-T_C cuprates.

Let us discuss the effects of grain-boundary-induced dilatation stresses on stoichiometry in the exemplary cases of YBaCuO and BiSrCaCuO superconductors, which are of high importance in applications. High-T_C superconducting cuprates are polyatomic solids whose lattices consist of negatively charged ions (anions) of oxygen O^{2-} and positively charged ions (cations) of other chemical elements (Y, Ba and Cu in the case of YBaCuO superconductors; Bi, Sr, Ca and Cu in the case of BiSrCaCuO superconductors) composing the cuprates. Oxygen ions are small compared to the cations composing crystalline lattices of cuprates, in which case small oxygen ions tend to move to regions where compressive stresses exist, while comparatively large cations tend to move to regions where tensile stresses exist.

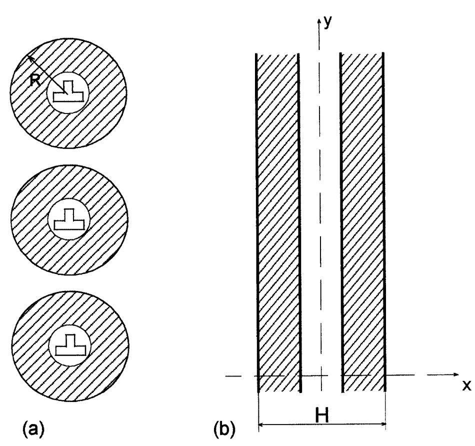

Fig. 1. Stoichiometric and electric-charge inhomogeneties within and near tilt boundaries. (a) Cores of lattice dislocations (composing low-angle boundaries) and (b) cores of high-angle boundaries are characterized by deviations from bulk stoichiometry and an excess positive charge density Q_{gb}. Their vicinities (shaded regions) are hole depletion zones.

More precisely, according to the general theory of diffusion in stressed solids [22], the elastic interaction between dilatation stresses ($\sigma_{xx}(x, y, z)$, $\sigma_{yy}(x, y, z)$, $\sigma_{zz}(x, y, z)$) and diffusing species (small and large ions in our case) in a stressed solid is specified by the interaction energy given as:

$$E_{int} = (\sigma_{xx} + \sigma_{yy} + \sigma_{zz})\Delta V/3 \quad (1)$$

Here, ΔV is the atomic volume difference that characterizes the diffusing species. In the discussed situation with stress-driven diffusion in polyatomic cuprates, diffusional exchange of small oxygen anions and large cations occurs. In this case, values of ΔV that figure on the right hand side of Eq. (1) are as follows:

$$\Delta V_k = [V_k - (V_k + V_O)/2] > 0 \quad (2)$$

$$\Delta V_O = [V_O - (V_k + V_O)/2] < 0 \quad (3)$$

where V_k denotes the atomic volume of cation of the kth type, V_O the atomic volume of oxygen ($V_k > V_O$). The elastic interaction in question forces large cations (small anions O^{2-}, respectively) to move to regions where tensile (compressive, respectively) stresses exist.

GB core regions — plate-like cores (with thickness of the order of 1 nm) of high-angle boundaries and cylinder-like cores (with diameter of the order of 1 nm) of lattice dislocations composing low-angle boundaries (Fig. 1) — are characterized by a low atomic density compared to the bulk or, in other terms, by a positive dilatation [32–34]. The component of the rigid body translation normal to a high-angle boundary, that characterizes the excess free volume of the boundary, is in the order of 1/10 to 1/100 of the lattice parameter [32,33]. In accordance with calculations [34], the excess free volume of dislocation cores (that compose low-angle boundaries) can be estimated as being in the order of 1/10 of magnitude of one or two atomic volumes. In doing so, the excess free volume of edge dislocations is larger than that of screw dislocations [34]. In addition, due to the low atomic density of GB cores, they exhibit enhanced diffusional properties; coefficients of self-diffusion, that characterize high-angle boundary cores and dislocation cores (composing low-angle boundaries), are higher, by several orders, than the bulk diffusion coefficient [32].

Thus, in the context of our previous discussion, boundary core regions are regions where tensile stresses exist, in which case the elastic interaction Eq. (1) gives rise to enhanced-diffusion-mediated deviations from bulk stoichiometry within these regions. According to Eq. (1), Eq. (2) and Eq. (3), large cations substitute small anions O^{2-} within GB cores (Fig. 1), resulting in creation of an excess positive charge density Q_{gb} (>0) of such cores. The concentration of cations within GB cores, resulted from the substitution processes in question, weakly deviates from the bulk concentration. Actually, if the substitution does not occur, the concentration of cations within GB cores is lower than that in the bulk phase due to a low atomic density inherent to the GB phase. The substitution of small oxygen anions by relatively large cations of other cuprate elements results in an increase of the initially low concentration of cations up to values close to the bulk concentration of cations. This is why experiments often do not reveal deviations from the bulk concentration of cations within GB cores. At the same time, in accordance with our model, GB cores should be deficient in oxygen, giving rise to the excess positive charge density Q_{gb}. The oxygen concentration is experimentally measured with large errors [5] (due to low atomic weight of oxygen), in which case deviations from bulk concentration of oxygen within grain boundary cores may be not detectable in experiments.

In order to screen the excess positive charge density Q_{gb}, hole depletion zones characterized by an excess negative charge density Q_z (<0) are formed in the vicinity of GB cores (Fig. 1). Such hole depletion zones have been detected in electron energy loss spectroscopy experiments [7,24,25]. Due to high sensitivity of high-T_C superconductivity to stoichiometry and hole concentration [31], it is natural to treat the GB cores and hole depletion zones in their vicinities as non-superconducting regions responsible for suppression of critical current density J_c across GBs in cuprates.

To summarize, the key points of the suggested model are as follows:

1. Dilatation stresses associated with GB cores induce deviations from bulk stoichiometry within such cores, mediated by enhanced GB diffusion.
2. Due to process (1), the ratio of the number of cations to the number of oxygen anions is high within GB cores compared to bulk, in which case any GB core in a cuprate possesses an excess positive charge density Q_{gb}.
3. Hole depletion zones are formed in the vicinity of GB cores, which are characterized by an excess negative charge density Q_a screening Q_{gb}.
4. Stoichiometric and electric-charge inhomogeneities induced by dilatation stresses of GB cores suppress the critical current density J_c across GBs.

3. Pre-existent models of the effect of grain boundaries on high-T_C superconductivity

It should be noted that the ideas on the effects of dilatation stresses, stoichiometric and electric-charge inhomogeneities on high-T_C superconductivity have been discussed earlier [7–21]. However, these models are different in several aspects from the model suggested in this paper. Let us briefly discuss the key points of each of these models, compare them with those of the model suggested in this paper, and analyze correspondence of predictions of these models to data of experiments with polycrystalline cuprates.

In [18,19], compositional variations at GBs are treated as those responsible for reduction of the critical current density J_c across boundaries. However, fabrication of (thin-film) samples where non-stoichiometry is not exhibited (or, at least, is non-detectable in experiments), but GBs show the same transport behavior, makes the idea [18,19] on the critical role of compositional variations to be discussive.

In [8], the crystallographic disorder within GB cores and strains induced by grain boundary dislocations is taken as being responsible for the superconducting-to-insulating phase transition in strained regions within and near GB cores. In doing so, the value of strain $\varepsilon_c \approx 0.01$ along the a or b axis is assumed to be critical, that is, the transition occurs in regions characterized by strain (along the a or b axis) $\varepsilon > \varepsilon_c$ [8]. This approach, and similar versions [10–12], do not take into account stoichiometric and electric-charge inhomogeneities at GBs. In particular, this model does not explain the experimentally detected [7,24,25] existence of hole-depletion regions in vicinities of GBs. Also, it meets the question related to the fact that strain fields of periodic dislocation walls (which serve as models of low-angle symmetric tilt boundaries; Fig. 1a) drop as $\exp(-x/d)$, where x denotes the distance from the GB plane, and d the period that characterizes arrangement of dislocations composing the dislocation wall. The period d is in following relationship with tilt boundary misorientation α: $b/d = 2\sin\alpha/2)$ [32], where b is the magnitude of the dislocation Burgers vector. With this relationship taken into consideration, the strain fields discussed should give rise to shrinkage of strained regions in vicinities of low-angle tilt boundaries and, therefore, to increase of the critical current density J_c (α) across tilt boundaries with increase of misorientation angle α from 0° to tentatively 10°. This is in contradiction with experimental data [1–6].

In [20,21], the approach [8] has been modified to a situation with chaotically arranged dislocations at tilt boundaries in high-T_C superconductors. Disordered arrays of dislocations create stress fields whose dispersion increases with rising α in the low-angle range [20,21]. This, in the framework of the approach in [8], should result in a decrease of the critical current density J_c across disordered tilt boundaries with rising α for $\alpha \leq 10°$. In addition, in a description of the transport properties of low-angle tilt boundaries one should take into account transformations (splitting and amorphization) of GB dislocations, that occur at some values of misorientation angle and are capable of strongly influencing the critical current density J_c (α); see experimental data [35] and a theoretical model [36]. However, GBs that contain chaotically arranged dislocations

[20,21] as well as dislocations with split and amorphous cores [35,36] are partial cases of GB structures in cuprates. Therefore, the discussed modifications of the approach in [8] cannot answer all questions regarding its validity in the general situation.

The idea of stress-induced suppression of high T_C superconductivity has been exploited also in [9]. Authors of this paper have theoretically described (in terms of the Ginzburg–Landau formalism) the angular dependence of the critical current density J_c across low-angle tilt boundaries in cuprates, taking into consideration the effect of crystal lattice anisotropy on stress fields of GB dislocations as well as the effect of electric-charge inhomogeneities caused by stress fields of the dislocations on the superconducting order parameter. In doing so, however, the model in [9] focused on the range of α from 5° to tentatively 25°, leaving the dependence J_c unclear, for $\alpha < 5°$ and $\alpha >$ 25°. In addition, the electric-charge inhomogeneities have been described as those associated with stress-induced inhomogeneities of the averaged (over an elementary cell) ion density, in which case the polyatomic structure of cuprates has been ignored. In the framework of this model description, the role of stoichiometric inhomogeneities driven by stress fields has been not taken into account.

In [13–16] the excess electric charge within high-angle boundary cores has been treated as being responsible for the formation of hole-depletion zones in the vicinity of GBs. In doing so, the origin of the excess charge in question has been attributed to crystallographic disorder existing within high-angle boundary cores as well as to the *d*-wave type symmetry of the superconducting order parameter. However, in the framework of the model description [13–16], the excess electric charge can be either positive or negative, in which case hole-enhancement zones in vicinities of GBs would be expected to be as likely to form as hole-depletion zones [16]. At the same time, only hole-depletion layers have been detected in electron energy loss spectroscopy experiments [7,24,25]. This raises questions about the origin of the excess electric charge, given by the model in [13–16]. A modified version of the approach in [13–16] is the so-called bond-valence model [7], which attributes the excess electric charge of grain boundary cores to variations of valency of copper atoms in atomic chains (existing between Cu–O planes) in YBaCuO superconductors. This model is based on results of computer simulations of the atomic structure of GBs in YBaCuO materials, in which case the atomic potentials are empirical and can, therefore, lead to errors. Finally, an uncertainty of the origin of hole-depletion layers is inherent to model described in [17], which uses the existence of such layers and their experimentally measured characteristics as an experimentally documented input of the Ginzburg–Landau-formalism-based description of the transport properties of tilt boundaries in YBaCuO superconductors.

The model suggested in this paper explains the GB effect on high-T_C superconductivity as that caused by electric-charge inhomogeneities within and near GB cores (Fig. 1), which are associated with stoichiometric inhomogeneities induced by dilatation stresses and mediated by enhanced grain boundary diffusion. This model, in fact, treats the combined effects of dilatation stress fields, stoichiometric and electric-charge inhomogeneities on the critical current density across GBs as those responsible for the experimentally detected features, 1–5 (see Introduction), of high-T_C polycrystalline cuprates. Also, the model allows one to make predictions of the doping elements capable of enhancing across GBs in doped high-T_C cuprates (Section 6) and to give a qualitative explanation of the specific transport properties of high-quality *c*-axis twist boundaries in BiSrCaCu superconductors (Section 7).

4. Dependence of critical current density across tilt boundaries on boundary misorientation

Let us discuss the effect of stoichiometric and electric-charge inhomogeneities within and near GB cores (Fig. 1) on the critical current density J_c across tilt boundaries in cuprates. The superconducting properties of cuprates are experimentally revealed to be suppressed by even weak deviations from the ideal (optimum) stoichiometry corresponding to maximum critical transition temperature T_C [31,37]. Also, electric-charge inhomogeneities strongly suppress the superconducting critical current; see discussion in papers [9,13–16]. Therefore, it is natural to treat the regions with "non-ideal" stoichiometry and electric-charge inhomogeneities within and near grain boundary cores (Fig. 1) as non-superconducting ones.

According to the theory of electron pairs tunneling in superconductors (see, e.g. [38,39]), reduction of the critical-current-density across a non-superconducting layer is approximately described by factor $\exp\{-h/\xi\}$, where h denotes the layer thickness and ξ the characteristic decay length which can be the tunneling length for insulating GBs or the proximity length for metallic GBs (which is close to the coherence length). With the aforesaid taken into account, we find the following formula which describes, in the first approximation, the effect of stoichiometric and electric-charge inhomogeneities at GBs on the angular dependence of the critical-current density across [001] tilt boundaries:

$$J_c(\alpha)/J_c(0°) \approx \frac{1}{S}\int_s \exp\{-h(y,z)/\xi\}\mathrm{d}y\ \mathrm{d}z \qquad (4)$$

Here, h plays the role as the thickness of the regions with "non-ideal" stoichiometry and charge distribution

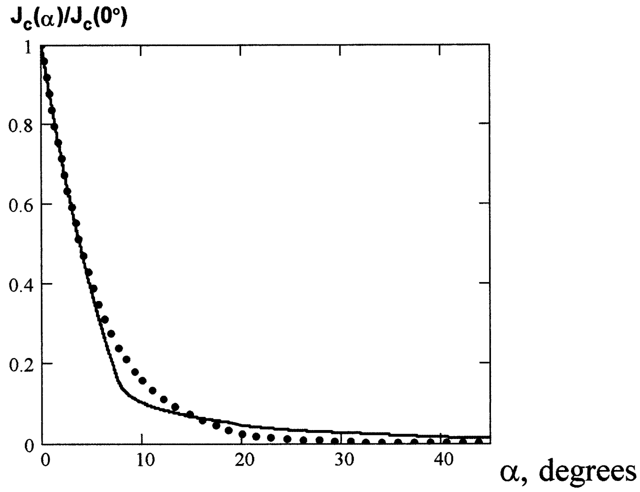

Fig. 2. Angular dependence of the critical current density J_c across [001] tilt boundaries. Curve 1, according to Eq. (6), is attributed to the combined effects of dilatation-induced stoichiometric and electric-charge inhomogeneities, $d_{x^2-y^2}$ symmetry and the faceted microstructure of tilt boundaries. The experimentally detected [1–4] angular dependence of J_c is shown, according to [48], as curve 2.

(Fig. 1). In general, h depends on coordinates, y and z, of boundary plane with area S.

Parallel with stoichiometric and electric-charge inhomogeneities, additional geometric factors are capable of affecting the critical current density across tilt boundaries. Thus, recently, the symmetry of the order parameter in many high-T_C superconductors has been experimentally recognized as being $d_{x^2-y^2}$ [40–45] or $d_{x^2-y^2}$ mixed with an s-wave component [46]. The $d_{x^2-y^2}$ symmetry causes a dependence of the critical-current density $J_c(\alpha)$ across a GB on boundary misorientation α and orientation of boundary plane relative to adjacent grains [47,48]. With this taken into consideration, the faceted microstructure of tilt boundaries, that is often detected experimentally (see, for instance, [49–52]), also influences the transport properties of tilt boundaries in high-T_C superconductors. According to [48], the combined effects of $d_{x^2-y^2}$ symmetry and faceting give rise to the following angular dependence of the critical current density J_c across [001] tilt boundaries (most commonly examined in experiments with polycrystalline high-T_C superconductors):

$$J_c(\alpha)/J_c(0°) = \left\langle \prod_{m=1,2} [(\sin\alpha_m)^2 - (\cos\alpha_m)^2] \right\rangle_F \quad (5)$$

Here α_m (m = 1,2) is the smallest angle between the grain boundary plane and a principal crystallographic axis ($\vec{a}$ or $\vec{b}$) of adjacent grain m ($\alpha_1 + \alpha_2 = \alpha$), $\langle ... \rangle_F$ denotes the averaging that takes into account the faceted boundary microstructure.

From Eq. (4) and Eq. (5) we find that the combined effects of the dilatation-induced and electric-charge stoichiometric inhomogeneities, $d_{x^2-y^2}$ symmetry and faceting cause the following angular dependence of J_c in the case of [001] tilt boundaries:

$$J_c(\alpha)/J_c(0°) \approx \frac{1}{S} \left\langle \prod_{m=1,2} [(\sin\alpha_m)^2 - (\cos\alpha_m)^2] \right\rangle_F \int_S \exp\{-h(y,z)/\xi\} \, dy \, dz \quad (6)$$

We have calculated with the help of Eq. (6) the angular dependence of J_c — curve 1 in Fig. 2 — attributed with the combined effects in question. In doing so, for high-angle ($\alpha \geq 20°$) boundaries, h is taken as $H \approx 3$ nm. For low-angle ($\alpha \leq 19°$) boundaries, the scale h is chosen, according to Fig. 1 a, as the y-dependent thickness of boundaries composed of lattice dislocation cores and their vicinities with "non-ideal" stoichiometry and charge density, that are characterized by diameters $2R \approx 3$ nm. (In this case interspacing between periodically arranged lattice dislocations composing low-angle boundaries (Fig. 1 a) depends on α as [32]: $2d = b\sin(\alpha/2)$, where b is the magnitude of the dislocation Burgers vector.) In the intermediate range of α from 19° to 20°, h is taken as interpolation of corresponding values of h for low- and high-angle boundaries. Results of the averaging $\langle ... \rangle_F$ are taken from paper [48], ξ is taken as 1.5 nm. (This corresponds to the coherence length in [001] planes that carry the current [5].)

The calculated dependence $J_c(\alpha)$ (curve 1 in Fig. 2) is in a satisfactory agreement with experimental data [1–4] (curve 2 in Fig. 2) for YBaCuO superconductors. This allows us to think that the idea on the combined effects of dilatation-induced stoichiometric and electric-charge inhomogeneities, $d_{x^2-y^2}$ symmetry of the superconducting order parameter and the faceted microstructure of GBs is effective in description of the dramatic distinction between the transport properties of low- and high-angle boundaries. (Generally speaking, there are weak changes of high-angle boundary core structures with rising α, for $\alpha > 20°$, which are not taken into account in our first-approximation model of high-angle boundary cores; see Fig. 1 b. The theoretical analysis of such changes will be the subject of further investigation).

Thus, GB cores cause the transport properties of tilt boundaries, because such cores are regions where (1) high tensile stresses exist which drive stoichiometric inhomogeneities within cores and electric-charge inhomogeneities within and near cores; and (2) enhanced diffusion occurs which mediates the stoichiometric inhomogeneities. Notice that GB dislocations also create dilatation stresses in the bulk phase adjacent to GBs [32]. However, the bulk diffusion is low compared to the GB diffusion and, therefore, can not effectively mediate dilatation-driven stoichiometric inhomogeneities in the bulk phase (especially in the case of quasiequilibrium formation of high-quality cuprate samples). In addition, values of dilatation in stressed regions of the bulk phase commonly are small compared to those characterizing GB cores. As a corollary, parameters of regions in the bulk phase, where dislocation-induced stresses exist, in most cases weakly affect the transport properties of GBs.

5. Effects of doping on the transport properties of grain boundaries

The doping of polycrystalline high-T_C cuprates is capable of strongly influencing the critical current densities J_c across GBs. The remarkable experimental fact in this area is a record enhancement of J_c in Ca-doped YBaCuO superconductors [16]. In particular, following data [16], J_c across a symmetric 24° [001] tilt boundary in $Y_{0.7}Ca_{0.3}Ba_2Cu_3O_{7-\delta}$ superconducting film, exceeds J_c across the same GB in "conventional" $YBa_2Cu_3O_{7-\delta}$ film by about a factor of eight. In the framework of model described in [13–16] treating the excess charge density Q_{gb} at GB cores as that responsible for the existence of hole-depletion zones causing values of J_c, the J_c enhancement due to doping is related to doping-induced decrease of the excess charge density Q_{gb} [16]. That is, dopants substitute host ions within GB cores, resulting in a decrease of Q_{gb} and, therefore, to a decrease of the hole-depletion layer thickness. This gives rise to enhancement of J_c. However, the model [13–16] treats the origin of Q_{gb} to be related to the crystallographic disorder and $d_{x^2-y^2}$-symmetry of the superconducting order parameter within GB cores, in which case the model can not explain the fact that the sign of Q_{gb} is always positive which manifests itself in the existence of experimentally detected hole depletion zones (see discussion in [16]). In these circumstances, the model in [13–16] does not allow one to predict dopants that enhance J_c in polycrystalline high-T_C cuprates.

In contrast to the approach in [13–16], the model developed in this paper gives a description of the positive charge of Q_{gb}. It treats the charge density inhomogeneities at GB cores, characterized by the excess charge density Q_{gb}, to be related to dilatation-induced depletion of oxygen anions within cores. In the framework of the suggested model, doping can decrease value of Q_{gb} and, therefore, enhance values of J_c across GBs in the following cases: (A) valency $\tilde{\upsilon}$ of dopant cations is lower than that (υ_k) of host cations; (B) radius $\tilde{r}$ of dopant cations is larger than that (r) of host cations. In case (A), the excess charge Q_{gb} decreases directly due to the doping-induced decrease of the sum charge of cations within GB core. In case (B), doping decreases the effect of dilatation stresses. That is, substitution of relatively small host cations by relatively large substitute cations results in an increase of the atomic density within GB cores. As a corollary, dilatation stresses associated with GB cores are decreased, in which case so is the driving force for diffusional exchange of cations and oxygen anions within boundary cores, thus decreasing Q_{gb}. In context of mechanism (A) of the influence of doping on the critical current density J_c across GBs, the Ca-doping of $YBa_2Cu_3O_{7-\delta}$ bi-crystals results in the J_c enhancement due to the following. Commonly Ca^{2+} cations substitute Y^{3+} cations in YBaCuO cuprates [53]. This gives rise to a decrease of the excess positive charge density Q_{gb}, associated with $Ca^{2+} \rightarrow Y^{3+}$ substitution in GB cores, and, as a corollary, to the J_c enhancement.

In the context of point (A), it is potentially interesting to experimentally test the effect of Na-doping on J_c in bi-crystalline YBaCuO. Actually, Na^{1+} cations substitute Y^{3+} cations in $YBa_2Cu_3O_{7-\delta}$ cuprates [54]. This non-isovalent doping can substantially decrease Q_{gb} and, therefore increase J_c in bi-crystalline $Y_{1-x}Na_xBa_2Cu_3O_{7-\delta}$ superconducting cuprates.

6. Grain boundary dislocations and stoichiometric inhomogeneties along tilt boundaries in high-T_C superconductors.

Let us briefly discuss the effect of GB dislocations on stoichiometric inhomogeneties in the vicinity of GBs along boundary planes. We think that such inhomogeneities in the bulk phase adjacent to GB cores result from the bulk diffusion driven by stress fields of GB dislocations. To do so, the bulk diffusion should be intensive, at least, during the synthesis of a cuprate. The situation in question comes into play, in particular, if a cuprate is synthesized at highly non-equilibrium conditions (say, by fast sintering technique). In doing so, the most mobile ions in the bulk of high-T_C superconducting YBaCuO and BiSrCaCuO cuprates are ions, Cu and O, because they are small compared to other ions and, therefore, are characterized by a low activation energy for their motion in a crystalline lattice. The ion volume of copper V_{Cu} is larger than that of oxygen V_O, in which case comparatively large ions Cu (small ions O, respectively) move to regions where tensile (compressive, respectively) stresses exist that are created by GB dislocations.

As with low-angle boundaries consisting of lattice dislocations (Fig. 1 a), high-angle boundaries (Fig. 1 b) commonly contain GB dislocations, e.g. [32]. Both low- and high-angle boundary dislocations create spatially inhomogeneous dilatation stress fields outside grain boundary cores. Spatial arrangements of GB dislocations in solids, after relaxation of their GB structures, are close to periodic; they are periodic, quasiperiodic [32,55,56] or weakly disordered [57,58]. Dislocations arranged in a tentatively periodic way along GB force tentatively periodic modulations of dilatation stress fields and, therefore, tentatively periodic modulations of "non-ideal" stoichiometry in the case of intensive bulk diffusion. Such tentatively periodic modulations of "non-ideal" stoichiometry have been observed experimentally in the vicinity of GBs in YBaCuO superconductors fabricated by the sintering technique [23].

For illustration of the effect of GB dislocations on stoichiometric inhomogeneties, let us analyze, with the help of Eq. (1), Eq. (2) and Eq. (3), the distribution of oxygen and copper in the vicinity of a high-angle tilt boundary that contains periodically-arranged boundary dislocations of the edge type (Fig. 3) in YBaCuO superconducting materials. In doing so, for definiteness, we will focus on the situation where the diffusional exchange of Cu and O ions dominates near and within GB cores, with diffusion of ions Y and Ba being neglected. In the coordinate system shown in Fig. 3 (with dislocation lines being parallel with z axis), stress fields, σ_{xx}, σ_{yy} and σ_{zz}, created by the periodic ensemble of boundary dislocations are given as [32,59]:

$$\sigma_{xx} = \frac{Gb}{2\pi(1-\nu)\lambda} \cdot \pi \sin 2\pi y^* \frac{\cosh 2\pi x^* - \cos 2\pi y^* + 2\pi x^* \sinh 2\pi x^*}{(\cosh 2\pi x^* - \cos 2\pi y^*)^2} \quad (7)$$

$$\sigma_{yy} = \frac{Gb}{2\pi(1-\nu)\lambda} \cdot \pi \sin 2\pi y^* \frac{\cosh 2\pi x^* - \cos 2\pi y^* - 2\pi x^* \sinh 2\pi x^*}{(\cosh 2\pi x^* - \cos 2\pi y^*)^2} \quad (8)$$

$$\sigma_{zz} = \nu(\sigma_{xx} + \sigma_{yy}) \quad (9)$$

where G denotes the shear modulus, ν the Poisson ratio, λ the period of the dislocation ensemble distribution, b the magnitude of the dislocation Burgers vector[1], $x^* = x/\lambda$ and $y^* = y/\lambda$. In Fig. 3 the distribution of oxygen-rich and oxygen-deficient zones is shown in vicinity of a tilt boundary (outside the layer-like boundary core) with the dislocation ensemble period $\lambda \approx 30$ nm, which is calculated in accordance with Eq. (1), Eq. (2), Eq. (3), Eq. (7), Eq. (8) and Eq. (9). In our calculations, the layer-like boundary core characterized by a low atomic density is assumed to be the region with a positive dilatation being insensitive to dislocation stress fields. As a corollary, the boundary region is treated as that deficient in oxygen and rich in copper.

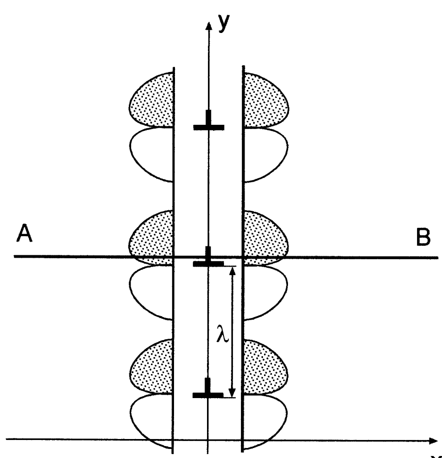

Fig. 3. High-angle tilt boundary with periodically arranged grain boundary dislocations. Oxygen- and copper-rich zones in the vicinity of tilt boundary are schematically shown as dotted and white regions, respectively.

Let us compare results of our theoretical calculations with data [23] of experiments detecting stoichiometric inhomogeneties in vicinities of GBs in YBaCuO superconductors. For a quantitative comparison, the number N_O of oxygen atoms in any elementary cell within oxygen-deficient (oxygen-rich, respectively) zones is assumed to be lower (larger, respectively) by 1 than $7-\delta = 6.95$[2], because chemical forces existing in cuprates, in general, prevent high deviations of stoichiometry from some "equilibrium" stoichiometry. With this assumption taken into account, the averaged oxygen concentration profiles $\langle n_O(x,y) \rangle$ have been calculated in vicinity of the high-angle tilt boundary shown in Fig. 3. The averaging $\langle \ldots \rangle$ corresponds to the conditions of experiments [23] in which the averaged concentration profiles were measured with spatial resolution being in the range 5–8 nm for each profile. In these circumstances, in our model calculations we deal with the averaged profiles defined as follows:

$$\langle n_O(x,y) \rangle = \frac{1}{q^2} \int_{-q/2}^{q/2} dx' \int_{-q/2}^{q/2} dy' \, n_O(x+x', y+y') \quad (10)$$

where $n_O(x,y)$ is the local concentration of oxygen, and $q = 5$ nm. That is, value $\langle n_O(x,y) \rangle$ of the averaged profile in point (x,y) is the oxygen concentration averaged on a square with center at point (x,y) and edges of 5 nm.

In Fig. 4 is plotted the calculated oxygen concentration profile $\langle n_O(x,y) \rangle$ (see curve 1) perpendicular to the tilt boundary along line AB shown in Fig. 3. This profile is in a satisfactory agreement with the profile (open circles in Fig. 4) measured in experiment [23]. Also, the corresponding copper profile $\langle n_{Cu}(x,y) \rangle$ (defined by Eq. (10) with $n_O(x,y)$ being replaced by $n_{Cu}(x,y)$) is shown along line AB. The copper profile is in a satisfactory agreement with the experimentally measured [23] profile (open boxes in Fig. 4).

[1] The magnitude of Burgers vectors of dislocations at high-angle GBs commonly are small compared to those of lattice dislocations composing low-angle boundaries [32].

[2] Value $\delta = 0.05$ characterizes $YBa_2Cu_3O_{7-\delta}$ superconductors with maximum critical transition temperature T_C; see, for example, [35], [36], [37].

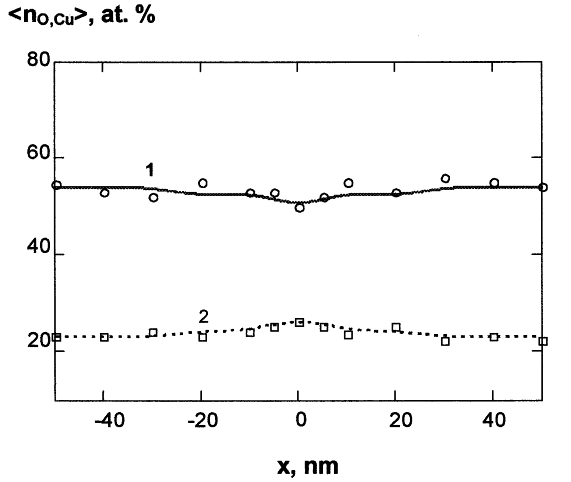

Fig. 4. Calculated oxygen concentration (curve 1) and copper concentration (curve 2) profiles along line AB perpendicular to the grain boundary of Fig. 3. The corresponding experimental data [23] are exhibited as open circles and boxes for oxygen and copper, respectively.

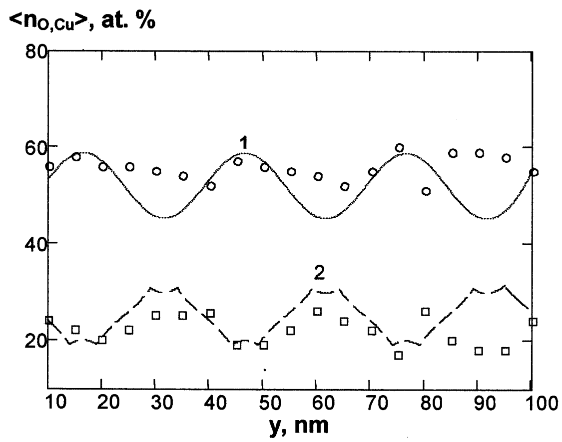

Fig. 5. Calculated oxygen concentration (curve 1) and copper concentration (curve 2) profiles along grain boundary, corresponding to $x = 0$. The corresponding experimental data [23] are exhibited as open circles and boxes for oxygen and copper, respectively.

In Fig. 5 the calculated oxygen profile (curve 1) and the corresponding copper profile (curve 2) are shown along the tilt boundary of Fig. 3 at position corresponding to x = 0. They are in a satisfactory agreement with the corresponding profiles (respectively open circles and boxes in Fig. 5) measured in experiments [23].

The agreement between results of our model calculations and experimental data [23] (see Figs. 4 and 5) allows one to think that GB dislocations are capable of essentially influencing stoichiometric inhomogeneties in vicinities of tilt boundaries in high-T_C superconductors, if the bulk diffusion is intensive. This, in spirit of our analysis of the relationship between stoichiometry and the transport properties of GBs (see discussion in previous sections), serves as an explanation of the experimentally observed [26–30] inhomogeneties of the supercurrent along GBs in high-T_C superconductors.

7. Critical current density across twist boundaries in high-T_C superconductors

Low-angle twist boundaries commonly are modeled as networks of lattice dislocations of the screw type (Fig. 6 a), e.g. [32,59]. Following analysis [34], cylinder-like cores of screw dislocations are characterized by a non-zero dilatation. At the same time, screw dislocations do not create any dilatation stress fields outside their cores [59]. In a conventional situation, cores of high-angle twist boundaries are layer-like regions (Fig. 6 b) with a low atomic density compared to the bulk, in which case they also are characterized by a non-zero dilatation [32]. In contrast to the conventional situation, recently, high-quality c-axis twist boundaries with "cores of zero thickness" and enhanced transport properties have been fabricated in BiSrCaCuO superconductors, e.g., [60–62]. In this section, first, we will consider conventional twist boundaries. Then the specific structural and behavioral features of high-quality twist boundaries will be discussed.

As with tilt boundaries (see Section 2), let us model stoichiometric inhomogeneties occurring due to dilatation stresses associated with conventional twist boundary cores as those existing within cylinder-like cores of screw dislocations in the case of low-angle boundaries (Fig. 6 a) as well as within layer-like GB cores in the case of high-angle boundaries (Fig. 6 b). GB cores are characterized by an excess positive charge density Q_{gb} (related to deviations from bulk stoichiometry) which is screened owing to formation of hole

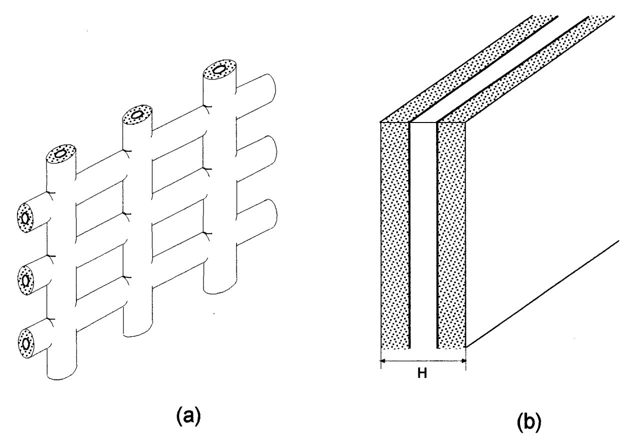

Fig. 6. Stoichiometric and electric-charge inhomogeneities within and near conventional twist boundaries. (a) Cores of lattice dislocations (composing low-angle boundaries) and (b) cores of high-angle boundaries are characterized by deviations from bulk stoichiometry and an excess positive charge density Q_{gb}. Their vicinities (shaded regions) are hole depletion zones.

depletion zones in vicinities of boundary cores (Fig. 6). The regions with "non-ideal" stoichiometry and charge density (Fig. 6) are assumed to be non-superconducting, in which case their effect on the critical current density is described by factor $\exp\{-h/\xi\}$. For geometry of regions with "non-ideal" stoichiometry and charge density in the case of twist boundaries (see Fig. 6), we have calculated with the help of Eq. (4) the dependence of the critical current density J_c across [100] twist boundaries on boundary misorientation α (see solid curve in Fig. 7). In doing so, for high-angle ($\alpha \geq 20°$) boundaries, h is taken as $H \approx 1.5$ nm. For low-angle ($\alpha \leq 19°$) boundaries, the scale h is chosen, according to Fig. 6 a, as y- and z-dependent thickness of boundaries composed of screw dislocation cores and their vicinities with "non-ideal" stoichiometry and charge density that are characterized by diameters $2R = H \approx 1.5$ nm. (In this case period d of the dislocation network composing a low-angle twist boundary with misorientation α (Fig. 6 a) depends on α as [32]: $2d = b\sin(\alpha/2)$, where b is the magnitude of the screw dislocation Burgers vector.) In the intermediate range of α,from 19° to 20°, h is taken as interpolation of corresponding values of h for low- and high-angle twist boundaries. ξ is chosen as 0.3 nm. (This corresponds to coherence length along the [001] direction [5]).

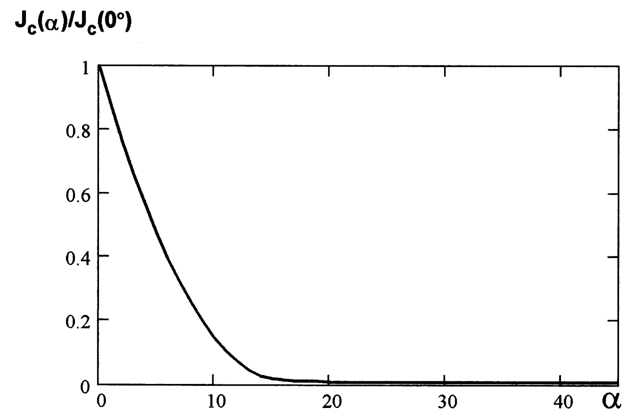

Fig. 7. Angular dependence of the critical current density J_c across conventional [100] twist boundaries, attributed, according to Eq. (4), to the effect of dilatation-induced stoichiometric and electric-charge inhomogeneities.

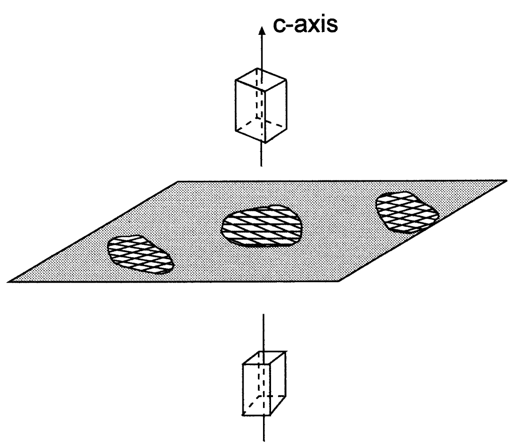

Fig. 8. Partly incoherent twist boundary as a model of high-quality c-axis twist boundaries in cuprates. Coherent and incoherent boundary fragments are schematically shown as cross-hatched and shaded and regions, respectively. Boxes above and below the boundary plane illustrate misorientation of adjacent grains.

The calculated dependence $J_c(\alpha)$ (see Fig. 7) has the same character as that in the case of tilt boundaries (see Fig. 2). This corresponds to data of experiments with conventional twist boundaries in high-T_C superconductors, reported in scientific literature (see review [5] and references therein).

Recently, high-quality c-axis twist boundaries have been fabricated in BiSrCaCuO superconductors, that exhibit the enhanced transport properties [60–62]. These twist boundaries carried critical current as high as their constituent single crystals (adjacent grains), regardless of the misorientation angle [60–62]. The specific structural feature of high-quality twist boundaries is the zero thickness of boundary core. That is, the twist boundaries are atomically intact and clean, with no detectable c-axis spacing increase or chemical changes between the BiO double layers [60,61].

The specific structural feature in question allows us to treat high-quality twist boundaries in BiSrCaCuO superconductors as analogs of partially incoherent interphase boundaries [63,64] in film/substrate systems. In this context, any high-quality twist boundary is thought of as that consisting of both coherent and incoherent fragments (Fig. 8). Coherent fragments are characterized by strong chemical binding between atoms of adjacent grains, in which case orientational mismatch between twisted crystalline lattices of the adjacent grains is accommodated by straining the lattices. Incoherent fragments of high-quality twist boundaries can be treated as having resulted from a rigid body contact of two crystalline lattices, associated with faults in chemical binding at the boundary. Incoherent fragments of a boundary do not induce any stress fields.

Any partly incoherent twist boundary is characterized by ratio $P = S_c/S_i$, where S_c (S_i, respectively) is the total area of its coherent (incoherent, respectively) fragments. In general, this ratio depends on misorientation and anisotropy of a material. However, for any P (≥ 0), partly incoherent twist boundaries have plane cores of "zero thickness". This corresponds to data [60,61] of structural characterization of high-quality twist boundaries in BiSrCaCuO superconductors.

A detailed description of partly incoherent twist boundaries as models of high-quality c-axis twist boundaries in cuprates is beyond the scope of this paper. Here we will focus on only the specific feature of high-quality twist boundaries as those having plane cores of zero thickness. A direct consequence of this feature is the fact that there are no dilatation stresses associated with grain boundary cores. In context of our previous analysis, it means that high-quality twist boundaries should not affect the critical current density (if the effect of d-symmetry of the superconducting order parameter is not taken into account). The aforesaid is in correspondence with experimental data [60–62].

8. Concluding remarks

In this paper a theoretical model has been developed relating the GB effect on high-T_C superconductivity to stoichiometric and electric-charge inhomogeneties induced by dilatation stresses of GB cores and mediated by enhanced boundary diffusion in superconducting cuprates. Within the framework of the model, dilatation stresses associated with a low atomic density of GB cores cause such cores to be characterized by high ratio of the number of cations to that of oxygen anions, compared to bulk. This results in an excess positive charge density Q_{gb} of GB cores. In order to screen Q_{gb}, hole depletion zones with charge density $Q_z < 0$ are formed near boundary cores.

Deviations from bulk stoichiometry within GB cores and formation of hole depletion zones in their vicinities are capable of strongly influencing the critical current density across GBs. So, the experimentally observed [1–6] dramatic distinction between the superconducting properties of low- and high-angle GBs is effectively described as that related to stoichiometric inhomogeneties within GB core regions and formation of hole depletion zones in their vicinities (see Section 2). The volume fraction of regions with "non-ideal" charge density (GB cores and hole depletion zones) rapidly grows with boundary misorientation α in the case of low-angle boundaries ($0° < \alpha < 20°$) and is tentatively constant in the case of high-angle boundaries ($\alpha > 20°$). This factor, in the framework of the suggested model description, causes the dramatic distinction between the superconducting properties of low- and high-angle boundaries. The dependence of the critical current density J_c on boundary misorientation α (see Figs. 2 and 7), calculated with the help of our model representations, are in satisfactory agreement with experimental data for tilt boundaries and twist boundaries. The transition from a high to a weak sensitivity of J_c to boundary misorientation α corresponds to misorientation at which boundary dislocation cores overlap.

The effect of doping on the critical current density J_c across GBs is analysed with the help of representations of the suggested model (see Section 5). The specific features of dopants that are capable of enhancing J_c in polycrystalline cuprates are discussed. The Na-doping of YBaCuO superconductors is suggested as a promising method to increase values of J_c across GBs in polycrystalline YBaCuO superconductors.

GB dislocations of the edge type create spatially inhomogeneous dilatation stress fields in the vicinity of GBs. With this taken into account, we have modeled stoichiometric inhomogeneties in the vicinity of GBs as those induced by dislocation stress fields in the situation where the bulk diffusion is intensive (see Section 6). Results of our model calculations are in satisfactory agreement with experimental data [23] (see Figs. 4 and 5).

Dilatation stresses associated with GB cores are capable of causing suppression of the critical current density across conventional twist boundaries (see Fig. 7). High-quality c-axis twist boundaries in BiSrCaCuO superconductors have plane cores of zero thickness, in which case dilatation stresses associated with their cores are absent. This explains the experimentally documented [60–62] fact that high-quality twist boundaries carry almost the same critical current as their constituent single crystals (grains), regardless of boundary misorientation.

Thus the above analysis allows us to treat dilatation stresses of GBs in high-T_C superconductors as the key factor causing stoichiometric and electric-charge inhomogeneities which are mediated by enhanced boundary diffusion and strongly influence the superconducting properties of GBs. The effect of dilatation-induced stoichiometric and electric-charge inhomogeneities on high-T_C superconductivity, on the one hand, accounts for many experimentally documented facts (see points 1–5 in Introduction) related to the behavior of polycrystalline cuprates and, on the other hand, can be used in technologically controlled enhancement of the high-T_C superconducting properties of GBs in cuprates.

Acknowledgements

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