Properties of Grain Boundaries in High-\(T_c\) Superconductors

– Notes on a Recent Presentation –

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Abstract

The purpose of this article is to discuss a view concerning key datasets of the properties of grain boundaries in high-\(T_c\) superconductors that was recently expressed in Ref. [1]. The reference also criticizes our research. Using examples I disprove this criticism.
Grain boundaries in high-$T_c$ superconductors are a topic of intense interest. Their low critical current densities provide a severe obstacle for the realization of superconducting wires that operate at 77 K. It is therefore important that the critical currents can be significantly enhanced if the grains are aligned, as reported by my colleagues and myself, for example in [2, 3]. Our finding provides the basis for the development of state-of-the-art high-$T_c$ wires, the so-called coated conductors. This field is very active, and many groups have made outstanding contributions.

In the following I will refer to the publications [2, 3] as “ours”. The author of [1] did not contribute to our studies.

The recent Ref. [1] comments on our work, and in doing so caused misunderstandings in the community. Aiming to provide clarifications, I will address two problems: First, in [1] its author admits that over many years he published our results without proper references, such as if they had been obtained by him or by his group ([1], p. 107,109). While he identifies only in exceptional cases the figures in which he displays our results, he recognizes that by publishing our data without naming their source he did not meet scientific standards ([1], p. 109). However, it also seems that he has misinterpreted some of our data. Second, Ref. [1] contains a number of invalid or misleading statements.

The following discussion refers only to part of the work of the author of [1]. He contributed to the progress of the field with very good studies that are not subject to the problems discussed here.

His coauthors had little opportunity to note the problems in their papers, since their articles usually refer to his publications as sources of the data under concern. To the coauthors, the citation of these papers, which appear to correctly show original measurements, must have seemed perfectly appropriate.
1. Analysis of our Data

As I will illustrate in the following with an example, it is not always possible for me to reproduce the analysis of our data published by the author of [1].

In the community it is controversially debated whether the boundaries’ critical current densities \( J_c \) and normal state resistivities \( \rho_n \) scale, for example like \( J_c \rho_n \propto 1/\rho_n^{3/2} \), as argued by him. This scaling is predicted by a model of the grain boundary mechanism, the intrinsically-shunted-junction (ISJ) model which he supports.

Figure 1 presents the \( J_c \rho_n \) products of bicrystals plotted as a function of \( J_c \) as published by him in [1], where he explains that the black datapoints of Fig. 1 are our data of table I [3]. I agree that several of these are our data: their \( J_c \) values, their \( J_c \rho_n \) products, and thereby also their \( \rho_n \) values are those that we have measured and published. Furthermore, other data of Fig. 1 (7.9 \( \times 10^4 \) A/cm\(^2\), 3.75 mV; 4.7 \( \times 10^5 \) A/cm\(^2\), 5.4 mV) that are not identified by black dots as ours, are the same as remaining data of table I [3].

If the data are plotted as a function of \( 1/\rho_n \), do they follow the proclaimed scaling behavior? According to the author of [1], our data scale well (Fig. 2) and thus provide evidence for the ISJ-model. However, it is not possible for me to reproduce the presentation of our data he is showing. The data that we measured and published are in clear disagreement with the scaling rule (Fig. 3) [4]. Why is it, that in his figures our data follow the scaling?

In Ref. [1] p. 108 its author mentions for the first time that he had ”reevaluated” our data of Fig. 3, selecting some and disregarding other data. He did not contribute to our studies, but he is of course welcome to use the data. Because we have evaluated our results correctly, I do not, however, see a possibility to improve the evaluation, and I am interested to know how he tried to do so. Unfortunately, [1] is very brief in this respect.

As a comparison of Fig. 2 with Fig. 3 shows, he furthermore apparently misinterpreted our data. To a large extent, the agreement with his model seems to originate from modifications of the \( \rho_n \) values of our data published in table I [3]. Because the apparent modifications are
extensive and change the conclusions to be drawn from our measurements, it is important
to know how he derived the specific values of his datapoints (Fig. 2) from our measurements
(Fig. 3). It is also important to understand why he still uses for his $J_c \rho_n(J_c)$ plots our $\rho_n$
values with little changes only (see Fig. 1 of [1]). His publications do not provide answers
to these questions. Refs. [5, 6], for example, present the data with the altered values,
but do not apprise the reader of the data’s origin or the interpretation they were subjected to.

Ref. [1] does not clarify the modifications of our data either, but criticizes our publication
[7] for not stating that the author of [1] found the scaling behavior at approximately the
same time as another group [8] did (see the comment presented as Ref. 17 in [1]). As we
describe in [7] on p. 506, Ref. [8] provided evidence that the $I_c R_n$ products of $45^\circ$ boundaries
scale with $J_c$ and $\rho_n$, in particular the $I_c R_n$ products of individual boundaries that are
gradually depleted from oxygen. R. Gross proposes a more universal scaling: he expects
the $I_c R_n$ products to scale with $J_c$ and $\rho_n$ if many boundaries with a variety of boundary
angles are compared. However, he has not provided convincing evidence that this universal
scaling exists. As discussed above, the datasets he presented as evidence are, for example,
not consistent with each other. His complaint is therefore unwarranted.

2. Comments on our Work Made in [1]

A large number of statements on our work presented in Ref. [1] are invalid or misleading,
as I will illustrate with a second example:

Fig. 4 presents data on the angular dependence of the grain boundary critical current
density $J_c$, plotted such that the datasets can be easily compared. It is evident that many
data of R. Gross shown in b) and d) are the same as the data of us and Z. Ivanov et al.
shown in a) and c), respectively. However, Refs. [5, 6, 9, in which b), d) or closely related
figures were published, do not use appropriate citations. They present the figures as if all
data were his.

My coauthor and I therefore cautioned in [7] (p. 498): “Data showing an exponential $J_c(\theta)$
dependence and critical current densities of [100]-tilt and [100]-twist boundaries have also been published by Gross and Mayer (1991) \cite{5} and Gross (1994a) \cite{6}. It seems that many of these data are reproductions of data given by Dimos et al. (1990) \cite{3} and Ivanov et al. (1991) \cite{10}.” The numbering of the references was adapted to fit the present manuscript. As Fig. 4 shows, our comment is correct and points towards a serious problem.

Instead of resolving this problem, the author of \cite{1} lists in the Ref. 43 of \cite{1} our statement in an altered form. He ignores that in addition to his paper Gross and Mayer (1991) \cite{5} our statement also concerns his article Gross (1994a) \cite{6}. Although modified by him only slightly, the statement now conveys an incorrect message, against which he is then arguing:

“In the recent review \cite{7} it is stated with respect Ref. \cite{3}: It seems that large part of the data are reproductions of data given by Dimos et al.1990 (\cite{3}) and Ivanov et al.1991 (\cite{10}). This is indeed true with respect to the data of Dimos et al. that have been included by courtesy of Dimos and Chaudhari. However, this is not true with respect to data of Ivanov et al. The paper by Ivanov et al. \cite{10} has been submitted about half a year later than the paper by Gross and Mayer \cite{5}. The data by Ivanov et al. have been included in a subsequent review \cite{11} by courtesy of Ivanov et al.” (cited from Ref. 43 of \cite{1}). The numbering of the references was adapted to fit the present manuscript.

The sequence of the submission and publication dates is as follows: The data of Dimos et al. were published first by Dimos et al. in \cite{3} (submitted 1989, published 1990) and afterwards by the author of \cite{1}, beginning with \cite{5} (published in 1991 without note to the submission date). The data of Ivanov et al. were published first by Ivanov et al. in \cite{10} (submitted and published 1991). The author of \cite{1} published afterwards the data of Fig. 4d in \cite{6, 9}, which both appeared without published submission date in 1994.

Although neither \cite{6, 9}, nor \cite{1} state so, it has to be concluded that the data of \cite{6, 9} shown in Fig. 4d are the data of Ivanov et al. \cite{10}. Our cautionary comment made in \cite{7} on p. 498 is therefore correct.

In summary, a large number of comments on our work presented in \cite{1} are invalid. I disproved some of them. By correcting, where possible, his articles in which he incorrectly
published our results or those of others, the author of [1] can resolve and settle the problems he started to address in [1].

I am grateful to all who supported me in writing this note.
References

[1] R. Gross, Physica C 432 (2005) 105.
[2] D. Dimos, P. Chaudhari, J. Mannhart, F. K. LeGoues, Phys. Rev. Lett. 61 (1988) 219.
[3] D. Dimos, P. Chaudhari, J. Mannhart, Phys. Rev. B 41 (1990) 4038.
[4] Ref. [1] proposes on p. 107 that a presumed low quality of our samples and problems of our measurements are the reason why our data do not scale. We indeed performed these experiments very early and the sample quality was improving rapidly in this time. However, to reconcile Figs. 2 and 3 by invoking experimental errors implies to suggest that for several samples we obtained from wrong $J_c$ and $\rho_n$ measurements correct $J_c\rho_n$ products. For the sample with the measured $1/\rho_n = 133/\mu\Omega cm^2$ and $J_c\rho_n = 1.2 mV$, for example, our measured $J_c$ and $\rho_n$ values would need to be a factor of 2.5 too large and too low, respectively. The only possible reason would be a hypothetical mistake in the measurement of the crosssection of the boundary bridges, such that the measured crosssection is a factor of 2.5 too small. Because we determined the crosssection of each bridge by optical microscopy and profilometry to an accuracy better than 30%, we can rule out that such errors occurred and that measurement problems are the reason for the differences between Figs. 2 and 3.

Note that other groups in later work did also not find the scaling behavior that was proposed by the author of [1] (for references see [7, 13]).

[5] R. Gross, B. Mayer, Physica C 180 (1991) 235.
[6] R. Gross, “Grain Boundary Josephson Junctions in the High Temperature Superconductors”, in Interfaces in High-Tc Superconducting Systems, edited by S. L. Shinde and D. A. Rudman (Springer Verlag), 1994, pp. 176-209.
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[9] R. Gross, “Structural and Electrical Transport Properties of Grain Boundaries in High Temperature Superconductors”, in Polycrystalline Semiconductors III, Solid State Phenomena, Vols. 37-38 (1994), Scitec Publications Ltd, Switzerland, pp. 107-118.
[10] Z. G. Ivanov, P. A. Nilsson, D. Winkler, J. A. Alarco, T. Claeson, E. A. Stepantsov, A. Ya. Tzalenchuk, Appl. Phys. Lett. 59 (1991) 3030.
Why [1] cites the paper in this context is unclear. The publication seems fine and I am not aware that it presents the data of Ivanov et al. More information by the author of [1] may reveal that he simply mistook the reference for another one.

[11] R. Gross, D. Koelle, Rep. Prog. Phys. 57 (1994) 651.

[12] R. Gross, P. Chaudhari, M. Kawasaki, A. Gupta, Phys. Rev. B 42 (1990) 10735.

[13] F. Tafuri, J.R. Kirtley, Rep. Prog. Phys. 68 (2005) 2573.
**Figure Legends**

**Fig. 1**
Data from an article by R. Gross, showing $J_c\rho_n$ products of bicrystal grain boundaries plotted as a function of the grain boundary critical current density $J_c$. As is evident from \[12\] and as he explains in \[1\], the solid data are our data from table I of \[3\]. The line shows the scaling behavior predicted by the ISJ-model. The data and this line are from \[12\], Fig. 3.

**Fig. 2**
Similar dataset as Fig. 1, taken from another publication of R. Gross \[5\]. The $J_c\rho_n$ products are plotted here as a function of the inverse boundary resistivity $1/\rho_n$. The match between this dataset and the dataset of Fig. 1 proves that both were obtained from the same samples. The solid data are therefore our data published in table I of \[3\]. The line shows the expected scaling behavior predicted by the ISJ-model. In this plot, our data follow the scaling behavior. The data and this line are from \[5\], Fig. 3a (see also \[6\], Fig. 6.5).

**Fig. 3**
The $J_c\rho_n$ products of our samples as measured and published (\[3\], table I), plotted as a function of $1/\rho_n$. Because the seven solid data of Fig. 2 are from the same dataset (\[3\], table I), they have to be identical to seven of the datapoints shown here, which is not the case. The line shows the scaling behavior according to Fig. 3a of \[5\]. Our data do not scale. One datapoint of table I, \[3\] ($I_cR_n=0.06$ mV, $\rho_n=0.02\,\Omega\mu\text{m}^2$) has not been plotted (see the comment given in \[12\] as Ref. 14).
Fig. 4
Grain boundary critical current densities $J_c$ measured as a function of the boundary angle.

a) Our data on the angular dependence of $J_c$, taken from table II of [3].

b) Data on the angular dependence of $J_c$ taken from R. Gross et al. (Fig. 2 of [5]). Together with additional data, these data were also published in Fig. 6.4a of [6] and in Fig. 3a of [9], always as if all data were his. A comparison with a) shows, however, that part of the data result from our work [3], which is now confirmed by [1].

c) Data from Z. Ivanov et al. (Fig. 4 of [10]), showing the $J_c$ of their samples measured as a function of the boundary angle.

d) Data from R. Gross (Fig. 6.4b of [6], also published in Fig. 3b of [9]), showing again $J_c$ as function of the angle. In these publications the figure is presented as if all data were data of his. It seems, however, that they are reproductions of the data of Z. Ivanov et al. shown in c).

The data shown in a) and b) have been measured at 4.2 K, those of c) and d) at 77 K. The hatched lines depict exponential $J_c(\theta)$-dependences. In a) one datapoint with $\Theta = 4^\circ$ and $\Phi = 4^\circ$ has not been plotted, because this boundary has two equally strong tilt-components.
Figure 3
Figure 4
Figure 4