Small grains: a key to high-field applications of granular Ba-122 superconductors?

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Abstract
The grain boundaries (GBs) of high-temperature superconductors (HTSs) intrinsically limit the maximum achievable inter-grain current density ($J_c$), when the misalignment between the crystallographic axes of adjacent grains exceeds a certain value. A prominent effect resulting from large-angle GBs is a hysteresis of $J_c$ between the increasing and decreasing field branches. Here, we investigate this feature for K- and Co-doped Ba-122 polycrystalline bulks with systematically varied grain size and find that the widely accepted explanation for this effect—the return field of the grains—fails. We use large-area scanning Hall-probe microscopy to distinguish $J_c$ from the intra-granular current density ($J_G$) in order to clarify their interactions. Measurements on Ba-122 bulks reveal that a large $J_c$ results from a small $J_G$ as well as small grains. An extended version of the model proposed by Svistunov and D’yachenko is successfully applied to quantitatively evaluate this behavior. The excellent agreement between the model and experiments suggests that the GBs limit the macroscopic current in all of the samples and that the inter-grain coupling is governed by Josephson tunneling. The predictions of the model are promising in view of realizing high-field HTS magnets. Our main result is that the field dependence of the $J_c$ of an untextured wire can be significantly reduced by reducing the grain size, which results in much higher currents at high magnetic fields. This result is not limited to the investigated iron-based materials and is therefore of interest in the context of other HTS materials.

Keywords: critical currents, granularity, iron-based superconductors, Josephson coupling, grain boundary currents, scanning Hall-probe microscopy

(Some figures may appear in colour only in the online journal)

1. Introduction
Round multifilamentary superconducting wires are the preferred choice for building high-field magnets such as those used in particle accelerators and fusion reactors [1]. Currently classic compounds, i.e. Nb–Ti and Nb₃Sn, are used for these applications since wires and tapes based on high-temperature superconductor (HTS) materials are not competitive so far. Their development is closely related to the optimization of the growth process and the grain boundary (GB) properties. Great efforts have been made to reduce the misalignment between the crystallographic axes of the grains in HTSs to prevent the weak-link character of large-angle GBs and thereby increase...
the maximum transport current. This has been successfully achieved for HTS tapes [2], but their geometry is less preferable than the round cross section of wires and their fabrication process is complicated and expensive.

The discovery of iron-based superconductors excited much interest because of their relatively high transition temperatures, small anisotropy, and high upper critical fields \((H_{c2} > 100 \text{T})\) [3]. Unfortunately, measurements of the critical current density across \([001]\)-tilt GBs in thin-film Co-doped Ba-122 bicrystals [4, 5], as well as magnetization and critical current measurements on polycrystalline iron-based compounds [6–8], indicate weakly linked GBs in these superconductors [9, 10], but their limiting effect on \(J_c\) is less pronounced compared to other HTSs [4]. Recently, results on Bi-2212 wires [11] have been promising for future HTS magnet applications. The large \(J_c\) of these wires is ascribed to a local biaxial texture of the grains [12]. The iron-based HTSs also have the potential for the production of superconducting wires. In contrast to Bi-2212, the wires based on Ba-122 in [13] are untextured, nonetheless, the achieved performance is much better compared to other untextured HTS wires. Still, major improvements of the iron-based materials are needed in order to become competitive with the long established Nb–Ti and Nb₃Sn wires currently used in superconducting magnets.

A feature of the weak-link character of the GBs is the hysteresis of the inter-grain critical current density \(J_c\) [14–18], which appears as an asymmetry in magnetization loops (see section 5 for details). The values of \(J_c\) are found to be smaller when the absolute value of the external field, \(|H_{\text{ext}}|\), is increased (referred to as the increasing field branch) compared to the case when \(|H_{\text{ext}}|\) is decreased (the decreasing field branch). The commonly accepted explanation of this effect is based on the reverse field \(H_{\text{return}}\) at the GBs arising from the intra-grain current density \(J^G\) [15, 19, 20]. The local magnetic induction \(B\) at the GBs is given by the vector sum of \(H_{\text{ext}}\) and \(H_{\text{return}}\). As a result the magnetization curve is shifted by the value of \(H_{\text{return}}\) relative to \(H_{\text{ext}}\). Therefore, \(H_{\text{return}}\) can be estimated from the field necessary to compensate this shift [20]. However, as shown later, the \(J^G\) necessary to explain the observed shift in a K-doped Ba-122 bulk with a grain radius of approximately 0.1 \(\mu\text{m}\) is about \(10^{12} \text{A/m}^2\), which is far larger than the maximum current density found in K-doped Ba-122 single crystals [21]. Although \(H_{\text{return}}\) contributes to the hysteresis of \(J_c\) to some extent, another effect has to be responsible for the observed shift in the magnetization curve.

An alternative description of the critical current hysterisis was suggested by Svistunov and D’yachenko [22], which is based on the results of the Josephson current density of a single Josephson junction [23]:

\[
J_f(B) = J_0 \frac{\sin(k(B)s)}{k(B)s}. \tag{1}
\]

Equation (1) is a different formulation for the Fraunhofer pattern, where the (normalized) flux through the junction \((\varphi/\phi_0)\) with \(\phi_0\) the magnetic flux quantum) is replaced by \(k(B)s\), which explicitly considers the length of the junction: \(2s\). \(J_0\) defines the maximum Josephson current density across the junction. Equation (1) already suggests the favorable property of a small junction length. The envelope of the Josephson current density \((J_f \sim J_0/|k(B)|)\) indicates that a small value of \(s\) (and also \(k\)) leads to higher values of \(J_f\) [24].

The field dependent variable \(k\) is derived by carrying out an integration over a closed path parallel to and across the GB [23], as visualized in figure 1. It defines the variation of the phase \(\varphi\) of the order parameter [23–25]:

\[
k(B) = \frac{\text{d}\varphi}{\text{d}y} = \frac{4\pi\mu_0}{\phi_0} \left( \chi J^G(B) + \frac{d}{\mu_0} B \right), \tag{2}
\]

Here, \(B\) is the magnetic induction which is parallel to the \(z\)-axis while the integration path is in the \(xy\)-plane, \(d\) is half of the thickness of the junction, and \(\lambda\) denotes the magnetic penetration depth of the material.

Svistunov and D’yachenko proposed that the intra-grain current density \(J^G\) is composed of a reversible contribution \(J^G_{\text{rev}}\), stemming from the reversible magnetization of the grain, and of an irreversible contribution \(J^G_{\text{irr}}\), stemming from the pinning of the flux-lines (e.g. surface and bulk pinning) [22]:

\[
J^G = J^G_{\text{rev}} + J^G_{\text{irr}}. \tag{3}
\]

If the local magnetic induction at the GB is smaller than the lower critical field \((|B|/\mu_0 < H_{c1})\) and no flux-lines are present inside the grains \((J^G_{\text{irr}} = 0)\), the current density at the GB is the Meissner shielding current density: \(J^G \sim B/|\mu_0\lambda\) (London model), and thus \(k\lambda = 4\pi(\lambda + d)B\phi_0/\phi_0\) is equivalent to the term \(\pi\varphi/\phi_0\), which is a more common representation for \(k\lambda\) in the literature [23]. Equation (2) allows the extension of the description of Josephson junctions to the mixed state \((H_{c1} < |B|/\mu_0 < H_{c2})\), where flux-lines have penetrated the superconductor [24, 25]. \(J^G_{\text{rev}}\) is defined by the distribution of the flux-lines when no pinning is present inside the grain. With pinning, this flux-line configuration is modified compared to the ideal case, i.e. a field gradient develops, which is described by \(J^G_{\text{irr}}\). The value of \(J^G_{\text{irr}}\) depends on the history of the external field, while \(J^G_{\text{rev}}\) is determined by

![Figure 1. Visualization of the geometry and the parameters used to model the Josephson current density across a weak link.](image-url)
the external field itself (see section 2). These different dependencies are responsible for the hysteresis of $I_c$ [22].

The attempt to verify the model proposed by Svitunov and D’yachenko requires a measurement technique which is able to quantify $J^G$ and $I_c$. Magnetization measurements probe the global magnetic response of a sample. They provide very limited insight into the spatial distribution of the local magnetic field, or the values and interactions of the inter- and intra-granular currents that are essential to test the predictions of the model. We measured the history dependence of $J^G$ and its impact on $I_c$, i.e. the $I_c$-hysteresis, with scanning Hall-probe microscopy (SHPM). This technique has the advantage of allowing the detection of $J^G$ and $I_c$ simultaneously.

2. Model extensions and predictions

Svitunov and D’yachenko used the equations describing a single Josephson junction to explain the hysteresis of $I_c$ [25]. However, in a polycrystal the inter-grain current has to cross multiple junctions (i.e. GBs) whose lengths vary statistically. In order to account for this, the Josephson current density of a single junction (1) has to be integrated over the distribution density of the junction lengths, i.e. the grain radii $s$, which is approximated by:

$$P(s, s_0, m) = \frac{1}{\Gamma(m)} \frac{s^m}{s_0^m} \exp\left(-\frac{ms}{s_0}\right),$$

with $s \in [0, \infty)$ and $\Gamma$ the gamma function. The parameter $s_0$ denotes the mode of the distribution density function, i.e. the value where $P$ has a maximum, which is referred to as characteristic grain size in the following. A large value of the dimensionless variable $m$ corresponds to a smaller width of the distribution. Following the calculations by Gonzalez et al. [26] we obtain an estimate for the inter-granular critical current density of the sample by integrating equation (1) multiplied by $P(s, s_0, m)$ over $s$:

$$J_c(\alpha) = J_0 \frac{\alpha^{m+1}}{\Gamma(m)} (-1)^{m-1} \frac{\partial \alpha^{m-1}}{\partial \alpha^{m-1}} \coth(\alpha/2),$$

where $\alpha = m \pi / (k |s_0|)$. Our definition of $\alpha$ is slightly different from that in [26], because we choose a different distribution density function (4) where the value of $s_0$ is independent of $m$. In principle $m$ can take any positive value, but it must be an integer in (5). The Fraunhofer pattern from (1) disappears in (5) and is replaced by a smooth function, which can be applied to quantitatively evaluate the current densities in polycrystalline HTS materials. Accordingly, the value of $J_c$ is larger if $\alpha$ is large, which means that small values of $|k|$ and $s_0$ are preferable.

The influence of the current densities inside the grains on $J_c$ is sketched in figure 2 based on (2), (3) and (5). Plots A and B show the simplified behavior of $J^G_c$ and $J^G_{rev}$ when the external field is ramped from positive to negative values. The sign of $J^G_c$ is always negative because the pinning force preserves the field gradient inside the grains, while the sign of $J^G_{rev}$ changes from positive to negative when passing through zero field. The superposition of the two current densities is illustrated by the insets of plot C for the respective field branches. They can be pictured as flowing in opposite directions in the decreasing field branch so that the magnitude of $J^G$ is smaller than in the increasing field branch where they flow in the same direction. In the increasing field branch $B$, $J^G_P$ and $J^G_{rev}$ are all negative so that $|k|$ is larger than in the decreasing field branch where $J^G_{rev}$ is positive. Hence, $I_c$ is smaller in the increasing field branch than in the decreasing field branch.

According to the model, the shift of the inter-grain magnetization peak near zero field is determined by the values of $J^G_{rev}$ and $J^G_P$. This is seen in figure 2C, where $I_c(B)$ is calculated from (5) from which we know that the maximum current density $J_0$ occurs at $k = 0$. The dotted curve shows the case $J^G_{rev} = J^G_P = 0$, where the peak is located at $B = 0$. If $|J^G_{rev}| < |J^G_P|$ (see figure 2A), the maximum of $I_c$ occurs in the decreasing field branch, because $J^G$ is negative in the decreasing field branch and thus $\chi^2 |J^G|$ subtracts from $dB/\mu_0$ in (2). The larger $|J^G_{rev}|$, the further the peak (i.e. $k = 0$) is shifted to higher fields of the decreasing field branch. If the absolute values of $J^G_{rev}$ and $J^G_P$ are exchanged (see figure 2B), the maximum of $I_c$ is located at $B = 0$ but is smaller than $J_0$ because $J^G$ is positive in the decreasing field branch and therefore: $k > 0$. The behavior is unchanged in the increasing field branch because $J^G_{rev}$ and $J^G_P$ flow in the same direction, thus the value of $J^G$ is the same although the values of $J^G_{rev}$ and $J^G_P$ are exchanged.

The return field effect [15, 20] and the effect just described always occur together because both originate from $J^G$. Nonetheless, for small grains with a moderate aspect ratio, the latter is far more important.

3. Samples and experiments

We investigated three optimally K-doped and three optimally Co-doped polycrystalline Ba-122 bulk samples. The K-doped samples were synthesized using a mechano-chemical reaction path described in [27]. The grain size was controlled by applying different temperatures during the final heat treatment. The samples were sintered for 10 hours in a hot isostatic press at 600 °C, 900 °C, and 1000 °C, where lower temperatures result in a smaller grain size.

The Co-doped Ba-122 polycrystalline bulk samples were synthesized using different starting powders and processing techniques [28]. For the large- and medium-grained samples, the starting materials were Ba, FeAs, and CoAs, while elemental powders were used for the small-grained sample. The powder of the large-grained sample was mixed and ground in an agate mortar and the other two samples were mixed by high-energy ball-milling. All samples were heat treated for 48 hours at 900°C for the large- and medium-grained sample and at 600 °C for the small-grained sample.

The magnetization of all samples was measured in a 7 T SQUID magnetometer and a 5 T vibrating sample magnetometer. The field profiles of the samples were recorded using a SHPM set-up with a spatial resolution of approximately
1 μm, and a scan range of 3 × 3 mm², which is located in an 8 T cryostat with an operable temperature range of approximately 3–300 K. The size of the active area of the Hall-probes is 400 × 400 nm².

The size of the grains is statistically distributed, which is described by the distribution density function \( P(s, s_0, m) \) (4). The value \( s_0 \) defines the grain size where the distribution density function has a maximum, which is approximately 0.1, 1, and 3 μm for the different K- and Co-doped samples. These characteristic grain radii are referred to as small, medium, and large. As an example, figure 3 shows the grain radii distribution density and its fit for the medium-grained K-doped sample, which was evaluated from polarized light images of the sample surface by choosing multiple lines of the image and measuring the distance from one GB to another. The grain size distribution of the large-grained sample was evaluated in the same way while \( s_0 \) of the small-grained sample was roughly estimated from transmission electron microscopy images utilizing the intercept technique [13].

4. Field profile evaluation

The inter- (\( J_I \)) and intra-grain current density (\( J^G \)) are extracted from SHPM data, i.e. the field profiles above the samples. We evaluate \( J_c \) by fitting an analytical function derived in [29] to the global field profile, where the spatially

Figure 2. Simplified dependence of \( J^G \), \( J^G_\text{rev} \), and \( J_\text{P}^G \) on an external field which is ramped from positive to negative values (A and B), and the impact on \( J_c \) predicted by (5) (C).

Figure 3. Grain size distribution density of the K-doped sample with medium-sized grains and the fit with equation (4), where \( m = 3 \). The bars represent the normalized number of grains with a radius in the interval \( \Delta s \), which is given by the width of the bars.
constant parameter \( J_c \) is the only free parameter. Figure 4A shows an example for a field profile generated by \( J_c \), \( J_c \) crosses the GBs as illustrated in figure 4C, leading to the global field profile shown in figure 4A.

The intra-grain current density \( J^G \) is confined to the individual grains as indicated in figure 4D by the small arrows. If the distance between the Hall-probe and sample surface is larger than the dimensions of the grains, the fields, which are generated by the intra-grain current densities of neighboring grains, cancel each other in a first order approximation (hatched area), except for a narrow region at the sample edge. Thus, the intra-granular field profile can be approximated by a field profile resulting from a current density, \( J^G \), flowing in a thin surface layer of thickness \( s_0 \) (large dashed arrows). The resulting field profile for this case is plotted in figure 4B. To fit the field profile originating from \( J^G \) we again utilize the equation from reference [29]. With this technique we are able to evaluate \( J^G \) from SHPM data quantitatively.

Figure 5 shows examples of field profiles obtained from the medium-grained K-doped sample and fits of the aforementioned analytical function. The field profiles were recorded along a field run from 3 to \(-3\) T. First the profile was measured in the decreasing field branch at \( \mu_0 H_{\text{ext}} = 30 \) mT and then in the increasing field branch at \( \mu_0 H_{\text{ext}} = -30 \) mT. \( J^G \) can be identified as the slopes at the sample edges in SHPM measurements, while \( J_c \) is proportional to the slopes in the remaining sample space, as indicated in figure 5A.

In order to make the SHPM results comparable to the magnetometry data, the obtained values for \( J_c \) and \( J^G \) are converted to a magnetization \( M \) using the standard formula for cubic samples [30] (compare to section 5.2, e.g. figure 8).

5. Results and discussion

5.1. Qualitative discussion

Figure 6A shows the hysteresis of the transport current density \( J_c \) of a K-doped Ba-122 wire. The radius of the grains is approximately 0.1 \( \mu \)m. This hysteresis corresponds to an asymmetry in the magnetization \( M \) of a bulk sample with a similar grain size, which is highlighted in figure 6B by the shaded areas. The asymmetry is given by the difference between the decreasing field branch \( \langle H_{\text{ext}} \rangle \) (ramped to smaller values) and the increasing field branch \( \langle H_{\text{ext}} \rangle \) (ramped to larger
values) after mirroring the latter about the reversible magnetization $M_{rev}$, as defined in [31] with $\lambda = 190$ nm and $\kappa = 100$. The comparison between $J_c$ in figure 6A and the $J_c$ evaluated from magnetization measurements shows good agreement [13].

Another distinct feature is the different behavior of the samples in figure 6B. The curve of the large-grained sample (dash-dotted curve) is very similar to the magnetization curve of single crystals with the maximum value of $M$ in the increasing field branch and its asymmetry is small. The asymmetry increases with decreasing grain size and another peak near $H_{ext} = 0$ emerges for a grain radius of 1 $\mu$m (dashed line). This peak is located in the decreasing field branch and becomes larger and wider when the grain radius is decreased to 0.1 $\mu$m (solid line), while the peak located in the increasing field branch disappears.

The Co-doped samples exhibit a similar behavior with a smaller magnitude of the magnetization curves. We observe the largest asymmetry in the sample with the smallest grain size, the two peaks in the magnetization curve of the medium-

Figure 5. Examples of fits to the field profiles of the medium-grained K-doped sample. The dotted vertical lines indicate the sample edges. The scans were performed across the sample center ($y = 0$) as defined by the inset in the upper right corner of panel B. Note the much larger trapped field in the decreasing field branch.

Figure 6. Hysteresis of the critical current density between the increasing and decreasing field branches of a K-doped Ba-122 wire with a modal grain radius ($s_0$) of approximately 0.1 $\mu$m measured by transport current (panel A) at 4.2 K. Magnetization loops of polycrystals with different grain sizes measured in a vibrating sample magnetometer (panel B) at 5 K.
grained sample, and only one peak in the increasing field branch for the large-grained sample. This indicates that the asymmetry effects are governed by the grain size of the individual polycrystals and not by the dopant, therefore we concentrate on the results obtained from the K-doped samples.

Figure 7 displays the field profiles of the K-doped samples measured at various constant external fields along a run from 3 to −3 T. In the small-grained sample (figure 7A), the field profiles in the decreasing field branch are shaped roughly in accordance with Bean’s critical state model, i.e. a constant slope of the field profile corresponding to a spatially uniform $I_c$. Hardly any contribution of the intra-grain currents is observed when looking at the sample edges, which indicates that the peak in the decreasing field branch in figure 6B originates from the inter-granular currents. When the field is ramped through zero, the field profile starts to ‘collapse’ from the sample edges towards the center. The difference in the magnitude of the slopes at comparable positive and negative fields is striking. For instance, the slope at 0.5 T (decreasing field branch) is 3 times larger than at −0.5 T (increasing field branch).

No Bean-like field profile is observed for the large-grained sample (figure 7C). Instead, a rather flat field distribution across the sample is present, except for a large field gradient at the sample edges. This is characteristic of a field profile dominated by the contribution of the individual grains and thus the peak in the increasing field branch in figure 6B is caused by the intra-granular current of the grains.

The measured field profiles of the sample with the medium-sized grains show an intermediate behavior between the large- and small-grained samples (figure 7B). When the field is ramped from the maximum applied field to zero, an inter-granular field profile builds up, while hardly any contribution of the intra-grain currents is observed. When the field passes through zero, the sample behaves like the small-grained sample, i.e. the slope of the inter-granular field profile becomes smaller. Simultaneously, the intra-grain currents emerge and start to dominate, leading to a flat field distribution across the sample. The hysteresis of $I_c$ is clearly visible when a field profile of the increasing field branch is compared to one recorded in the decreasing field branch at the same $|H_{ext}|$. In the decreasing field branch the profiles exhibit a much steeper slope, whereas $|J_c|$ is noticeably larger in the increasing field branch.

The SHPM data in figure 7 reveal that the magnetization peak in the decreasing field branch in figure 6B originates from the inter-granular current density $I_c$, and the peak in the increasing field branch originates from the intra-granular current density $J_c$. Accordingly, the peak of the small-grained sample is determined by $I_c$. When the peak on the field axis is generally explained in the context of the return field of the grains, $H_{\text{return}}$, which results from $J_c$. Following [20], the peak occurs when the local magnetic
induction at the GBs is zero, i.e. when \( H_{\text{ext}} + H_{\text{return}} = 0 \), and therefore \( H_{\text{return}} \) coincides with the field at which the peak is found: \( \mu_0 H_{\text{return}} \approx 0.1 \, \text{T} \). The value of \( J^G \) necessary to generate this field is: \( |J^G| > |H_{\text{return}}|/\mu_0 \approx 10^2 \, \text{A m}^{-2} \), for a moderate aspect ratio of the grains (which is true for the investigated small-grained sample). If we compare this value to the maximum current density found in K-doped Ba-122 single crystals [21]: \( 5 \times 10^{10} \, \text{A m}^{-2} \), the return field on its own appears to be a highly questionable explanation for the observed behavior.

Clear evidence that \( H_{\text{return}} \) alone cannot be responsible for the \( J_c \)-hysteresis is observable in figure 7A. Simply put, the return field has to be of the order of the trapped field inside the grains which corresponds to the height of the field step at the sample edges (compare figures 4B and 5A). Since the step height barely depends on the actual geometry of the intragranular current flow, this argument includes scenarios where the magnetic grain size differs from the crystallographic grain size (\( \delta_0 \)), i.e. the formation of grain-clusters, characterized by a number of connected grains with low-angle GBs that do not hinder the current transport from one grain to another and therefore behave like a single grain. However, such a field step is not present in the SHPM measurements in figure 7A. Consequently, another effect has to contribute to the hysteresis of \( J_c \).

The model presented in this paper can account for this behavior using much smaller values of \( |J^G| \). As discussed in section 2, the peak position of \( J_c \) is determined by \( J^G_\text{rev} \) and \( J^G \) \( (|J^G_\text{rev}| < |J^G|) \). To shift the peak of the small-grained sample to the experimentally determined 0.1 T, \( |J^G| \) has to attain a value of approximately \( 3 \times 10^8 \, \text{A m}^{-2} \) in the decreasing field branch—a much more realistic current density. Our SHPM data directly confirm the predictions of the presented model. We will show this below for the medium-grained sample, which allows the simultaneous detection of inter- and intra-grain currents.

5.2. Quantitative evaluation

The magnetizations predicted from the SHPM measurements on the medium-grained sample, i.e. \( M_{\text{SHPM}} \), stemming from the inter-grain current density (\( J_c \)), and \( M_{\text{SHPM}} \), stemming from the intra-grain current density (\( J^G \)), are shown in figure 8. The values are extracted from the data shown in figure 7B as described in section 4. The sum of the two contributions agrees well with the SQUID data, which supports the reliability of our evaluation procedure. The equally evaluated data of the small- and the large-grained samples (figure 7A and B) also show agreement between the magnetometry and SHPM measurements.

At \( \mu_0 H_{\text{ext}} = 0.03 \, \text{T} \), the applied field where the maximum magnetization is observed, the slope of the global field profile corresponds to a \( J_c \) of approximately \( 1.9 \times 10^8 \, \text{A m}^{-2} \). The value of \( |J^G| \) at the intra-grain peak position (\( -0.17 \, \text{T} \)) is approximately \( 3.7 \times 10^{10} \, \text{A m}^{-2} \).

The \( J_c \) values, obtained from the SHPM data, together with equation (5) are used to calculate a value of \( J_c \) and consequently \( M_{\text{Model}} \), the magnetization corresponding to the predicted \( J_c \). The grain radius distribution parameters \( m = 3 \) and \( \delta_0 = 1 \, \mu\text{m} \) are evaluated from images of the sample surface (see section 3). The self-field of the sample is taken into account by substituting \( B = \mu_0 H_{\text{ext}} + \Delta H_{\text{profile}}/2 \) in (2), where \( \Delta H_{\text{profile}} \) is the difference between the maximum and minimum field values of the (measured) field profile. The thickness of the GBs, \( d \), the penetration depth \( \lambda \), and the maximum current density, \( J_0 \), are the fitted parameters. The obtained values of \( M_{\text{Model}} \) are plotted in figure 8 for comparison with \( M_{\text{SHPM}} \).

Figure 8 shows the advantage of large-area SHPM scans. The sum of \( M_{\text{SHPM}} \) and \( M_{\text{SHPM}} \) clearly reproduced the unconventional double-peak in the magnetization curve \( M_{\text{SQUID}} \), and \( M_{\text{Model}} \) closely follows \( M_{\text{SHPM}} \). The fitted value of \( J_c = (1.9 \pm 0.4) \times 10^8 \, \text{A m}^{-2} \) corresponds to the maximum inter-grain current density extracted from the field profiles in figure 7B (\( J_c = 1.9 \times 10^8 \, \text{A m}^{-2} \)). The penetration depth \( \lambda = 180 \pm 20 \, \text{nm} \) is comparable to the results obtained in [32], where the authors found a value of 200 nm at 10 K. In [33] the authors investigated identically synthesized samples by atom-probe tomography. Their data show that the length scale of the composition variation of Ba, K, Fe, As, and O across the GBs is about 5–15 nm. This length is comparable to the fitted total thickness of the GBs: \( 2d = (5.4 \pm 1.5) \, \text{nm} \).

6. Implications

We discussed the hysteresis of \( J_c \) in dependence of \( J^G \) in figure 2. In the increasing field branch \( J^G_\text{rev} \) and \( J^G \) flow in the same direction and \( J^G_\text{rev} \) is large while \( J^G \) is small. According to the presented model, \( J_c \) in the increasing field branch should decrease further if \( J^G \) becomes larger. To validate this prediction, a bulk sample with an approximate grain radius of 0.1 \( \mu\text{m} \) was exposed to fast neutron irradiation. This
procedure induces defects in the crystallographic structure of the grains which are capable of pinning flux lines, thus increasing $|J_0^0|$ [34]. Magnetization measurements performed after neutron irradiation confirm the decrease of $I_c$ in the increasing field branch. This implies that a smaller value of $J^0$ would increase $I_c$, i.e. the pinning in polycrystalline HTS with large-angle GBs should be minimized.

The impact of the modal grain radius, $s_0$, on the inter-grain critical current density, $I_c$, is most interesting in view of the production of technical superconductors. In contrast to $k$, $s_0$ is independent of the field and allows a more precise interpretation. Figure 9 visualizes the dependence of $I_c$ of a Josephson junction network with different $s_0$ in the decreasing (A) and increasing (B) field branches. A small $s_0$ in equation (5) entails a diminished field dependence of $I_c$. The dashed arrows in the graphs highlight the corresponding increase of $I_c$.

In the framework of the presented model the most advantageous grain size of a weak-linked superconductor is as small as possible. For the examined iron-based superconductors the grain size can be changed by milling the starting materials and by varying the heat treatment of the bulks. The model neglects the influence of the grain size on other superconducting properties, such as the transition temperature, the upper critical field, or the anisotropy. In reality the optimal/minimal grain size will be determined by the deterioration of the superconducting properties of the grains.

Neglecting this deterioration, a magnetic induction $B_{\text{max}}$ can be extracted from figure 9B up to which a wire with a certain grain size can be used in typical applications. The value of $B_{\text{max}}$ is defined by the point where $|J_c|$ drops below a minimum value $J_{\text{app}}$, which is determined by the required critical current density of a certain application (see figure 9B). $B_{\text{max}}$ can be derived from (5). For typical values of $\lambda$ ($\approx 200$ nm), $d$ ($\approx 2$ nm), and $|J^0|$ ($\approx 10^{10}$ A m$^{-2}$) the field dependence of $k$ is mainly determined by the term $4\pi dB/\phi_0$ for $B > 1$ T, which is true for high-field applications. In this case $\alpha = m\gamma/(k|s_0|)$ is smaller than 1 and (5) can be expanded into a Taylor series at $\alpha = 0$, yielding the first order approximation:

$$B_{\text{max}} \approx \frac{\phi_0}{2\pi} \frac{|J_0|}{2} \frac{1}{J_{\text{app}} s_0 d}. \quad (6)$$

The value $J_{\text{app}}$ typically lies in the range of $10^8$–$10^9$ A m$^{-2}$, depending on the filling factor of the wire and the envisioned application. Equation (6) indicates that $B_{\text{max}}$, which defines the operable field range of a wire, can be increased by one order of magnitude by reducing the grain size by one order of magnitude.

7. Conclusions

We presented a model describing the history dependence of $I_c$ found in polycrystalline HTSs. The irreversible currents in the grains reduce $I_c$ in the increasing field branch, while they enhance it in the decreasing field branch. We investigated K- and Co-doped Ba-122 polycrystals of three different grain sizes via magnetization and SHPM measurements. With SHPM, both the inter- and intra-granular current density are accessible, which makes SHPM measurements superior to magnetization and transport measurements in order to understand the current flow in granular materials. The model is able to describe the obtained inter-granular current densities based on the intra-granular current densities both qualitatively and quantitatively. It allows the estimation of the field range in which a wire can be operated from the size of the grains inside the wire. This field range increases significantly as the grain size is reduced, a feature which is confirmed experimentally. Our results show that the grain size is a key parameter for increasing the maximum operable field range of polycrystalline HTS materials that are governed by Josephson tunneling. In its essence the model is independent of the material, and therefore our findings are also relevant to other HTS materials.

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References

[1] Bottura L, de Rijk G, Rossi L and Todesco E 2012 IEEE Trans. Appl. Supercond. 22 4002008
[2] Shiohara Y, Yoshizumi M, Takagi Y and I T 2013 Physica C 484 1
[3] Aswathy P M, Anooja J B, Sarun P M and Syamaprasad U 2010 Supercond. Sci. Technol. 23 073001
[4] Katase T, Ishimaru Y, Tsukamoto A, Hiramatsu H, Kamiya T, Tanabe K and Hosono H 2011 Nat. Commun. 2 409
[5] Lee S et al 2009 Appl. Phys. Lett. 95 21
[6] Eisterer M, Zehetmayer M, Weber H W, Jiang J, Weiss J D, Yamamoto A, Hellstrom E E, Larbalestier D C, Zhigadlo N D and Karpinski J 2010 Supercond. Sci. Technol. 23 054006
[7] Haindl S et al 2010 Phys. Rev. Lett. 104 077001
[8] Yamamoto A et al 2008 Supercond. Sci. Technol. 21 095008
[9] Durrell J H, Eom C-B, Gurevich A, Hellstrom E E, Tarantini C, Yamamoto A and Larbalestier D C 2011 Rep. Prog. Phys. 74 124511
[10] Seidel P 2011 Supercond. Sci. Technol. 24 043001
[11] Larbalestier D C et al 1997 Nat. Mater. 13 375
[12] Kametani F, Jiang J, Matras M, Abraimov D, Hellstrom E E and Larbalestier D C 2015 Sci. Rep. 5 8285
[13] Weiss J D, Tarantini C, Jiang J, Kametani F, Polyanskii A A, Larbalestier D C and Hellstrom E E 2012 Nat. Mater. 11 682
[14] Däumling M, Sarnelli E, Chaudhari P, Gupta A and Lacey J 1992 Appl. Phys. Lett. 61 1355
[15] Evetts J E and Glowacki B A 1988 Cryogenics 28 641
[16] Kunchur M N and Askew T R 1998 J. Appl. Phys. 84 6763
[17] List F, Kroeger D and Selvamanickam V 1997 Physica C 275 220
[18] McHenry M E, Maley M P and Willis J O 1989 Phys. Rev. B 40 2666
[19] Palau A et al 2004 Appl. Phys. Lett. 84 230
[20] Palau A, Puig T, Obradors X and Jooss C 2007 Phys. Rev. B 75 054517
[21] Kihlstrom K J et al 2013 Appl. Phys. Lett. 103 202601
[22] Svistunov V M and Dayachenko A I 1992 Supercond. Sci. Technol. 5 98
[23] Barone A and Paterno G 1982 Physics and Applications of the Josephson Effect (New York: Wiley)
[24] Bulaevskii L N, Clem J R, Glazman L I and Malozemoff A P 1992 Phys. Rev. B 45 2845
[25] Svistunov V M, Nomine T, Fukami T, Yamamoto T and D’Yachenko A I 1997 Mod. Phys. Lett. B 11 1133
[26] Gonzalez J L, Mello E V L, Orlando M T D, Yuge E S and Baggio-Saitovitch E 2001 Physica C 364-5 347
[27] Weiss J D, Jiang J, Polyanskii A A and Hellstrom E E 2013 Supercond. Sci. Technol. 26 074003
[28] Hayashi Y, Yamamoto A, Ogino H, Shimoyama J and Kishio K 2014 Physica C 504 28
[29] Forkl A and Kronmüller H 1995 Phys. Rev. B 52 16130
[30] Wiesinger H P, Sauerzopf F M and Weber H W 1992 Physica C 203 121
[31] Brandt E H 2003 Phys. Rev. B 68 054506
[32] Li G, Hu W Z, Dong J, Li Z, Zheng P, Chen G F, Luo J L and Wang N L 2008 Phys. Rev. Lett. 101 107004
[33] Kim Y, Weiss J D, Hellstrom E E, Larbalestier D C and Seidman D N 2014 Appl. Phys. Lett. 105 162604
[34] Eisterer M, Weber H W, Jiang J, Weiss J D, Yamamoto A, Polyanskii A A, Hellstrom E E and Larbalestier D C 2009 Supercond. Sci. Technol. 22 065015