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Microscopic origin of highly enhanced supercurrent in 122 pnictide superconductor

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ABSTRACT

By a combination of microstructure analysis techniques, we reveal the structural origin of the extremely high supercurrent (up to the practical level of 0.1 MA/cm^2 at 10 T, 4.2 K) in $Sr_{0.6}K_{0.4}Fe_2As_2$ (122) tape. Transmission Kikuchi diffraction analysis reveals that hot pressing promotes a very high fraction of low-angle grain boundaries and texturing of the crystals, which is beneficial for the intergrain physical properties. Moreover, the unique characteristics of low-angle grain boundaries favor both long-range dislocations and short-range dislocations that totally change the pinning mechanism of the bulk 122 system. These defects combined with the grain texturing are not only effective for pinning vortices in the superconducting state, but also improve inter-granular supercurrent degradation, leading to substantially enhanced supercurrent over a wide range of magnetic fields.

1. Introduction

The competitive advantage of superconducting materials is the ability to carry high current, thus generating strong magnetic fields in significant volumes. Due to the small size of a single crystal, the superconductors used for applications are polycrystalline wires or tapes, where many grain boundaries (GBs) are inevitable. The GBs coexist with the superconducting matrix in the form of a network, across which, long-range supercurrent has to pass, evoking the importance of GB superconducting properties. It is universally found that the critical current density, J_c, is proportional to the density of GBs for type-I low temperature superconductors, such as Nb-Ti, $SnMo_6S_8$, and MgB_2 [1–4]. In this type of superconductor, GBs are not intrinsic barriers to supercurrent flow, and the low binding energy of vortices to GBs leads to the enhancement of vortex pinning capacity [5–7]. Nevertheless, type-II high temperature superconductors, which are expected to have a larger application market, suffer from obvious weak-link behaviour of GBs. The weak-link behaviour is caused by the quasi-two-dimensional (2D) phase, which produces weak growth texture, with almost randomly distributed values of the GB misorientation angle, θ. The GB critical current density ($J_{cgb}(\theta)$) falls off exponentially when the GB misorientation angle θ exceeds the critical angle θ_c, which has a value of 3–5° [8,9]. Recently, in the newly discovered iron-based superconductors (IBSs), in the $BaFe_2As_2$-122 system and the Fe(Se,Te)-11 system, the value of θ_c could be improved up to 9°, making IBSs more promising materials for application in high J_c superconducting tapes [10,11]. Since high J_c of more than 1 MA/cm [2] has been reported in single crystal and epitaxial thin films of IBSs, the next step is achieving special grain-to-grain low-angle misorientation and improving the texture of the whole sample [12–15]. In a recent breakthrough, transport J_c at a practical level (0.1 MA/cm^2) at 4.2 K and 10 T was realized by the improvement of *c*-axis textured superconducting tape [15,16]. Detailed analysis of the microstructure to reveal the origin of the high J_c is still ambiguous, however.

In this work, we carried out transmission Kikuchi diffraction (TKD) using a Zeiss Auriga scanning electron microscope to quantitatively describe the crystal phase formed at each point of the micrograph with high resolution. It was found that special GBs with

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low angle misorientation combined with high texture along the *c*-axis are promoted by using the hot pressing (HP) method. Transmission electron microscopy (TEM) with weak beam dark field (WBDF) images reveals that both long-range dislocations and short-range dislocations are assembled in low-angle GBs, which totally change the pinning mechanism of the bulk 122 system. Textured crystal structure, special low-angle GBs, and a high density of defects are leading to the improvement of supercurrent to a practical level over a wide range of magnetic fields.

2. Experimental section

Preparation of Sr-122 tape: Ag-clad $Sr_{0.6}K_{0.4}Fe_2As_2$ tapes containing Sn as an additive were fabricated by the *ex-situ* powder-in-tube (PIT) method. Sr filings, K pieces, and Fe and As powders in the ratio of Sr: K: Fe: As = 0.6: 0.5: 2: 2.05 were mixed for 12 h by the ball-milling method. The milled powders were packed into Nb tubes and then sintered at 900 °C for 35 h. The as-prepared Sr-122 superconducting precursors were then ground into powders under Ar atmosphere. In order to increase the grain connectivity, the precursors were mixed with 5 wt% Sn by hand with an agate mortar. Then, the fine powders were packed into Ag tubes with outer diameter (OD) of 8 mm and inner diameter (ID) of 5 mm. These tubes were sealed and then cold worked into tapes (~0.4 mm thickness) by swaging, drawing, and flat rolling. Finally, hot pressing was performed on the 60 mm long tapes under ~30 MPa at the sintering temperature of 850 °C for 30 min. More detailed information on the fabrication can be found elsewhere [16,19].

Superconducting properties measurements and microstructure characterization of Sr-122 tape: Transport and magnetization measurements under different magnetic fields were carried out on a 14 T physical properties measurement system (PPMS). The transmission Kikuchi diffraction was carried out using a Zeiss Auriga scanning electron microscope, which enabled us to quantitatively describe the crystal phase formed at each point of the micrograph with a minimum resolution of about 1 nm. The orientation of the crystals at each point was determined by using a HKL Nordlys 2 system. Electron microscopy studies were carried out using a JEOL 2200FS instrument equipped with electron energy loss spectroscopy, with a 200 kV field emission gun and an in-column omega type energy filter.

3. Results and discussion

3.1. Superconducting properties of HP Sr-122 tape

Fig. 1(a) displays the comparative results of investigations of the magnetic field dependence of transport J_c at 4.2 K for tapes or wires fabricated by different methods. The $(Sr,K)Fe_2As_2$ wires produced by the simple rolling technique exhibit the lowest J_c (less than 10^4 A/cm^2) among all the results [17]. The value of J_c was improved up to more than 10^4 A/cm^2 by cold pressing with enhanced pressure in tape samples [17]. For the wires synthesised by cold isostatic pressure combined with a hot isostatic pressure process, the self-field J_c reached as high as 0.1 MA/cm^2, which is a criterion for practical application [18]. J_c shows a rapid decrease in low field, however, reflecting the typical weak-link behaviour between GBs [11]. Such a dip could be related to the large GB misorientation induced by randomly distributed grains and the FeAs wetting phase formed in the GBs, both of which decrease the J_c in GBs [18]. J_c of 0.1 MA/cm^2 at 10 T has been realized in hot pressed tapes with high density and *c*-axis orientation [19]. It should be noted that this is the first time that J_c of IBS tapes has reached the practical level of 0.1 MA/cm^2 at 10 T. Another significant phenomenon is that J_c shows almost field independence in the measured field range, indicating strong pinning in the tape. Fig. 1(b) shows the temperature dependence of the upper critical field, B_{c2}, for both $B//ab$ and $B//c$. A small anisotropy is calculated by using $\gamma = B_{c2}^{B//ab}/B_{c2}^{B//c}$ ~1.5, which is comparable to the value for 122 single crystals, but larger than that for polycrystalline samples (~1) [20], indicating that the tape has textured structure. Moreover, the smaller anisotropy value compared to those of other Fe-based superconductors (>3) is suitable for potential magnetic applications.

3.2. Grain orientation map of HP Sr-122 tape

The transmission Kikuchi diffraction (TKD) technique is used to analyse the distribution of grain orientations. Fig. 2(a)-(c) provides examples of microstructure, band contrast, and inverse pole figure images derived from the TKD map after noise removal, respectively. The grain size variation is huge, from 2 μm down to as small as 10 nm. Nevertheless, the smaller grains are the dominant group, with average grain size of 200 nm, which is similar to the findings of a previous report on Ba-122 wires [18]. There is a strong correspondence between the different coloured regions (representing different orientations) in the inverse pole figures and the band contrast image.

The angles of GB misorientation are classified into three types: high-angle GBs with ≥ 10° misorientation with respect to adjacent grains are denoted by the red lines; low-angle GBs denoted by the green lines exhibit only 2–10° misorientation; and super low-angle GBs denoted by grey lines show very low (<2°) misorientation. A quantitative analysis reveals that around 66% of grains have misorientation angles below 15°. Furthermore, nearly half of the GBs possess misorientation angles less than the critical angle θ_c (9°) in iron-based superconductors. Since these GBs (misorientation

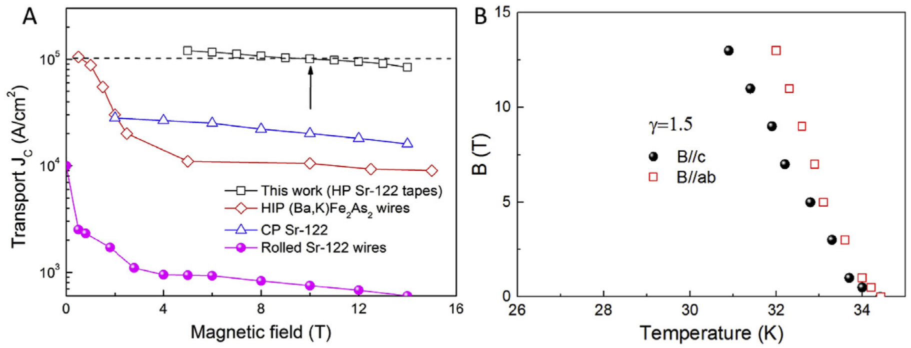

Fig. 1. (a) Comparative study of transport J_c as a function of magnetic field at 4.2 K for tapes or wires fabricated by different methods [17–19]. The black arrow points at the criterion for practical application. (b) Temperature dependence of the upper critical field B_{c2} for both $B//ab$ and $B//c$.