

Upper Critical Fields and Critical Current Densities of Fe-based Superconductors as Compared to Those of Other Technical Superconductors

I.Pallecchi^{1,*}, M.Tropeano², G.Lamura¹, M.Pani³, M.Palombo^{2,3,4},
A.Palenzona³, M.Putti^{1,4}

¹ CNR-SPIN, Corso Perrone 24, 16152 Genova, Italy

² Columbus Superconductors S.p.A, Via delle Terre Rosse 30, 16133 Genova, Italy

³ Dipartimento di Chimica e Chimica Industriale, Università di Genova, Via Dodecaneso 31, 16146 Genova, Italy

⁴ Dipartimento di Fisica, Università di Genova, Via Dodecaneso 33, 16146 Genova, Italy

* E-mail: ilaria.pallecchi@spin.cnr.it

Abstract - Three years since the discovery by the Hosono's group of Fe-based superconductors, an enormous number of compounds, belonging to several different families have been discovered and fundamental properties have been deeply investigated in order to clarify the interplay between magnetisms and superconductivity in these compounds. Indeed, the actual potential of these compounds for practical applications remains still unclear.

Fe-based superconductors are midway between high temperature superconductors (HTSC) and MgB₂. In Fe-based superconductors the critical current is rather independent of the field, similarly to HTSCs, as a consequence of the exceptionally high upper critical field and strong pinning associated with nm-scale local modulations of the order parameter. They exhibit low anisotropy of the critical current with respect to the crystalline directions, as in the case of MgB₂, which allows current flow along the c-axis. However, Fe-based superconductor polycrystalline materials currently available still exhibit electromagnetic granularity, like the HTSCs, which suppresses superconducting current flow over long length. Whether the nature of such granularity is extrinsic, as due to spurious phases or cracks between grains or intrinsic, as related to misalignment of adjacent grains, is under debate. These aspects will be review in the light of the recent literature.

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I. INTRODUCTION

The modern age of superconductivity started in 1986 with the discovery by Bednorz and Müller of high-temperature superconductors (HTSC), where an exotic coupling mechanism yields a critical temperature, T_c , above liquid nitrogen temperature (77 K). Till now many of the promises of these materials have not been kept: in fact, on the one hand we are still far from a full understanding of the coupling mechanisms and, on the other hand, due to the structural complexity, anisotropy, and bad metallicity of these compounds, only a few niche applications have been realized.

The new century has started with the discovery of MgB₂¹. In this case it was soon understood that conventional electron-phonon coupling in a complex multiband framework determines superconductivity at $T_c=40$ K. This simple, metallic, cheap, light compound has just proved to be suitable for cryogen-free applications.

Finally, in February 2008, the discovery of a Fe-based superconductor (FeSC), LaFeAs(O_{1-x}F_x) with T_c of 26 K, was announced by Hosono *et al.*². This event prompted an army of physicists and

chemists to discover other different families (the most studied are called “1111” for REFeAsO, “122” for AFe₂As₂, “111” for AFeAs, “11” for Fe(Te,Se)) with critical temperatures, in the optimally doped compounds, ranging from 55 K³ to 19 K and incredibly high upper critical fields. Two years after Hosono’s discovery two crucial questions arise: the first is, will FeSC be instrumental in deciphering the 22-year-old mystery behind high-T_c superconductivity? The second is, will FeSCs be simpler and more suitable materials for applications than the HTSCs? In this paper we will try to address the latter issue, reviewing the most recent results on the critical current density of single crystals, films, polycrystals wires and tapes of FeSCs, also in comparison with data on high-T_c cuprates and technical superconductors. At the present stage, no conclusive answer can be given yet on the actual application potential of these compounds and further studies are clearly needed, on all FeSC families.

II. BASIC PROPERTIES OF Fe-BASED SUPERCONDUCTORS

FeSCs share several characteristics with HTSCs, such as the layered structure, the coexistence of different orderings, the occurrence of superconductivity upon doping, the small coherence length, and non-conventional pairing. Some of these aspects have shown to be unsuitable for practical application. On the other hand FeSCs exhibit several advantages with respect to HTSCs; namely, they are metallic in the parent compounds, the anisotropy is generally smaller and not strongly dependent on the level of doping, the supposed order parameter symmetry seems to be different, and in principle not so detrimental to current transmission across grain boundaries (GBs), impurities do not significantly affect T_c.

FeSCs share characteristics also with the more conventional MgB₂. The most remarkable one is the multiband nature that has offered unprecedented tools in MgB₂ to tune and improve the superconducting properties and needs to be carefully investigated also in FeSC.

Moreover, FeSCs appear to be extremely versatile in terms of chemical composition, as they belong to a comprehensive class of materials, where many chemical substitutions are possible and their layered structure allows designing new FeSC with composite structures or even artificial multilayers. This versatility could enable the superconducting properties to be tailored for commercial technologies.

We will now review the most relevant properties for applications.

III. UPPER CRITICAL FIELDS

Since the earliest stages of the research on FeSCs, it has been apparent that they are characterized by very high **upper critical field** values (H_{c2}), which is one of the main requirements for having a good in-field behaviour of the critical current density. The first magnetotransport measurements in high fields have been carried out on the 1111 phase^{4,5,6}. The shape of the 1111 resistive transition is significantly broadened by the magnetic field^{5,4}, but to a smaller extent than in the high-T_c cuprates. The extracted upper critical field curves exhibit a distinctive upward curvature, reminiscent of the MgB₂ behaviour⁷, which is a signature of the multiband character. H_{c2} values of up to 60 T and H_{c2}-slopes close to T_c between -10 T/K and -15 T/K have been measured^{5,8}. The H_{c2} anisotropy is between 5 and 9 close to T_c^{5,8} and its slight temperature dependence is again indicative of multiband behaviour. On the other hand, as a consequence of the broadening of the transition, the irreversibility field H_{irr} (defined as the field where the resistivity becomes zero) is much smaller than H_{c2} (defined at the onset of the transition). This feature, which is an obvious drawback for applications, is very common in HTSC cuprates and strongly related to their anisotropic nature, the nearly 2D-character of these compounds and a signature of the weakness of flux pinning and/or the significance of thermal fluctuations.

A strikingly different H_{c2} behaviour has been observed in the 122 phase. The in-field 122 resistive transition exhibits no broadening⁹, much like the behaviour of low- T_c superconductors¹⁰, where the effect of the magnetic field is an almost rigid shift of the transition to lower temperatures. Consequently, the H_{irr} curve closely follows the H_{c2} curve. The H_{c2} anisotropy is 1.5-2 close to T_c and rapidly approaches unity with decreasing temperature^{11,12}. This behaviour of the anisotropy is a consequence of approaching the paramagnetic limit, responsible for the downward curvature of H_{c2} for H parallel to the Fe planes. On the whole, the H_{c2} values of this family are smaller than those of the 1111 family, with H_{c2} slopes close to T_c around -5 T/K^{9,11,12}.

The magnetoresistance behaviour of the 11 family is midway between the fan-shaped one of the 1111 phase and the rigidly shifted one of the 122 phase^{8,13}. The H_{c2} slopes close to T_c are the largest among FeSCs, ranging from -10 T/K to -30 T/K^{14,15,13,8}. Very recently, huge values of H_{c2} slopes as large as -500 T/K have been measured in thin films¹⁶. Such values indicate an almost complete suppression of the orbital pair-breaking, possibly as a consequence of the inhomogeneous Fulde-Ferrel-Larkin-Ovchinnikov (FFLO) state which forms below 5-6 K by applied strain. This finding is of crucial importance in driving future experimental investigations on H_{c2} , as it may suggest that the H_{c2} enhancement can be better tuned by doping and strain than by the conventional disorder tuning. The H_{c2} anisotropy in the 11 family quickly decreases to unity with decreasing temperature and even becomes smaller than unity at the lowest temperatures¹⁵ as a consequence of approaching the paramagnetic limit, which bends the H_{c2} curves to downward curvatures for both field directions.

In Figure 1, typical H_{c2} curves of the 1111, 122 and 11 families for field parallel and perpendicular to Fe planes are shown.

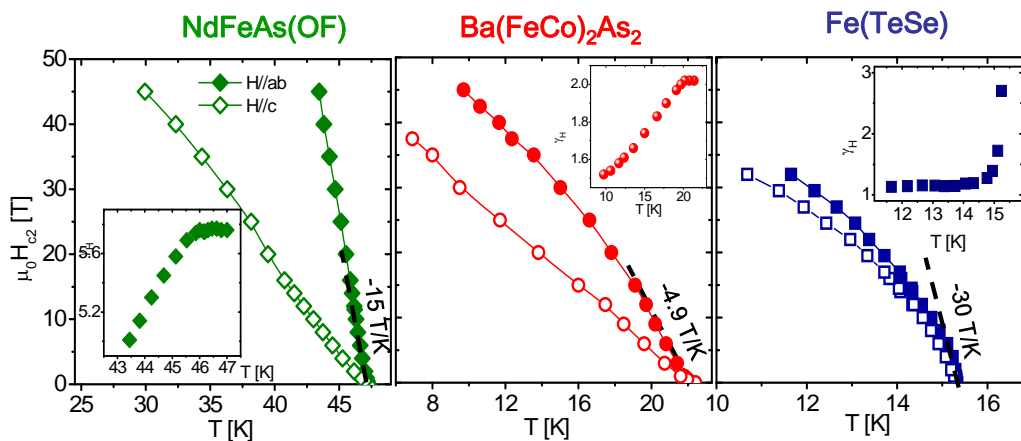


Fig. 1. Temperature dependences of $H_{c2||ab}$ (full symbols) and $H_{c2\perp ab}$ (open symbols) for NdFeAs(OF) (left panel), Ba(FeCo)₂As₂ (middle panel) and Fe(SeTe) (right panel), representative of the 1111, 122 and 11 families. In the inset, the H_{c2} anisotropy versus temperature curves are plotted. All the data are taken from ref. 8).

Table I: Typical values of superconducting state parameters of the FeSC families⁸⁻¹⁷, YBCO^{18,19,20,21} and MgB₂^{22,23}.

	1111	122	11	YBCO	MgB ₂
T_c [K]	55	38	16	93	39
$\mu_0 H_{c2}(T=0)$ [T]	>50	60	55	>50	30
γ_H	5	2	2-3	4-14	3-5
ξ_{ab} [nm]	2.5	3	1.5	2.2	10
ξ_c [nm]	0.6	1.5	0.6	0.4	2

λ_{ab} [nm]	200	200	500	120	50
Ginzburg number G_i	$4 \cdot 10^{-4}$	$1.5 \cdot 10^{-5}$	$1 \cdot 10^{-3}$	$5 \cdot 10^{-4}$	10^{-5}

As can be seen in Table I, FeSCs show the largest H_{c2} slopes close to T_c in comparison with all other HTSC. This is a point of strength of FeSCs making them suitable for high field applications.

On the whole, the **anisotropy** of FeSCs is smaller than that of the $YBa_2Cu_3O_{7-\delta}$ (YBCO) family, where values from 4 to 14 have been reported¹⁸ and much smaller than that of the $Bi_2Sr_2CaCu_2O_x$ (BSCCO) family, where it is typically in the range 50-60²⁴. This is a very important point because the anisotropic, nearly two-dimensional nature of high- T_c cuprates is the main reason for the weakness of the pinning and the significance of thermal fluctuations, which cause the broadening of the transition.

The different behaviour of the broadening of the transitions in these superconductors is understood if the **coherence lengths** ξ are considered. Rough estimates of ξ obtained from the H_{c2} slopes at T_c are reported in Table I (2.1, 2.9 and 1.5 nm along the Fe planes, for the 1111, 122 and 11 families, respectively)⁸. These values must be compared with the distance d between the Fe-As planes, in order to establish the two-dimensional or three-dimensional character of superconductivity and consequently the degree of dissipation in a magnetic field. The values of d are around 0.86, 0.65 and 0.6 nm in the 1111, 122 and 11 families, respectively. Therefore, the 122 family appears to be the most promising for applications in this respect, with the largest ξ/d ratio among the FeSCs.

Broadening of the transition in zero field is caused by **thermal fluctuations**, which can be parameterized by the Ginzburg number $G_i = (\pi \lambda_0^2 k_B T_c \mu_0 / 2 \xi_c \Phi_0^2)^2$, where λ_0 is the London penetration depth at zero temperature, k_B the Boltzmann constant, Φ_0 the magnetic flux quantum. Typical values of $G_i \approx 4 \cdot 10^{-4}$, $1.5 \cdot 10^{-5}$, $1 \cdot 10^{-3}$ for the 1111, 122 and 11 families⁸ should be compared to $G_i \approx 5 \cdot 10^{-4}$ for $YBa_2Cu_3O_{7-\delta}$ and 10^{-5} for MgB_2 . Again the 122 family seems to be the most promising for applications, as its slightly lower T_c and lower anisotropy yield reduced thermal fluctuations.

IV. J_c IN SINGLE CRYSTALS AND THIN FILMS

Measurements of the critical current density J_c in **single crystal samples** of FeSCs have revealed a promising combination of high and nearly isotropic critical current densities along all crystal directions. Moreover, J_c is rather independent of the field at low temperatures, similarly to observations in $YBa_2Cu_3O_{7-\delta}$ ²⁵. Such a behaviour is consistent with the nm-scale coherence lengths, the exceptionally high H_{c2} values and the pinning associated with atomic-scale defects, resulting from chemical doping or nm-scale local modulations of the order parameter²⁶. For the 1111 family, a high in-plane J_c of $2 \cdot 10^6$ A cm⁻² at 5 K in a $SmFeAsO_{1-x}F_x$ crystal, almost field-independent up to 7 T at 5 K, has been reported^{27,28}. Many single crystal results have been reported for the 122 system, since larger crystals can easily be grown. Significant fishtail peak effects and large current carrying capability up to $5 \cdot 10^6$ A cm⁻² at 4.2 K have been found in a K-doped $Ba_{0.6}K_{0.4}Fe_2As_2$ single crystal²⁹. Fishtail effect and currents in the range 10^5 A cm⁻² at low temperature have been also reported for the 122 family in ref.^{30,9}. As for the 11 system, J_c of $FeTe_{0.61}Se_{0.39}$ crystals with $T_c \approx 14$ K exceeding 10^5 A cm⁻² at low temperatures has been reported³¹. Irradiation with Au ions³² and neutrons³³ have emphasized that pinning can be further increased by introducing defects without affecting T_c . Typical $J_c(H)$ curves measured on single crystals of the 1111, 122 and 11 families are shown in Figure 2.

However, in general, the largest critical current values are measured in **thin films**, which on one hand are free from grain boundaries and on the other hand may have strong pinning centers such as surface roughness, inclusions, defects related to the growth mode. In superconductors such as MgB_2

³⁴ and YBCO ³⁵, the critical current measured in thin films is larger than that measured in polycrystals by orders of magnitude and it even achieves 20% of the ultimate intrinsic depairing limit. In iron based superconductors as well the critical current measured in thin films is promising and its investigation may yield important clues to enhance the performances of bulk samples, tapes and wires. Noteworthy results have been obtained in films of the 122 phase with Ba(Co,Fe)₂As₂ thin films, where columnar defects grown from the template act as effective pinning centers for H parallel to the c axis ^{41,36}. Critical current densities up to 4.5 MA/cm² at 4 K have been measured ³⁶. The resulting pinning force turns out to be better than in optimized Nb₃Sn at 4.2 K ⁴¹. High critical current densities of 4 MA/cm² at 4 K have been also obtained in Co-doped BaFe₂As₂ epitaxial films grown directly on (La,Sr)(Al,Ta)O₃ substrates by pulsed laser deposition ³⁷. Epitaxial films of the 1111 phase (LaFeAs(O,F)) have been deposited by pulsed laser ablation and their critical current density has been shown to obey the anisotropic Ginzburg-Landau angular dependence, with a maximum J_c for H parallel to the film around to 10⁵ A/cm² ³⁸. As for films of the 11 phase, the J_c's of FeTe_{0.5}Se_{0.5} films deposited on LaAlO₃ substrates at T ≤ 4 K is around 5·10⁵ A/cm² in self-field and it remains above 10⁴ A/cm² up to 35 T ³⁹. Hence, in general, it can be said that, apart from the 1111 family whose complex chemical composition makes the epitaxial film growth more difficult, the J_c results on thin films are larger than those on single crystals, even if still two orders of magnitude smaller than the respective depairing limits.

J_c values measured on FeSC single crystals and thin films are summarized in Table II, in comparison with other technical superconductors. It can be concluded that the single crystal properties are excellent, indeed J_c is high and rather **field independent** as in the high-T_c cuprates, whereas the **J_c anisotropy**, defined as the ratio between the current flowing in plane and out of plane J_c^{(ab)l}/J_c^(c), is around 2 in all FeSC families ^{8,40,41}, which is much alike the anisotropy in MgB₂ and much lower than the values of up to 30-50 found in the cuprates ⁴². This is a very significant result, because it could imply that, contrary to the high-T_c cuprates, the FeSCs do not require complex texturing processes for the fabrication of wires or tapes. Actually, the low J_c anisotropy is only a prerequisite for the current transport in polycrystalline materials. Indeed, the second requirement is the transparency of the GBs to current flow. This issue is considered in the next section.

Table II. Indicative values of J_c for various superconductors.

		1111	122	11	YBCO	MgB ₂
Single crystals	J _c (0) at 5K (A/cm ²)	2·10 ⁶ _{43,28}	3·10 ⁵ _{30,9}	10 ⁵ ₃₁	3·10 ⁶ ₄₄	10 ⁵ ₄₅
	J _c ^{(ab)l} /J _c ^(c)	2.5 ²⁸	2 ⁴⁰		10-50 ^{46,42}	1- 2
	J _c (5T)/J _c (0)	0.8 ²⁸	0.5 ⁹	0.3 ³¹	0.5 ⁴⁴	<0.01
Thin films	J _c (0) at 5K (A/cm ²)	10 ⁴ ₃₈	4·10 ⁶ _{36,37,41}	7·10 ⁵ ₃₉	10 ⁷ -10 ⁸ ₃₅	10 ⁷ ₃₄
	J _c (5T)/J _c (0)	0.2	0.5	0.5	0.2-0.5	0.003-0.1

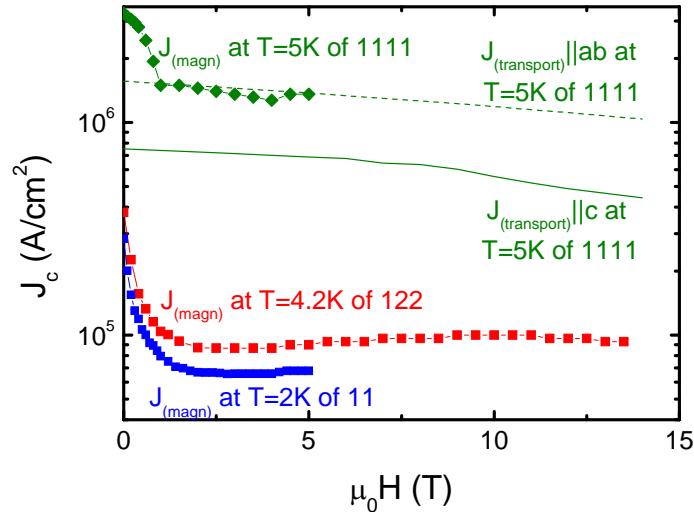


Fig. 2. Critical current density curves extracted from magnetization and transport measurements on single crystals of the 1111²⁸, 122⁹ and 11³¹ families. In all cases H is parallel to the c axis.

V. THE ISSUE OF GRAIN BOUNDARIES

One of the essential requirements for large scale application of superconductivity is the capability to carry currents over long lengths, i.e. the superconducting currents need to flow in polycrystalline materials across many grain boundaries (GB) regions. The “magical” ability of Cooper pairs to cross non-superconducting regions or areas with depressed order parameter is due to their long coherence length, of the order of several ten nm in conventional superconductors. The discovery of the cuprates with high T_c , high H_{c2} , enhanced anisotropy and, consequently, low coherence lengths (1-2 nm in the ab-plane and much lower along the c-axis) has shown the limits of this capability and the “issue of grain boundaries” has clearly emerged for the first time. In the past twenty-four years enormous efforts have been made to grow textured superconducting tapes with small-angle GBs, but this approach is not yet commercially viable for most applications. Very recently a theoretical understanding of why the currents in the HTSC cuprates are so sensitive to the GB mismatch has been proposed⁴⁷ that will hopefully suggest how to make technologically viable wires.

A very different story is that of MgB_2 , discovered less than ten years ago, with not evident GB problem⁴⁸. Indeed, MgB_2 has already been successfully used for fabricating magnets for commercial MRI systems.

These premises highlight the importance of exploring the nature of GBs in novel superconducting materials.

Regarding FeSCs, as shown in Table I, the large H_{c2} values imply coherence lengths varying between 1 and 3 nm among the families. These values are reminiscent of those in the cuprates, but the reduced J_c anisotropy of FeSCs compared to the cuprates (see Table II), allows current flow along the c-axis, thus making the requirement of texturing less stringent.

Early studies of the critical current density of 1111 polycrystals have emphasized the **strong granularity** of these compounds, which limits the global J_c flow across all the sample to very low values (see ref. 8 for a review). Two distinct scales of current flow have been found in polycrystalline Sm- and Nd-iron-oxypnictides using magneto-optical imaging and studying the field dependence of the remnant magnetization⁴⁹. Low temperature laser scanning microscopy (LTLSM) and scanning electron microscopy (SEM) observations on polycrystalline Sm-1111 samples⁵⁰ emphasized cracks and wetting Fe-As phase at the GBs as the main mechanisms of current blocking in polycrystalline materials.

Up to date the difficulty to obtain fully dense single-phase polycrystalline materials seems to be one of the main reasons for the granular behaviour of bulk materials. However, considering the strong

similarities between HTSC and FeSCs, the existence of intrinsic mechanisms limiting the transmission of current in misaligned GB needs to be carefully considered.⁵¹

A pioneering experiment has investigated the nature of GB weak links in $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ **films grown on bicrystals** with different misorientation angles θ . It has been shown there that the critical current is severely depressed for θ larger than 3° , whereas at higher fields the GBs are lesser obstacles to J_c ⁵². This is reminiscent of the behaviour of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ⁵³, even if a close comparison suggests that in FeSCs the suppression is less severe than in the HTSC: as shown in Figure 3 for θ from 0° to 24° J_c decreases by one order of magnitude, while in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ by two orders of magnitude. Another similar experiment on $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ films grown on bicrystals, whose results are also reported in Figure 3, has been carried out by Katase⁵⁴. They have found a slightly larger critical angle $\theta \approx 9^\circ$ and they have concluded that Josephson-coupled grain boundaries show SNS rather than SIS behaviour, a result which points to higher angle GBs being metallic rather than insulating, as in the cuprates. On the whole, the studies by⁵² and⁵⁴ are pretty consistent, both showing a similar exponential fall off of J_c with increasing misorientation angle above a certain critical value. However, these preliminary results needs to be confirmed to rule out possible extrinsic effects, such as diffusion of oxygen from the substrate. This requires the fabrication of ideal GBs, which in turn needs wide-ranging investigations, including different kinds of compounds, the use of buffer layers, and improvements of the deposition processes.

Very recent results⁵⁵ indicate that the misorientation angle is not the only parameter that determines whether or not GBs are transparent to the supercurrent, but also that the orientation of the field with respect to the GB has also to be taken into account. Indeed, it has been found that the inter-grain J_c degrades only when a significant portion of the vortex lies in the GB, while when the vortex lies obliquely across the GB there is little or no suppression of the inter-grain J_c .

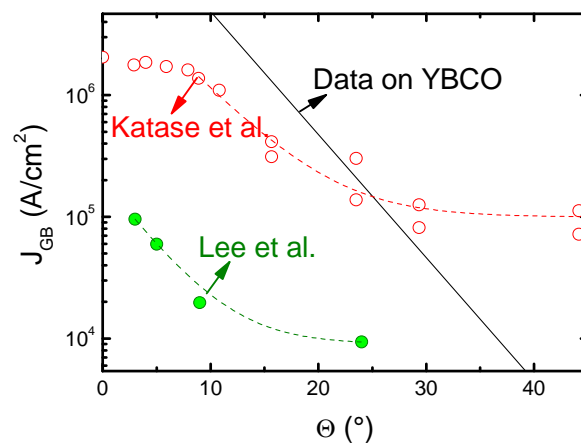


Fig. 3. Dependence of the critical current density across GBs, J_{gb} at 12 K and 0.5 T as a function of the misorientation angle θ measured on $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ films grown on bicrystals with different misorientation angles⁵². The dotted lines are exponential fits. The continuous line shows the behaviour of YBCO GBs according to ref.⁵³.

VI. TRANSPORT J_c IN WIRES AND POLYCRYSTALS

Even with substantial blocking by such GB phases, the intergranular current densities in FeSCs appear to be more than one order of magnitude larger at 4 K than in early results on randomly oriented polycrystalline cuprates⁵⁶: global currents flowing in the whole sample of the order of 4000 A cm^{-2} have been evaluated at 4 K in self-field in 1111 samples⁴⁹. Also polycrystals of the 11 family synthesized at low temperature have shown global current patterns in magneto-optical images and values of transport currents up to 700 A/cm^2 have been measured⁵⁷.

Granularity has so far limited the properties of pnictide wires. Yet, very recently, wires of the 11 family have been fabricated by powder-in-tube (PIT) method, exhibiting J_c values from 200 A/cm^2 ⁵⁸ to 600 A/cm^2 ^{59,60} and, very recently, even 1000 A/cm^2 ⁶¹. Scanning electron microscope (SEM) images of mono- and multi-filamentary Fe(Se,Te) wires prepared by *ex situ* PIT method are displayed in Figure 4, where also the resistive transition of a filament is shown. As for wires of the 1111 family, values up to $4 \cdot 10^3 \text{ A/cm}^2$ ^{62,63} have been obtained. Finally, regarding the 122 family, the highest J_c values in wires fabricated by PIT are slightly above 10^4 A/cm^2 ^{64,65}. The time evolution of the maximum transport critical current density measured in FeSc wires is displayed in Figure 5. It can be seen that the 122 family appears to be the most promising, but also the other families seem to be catching up, particularly the 11 family. Most importantly, for all the three families the maximum obtained J_c has been steadily increasing, suggesting that there is likely still a large edge of improvement.

Finally it is worth mentioning that 122 and 11 films have been deposited on buffered metal-tape flexible substrates with outstanding results, namely J_c values 10^6 A/cm^2 and 10^4 A/cm^2 for the 122⁶⁶ and 11⁶⁷ families, respectively. These encouraging results are likely due to the reduced J_c anisotropy of FeSCs, which allows current flow along the c-axis and makes the requirements on texturing less severe.

In conclusion, as also pointed out in the review⁵⁵, the inter-granular J_c of randomly oriented FeSC polycrystals is significantly higher than in cuprates. Indeed, transport J_c values of 10^3 – 10^4 A/cm^2 have been reported in randomly oriented FeSCs bulks and wires, as compared to the typical $\sim 100 \text{ A/cm}^2$ of random cuprate bulks. This datum, joined with the low anisotropy of J_c and its in-field behaviour, makes FeSC potential still worth of investigation and serious consideration for future technology.

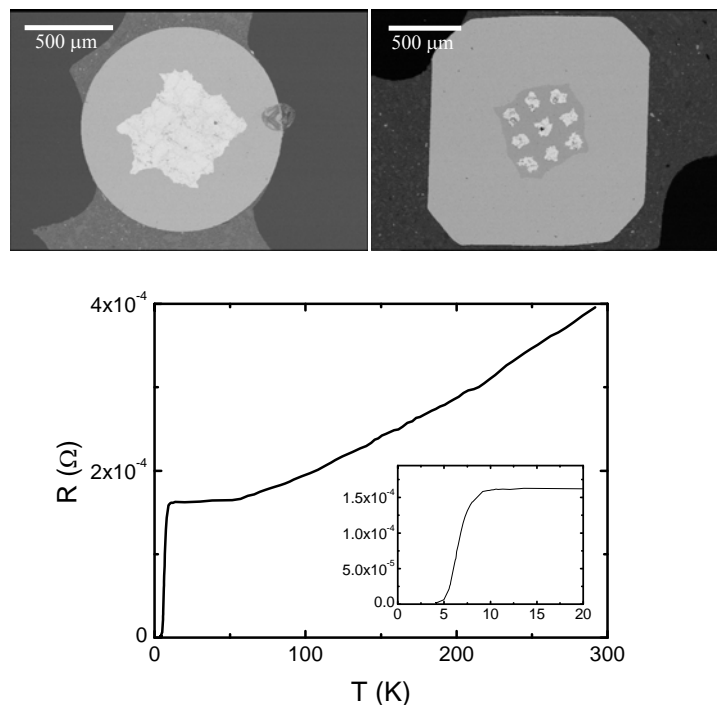


Fig. 4. In the two upper panels, SEM images in back scattered mode of mono- and multi-filamentary $\text{FeTe}_{0.5}\text{Se}_{0.5}$ wires are shown. The mono-filamentary wire has a Fe sheath, whereas the multi-filamentary one is composed by an external Ni sheath and nine Fe sheathed filaments. A resistive transition of a filament is shown in the lower panel, exhibiting a T_c around 8K.

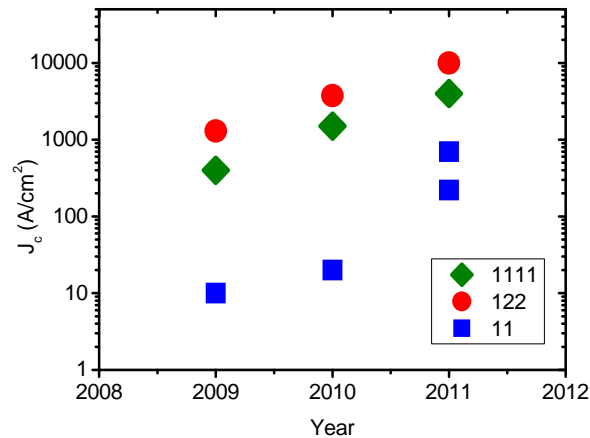


Fig. 5. Transport J_c values measured in FeSC wires and polycrystals since their discovery, plotted versus the year of publication. The data are taken for the 11 family from ⁶⁸ (2009), our own datum obtained in polycrystals (2010) and ^{61,57} (2011); for the 1111 family from ⁶⁹ (2009), ⁶³ (2010) and ⁶² (2011); for the 122 family from ⁷⁰ (2009), ⁷¹ (2010) and ^{65,64} (2011).

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- E-mail addresses of co-authors: tropeano@fisica.unige.it; gianrico.lamura@spin.cnr.it; marcella@chimica.unige.it; palombomarco@gmail.com; metalli@chimica.unige.it; putti@fisica.unige.it.
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