

PREFACE

Preface to the special issue 'Focus on 10 Years of Iron-Based Superconductors'

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Preface

Preface to the special issue ‘Focus on 10 Years of Iron-Based Superconductors’

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In historical or archaeological terms, the iron age began around the 12th century BC and lasted for over a millennium, up to the onset of historiographical records. The key role of iron in improving the life of human beings was established with the production of tools by ferrous metallurgy. According to Greek mythology, among the five Ages of Man representing the stages of human existence on Earth, the last one is the mythological iron age, where moral values and well-being eventually decline.

Moving away from such awesome themes and blurred chronology, we instead focus on the well-defined era of iron superconductivity: it started in early 2008, with the discovery of a superconducting transition at 26 K in the $\text{LaFeAsO}_{1-x}\text{F}_x$ compound by the Hideo Hosono group [1] and has now passed its tenth anniversary, with no sign of fading vitality. Also in this context, it was clear since the very beginning that iron was bound to play a primary role: On one hand it defied the shared belief about the antagonistic relationship between magnetism and superconductivity, and on the other hand it reignited new excitement about the mechanisms and perspectives of unconventional superconductivity, over 30 years after the discovery of high- T_c in copper oxides [2].

Such a ten-year period may represent infancy from a commercial and technological application perspective, though early maturity from the scientific research point of view. Certainly, it is a milestone, which first of all deserves celebration, secondly calls for an assessment of the worldwide status of research on this topic and finally allows a realistic, yet still tentative evaluation of the prospective potential in specific applications. This multifold aim is addressed by this focus issue, whose scope is to collect contributions from acknowledged researchers in the scientific community about the most relevant topics related to iron-based superconductors, including state-of-the-art results and reviews covering fundamental issues, applications, physical mechanisms, properties, and compounds.

1. Fe-based superconductors: a 10-year story

After an early report on superconductivity in Fe-based LaFePO and LaFeP(O,F) at low temperature $T_c \approx 5$ K [3], high-temperature superconductivity was discovered in the so called 1111 [1], 122 [4] and 11 [5, 6] main Fe-based families.

The most accredited scenario for pairing effects and wave symmetry are those related to antiferromagnetic spin fluctuation and $s \pm$ symmetry, with a sign change in the phase of the order parameter in different sheets of the Fermi surface, yet the debate on this is still open [7–10].

Pretty soon it was clear that these compounds exhibited interesting properties in view of potential applications, namely high T_c 's up to 58 K in 1111 [11] and up to 38 K in 122 [12] groups, large upper critical fields H_{c2} [13, 14], moderate-to-low H_{c2} and J_c anisotropies [15–19], especially low at low temperatures and in the 122 and 11 families [16, 17]. Their small coherence lengths in the nm scale and related weak link behavior of the critical current at grain boundaries made them

similar to high- T_c cuprates, however their critical intergrain misalignment angle was found to be larger than that of cuprates [20–23].

These findings triggered large scale application-oriented research, whose progress has evolved to demonstrate remarkable technological achievements and shows no sign of slowing down. Reports have appeared about the effectiveness of introducing pinning centers [24, 25], fabrication of wires and tapes [26] with J_c exceeding the application threshold of 10^5 A cm⁻² [27, 28], even at high fields [29, 30], fabrication of a 100 m long powder-in-tube (PIT) wire with J_c exceeding 10^4 A cm⁻² at 4.2 K and 10 T via a scalable rolling process [31], demonstration of bulk compact magnet trapping over 1 T [32], fabrication of 122 and 11 coated conductors, with $J_c \sim 10^6$ A cm⁻² at 4.2 K and 9 T [33–35], as well as proof-of-principle experiments regarding coated conductor architectures [36]. Also, potential electronic applications have been addressed, with deposition of films [37] and fabrication of electronic devices, such as functional multilayers [37], Josephson junctions [38, 39] and quantum interference devices [40].

In parallel, fundamental research has proceeded, investigating topics and mechanisms in these compounds, such as phase diagrams [41], quantum criticality [42], Lifshitz transitions, nesting, multiband character [43], pressure/strain effects [44, 45], and disorder effects [46]. Deep understanding of such topics could not only cast light on fundamental issues of superconductivity and condensed matter physics in general, but also provide useful hints to drive the application-oriented and technological research.

Ever new iron-based superconductor families have been discovered such as 111 [47], 32225 [48], 21311 [49], 22438 [50], 112 [51], 12442 [52] and 1144 [53]. High-temperature superconductivity at impressively enhanced temperatures has been discovered in single-layer or electric-field-applied FeSe films [54–57].

Research is continuing, more intensively than ever, stimulated by the potential large-scale applications at low-to-moderate temperatures (up to 20 K) and high-to-very-high fields (up to 30 T), where these compounds can be advantageous compared to cuprates thanks to their lower anisotropies and fabrication costs.

2. This focus issue

10 years after the discovery of superconducting properties in iron-based compounds, this special issue of *Superconductor Science and Technology* is focused on research development toward applications, with particular attention to the inter- and intra-granular critical current density [58–66] and the exploration of strategies to improve it [59, 60, 67, 68].

Soon after the discovery of unconventional high-temperature superconductivity in iron-based compounds, the richness of possibilities to synthesize such compounds became apparent. Different families of iron-based superconductors are represented in this focus issue, both the most commonly studied ones, such as 122 chalcogenides [69] and pnictides [59–62, 67], 1111 oxypnictides [66, 70] and 11 chalcogenides [64, 67, 71], as well as the less-studied 21311 pnictides [72].

A decade since the seminal work by the Hideo Hosono group, the technology is maturing in the fabrication of different kinds of samples, all of which are considered in this focus issue, namely thin films [58, 59, 66, 70, 71], coated conductors [64], single crystals [61, 67, 69], polycrystals [60, 72] and tapes [62].

Thin films are arguably a good platform for both fundamental and applied superconductivity research, as they offer the possibility of studying intrinsic anisotropic physical properties, just like single crystals, with the further benefit of macroscopic size and mechanical robustness. Indeed, high-quality epitaxial thin films of the main iron-based families are grown by pulsed laser ablation (PLD) and molecular beam epitaxy (MBE). The extensive studies carried out to

investigate the influence of substrates or buffered templates in determining the key superconducting properties (like critical temperature T_c , upper critical fields H_{c2} , and critical current density J_c) are reviewed in the opening paper of this focus issue [58], where the roles of misfit, thermal expansion, and chemical stability are discussed.

Thin-film technology also offers multifold tuning possibilities, such as building artificial heterostructures that combine multiple phases, stabilizing metastable phases, relying on the optimization of growth parameters and on epitaxial constraints, enhancing pinning properties by nanoparticle inclusion and growth defects, as well as studying the weak-link behavior as a function of the intergrain misorientation angle in films on bicrystal substrates. Regarding the possibility of fabricating artificial heterostructures, in the work by Haindl and coworkers [70], by simply varying the temperature and deposition time in $\text{SmFeAsO}_{1-x}\text{F}_x$ thin films grown by PLD, the fluorine diffusion process was controlled and a fluorine content gradient along the thickness was created. In such samples, T_c 's up to ~ 43 K and high upper critical fields with low anisotropy ($\gamma < 2.25$ at low temperature) were obtained. Regarding the use of non-equilibrium film growth techniques to stabilize metastable phases, tetragonal iron sulfide (FeS) films were deposited by Hanzawa and coworkers on different substrates and characterized in terms of structural and transport properties under high-density carrier doping by ionic liquid gating [71]. Regarding the strategies to improve pinning properties, Miura *et al* demonstrated further enhancement of J_c , decreased J_c anisotropy, and limited creep rates by incoherent BaZrO_3 nanoparticles with tunable density and size in $\text{BaFe}_2(\text{As}_{0.66}\text{P}_{0.33})_2$ films over a wide range of temperatures and magnetic field. They achieved a self-field $J_c \sim 7.2 \text{ MA cm}^{-2}$ at 5 K, which is a sizeable 15% of the depairing current, and $J_c \sim 2.1 \text{ MA cm}^{-2}$ at 5 K and 9 T ($\mu_0 H_{llc}$) [59]. Regarding the grain boundary angle θ_{GB} dependence of transport properties, Iida and coworkers [66] carried out a study on $\text{NdFeAs}(\text{O},\text{F})$ films on MgO bicrystals. By limiting the extrinsic effects related to damage by excess F-diffusion along the grain boundaries, they determined a critical angle of 8.5° , above which J_c starts to decrease exponentially for this 1111 compound, similar to the values of other iron-based superconductor families.

Thin film technology deploys its application potential in the fabrication of coated conductors. $\text{Fe}(\text{Se},\text{Te})$ deposited on a CeO_2 buffered rolling-assisted biaxially textured substrate (RABiTS) template by Sylva *et al* exhibited an almost isotropic J_c of $1.7 \times 10^5 \text{ A cm}^{-2}$, which is reduced by less than one order of magnitude in fields of 18 T [64]. Considering the moderate T_c of 16 K, the high upper critical fields, the relative ease of fabrication and the absence of the more toxic arsenic compared to selenite, this compound is particularly interesting, extending the application ranges of MgB_2 and Nb_3Sn at low-temperatures and high-to-very-high fields ($T < 30$ K and $\mu_0 H > 10$ T).

Extensive experimental studies have been carried out on samples of different form and composition in order to explore the effects of many factors on the critical current density J_c and to develop strategies to improve it. Effects of chemical doping [61, 67], irradiation [67], fabrication parameters [60, 62], defects [60], external pressure [61] and weak links at grain boundaries [63, 64] are featured in this focus issue.

Nanometric defects induced by fast neutron irradiation in $(\text{Ba},\text{K})\text{Fe}_2\text{As}_2$ single crystals drastically change the pinning landscape that dominates the flux pinning properties, enhancing J_c toward the depairing current density limit and modifying the doping dependence of J_c , as shown by Kagerbauer *et al* [67]. Critical currents and pinning mechanisms were studied on $\text{Ba}(\text{Fe}_{1-x}\text{Ni}_x)_2\text{As}_2$ single crystals as a function of doping x and applied pressure p by Bioletti and coworkers [61]. The richness of physical mechanisms in play is apparent in the non-monotonic dependence of J_c on pressure and in a possible role of the proximity to a quantum

critical point in the phase diagram. Uhrig and coworkers found that annealing of $\text{FeSe}_{1-x}\text{Te}_x$ single crystals in air was a very simple strategy to increase T_c from 7 to 14 K and the critical current density J_c by up to one order of magnitude at all the applied magnetic fields [67]. The optimized annealing conditions were thickness dependent, and the related changes were attributed to the control of the interstitial excess iron by annealing, as well as to the emergence of a surface barrier, related to structural changes and oxide formation at the sample surface. In the study by Shimada *et al* [60], the microstructure of $\text{Ba}(\text{Co,Fe})_2\text{As}$ polycrystals was controlled by the preparation parameters in terms of grain size and formation of defects, such as stacking faults, intra- and inter-granular cracks, and secondary phases at the grain boundaries, with a sizeable effect on the inter- and intra-granular current.

The role of weak links at the grain boundaries in quasi-two-dimensional (quasi-2D) superconductors with low coherence length was addressed by Talantsev and Crump [63]. They proposed a criterion to reveal the presence or absence of weak links based on T_c and self-field J_c and comparatively applied it to different families of iron pnictides and cuprates. With this criterion, a number of iron based compounds were identified as promising weak-link free superconductors for the fabrication of tapes with J_c values in the range $1\text{--}3 \text{ MA cm}^{-2}$, including $\text{BaFe}_2(\text{As}_{1.72}\text{P}_{0.28})_2$, $\text{Ba}(\text{Co,Fe})_2\text{As}_2$, $(\text{Ba,K})\text{Fe}_2\text{As}_2$, $(\text{Ba,Lu})\text{Fe}_2\text{As}_2$, and $\text{CaKFe}_4\text{As}_4$, as well as intercalated FeSe [64].

For samples in the form of tapes, the research target is not only enhancing the current carrying capability, but also the development of cost-effective fabrication recipes. $\text{Ba}_{0.6}\text{K}_{0.4}\text{Fe}_2\text{As}_2$ tapes fabricated via a hot isostatic pressing method by Liu *et al* [62] exhibited J_c 's up to $5.8 \times 10^4 \text{ A cm}^{-2}$ at 10 T and low temperature, thanks to their phase purity, homogeneous element distribution, oriented grains, and good grain connectivity, despite the sheath material being a Cu/Ag composite rather than the optimal but expensive Ag.

In this focus issue, further specific aspects are addressed. Dudin *et al* [69] present investigations of the local chemical, electronic, and magnetic structure of the co-existing superconducting and antiferromagnetic phases in $\text{Rb}_x\text{Fe}_{2-y}\text{Se}_2$ single crystals by scanning microscopy techniques. Wakimura and coworkers studied the effect of electron doping by Cr substitution in $\text{Sr}_2\text{VFeAsO}_3$ and observed a moderate suppression of T_c and an increase in the residual resistivity ratio (RRR) due to the introduction of disorder in the blocking layer [72].

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