A view from inside iron-based superconductors

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Abstract
Muon spin spectroscopy is one of the most powerful tools to investigate the microscopic properties of superconductors. In this paper, an overview on some of the main achievements obtained by this technique in iron-based superconductors (IBS) are presented. It is shown how the muons allow to probe the whole phase diagram of IBS, from the magnetic to the superconducting phase, along with their sensitivity to unravel the modifications of the magnetic and the superconducting order parameters, as the phase diagram is spanned either by charge doping, by an external pressure or by introducing magnetic and non-magnetic impurities. Moreover, it is highlighted that the muons are unique probes for the study of the nanoscopic coexistence between magnetism and superconductivity taking place at the crossover between the two ground-states.

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

1. Introduction

More than 20 years after the discovery of high-temperature superconductivity (HTSC) in the cuprates [1, 2], the observation of critical temperatures ($T_c$) approaching 55 K in iron-based pnictides [3, 4] has renewed the interest for superconductivity (SC) and for the fascinating phenomenology it gives rise to⁴. Remarkably, the iron-based superconductors (IBS hereafter) share many similarities with the cuprates [5–9]. First of all, both families of materials are characterized by layered structures, involving FePn layers (with Pn = As, P or Se) in the IBS [10] and CuO$_2$ planes in the cuprates [11] (see figures 1 and 2 for the structure of these two classes of materials, respectively). The layered structure gives rise to a sizeable anisotropy in the transport and magnetic properties, which is more marked in the latter compounds. The high anisotropy is known to cause an enhancement of the flux lines lattice (FLL) mobility and to yield detrimental dissipative phenomena [13, 14] which could hinder the technological applicability of these materials. Still, the IBS are characterized by an anisotropy which is not so pronounced and, accordingly, by a lower FLL mobility [15–19]. At the same time they show upper critical fields often exceeding 60 T [20, 21], making the IBS rather promising for applications. The high upper critical fields imply rather short coherence lengths, reaching a few nanometers along the FeAs planes, so that these materials are weakly sensitive to disorder and, in particular, to non-magnetic impurities. Another relevant similarity between the IBS and the cuprates is that SC is obtained by charge-doping a strongly correlated electron system with a magnetic ground-state in both families of compounds [2, 9, 22]. This is shown in the typical electronic phase diagrams reported in figures 3 and 4 for the IBS and for the cuprates, respectively.

⁴ Visit the site www.superconductivity.eu to have a nice overview on the fascinating world of superconductors.

The correlations are certainly more relevant in the cuprates where the significant Coulomb repulsion yields to an insulating ground-state, while in the IBS the magnetic precursors are metallic. In particular, a multi-band scenario applies to pnictides where the five bands involving all the Fe 3d orbitals contribute to the density of states at the Fermi level [23–25]. Notice that a metallic behavior with reconstruction of the Fermi surface (FS) is also observed in the cuprates after
a few per cent of charge doping, prior to the onset of the superconducting ground-state [26]. At the same time, in the pnictides the scenario of an antiferromagnetic (AFM) phase arising just from the FS nesting remains controversial, since non-negligible correlations, bringing those samples close to a Mott-like transition, should also be considered [27–31]. As a result, several authors have described the magnetism in pnictides in terms of effective Hamiltonians for localized magnetic moments in the presence of competing frustrating interactions [27, 28, 32–34].

The phase diagrams described above are obtained by doping the parent compound with charges (either holes or electrons). Several chemical substitutions have been investigated in order to achieve such a goal. However, as far as the substitution of the Fe ions with other transition metal elements is considered, the effectiveness of charge doping is still under strong debate and has been the subject of recent intense efforts both on the computational and on the experimental sides, particularly in the case of Co for Fe substitution. Contrasting results have been reported from density-functional theory (DFT) calculations [35, 36] as well as from measurements via x-rays absorption [37, 38] and photoemission [39] spectroscopies. At the same time, the role of isovalent substitutions such as $\text{Fe}_{1-x}\text{Ru}_x$ attracted great interest both in $\text{LnFeAsO}_{1-x}\text{F}_x$ (the so-called 1111 family of IBS, with Ln a lanthanide ion) and in $\text{AFe}_2\text{As}_2$ (the 122 IBS, with A an alkali ion) compounds. The $\text{Fe}_{1-x}\text{Ru}_x$ dilution does not induce a superconducting phase in $\text{LaFeAsO}$ [34], at variance with what is observed in $\text{BaFe}_2\text{As}_2$ [40, 41]. Still, it induces a re-entrant static magnetic phase nanoscopically coexisting with SC in the optimally Fluorine-doped 1111 IBS for different Ln ions [42, 43] (see figure 5).

It should be mentioned that, as an alternative to chemical doping of the parental compounds, the application of an external [44–47] or of a chemical [48] pressure has been reported to be a useful tool to span through the different electronic ground-states of these materials. However, the effects of chemical doping and pressures are not equivalent at all. In particular, the increase of pressure is known to also involve the modification of more subtle many-body effects like the Kondo screening in the presence of conduction carriers hybridized with unpaired f electrons [49]. It has been established theoretically that the Kondo effect and SC strongly
compete in 1111 pnictide compounds [50]. For instance, this is clearly the case for Ce-based 1111 compounds where SC is induced by charge doping [51–53] but bulk SC cannot be induced neither by external [54] nor by chemical [55] pressures, which are naively expected to enhance the Kondo coupling.

The similarities among the IBS and cuprates discussed above suggests that the understanding of the microscopic mechanisms at work in IBS would have eventually lead to the clarification of the long debated origin of HTSC. The interplay between magnetism and SC have lead to the idea that in both classes of materials a pairing involving the spin excitations could be present [7, 56]. However, in spite of the huge efforts both on the experimental and theoretical sides, it is currently not yet clear whether such a pairing mechanism is indeed driving HTSC. On the other hand, it is well established that for the hole-doped cuprates the symmetry of the order parameter is d-wave [57], while it is s-wave for the electron-doped cuprates [58]. In the IBS, the symmetry of the order parameter is presently the subject of an intense debate. While it was initially suggested that the FS nesting would favor a magnetic coupling between hole-like and electron-like bands, yielding to an $s_{\pm}$ symmetry of the order parameter [59, 60], more recent studies suggested that a conventional s-wave symmetry would be more likely, with a pairing mediated by orbital current fluctuations [61, 62]. Even if the nature of the pairing mechanism is still undisclosed, it seems to be well established that a conventional phonon-mediated SC is unlikely in IBS [63]. Overall, the IBS appear as rather complex systems, and more than 5 years after their discovery their understanding still demands significant experimental investigation and a suitable theoretical modeling.

Muon spin spectroscopy ($\mu^+\text{SR}$) has played a major role in the clarification of the electronic and magnetic properties of cuprate materials firstly [64, 65] and, more recently, of the IBS. The muons act as nanoscopic Hall sensors which allow for mapping of the local fields inside those materials and to track their time evolution. In fact, they allow the probing of the spin dynamics, the local field arising from the onset of a magnetic order or the field distribution generated by the flux lines in the mixed state [65–70]. The local nature of the technique is particularly useful to evidence the intrinsic microscopic electronic inhomogeneities leading to the nanoscopic coexistence of magnetic and superconducting domains and/or of charge poor and charge rich regions, which characterize both the IBS [42, 71] and the cuprates [80, 81]. In this respect, $\mu^+\text{SR}$ is a rather unique tool, complementary to non-local techniques such as neutron scattering or to other local techniques as NMR, which can suffer from RF penetration and/or excessive line broadening problems. In the following, we present some of the main achievements obtained by means of $\mu^+\text{SR}$ in the IBS. First, it is shown how the muons allow for investigation of the whole phase diagram of these materials, from the magnetic to the superconducting phase, as well as the nature of the phase transition between the superconducting and magnetic ground-states. It is then shown how the study of the transverse field (TF) relaxation in the superconducting state provides useful information on the evolution of the superconducting transition temperature with the concentration of Cooper pairs. We shall mostly concentrate on the static properties of IBS by referring to the 1111 family and on the effect of hetero-valent and isovalent chemical substitutions on its phase diagram and on the superconducting properties. The scenario observed for the 122 family of IBS will also be briefly recalled, while for the $\mu^+\text{SR}$ studies carried out in other families of IBS, as the 11 chalcogenides and the 111 the reader can refer to [72–79].

2. The zero field muon spin polarization in the magnetic phase

The full spin-polarization of the positive muon ($\mu^+$) beams produced at meson factories, such as ISIS (Rutherford–Appleton Laboratories, UK) and S$\mu$S (Paul Scherrer Institut, PSI, Switzerland), is the prerequisite for running $\mu^+\text{SR}$ experiments. The possibility to work with fully polarized beams has the great advantage, in comparison to other local-probe techniques such as NMR, that there is no need to perturb the system under investigation with an external polarizing magnetic field. Accordingly, local magnetism can be investigated even in conditions of zero-magnetic field (ZF).

In general, at a given temperature ($T$), the ZF $\mu^+$ depolarization as a function of the time ($t$) elapsed after the implantation into the sample is described by

$$\frac{A_{ZF}(t)}{A_0} = (1 - a_s) G_{KT}(t) + a_s G_{ZF}(t). \quad (1)$$
Here $A_0$ is an instrument-dependent constant corresponding to the condition of full spin-polarization extrapolated at $t \to 0$, $a_s$ represents the fraction of $\mu^+\mu^-\text{SR}$ implanted into the investigated sample while $(1-a_s)$ is the background fraction. This latter quantity includes, for example, the $\mu^+$ implanted into the sample holder or into the crystal walls and probing weakly magnetic regions whose main contribution comes from magnetism of nuclear origin. This typically results in a Gaussian Kubo–Toyabe depolarization function $G_{KT}^M(t)$, which at early times shows a Gaussian trend $G_{KT}^M(t) \sim e^{-\left(\frac{a_{s}}{T}\right)^2}$, governed by a weakly $T$-dependent factor $\sigma_N$. Both MUSR and EMU, at ISIS, and GPS and Dolly, at PSI, are designed as low-background spectrometers, namely allowing one to achieve the condition $a_s \approx 1$. However, the background term in equation (1) is of crucial importance while analyzing data from measurements under applied pressure performed, for instance, at the GPD facility of S$\mu$S [46, 82, 83]. Here, the thick walls of the pressure cell are able to stop a sizeable fraction of $\mu^+\mu^-\text{SR}$ leading typically to $a_s \approx (1-a_s) \approx 0.5$. Nevertheless, since all the measurements to be discussed subsequently have been performed in the low-background spectrometers, from now on it is assumed that $a_s = 1$, so that $A_{ZE}(t)/A_0 = G_{ZE}(t)$, with $G_{ZE}(t)$ the spin-depolarization function of the $\mu^+\mu^-\text{SR}$ implanted into the sample. As will be explained in the following, in the IBS the ZF depolarization can be described rather well by

$$A_{ZE}(t)/A_0 = G_{ZE}(t) = \left[1 - V_{m}(T)\right] G_{KT}^M(t)$$

$$+ \sum_{i=1}^{N} \zeta_i \left[a_{i}^{Tr}(T) F_i(t) D_{i}^{Tr}(t) + a_{i}^{L}(T) D_{i}^{L}(t)\right], \quad (2)$$

where $G_{KT}^M(t) \approx e^{-\left(\frac{a_{s}}{T}\right)^2}$ for $a_{s}^{-1} \ll 1$, is the Kubo–Toyabe function describing the relaxation arising from the dipolar coupling with the nuclei in the sample. Equation (2) typically holds in the case of materials undergoing a phase transition to a magnetically ordered state at a temperature $T_N$. Accordingly, a set of ZF-$\mu$SR measurements at different $T$ values allows one to access several microscopic quantities associated both with the ordered and with the paramagnetic phases. The parameter $V_{m}(T)$ represents the fraction of $\mu^+$ implanted into the sample and feeling a spontaneous static magnetic field, namely the sample magnetic volume fraction, from which one can estimate $T_N$. The ideal case of a step-like behavior of $V_{m}(T)$ at $T_N$ is modified in real systems where a spatial distribution of $T_N(r)$ can be present. The assumption of a Gaussian-like distribution of local transition temperatures generally leads to the following phenomenological expression for $V_{m}(T)$:

$$V_{m}(T) = \frac{1}{2} \text{erfc} \left[ \frac{T - T_N}{\sqrt{2}\Delta T}\right], \quad (3)$$

where erfc$(x)$ is the complementary error function. A fitting procedure to the experimental $V_{m}(T)$ data according to equation (3) allows a precise definition both of $T_N$ and of the relative width $\Delta T$ of the transition itself. In particular, the $T_N$ value extracted from equation (3) is an average value defined as the $T$ value corresponding to 50% of the magnetic volume fraction.

Let us now consider the behavior of the depolarization function at $T \ll T_N$. In general, one should account for the possibility of different phases in the sample volume, i.e. for a macroscopic or nanoscopic (see section 3) segregation of magnetic and paramagnetic phases leading to the condition $V_{m}(T) < 1$. Here, it is assumed $V_{m}(T) = 1$ in the simplified assumption of a homogeneous fully magnetic sample. Then the first term in equation (2) drops out, leading to

$$A_{ZE}(t)/A_0 = \sum_{i=1}^{N} \zeta_i \left[a_{i}^{Tr}(T) F_i(t) D_{i}^{Tr}(t) + a_{i}^{L}(T) D_{i}^{L}(t)\right]. \quad (4)$$

The presence of $N$ crystallographically-inequalit stopping sites for $\mu^+$ is accounted for by the sum over the index $i$. Each stopping site is characterized by its corresponding stopping probability $\zeta_i$, with $\sum_i \zeta_i = 1$. Accordingly, the quantities $a_{i}^{Tr}(T)$ and $a_{i}^{L}(T)$ represent the fractions of all the implanted $\mu^+$ at the site $i$ which feel a static field in transversal (Tr) and longitudinal (L) directions with respect to the initial $\mu^+$ polarization, respectively. Thus, the following relation holds under general assumptions:

$$\sum_{i=1}^{N} \zeta_i [a_{i}^{Tr}(T) + a_{i}^{L}(T)] = V_{m}(T). \quad (5)$$

For powder samples, in the ideal condition that long-range magnetic order develops throughout the whole sample volume, the relations

$$\sum_{i=1}^{N} \zeta_i a_{i}^{Tr}(T \ll T_N) = \frac{2}{3} \quad \text{and} \quad \sum_{i=1}^{N} \zeta_i a_{i}^{L}(T \ll T_N) = \frac{1}{3}$$

follow from straightforward geometrical arguments. This is commonly called the $2/3$-$1/3$ rule. The function $F_i(t)$ in equation (4) describes the coherent oscillations associated with the Larmor precession of the $T$ fraction of muon spins around the local magnetic field $B_z$, generated by the long-range magnetic order. The distribution in the magnitude of the local fields give rise to the corresponding damping functions $D_{i}^{Tr}(t)$. On the other hand, $\mu^+$ belonging to the L fraction do not precess and usually probe longitudinal $T_1$-like dynamical processes resulting in a relaxing tail described by $D_{i}^{L}(t)$. Due to the limited time window, to the finite number of counts and to the comparable values of the relaxation constants, it is often difficult to distinguish the longitudinal relaxations of different $\mu^+$ sites, which eventually merge in a common $D^L(t)$. Typically, $D^L(t)$ is an exponentially relaxing component characterized by weak relaxation rates, typically of the order of $0.1 \mu s^{-1}$.

In the opposite temperature limit ($T \gg T_N$), namely in the paramagnetic phase, no static fields of electronic origin alter the initial $\mu^+$ polarization. In this case, $V_{m}(T) = \sum_i \zeta_i a_{i}^{Tr} = \sum_i \zeta_i a_{i}^{L} = 0$ and the whole second term in equation (2) vanishes, leading to

$$A_{ZE}(t) \approx A_0 e^{-\left(\frac{a_{s}}{T}\right)^2}, \quad (7)$$

namely the decay is determined by the dipolar field distribution arising from the nuclear magnetic moments.
The \( \mu^+ \) implanted in both sites give rise to a characteristic beat in the \( \mu^+ \) depolarization function of the LaFeAsO parental magnetic compound at \( T \ll T_N \) \cite{85}. This is not the case in compounds based on magnetic rare earth (RE) ions where only one oscillating signal is detected from site ‘1’ (see figure 7). In fact, site ‘2’ being very close to the large RE magnetic moments, which generate a broad field distribution, shows an extremely fast signal decay \cite{51}. Both in the case of LaFeAsO and CeFeAsO, the function \( F_1(t) = \cos (\gamma t B_{\mu \nu} t) \) describes the data quite well \((\gamma = 2\pi \times 1.355 \times 10^{-2} \text{ MHz} \text{ G}^{-1} \text{ being the } \mu^+ \text{ gyromagnetic ratio}). As reported in figure 8, also in the case of the parent compounds of the 122 family (e.g. BaFe\(_2\)As\(_2\)) one clearly observes beats arising from the signals of two muon sites \cite{86}.

When considering slightly charge-doped magnetic compounds, long-range magnetism is still probed by \( \mu^+ \) even if some qualitative changes in the spin-depolarization functions should be considered. First of all, the oscillating cosine-like function considered above in the case of the parent compounds should be modified to \( F_1(t) = J_0 (\gamma_{\mu} B_{\mu \nu} t) \) \((J_0 \text{ standing for a zeroth-order-first-kind Bessel function}). This function better describes the field distribution and the gradual modification of the spin density wave (SDW) phase commensurability upon increasing the charge doping \cite{87, 88}. On the other hand, the increase in the degree of magnetic disorder yields to an enhancement of the transversal damping, with respect to what is observed in the parental compounds. This is evident both in CeFeAsO\(_{1-x}\)Fe\(_x")\ CeFe\(_{1-x}\)Co\(_x\)AsO (see figure 7) and in Ba(Fe\(_{1-x}\)Co\(_x\))\(_2\)As\(_2\) (see figure 8).

By further increasing \( x \), the disorder is so strong that the oscillations are completely overdamped, hence reflecting an extremely broad distribution of static magnetic fields at the \( \mu^+ \) site. Even in this highly disordered configuration, one can give an estimate of the typical internal field of the magnetic phase by referring to the width of the field distribution (i.e. the squared root of the second moment). It should be remarked that in the case of overdamped transversal oscillations one should carefully exclude dynamical effects on the muon depolarization function. In these cases, one useful way to distinguish among the two scenarios is to perform a longitudinal-field scan. Only in the case of a static distribution of local fields the application of a strong enough magnetic field allows one to quench the spin-depolarization and to estimate the width of the static distribution (see \cite{51} for example).

In general, the three main microscopic contributions to the local field at the muon site \( B_\mu \) come from the dipolar, the transferred hyperfine and the Lorentz terms (see \cite{83} and references therein)

\[
B_\mu = |B_{\mu \text{ dip}}(r_\mu) + B_\text{hf}(r_\mu) + B_\text{L}|. \tag{8}
\]

The commensurate SDW actually corresponds to an AFM configuration and \( B_\mu(T) \) is proportional to the sublattice magnetization \( M_{\text{sl}}(T) \) (see \cite{82}). Owing to the crystallographic symmetry of site ‘1’, for an AFM order the only relevant contribution is the dipolar term, since the hyperfine contribution is almost entirely averaged out and \( B_\text{L} \) can be neglected because of the vanishing macroscopic magnetization. Accordingly, if the crystallographic position of site ‘1’ is precisely known it is possible to estimate the
Fe magnetic moment from dipolar sums. From the internal field at the $\mu^+$ site measured in the case of $\text{IIII}$ materials with no charge doping, one derives $\mu_{\text{Fe}} \simeq 0.65\mu_B$ for the Fe magnetic moment [82]. Even in the case of a sizeable degree of magnetic disorder, the order parameter can be estimated from the width of the distribution of static local magnetic fields, as explained above.

3. Electronic phase diagram and the coexistence between magnetism and superconductivity

The main typical outputs of ZF-$\mu^*$SR experiments on magnetic materials are the order parameter and the sample volume fraction $V_{\text{m}}$, which can be evaluated from equation (2) as two independent fitting parameters. The great advantage

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Figure 7. Experimental spin-depolarization at low-$T$ in ZF conditions for $\text{CeFe}_{1-x}\text{Co}_x\text{AsO}$ in the low-$x$ region of the electronic phase diagram [53]. The strong increase of the transversal damping with increasing $x$ is the experimental trademark of the progressive disordering of the magnetic phase toward the limit of short-range magnetic order coexisting with SC.

Figure 8. Experimental spin-depolarization at low-$T$ in ZF conditions for $\text{Ba}(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2$ in the low-$x$ regime (figure reprinted with permission from Bernhard et al [86]. Copyright (2012) by the American Physical Society).
of $\mu$SR when compared, for example, to neutron scattering is that it is a local probe technique which can detect a static magnetic order even when the coherence length is reduced to a few lattice steps and the system is quite inhomogeneous. Thus, $\mu^+\text{SR}$ is perfectly suited to investigate the details of the transition between the magnetic and the superconducting phases and it has been widely employed in pnictide superconductors to study the phase diagram obtained upon tuning in a controlled way, some key-parameter such as the level of iso- and/or hetero-valent chemical substitutions, the structural parameters and the external pressure.

The crossover region between the magnetic and superconducting electronic ground-states is of crucial relevance for the understanding of the intrinsic microscopic properties of pnictide materials. According to some theoretical models [89, 90], the experimental finding of coexistence between magnetism and SC over a certain region of the phase diagram is an indication of an unconventional pairing among the supercarriers. Still, one should clarify which is the spatial level of coexistence or, in other terms, the degree of spatial intertwining of the two different order parameters. This degree can be quantified by a characteristic length scale $d$ describing the typical distance among magnetic or, alternatively, superconducting domains. The strength of the claim of 'coexistence between magnetism and SC' is crucially dependent on the order of magnitude of $d$. In the case $d \sim 100 \text{nm} \sim 1 \mu\text{m}$, one typically refers to the so-called macroscopic or mesoscopic segregation of the two ground-states. No definitive conclusion can be obtained in this case concerning the symmetry of the superconducting order parameter since the ground-states occupy different spatial regions that are in principle mutually independent from each other. This condition can be driven for instance by trivial inhomogeneity of the chemical doping in the investigated material and it has typically to deal with extrinsic properties. The opposite case $d \sim 1 \text{–} 10 \text{nm}$ is by far more interesting since it implies a much finer intertwining of the two order parameters (nanoscopic segregation or nanoscopic coexistence) toward the limit of the so-called 'atomic coexistence' ideally realized in the same spatial position. This is clearly a much more interesting limit in order to draw conclusions on the intrinsic properties of the examined materials.

The experimental way of treating this problem is typically not trivial at all, and qualitatively different results can be obtained as a function both of the chemical homogeneity of the investigated sample and of the technique employed. In this respect, it should be stressed that $\mu^\text{+SR}$ measurements are of crucial importance for the reasons outlined above. The local nature of the technique allows one to be sensitive to the disordered magnetism that is typically realized in the case of nanoscopic coexistence. As a further advantage with respect to other local techniques like, e.g. NMR, only $\mu^\text{+SR}$ allows to precisely estimate $V_m$. This information is of major importance in order to quantify the order of magnitude of $d$. Let us refer to figure 9 where a sketchy representation of the phase segregation of two different order parameters is reported [91]. The global magnetic moment within the domains is null due to the AFM correlations characteristic of the SDW phase. Still, $\mu^+$ are able to feel the dipolar field generated by the uncompensated magnetic moments on the domain walls (see red arrows in the figure under the magnifying lens). In the case of the dipolar field generated by Fe in the case of 1111 oxy-pnictide materials, one can roughly deduce that the minimal distance required in order to probe magnetism under these conditions for $\mu^+$ implanted out of the domains is of the order of $1 \text{nm}$ [92]. In the case of the mesoscopic segregation of the order parameters, $\mu^+$ implanted out of the magnetic domains (and conventionally labeled as $\mu_1^+$) are on average too far away from the domains themselves to probe static magnetism and only $\mu^+$ implanted into the domains (and conventionally labeled as $\mu_2^+$) give rise to a magnetic signal. As a result, $V_m < 1$ in the case of mesoscopic segregation and a value $(1 - V_m) > 0$ is measured as a paramagnetic volume fraction, accordingly. This behavior was clearly visible in the case of La-based 1111 samples with the phase diagram being swept both by the increasing concentration of electrons triggered by the $\text{O}_{1-x}\text{Fe}$ substitution [93] or by the application of external hydrostatic pressure [46], at variance with what is typically reported for 1111 materials based on magnetic RE ions, as will be shown subsequently. At the same time, early reports on hole-doped 122 compounds like $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ seemed to confirm the picture of the mesoscopic segregation [91] at variance with what was reported about electron-doped compounds like $\text{Ba}_{(\text{Fe}_{1-x}\text{Co}_x)_2}\text{As}_2$ [86, 94, 95]. It should be noted that independent quantifications of the superconducting shielding fractions of samples displaying mesoscopic segregation (e.g. derived from dc magnetometry or ac susceptibility) show a complementary trend with respect to that of the magnetic volume fraction across the phase diagram. In particular,

Figure 9. Sketchy representation of the model of phase segregation of magnetism and SC (figure reprinted with permission from Park et al [91]. Copyright (2009) by the American Physical Society). The nanoscopic coexistence is realized whether the order-of-magnitude of the mean distance among magnetic domains (green droplets) is close to $d \sim 1 \text{nm}$, namely the spatial range of the magnetic dipolar field generated by uncompensated Fe moments on the domain walls (red arrows).
and now assuming that these quantities are typically reflected into a specular increase in the saturation value for the superconducting shielding fraction. This is clearly explained by figure 10 where these quantities are reported for LaFeAsO$_{0.94}$F$_{0.06}$ as a function of the external pressure. These results confirm the picture of segregation of the two different electronic environments into well-separated regions strongly competing for volume. It should be noted that such a picture for LaFeAsO$_{1-x}$F$_x$ is fully consistent with the first-order-like transition between the two electronic ground-states reported in [96].

On the other hand, the cases of Ce- and Sm-based 1111 oxy-pnictides were shown to be dramatically different from that observed in LaFeAsO$_{1-x}$F$_x$. In these materials, the coexistence region of magnetism and SC was shown to be characterized by $V_{m} \gtrsim 100\%$ with a bulk fraction of the sample being at the same time a superconductor, as illustrated in figure 11 in the case of CeFeAsO$_{0.94}$F$_{0.06}$ [92]. These findings are interpreted by coming back to figure 9 and now assuming that $d \sim 1-10$ nm. Under these circumstances, both $\mu_1^s$ and $\mu_2^s$ (namely, all the implanted $\mu^-$) probe static magnetism being the maximum distance between different magnetic domains of the same order of magnitude of $d$, namely the spatial range for the dipolar fields generated by domain walls. Accordingly, one has $V_{m} \simeq 100\%$ and it must be assumed that the interstitial regions between the different magnetic domains are superconducting since dc magnetometry confirms that a bulk fraction of the sample contributes to the diamagnetic shielding. Such fine intertwining of different order parameters was detected in SmFeAsO$_{1-x}$F$_x$ [97–99], CeFeAsO$_{1-x}$F$_x$ [51, 92] and CeFe$_{1-x}$Co$_x$AsO [53].

It should be noted that the picture of mesoscopic segregation described above for the case of Ba$_{1-x}$K$_x$Fe$_2$As$_2$ was modified after the investigation of samples with higher chemical homogeneity, demonstrating that the nanoscopic coexistence is a more suited framework for such materials [102]. At the sake of clarity, results relative to the hole-doped Ba$_{1-x}$K$_x$Fe$_2$As$_2$ are reported in figure 12 [102]. It is rather interesting to observe that a static magnetic ground-state is also recovered by diluting optimally electron-doped 1111 materials with Ru [42, 43]. Remarkably, the magnetic and superconducting ground-states coexist at the nanoscopic level and the latter is completely suppressed for a Ru content around 60% (see figure 5). The observation of such a tiny effect of diamagnetic impurities on the superconducting transition temperature would appear at first in contrast with a magnetic pairing mechanism and has lead to alternative explanations, such as the ones involving orbital currents [61, 62]. Still, it is possible that the subtle interplay between intraband and interband pairing processes can give rise to such a slow decrease of $T_c$ even in the framework of a magnetic pairing mechanism [100, 101]. Finally, it is important to point out that the different families of IBS show some significant differences. In figure 13 the phase diagram of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ [86] and of CeFe$_{1-x}$Co$_x$AsO [53] is reported. In spite of the quantitatively similar trend of the magnetic ordering temperature, which in the Ce-based 1111 compound was shown to be long-range for $x < 0.03$ and short-range at higher doping levels [53], one notices clear differences at the crossover between the magnetic and the superconducting ground-states. In the 122 compound the coexistence region between the two phases is quite extended while in the 1111 compound it is only marginal. Moreover, the superconducting transition temperature is lower in the 122 compound than in the 1111 compounds. This suggests that the differences in the band structure, namely in the FS nesting, and in the anisotropy of the two family of compounds could play a relevant role in determining the superconducting properties, particularly when the charge doping is associated with the introduction of impurities in the FeAs planes.
Figure 12. Nanoscopic coexistence of magnetism and SC in Ba$_{1-x}$K$_x$Fe$_2$As$_2$ (figure reprinted with permission from Wiesenmayer et al [102]. Copyright (2011) by the American Physical Society). (a) Superconducting shielding fractions. (b) Electronic phase diagram for Ba$_{1-x}$K$_x$Fe$_2$As$_2$. (c) Magnetic volume fractions.

Figure 13. The phase diagram of Ba$_{1-x}$CeFe$_x$AsO [86] and of CeFe$_{1-x}$Co$_x$AsO [53] are reported as a function of the Co content $x$. Open symbols show the behavior of $T_N(x)$ while the closed symbols show that of $T_c(x)$.

4. Transverse-field muon spin spectroscopy ($\mu$SR) in superconducting materials

When type II superconductors, such as the IBS, are field-cooled (FC) in an external field $H_0 < H_c < H_{c2}$ ($H_0$, $H_{c1}$ and $H_{c2}$ being the external, the lower and the upper critical field, respectively) the magnetic field is not homogeneously distributed throughout the sample but concentrated along tubes which form a FLL. Since the distance of two adjacent flux lines is much larger than the distance between the muon sites, the muons are able to finely probe the field distribution generated by the FLL. Such distribution leads to an enhancement of the muon depolarization rate which can be suitably probed in TF $\mu$SR experiments. The FLL field distribution is not symmetric around $H_0$ [14] since it reflects the minimum, the maximum and the saddle-point values of the magnetic field profile within the triangular FLL unit cell. In real samples, and in particular in powders, the FLL imperfections arising from randomly distributed pinning centers average out these asymmetries and the field distribution tends to a Gaussian one, centered around the average internal field $B_\mu$. Hence, the TF muon depolarization function can be approximated by $D_{TF}(t) \approx \exp[-(\sigma_{\text{SC}}^2 + \sigma_{\text{w}}^2) t^2/2]$, with $\sigma_{\text{w}}$ the weak relaxation due to the nuclear magnetic dipoles (see equation (2)) and $\sigma_{\text{SC}}$ the second moment of the FLL field distribution, namely $\sigma_{\text{SC}}/\gamma_\mu = (B_0^2 - B_\mu^2)^{1/2}$. The latter term is proportional to the inverse square of the London penetration depth $\lambda_L$, and, in turn, to the supercarrier density $n_s$. Accordingly, one can write that $\sigma_{\text{SC}}(T) \propto n_s/m^*$, with $m^*$ the electron effective mass. Since each flux line is surrounded by a screening current over a distance of the order of $\lambda_L$, the average field at the muon site $B_\mu$ is slightly lower than the applied field, namely $B_\mu = (1 + 4\pi \chi) H_0$ with $\chi < 0$. 

\[ \sigma_{\text{SC}}(T) \propto n_s/m^* \]
Throughout the whole temperature range $0 < T < 50$ K with a constant amplitude $\alpha_{TF}$. This ensures that all the muons probe the same field distribution, i.e. the sample is always single phase and fully superconducting. The depolarization rate for $T > T_c$ is Gaussian and its rather low value, $\sigma_N = 0.17 \mu s^{-1}$, indicates that the amount of dilute magnetic impurities, which are often present in IBS and cause an unwanted enhancement in the relaxation rate [97, 103], is negligible in that sample. The temperature evolution of the FLL contribution to the depolarization rate, $\sigma_{SC}(T)$, and the relative shift of the field at the muon site, $(B_{\mu}(T) - H_0)/H_0$, due to the diamagnetic shielding are shown in figures 15(b) and (c), respectively. The development of the FLL is clearly evidenced both by the increase of $\sigma_{SC}$ and by the diamagnetic shift of $B_{\mu}$ below $T_c = 28$ K, which coincide with that measured by $\chi(T)$. From the value of $\sigma_{SC}$ extrapolated at $T = 0$, using the relation $\sigma_{SC} \approx 7.58 \times 10^{-4} \lambda_L^2$, in CGS units [104, 105], the London penetration depth is estimated to be $\lambda_L \approx 2620$ Å. The temperature dependence of $\sigma_{SC}(T)$ follows the $s$-wave weak coupling Bardeen, Cooper and Schriffer (BCS) temperature dependence predicted by the two fluid model $\sigma_{SC}(T) \propto 1/\lambda^2(T) \propto [1 - (T/T_c)^2]^4$, as shown by the curve in figure 15(b). In particular, the flat behavior for $T < T_c/3$ is indicative of the absence of nodes in the gap function.

The $s$-wave-like $T$-dependence of $\sigma_{SC}$ was also observed in an La-based 1111 compound with $T_c = 23$ K [106]. However, this behavior is in conflict with the observed dependence of $\sigma_{SC}$ on the magnitude of the applied field $H_0$. In fact, as it is shown in figure 16 [106], $\sigma_{SC}$ displays a maximum for a field value $B^{\text{max}} \approx 200$–2000 G and at higher fields the relaxation rate decreases, which is usually indicative of SC with nodes in the gap function [69, 106]. It should be noted that this feature, also found in Sm-based 1111 compounds, could rather be due to the presence of different superconducting gaps [108], as can be expected for system with several bands crossing the Fermi level. This field dependence should be carefully considered when comparing results from different experiments in the so-called Uemura like plot, where $T_c$ is plotted as a function of $\sigma_{SC}(T = 0) \propto n_s(T = 0)$, in figure 17 this behavior is shown for the 1111 family as a function of the F doping for those values of $\sigma_{SC}(T = 0)$ measured at a field close to $B^{\text{max}}$. Although a theoretical description of the Uemura plot for the 1111 IBS is still missing, it is clear that the rate of the decrease of $T_c$ with $n_s(0)$ strongly depends on the symmetry of the order parameter, on the presence of nodes in the gap.
and on the occurrence of pair breaking processes. Hence, a suitable modeling of that plot would have significant implications in the clarification of the pairing mechanism for IBS. Nevertheless, it also has to be remarked that a temperature dependence of $\sigma_{SC}$ can be observed at low $T$ in underdoped La-based 1111 samples [107], leading to some uncertainty in the estimate of $\sigma_{SC}(T = 0)$. Indeed, while analyzing the behavior of $\sigma_{SC}(T)$ great care must be taken in order to discern those effects not associated with the $n_s$ $T$-dependence, as will be described in the following section.

5. Transverse-field $\mu^+\text{SR}$ in case of coexistence between magnetism and SC

In IBS, two main effects might alter the TF depolarization rates. One is extrinsic and comes from diluted ferromagnetic impurities which are often present in polycrystalline samples. These can be recognized in ZF experiments by an exponential behavior (figure reprinted with permission from Luethens et al [106]. Copyright (2008) by the American Physical Society).

In the 1111 compounds, where magnetism and SC coexist at the nanoscopic level, $B_i$ ranges from a maximum field of few hundreds of Gauss to nearly zero, since it originates from ordered moments varying over distance from less than one to four-five lattice steps. In this case, one may assume that a fraction of the muons experiences internal fields lowered by $4\pi|\chi|H_0$. Then, the typical TF signal of the FLL is roughly distinguishable, even if it is still slightly affected by the nearby $B_i$ which give rise to a moderate increase both of $B_\mu(T)$ and $\sigma_{SC}(T)$. For example, here the behavior of a NdFe$_{0.8}$Ru$_{0.2}$As$_{0.8}$O$_{0.8}$F$_{0.13}$ powder sample is considered. This sample has $T_N = 26$ K (figure 18(a)) and by ZF muon experiments it was shown to display a magnetic transition with an onset at $T_N^{\text{onset}} \approx 20$ K and a full magnetic volume fraction $V_m \approx 1$ below $T = 5$ K, with internal fields at the muon site of the order of 200 G (see figure 1 of [43]). The TF asymmetry data (not shown) fit quite well to the sum of two oscillating terms

$$\frac{A_{TF}(t)}{A_{TF}(0)} = a_{SC} \exp(-\Delta t) \exp[-(\sigma_{SC}t/2)] \cos(\gamma_\mu B_\mu t)$$

$$+ a_{mag} \exp[-\lambda_{mag}t] \cos(\gamma_\mu B_{mag}t), \quad (10)$$

where the first accounts for those regions with vanishing $B_i$, where the field distribution still reflects the FLL for $T > T_{N\text{onset}}$, while the second one accounts for those regions where the spontaneous internal field is of the order of $H_0$. The $\Lambda$ decay rate takes into account the exponential decay already measured at high temperature, close to the value $\Lambda \approx 0.8 \mu s^{-1}$ and nearly temperature independent, which is most probably due to the presence of a small fraction of diluted magnetic impurities. The temperature dependence of the TF parameters is shown in figure 18. SC is well reflected by the behavior both of the increase of $\sigma_{SC}(T)$ and of the diamagnetic shift (right axis of panels (c) and (d), respectively) below the same $T_C$ determined by the $\chi(T)$ curve. At low temperature they both deviate from the behavior expected for a single SC phase compound, roughly for $T < 10$ K where the ZF experiment displays a sizeable magnetic volume fraction [43], confirming
the IBS, in the understanding of the phase transition between the magnetic and the superconducting phases and of their possible coexistence at the nanoscopic level. The TF-$\mu$SR studies reported in section 4, performed in the mixed phase where the FLL sets in, have provided a description of the evolution of the superconducting carrier density $n_s$ as one spans through the phase diagram of IBS, and have shown how this quantity is correlated with $T_c$. The behavior of $n_s(T)$ and of $n_s(T \rightarrow 0)$ versus $T_c$ have given relevant hints on the symmetry of the superconducting order parameter and on the microscopic mechanisms underlying the SC in IBS. A remarkable experimental effort is still required to have a clear understanding of the physics of IBS and to distinguish the peculiarities of each family of compounds from the relevant features underlying a common description of the superconducting condensate.

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