Coexistence of superconductivity and ferromagnetism in iron pnictides

Guang-Han Cao, Wen-He Jiao, Yong-Kang Luo, Zhi Ren, Shuai Jiang and Zhu-An Xu
Department of Physics and Center for Correlated Matter, Zhejiang University, Hangzhou 310027, China
E-mail: ghcao@zju.edu.cn

Abstract. Superconductivity (SC) and long-range ferromagnetism (FM) are mutually antagonistic, thus in general SC does not coexist with FM. In this mini-review, however, we will show that such a coexistence can inhabit several iron pnictide systems, including EuFe$_2$(As$_{1-x}$P$_x$)$_2$, Eu(Fe$_{1-x}$Co$_x$)$_2$As$_2$, Eu(Fe$_{1-x}$Ru$_x$)$_2$As$_2$, CeFe(As$_{1-x}$P$_x$)O$_{0.95}$F$_{0.05}$ and Sr$_2$VFeAsO$_3$. We will briefly discuss the possible reasons and consequences of the coexistence of 3$d$-electron SC and 4$f$-electron FM.

1. Introduction
Superconductivity (SC) and ferromagnetism are two intriguing collective phenomena in condensed matter physics. Conventional SC is characterized by spin-singlet $s$-wave Cooper pairing, in which time-reversal symmetry is conserved. However, ferromagnetism breaks time-reversal symmetry. This fundamental difference makes the two phenomena antagonistic and competitive. One may easily see this point by the fact that all the magnetic elements do not superconduct. Ginzburg[1] first pointed out that SC can be destroyed in external magnetic field by electromagnetic interaction. The incompatible nature of SC and FM was demonstrated by the experiments followed, which showed the competition of the two collective phenomena in (La,Gd) and (Ce,Pr)Ru$_2$ solid solutions.[2] The suppression of FM in the superconducting regime was explained by Anderson and Suhl[3] in terms of the Ruderman-Kittel-Kasuya-Yosida (RKKY) interactions by the end of 1950s. Judging from the energy scale, however, FM wins SC in most cases. Thus it was suggested that in the superconducting state pure ferromagnetism should be modified in the form of cryptoferromagnetic alignment for localized spins. It was not until late 1970s that coexistence of SC and FM was evidenced in ErRh$_4$B$_4$[4] and Ho$_{1.2}$MoxS$_8$[5], in narrow regimes of temperature and external field. In late 1990s, SC and weak ferromagnetism were observed in high-$T_c$ rutheno-cuprates[6, 7]. In the examples above, SC and FM originate from obviously different electrons of different elements. However, there is a scenario that both SC and FM come from the same type of electrons, as in UGe$_2$[8] and URhGe,[9] where the SC emerges under the ferromagnetic background ($T_c < T_M$). It is widely believed that the spin structure of the Cooper pairs is in triplet states.

The recently discovered Fe-based superconductors[10] are characterized by multiband superconductivity as well as high transition temperatures. This feature makes it possible to see new phenomena including those on the interplay of SC and FM. The outstanding example comes...
from the Eu-based iron pnictides, EuFe$_2$As$_2$-related systems, in which the Eu$^{2+}$ ions show large local magnetic moments with $J = S = 7/2$. By the chemical doping either at the Fe-site or at the As-site, coexistence of SC and FM (or with significant ferromagnetic components) has been observed[11, 12, 13] and confirmed by various kind of method[14, 15, 16, 17, 18, 19]. Evidence of additional coexistence of SC and FM was also observed in other iron pnictide systems, e.g., SrFe$_2$As$_2$ with internal strain[20] CeFe(As$_{1-x}$P$_x$)O$_{0.95}$F$_{0.05}$[21] and Sr$_2$VFeAsO$_3$[22], which suggests common mechanism that supports the coexistence.

2. EuFe$_2$(As$_{1-x}$P$_x$)$_2$ system

The two end members in EuFe$_2$(As$_{1-x}$P$_x$)$_2$ system crystallize in the same ThCr$_2$Si$_2$-type structure with space group $I4/mmm$, however, they show contrasting physical properties. It was confirmed that EuFe$_2$As$_2$ underwent two magnetic transitions at $\sim$200 K and 20 K, respectively.[23] We first recognized the 200-K transition as a spin-density-wave (SDW) ordering associated with the iron sublattice, upon which superconductivity was predicted to realize through appropriate chemical doping.[23] Furthermore, we proposed an A-type antiferromagnetic (AFM) order for the Eu sublattice.[23, 24] which was then confirmed by magnetic resonant x-ray scattering[25] and neutron diffraction[26] experiments. On the other hand, the iron in EuFe$_2$P$_2$ is non-magnetic, and the Eu$^{2+}$ moments order ferromagnetically at 29 K with the spin direction tilted toward the c-axis.[27, 28]

Although neither EuFe$_2$As$_2$ nor EuFe$_2$P$_2$ superconduct, their solid solutions with certain range of P concentration do. For instance, in EuFe$_2$(As$_{0.7}$P$_{0.3}$)$_2$, SC transition occurs at $T_c=26$ K (see the inset of figure 1). Following the superconducting transition, surprisingly, the Eu sublattice becomes ferromagnetically ordered below 20 K.[11] Bulk nature for both SC and FM was confirmed by the heat capacity measurement, which demonstrates unambiguously the coexistence of SC and FM to the microscopic scale. The recent Mössbauer study[14, 19] shows the canting of the Eu spins toward the c-axis with the phosphorus doping. The spin canting angle and the $^{151}$Eu hyperfine field "saturate" at $x=0.2$, suggesting ferromagnetism for the Eu sublattice. The spin canting at lower doping level ($x = 0.12$) was also evidenced by the anisotropic magnetization,[18] indicating that there is ferromagnetic component along the c-axis.

Figure 1 shows the superconducting and magnetic phase diagram in EuFe$_2$(As$_{1-x}$P$_x$)$_2$.[29] With the P doping, the Fe-AFM is suppressed rapidly, and then SC emerges for $x \geq 0.2$. This phenomenon is similar to that in the BaFe$_2$(As$_{1-x}$P$_x$)$_2$ system.[30] The difference is that the
former has narrower superconducting window. Compared with the Fe sublattice, the magnetic transition temperature of the Eu sublattice changes very mildly. However, it changes from AFM to FM, accompanied with the tilting of Eu spin direction. SC coexists with FM for 0.2 ≤ x ≤ 0.4. A recent paper based on single crystal investigations shows a similar phase diagram, however, the SC regime varies a little.[17] Despite of the discrepancy, it is clear that in the superconducting region the Eu sublattice at least has spontaneous ferromagnetic components because of the Eu$^{2+}$ spins tilted along the same direction.

3. Eu(Fe$_{1-x}$M$_x$)$_2$As$_2$ (M=Co and Ru) system

As is known, Fe-site doping may also introduce SC. By doping with Ni, we observed ferromagnetism for the Eu$^{2+}$ moments, though SC transition was not able to be seen.[31] However, evidence of SC was given for the Co doping.[12] As can be seen in figure 2, the resistivity of Eu(Fe$_{0.89}$Co$_{0.11}$)$_2$As$_2$ crystals drops at ~21 K, suggestive of a SC transition. The corresponding SC diamagnetism can be detected only when the applied field is very small (see the inset of figure 2). There is a clear resistivity reentrance around 17 K, which was confirmed to be due to the magnetic ordering of Eu spins.

![Figure 2](image2.png)

**Figure 2.** (Adapted from Ref. [12]) Resistivity and magnetic susceptibility (lower inset) for Eu(Fe$_{0.89}$Co$_{0.11}$)$_2$As$_2$.

The Ru doping in EuFe$_2$As$_2$ gives stronger signal for SC, as shown in figure 3. Not only does the resistivity show sharp superconducting transition to zero, but also the magnetic susceptibility has strong diamagnetic signal for the zero-field-cooling mode (the step-like anomaly is due to the magnetic ordering of Eu spins). When changing the Ru content, interesting anisotropic SC was observed, which could be the result from either spontaneous vortices or a novel FFLO state.[13]

4. Discussion

Apart from the Eu-containing iron pnictide system, there are some other cases showing the evidence of coexistence of SC and FM. For example, In CeFe(As$_{1-x}$P$_x$)O$_{0.95}$F$_{0.05}$ system, a ferromagnetic transition (associated with Ce 4f electrons) occurs at ~5 K, meanwhile, SC transition happens at 13 K for x=0.5.[21] Another example is seen in Sr$_2$VFeAsO$_3$, where V 3d weak FM takes place at 55 K, followed by a SC transition at 24 K.[22] These facts suggest a common mechanism supporting the coexistence of SC and FM in iron pnictides.

Here we present a simple explanation. Since the superconducting upper critical fields $H_{c2}$ in Fe-based superconductors are very high (e.g., ~60 T for BaFe$_2$As$_2$[32]), which are probably
higher than the exchange fields among the Eu spins, thus SC may survive in the presence of a strong ferromagnetic internal fields. On the other hand, the multiband feature of Fe 3d electrons allows the effective RKKY interactions among the Eu$^{2+}$ spins even in the superconducting state. It is widely believed that $d_{xz}$ and $d_{yz}$ orbitals are most probably related to SC, and the $d_{z^2}$ electrons are supposed to be responsible for mediating RKKY interactions. Therefore, both SC and FM could be satisfied simultaneously. It is our point that the multiband effect plays an important role to facilitate the coexistence of SC and FM.

Finally we would like to talk about the possible consequences of the coexistence. Just like the modulation of FM by SC, proposed by Anderson and Suhl[3], SC should also be modified by the internal exchange fields. Spontaneous vortex state and FFLO phase are the possible candidates.[13] In most cases the Fe-based SCs have the following characteristic: 1)pauli-limiting $H_{c2}$ and large Maki parameters; 2)multi-Fermi pockets with small $E_F$ (or $E_F$ can be tuned to a small value); 3) quasi-two dimensionality. All these factors favor for the formation of FFLO state. Thus we expect a novel FFLO state at zero external fields in the SC/FM coexisted iron pnictide system.

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