Magnetism and superconductivity in single crystals $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$

Q. J. Zheng, Y. He, T. Wu, G. Wu, H. Chen, J. J. Ying, R. H. Liu, X. F. Wang, Y. L. Xie, Y. J. Yan, Q. J. Li and X. H. Chen

Hefei National Laboratory for Physical Science at Microscale and Department of Physics, University of Science and Technology of China, Hefei, Anhui 230026, People’s Republic of China

We systematically studied the transport properties of single crystals of $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$. Co doping can suppress the spin-density wave (SDW) ordering and induces a superconducting transition, but a resistivity reentrance due to the antiferromagnetic ordering of $Eu^{2+}$ spins is observed, indicating the competition between antiferromagnetism (AFM) and superconductivity. It is striking that the resistivity reentrance can be completely suppressed by external magnetic field (H) because a metamagnetic transition from antiferromagnetism to ferromagnetism for $Eu^{2+}$ spins is induced by magnetic field. Superconductivity without resistivity reentrance shows up by partial substitution of $Eu^{2+}$ with non-magnetic $Sr^{2+}$ to completely destroy the AFM ordering of $Eu^{2+}$ spins. These results suggest that the antiferromagnetism destroys the superconductivity, while the ferromagnetism can coexist with the superconductivity in the iron-based high-$T_c$ superconductors.

PACS numbers: 74.25.-q, 74.70.-b, 75.50.-y

Interplay between magnetism and superconductivity has long been an interesting issue in condensed matter physics, where in most cases, these two orders compete with each other. Theoretical work done by Ginzberg, Balsenperger and Straesler predicted that long range ferromagnetism would greatly damage superconductivity while antiferromagnetism could coexist with superconductivity to some extend [1, 2]. Indeed, in traditional magnetic superconductors like RMo$_6$O$_8$ and RRh$_4$B$_4$ (R = magnetic rare earth ions), superconductivity is found to be moderately robust coexisting with antiferromagnetism, yet fragile with ferromagnetism (FM). However, coexistence of ferromagnetism and superconductivity was observed in heavy fermion system UGe$_2$ and unconventional high-$T_c$ cuprates superconductor RuSr$_2$GdCu$_2$O$_{8+δ}$, where local moments locate far from conducting plane. Apart from all mentioned above, one of the most typical families of magnetic superconductor is RNi$_2$B$_2$C where R = Ho, Er, Tm, Yb, Lu, [6, 7] with ThCr$_2$Si$_2$-type structure. In these materials, a resistivity reentrance below superconducting transition temperature was observed due to the antiferromagnetic ordering of R ions. Therefore, the interaction between superconductivity and magnetic ordering is still puzzling. Here we report intriguing results that the antiferromagnetism destroys the superconductivity, while the ferromagnetism can coexist with the superconductivity in the iron-based high-$T_c$ superconductors. The antiferromagnetism and ferromagnetism can be manipulated by tiny magnetic field. These results are definitely significant to understand the interaction between superconductivity and magnetic ordering.

Since Fe ions are more likely to convey magnetic moment in various ways due to their nearly half filled 3d orbital and larger degree of freedom in electronic spin states than cuprates, the new iron-based family of high $T_c$ superconductors [8, 9] is well expected to be a promising category among which might exist new magnetic superconductors. In fact, an AFM SDW ordering of $Fe^{2+}$ is widely observed in the parent compounds of iron-based superconductors [10, 11]. The $ARFe_2As_2$ (AR=Ba, Sr, Ca, Eu) 122 family have a relatively simply-structured charge reservoir layer which may serve as an ideal candidate to embed magnetism into this system. The $Ba^{2+}$, $Sr^{2+}$ and $Ca^{2+}$ are non-magnetic ions, while $Eu^{2+}$ is magnetic ion with $s=7/2$. The magnetic ordering of $Eu^{2+}$ spin occurs at about 17 K. As illustrated in Fig.
1, an A-type antiferromagnetic structure for Eu$^{2+}$ sub-lattice is proposed by Wu et al. and Jiang et al. in parent compound EuFe$_2$As$_2$, where local moments align collinearly to form strong FM order within ab plane and weak AFM order along c axis (Fig.1a and 1b). The interlayer AFM coupling can be tuned to FM coupling (Fig.1c) by a tiny external magnetic field. In Ba122 system, the superconductivity can be induced by substitution of K for Ba or Co-doping on Fe site. Therefore, EuFe$_2$As$_2$ is a good system to study the interaction between superconductivity and magnetic ordering of Eu$^{2+}$ spins. Therefore, we systematically measure the transport properties to study the interaction between superconductivity and magnetism in the Co-doped Eu122 system in which the magnetic ordering for Eu$^{2+}$ sublattice nearly does not change with Co doping. Then, we substituted Eu with Sr to systematically suppress the magnetic ordering and study the effect of suppression of magnetic ordering on superconductivity. It is found that a resistivity reentrance due to the antiferromagnetic ordering of Eu$^{2+}$ spins is observed below $T_c$, indicating the competition between antiferromagnetism (AFM) and superconductivity. It is striking that the resistivity reentrance can be completely suppressed by external magnetic field (H) because of the metamagnetic transition from antiferromagnetism to ferromagnetism for Eu$^{2+}$ spins induced by H. These results suggest that the antiferromagnetism destroys the superconductivity, while the ferromagnetism favors the superconductivity in the iron-based high-$T_c$ superconductors.

The single crystals of EuFe$_{2-x}$Co$_x$As$_2$ and Eu$_{0.25}$Sr$_{1-x}$Fe$_{2-x}$Co$_x$As$_2$ were synthesized via conventional self-flux method. Figure 2 demonstrates the systematic evolution in temperature dependent resistivity in the temperature range from 290K down to 5K for EuFe$_{2-x}$Co$_x$As$_2$ single crystals. The left figure (Fig.2a) presents the resistivity data for the underdoped samples, and the right figure (Fig.2b) illustrates resistivity as a function of temperature for the optimal doped ($x=0.285$) and overdoped samples. In underdoped region, the resistive behavior largely resembles that of BaFe$_{2-x}$Co$_x$As$_2$ in terms of high temperature features. The only difference happens at about 17 K, where a dopant independent kink emerges, corresponding to the AFM ordering established among Eu$^{2+}$ layers. Further Co-doping introduces a superconducting transition in resistivity around 22 K, but consequently a resistance reentrance shows up just below the AFM ordering temperature, so that no zero resistivity is observed with the temperature down to 4.2 K in EuFe$_{2-x}$Co$_x$As$_2$ system. This is totally different from that observed in BaFe$_{2-x}$Co$_x$As$_2$ system. It suggests that the introduction of Co$^{2+}$ into FeAs layer fail to bring about superconductivity in EuFe$_{2-x}$Co$_x$As$_2$ system. Even the optimally doped single crystal at $x = 0.285$ just shows a resistivity decrease by less than 80% before the occurrence of resistivity reentrance. No superconducting transition and no resistivity reentrance are observed in the overdoped crystal with $x=0.5$.

As reported by Wu et al., Eu$^{2+}$ ions in

**FIG. 2:** (Color online). Temperature dependence of in-plane resistivity for the EuFe$_{2-x}$Co$_x$As$_2$ single crystals. (a): $x=0$, 0.1, 0.14, 0.2, 0.25 and 0.275 from 5 K to 300 K; (b): $x=0.275$, 0.285, 0.35, 0.4 and 0.5 from 5 K to 40 K. Note that all the dips in resistivity correspond to the same temperature as $T_N$ in parent EuFe$_2$As$_2$.

**FIG. 3:** (Color online). Temperature dependence of resistivity and susceptibility under different magnetic fields for optimal doped single crystal EuFe$_{1.75}$Co$_{0.25}$As$_2$. (a): H applied within ab-plane; (b): H applied along c-axis; (c): H applied within ab-plane; (d): H applied along c-axis; (e): Specific heat under different H applied along c-axis (enlarged from 9 K to 24 K).
FIG. 4: (Color online). (a): Temperature dependent resistivity in single crystals of Eu$_{1-y}$Ba$_y$Fe$_{1.8}$Co$_{0.2}$As$_2$. (b): Temperature dependent resistivity in single crystals of Eu$_{1-x}$Sr$_x$Fe$_{1.715}$Co$_{0.285}$As$_2$. (c) and (d): Temperature dependent resistivity under different magnetic fields for magnetically weakened Eu$_{0.7}$Sr$_{0.3}$Fe$_{1.715}$Co$_{0.285}$As$_2$ single crystals, where both superconducting transition and resistance reentrance emerge at low temperatures. (c): H applied within ab-plane; (d): H applied along c-axis.

EuFe$_2$As$_2$ would experience a metamagnetism from AFM to FM induced by magnetic field at low temperature. In order to study how superconductivity behaves together with AFM and FM order, we measured the temperature dependent resistivity under different magnetic field, specific heat and susceptibility of the optimal doped sample EuFe$_{1.715}$Co$_{0.285}$As$_2$. As shown in Fig.3a, the reentrance in resistivity is continuously suppressed both in terms of intensity and corresponding temperature with increasing H applied within ab-plane. When the H reaches to 1T, the resistivity reentrance is completely suppressed, accompanying by a conventional field-induced suppression on superconductivity. Figure 3b presents the resistivity data measured with H applied along c-axis for the same sample. Unlike the case in Fig.3a, the resistivity reentrance does not exhibit conspicuous sign of shift or weakening with increasing H, and the only change is the closing of the gap in resistivity except for the expected down-shift of $T_c$ and transition broadening induced by magnetic field. These results are consistent with the results of susceptibility as shown in Fig.3c and 3d. Hence we may reckon that the easy axis lies in ab-plane, which might have contributed to the insensitive behavior when applying H along c-axis. Susceptibility and specific heat measurements further confirm the antiferromagnetic transition at 17K. In Fig.3c, the Neel temperature monotonously decreases with increasing H. The similar suppression of the peak on specific heat could be observed in Fig.3e. After H goes beyond 0.5 T, neither response from resistivity nor susceptibility could be clearly observed. It is because a metamagnetism from AFM to FM has taken place. These results give strong evidence that a resistivity reentrance arises from the antiferromagnetic ordering of Eu$^{2+}$ spins, indicating the competition between antiferromagnetism (AFM) and superconductivity. It is striking that the resistivity reentrance can be completely suppressed by external magnetic field (H) due to the field-induced metamagnetic transition from antiferromagnetism to ferromagnetism for Eu$^{2+}$ spins. All above measurements concerning single crystals of EuFe$_{2-x}$Co$_x$As$_2$ have contributed to the picture that the spins of Eu$^{2+}$ ions tend to establish an antiferromagnetic order around 17 K, and are easily tuned to ferromagnetic order with small H. Such metamagnetism of Eu$^{2+}$ ions provides a good system to study the intriguing interaction between AFM/FM and superconductivity. Antiferromagnetism appears to have strongly counteracted superconductivity, while ferromagnetism could coexist rather at ease with the superconductivity.

In order to make sure that it is AFM from Eu$^{2+}$ sublattice that destroys superconductivity, we choose to partially substitute Eu$^{2+}$ with nonmagnetic Ba$^{2+}$/Sr$^{2+}$. As expected, superconductivity shows up at 23 K as shown in Fig.4a for the Ba-doped crystals, being consistent with that in BaFe$_{1.8}$Co$_{0.2}$As$_2$ crystal. Figure 4b clearly demonstrates the evolution in resistivity in Eu$_{1-x}$Sr$_x$Fe$_{1.715}$Co$_{0.285}$As$_2$ system. With increasing Sr-doping, the gap caused by reentrance is gradually narrowed along with the suppression of the reentrance’s peak. For the crystal with x=0.3, superconducting transition is sharp, and the resistivity reaches zero shortly before a resistivity reentrance takes place. Further Sr-doping eventually kills the resistivity reentrance, and stable superconducting phase can be achieved for the crystal with x=0.5. Figure 4c and 4d present the temperature dependent resistivity under different H applied within ab-plane and along c-axis for the Eu$_{0.7}$Sr$_{0.3}$Fe$_{1.715}$Co$_{0.285}$As$_2$ sample, respectively. The resistivity reentrance can be continuously suppressed with increasing magnetic field applied within ab-plane. However, we didn’t observe the same trend with H applied perpendicular to ab-plane. This could probably be understood in terms of the anisotropic magnetic structure and exchange integration within or between Eu$^{2+}$ layers. In Eu$_{1-x}$Sr$_x$Fe$_{2-y}$Co$_y$As$_2$ system, the antiferromagnetism appears to be more destructive to superconductivity, while the ferromagnetism can coexist with superconductivity. It should be pointed out that such metamagnetic transition from AFM to FM is induced by a small external magnetic field. Therefore, the observed behavior here is intrinsic.

In RNi$_2$B$_2$C system, a pronounced resistivity reentrance was observed when AFM transition in
RC layers happens below superconducting transition temperature, similar to the observation in Eu$_{0.7}$Sr$_{0.3}$Fe$_{1.715}$Cu$_{0.285}$As$_{2}$ system. If the AFM transition temperature $T_N$ is lower than critical superconducting temperature $T_c$, $T_c$ could be negatively scaled by $R^{3+}$ ions’ de Gennes factor (often referred to as DG factor) which quantifies the strength of local moment’s influence on conducting carriers. As suggested in s-wave superconductors, the presence of local magnetic moment tends to destabilize the bonding of spin singlet Cooper pairs. However, we find the linear DG scaling to be totally broken down in our double-doped Eu$_{1-x}$Sr$_x$Fe$_{2-y}$Co$_y$As$_2$ system. It resembles the result in Dy-doped HoNi$_2$B$_2$C system, suggesting potential scattering from collective magnetic excitations (magnons) rather than conventional long range magnetic order in terms of RKKY model. According to the calculations, FM state is always energetically favored than superconducting state at low temperature. On the other hand, if we take into consideration possible emergence of vortices, the advantageous energy state might change with respect to the density of the vortices like in ErNi$_2$B$_2$C and TmNi$_2$B$_2$C systems, suggesting a stir among the conventional ground state and excited states nearby.

In this Letter, we carefully measured the transport properties for EuFe$_{2-x}$Co$_x$As$_2$ and Eu$_{1-x}$Sr$_x$Fe$_{2-y}$Co$_y$As$_2$ single crystals. Co-doping suppresses the SDW transition and induces superconducting transition. In contrast to the case of BaFe$_{2-x}$Co$_x$As$_2$ system, a resistivity reentrance is observed at the temperature corresponding to the antiferromagnetic ordering of Eu$^{3+}$ ions and no zero resistivity is achieved in EuFe$_{2-x}$Co$_x$As$_2$ system. The resistivity reentrance can be completely suppressed by H due to metamagnetic transition from antiferromagnetism to ferromagnetism for Eu$^{2+}$ spins induced by H. These results suggest that the antiferromagnetism destroys the superconductivity, while the ferromagnetism can coexist with the superconductivity. The coexistence of superconductivity and ferromagnetism denotes curious property of p-wave superconductors.

We thank professor Lixin He and Yuwei Cui for helpful discussion in terms of first principle calculation. This work is supported by the Nature Science Foundation of China and by the Ministry of Science and Technology of China (973 project No: 2006CB601001) and by National Basic Research Program of China (2006CB922005).

---

[1] V. L. Ginzburg, Sov. Phys. JETP 4, 153 (1957).
[2] W. Baltensperger and S. Strasler, Z. Phys. B 1, 20 (1963).
[3] W. A. Fertig, D. C. Johnston, L. E. Delong, R. W. McCallum, M. B. Maple, B. T. Mattias, Phys. Rev. Lett. 38, 987 (1977).
[4] W. Ishikawa, Fisher, Solid State Commun. 23, 37 (1977).
[5] C. Bernhard, J. L. Tallon, Ch. Niedermayer, Th. Blassius, A. Golnik, E. Bruecher, R. K. Kremer, D. R. Noakes, C. E. Stronach, E. J. Ansald, Phys. Rev. B 59, 14099 (1999).
[6] H. Eisaki, H. Takagi, R. J. Cava, B. Batlogg, J. J. Krajewsky, W. F. Peck, Jr., K. Mizuhashi, J. O. Lee, S. Uchida et al., Phys. Rev. B 50, 647 (1994).
[7] Paul C. Canfield, Peter L. Gammel, David J. Bishop, Physics Today 51, 10 (1998).
[8] Y. Kamihara, T. Watanabe, M. Hirano, H. Hosono, J. Am. Chem. Soc. 130, 11 (2008).
[9] X. H. Chen, T. Wu, G. Wu, R. H. Liu, H. Chen, D. F. Fang, Nature, 453, 761-762 (2008).
[10] C. Cruz, Q. Huang, J. W. Lynn, J. Y. Li, W. Ratcliff II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang and P. C. Dai, Nature, 453, 899 (2008).
[11] Q. Huang, Y. Qiu, W. Bao, M. A. Green, J. W. Lynn, Y. C. Gasparovic, T. Wu, G. Wu and X. H. Chen, Phys. Rev. Lett., 101, 257003 (2008).
[12] T. Wu, G. Wu, H. Chen, Y. L. Xie, R. H. Liu, X. F. Wang, X. H. Chen, Journal of Magnetism and Magnetic Materials (in press), arXiv:0808.2277.
[13] Shuai Jiang, Yongkang Luo, Zhi Ren, Zengwei Zhu, Cao Wang, Xiangfan Xu, Qian Tao, Guanghan Cao, Zhan Xu, New J. Phys, 11 025007 (2009).
[14] M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006 (2008).
[15] H. Chen, Y. Ren, Y. Qiu, Wei Bao, R. H. Liu, G. Wu, T. Wu, Y. L. Xie, X. F. Wang, Q. Huang, and X. H. Chen, Europhys. Lett. 85, 17006 (2009).
[16] X. F. Wang, T. Wu, G. Wu, R. H. Liu, H. Chen, Y. L. Xie, X. H. Chen, New J. Phys. 11, 045003(2009).
[17] X. F. Wang, T. Wu, G. Wu, H. Chen, Y. L. Xie, J. J. Ying, Y. J. Yan, R. H. Liu, X. H. Chen, Phys. Rev. Lett. 102, 117005(2009).
[18] B. K. Cho, P. C. Canfield, D. C. Johnston, Phys. Rev. Lett. 77, 163(1996).
[19] A. Amici, P. Thalmeier, P. Fulde et al., Phys. Rev. Lett. 84, 1800(2000).
[20] H. S. Greenside, E. I. Blount, C. M. Varma, Phys. Rev. Lett. 49, 49(1981).
[21] P. C. Canfield, S. L. Bud’ko, B. K. Cho, Physica C 262, 249-254(1996).
[22] U. Gasser, P. Allenspach, A. Furrer, A.M. Mulders, J. Alloys Comp. 275 587C590(1998).
[23] Dai Aoki, Andrew Huxley, Eric Ressouche, Daniel Braithwaite, Jacques Flouquet, Jean-Pascal Brison, Elsa Lhotel, Carley Paulsen, Nature 413 613-616(2001).
[24] I. A. Luk’yanchuk, V. P. Mineev, JETP Lett. 44, 233-236 (1986).