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Evidence for competing magnetic and superconducting phases in superconducting $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ single crystals

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Abstract
In single crystals of $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$, Co doping suppresses spin-density wave (SDW) ordering and induces a superconducting transition. A resistivity reentrance due to the antiferromagnetic ordering of Eu^{2+} spins is observed, indicating the competition between antiferromagnetism (AFM) and superconductivity (SC). It is striking that the resistivity reentrance can be completely suppressed by a small magnetic field due to a field-induced metamagnetic transition from AFM to ferromagnetism (FM). The resistivity reentrance can also be suppressed by the substitution of Eu^{2+} ions with nonmagnetic Ba^{2+}/Sr^{2+} to completely destroy the AFM ordering. These results indicate that the AFM order appears destructive to SC, while FM can coexist with the superconductivity. Further we find that magnon excitation exists in AFM ordering and can be suppressed by an applied field. Coexistence of SC from the FeAs layer and the inner field produced by the ferromagnetic Eu^{2+} layer suggest a possible p-wave component in the superconducting order parameter.

(Some figures in this article are in colour only in the electronic version)

Interplay between magnetism and superconductivity has long been a fundamental issue where rich physical phenomena emerge. Pairing symmetry, superconducting phase diagram and gap energy all closely rely on a sample's magnetic property. Theoretical work predicted that long range FM would greatly damage SC, while AFM could coexist with SC to some extent [1, 2]. Indeed, SC is found to be moderately robust coexisting with AFM in traditional magnetic superconductors like RMo_6O_8 and RRh_4B_4 (R = magnetic rare earth ions) [3, 4], yet fragile with FM. However, coexistence of FM and SC was observed in the heavy fermion system UGe_2 [5] and the high-T_c cuprate superconductor $RuSr_2GdCu_2O_8$ [6], where local moments were located far from the conducting plane. Apart from all those mentioned above, one of the most typical families of magnetic superconductor is RNi_2B_2C, where R = Ho, Er, Tm, Yb, Lu [7, 8], with $ThCr_2Si_2$-type structure. In these materials, a resistivity reentrance below T_c was observed due to the AFM ordering of R ions. Therefore, the interaction between superconductivity and magnetic ordering is still puzzling.

The iron-based superconductors [9, 10] are the second family of high T_c superconductors and an AFM SDW ordering of Fe^{2+} is widely observed [11, 12]. In the parent compounds AFe_2As_2 (A = Ba, Sr, Ca, and Eu), Ba^{2+}, Sr^{2+} and Ca^{2+} are

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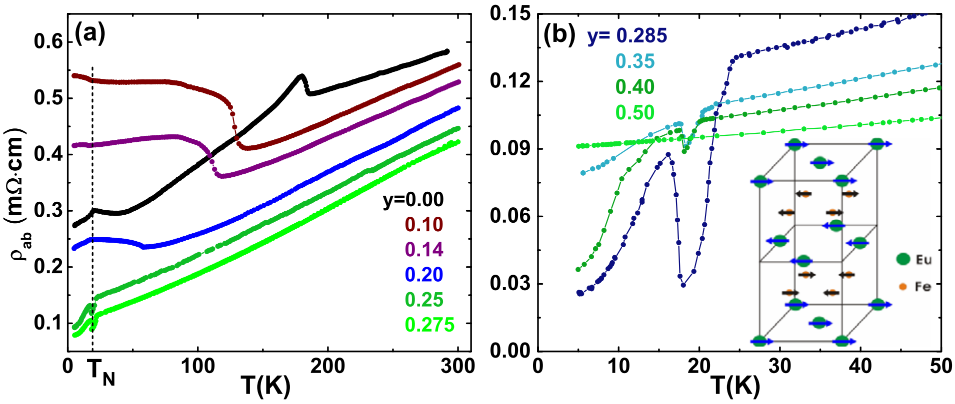

Figure 1. Temperature dependence of in-plane resistivity for the $EuFe_{2-y}Co_yAs_2$ single crystals. (a) $y = 0$, 0.1, 0.14, 0.2, 0.25 and 0.275 from 5 to 300 K; (b) $y = 0.285$, 0.35, 0.4 and 0.5 from 5 to 40 K. Note that all the dips in resistivity correspond to the same temperature as T_N in parent $EuFe_2As_2$. Inset of (b) is the A-type AFM structure of $EuFe_2As_2$ confirmed by neutron scattering result [21].

nonmagnetic ions, while Eu^{2+} is magnetic with $s = 7/2$. An A-type AFM ordering of Eu^{2+} ions occurs at about 17 K. As illustrated by the inset of figure 1(b), the A-type AFM structure for the Eu^{2+} sublattice is proposed by Wu *et al* and Jiang *et al* [13, 14], where local moments align collinearly to form strong FM order within the *ab* plane and weak AFM order along the *c* axis. The interlayer AFM coupling can be tuned to FM coupling by a small magnetic field. $EuFe_2As_2$ is a fantastic system to study the interplay between magnetism and SC because the AFM and FM can be manipulated just by a small external field, so that the AFM and FM can occur in the same superconductors [13, 14]. To our knowledge, $EuFe_2As_2$ is the only system to offer a chance to study the interaction between AFM/FM and SC in iron-based compounds. The Ba(Sr)122 system can be considered as a reference material, where the SC can be induced by substitution of K for Ba or Co doping on the Fe site [15–17]. Here we report intriguing results that the AFM acts negatively on SC, while the FM can coexist with SC in the iron-based high-T_c superconductors. These results are essentially significant in understanding the interaction between SC and AFM/FM. We systematically study the interaction between SC and magnetism in the $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ system. It is found that a resistivity reentrance due to the AFM ordering of Eu^{2+} is observed below T_c, indicating the competition between AFM and SC. Strikingly, the resistivity reentrance can be completely suppressed by an external field because of the field-induced metamagnetic transition from AFM to FM for Eu^{2+} spins. Evidence shows that this process is accompanied by the quenching of abnormal magnon excitation, and the magnon excitation could be the cause for the destruction of the superconducting state.

The single crystals of $EuFe_{2-y}Co_yAs_2$ and $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ were synthesized via the conventional self-flux method [18]. Figure 1 presents the resistivity as a function of temperature for the underdoped, the optimal doped ($x = 0.285$) and overdoped crystals of $EuFe_{2-y}Co_yAs_2$. In the underdoped region, the resistive behavior largely resembles that of $BaFe_{2-y}Co_yAs_2$ in terms of high temperature features [17]. The only difference happens at about 17 K, where a dopant independent kink emerges, corresponding to the AFM ordering established in Eu^{2+} layers. Further Co doping introduces a superconducting transition around 22 K, but subsequently a resistance reentrance shows up just below the AFM ordering temperature, so that no zero resistivity is observed with the temperature down to 2 K in the $EuFe_{2-y}Co_yAs_2$ system. This is totally different from that observed in the $BaFe_{2-y}Co_yAs_2$ system. It suggests that introduction of only Co^{2+} into the FeAs layer fails to get SC with zero resistance due to the AFM ordering of Eu^{2+} in the $EuFe_{2-y}Co_yAs_2$ system. The optimally doped single crystal with $y = 0.285$ shows a sharp superconducting transition in resistivity by ~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* [13, 14], Eu^{2+} ions in $EuFe_2As_2$ show metamagnetism from AFM to FM at low temperature. In order to study how to interact between SC and AFM/FM, we measured the resistivity, specific heat and susceptibility under different fields for the optimally doped sample $EuFe_{1.715}Co_{0.285}As_2$. As shown in figure 2(a), the resistivity reentrance is continuously suppressed both in terms of intensity and temperature with increasing field applied within the *ab* plane. When the field reaches 1 T, the resistivity reentrance is completely suppressed, accompanied by field-induced suppression of the superconducting transition. Figure 2(b) presents the resistivity data measured with a field applied along the *c* axis for the same sample. Unlike the case in figure 2(a), the resistivity reentrance does not exhibit a conspicuous down-shift with increasing magnetic field, and the only change is the closing of the gap in resistivity except for the expected down-shift of T_c induced by an applied field. These results are consistent with the results of susceptibility as shown in figures 2(c) and (d). This is because the spins align within the *ab* plane as shown in the inset of figure 1(b), and the field applied along the *c* axis has less effect on the magnetic ordering of Eu^{2+} relative to the field applied within the *ab* plane. Susceptibility and specific heat measurements further confirm the AFM transition at 17 K. In figure 2(c), the Néel temperature monotonically decreases with increasing field. The similar suppression of the peak in specific heat could

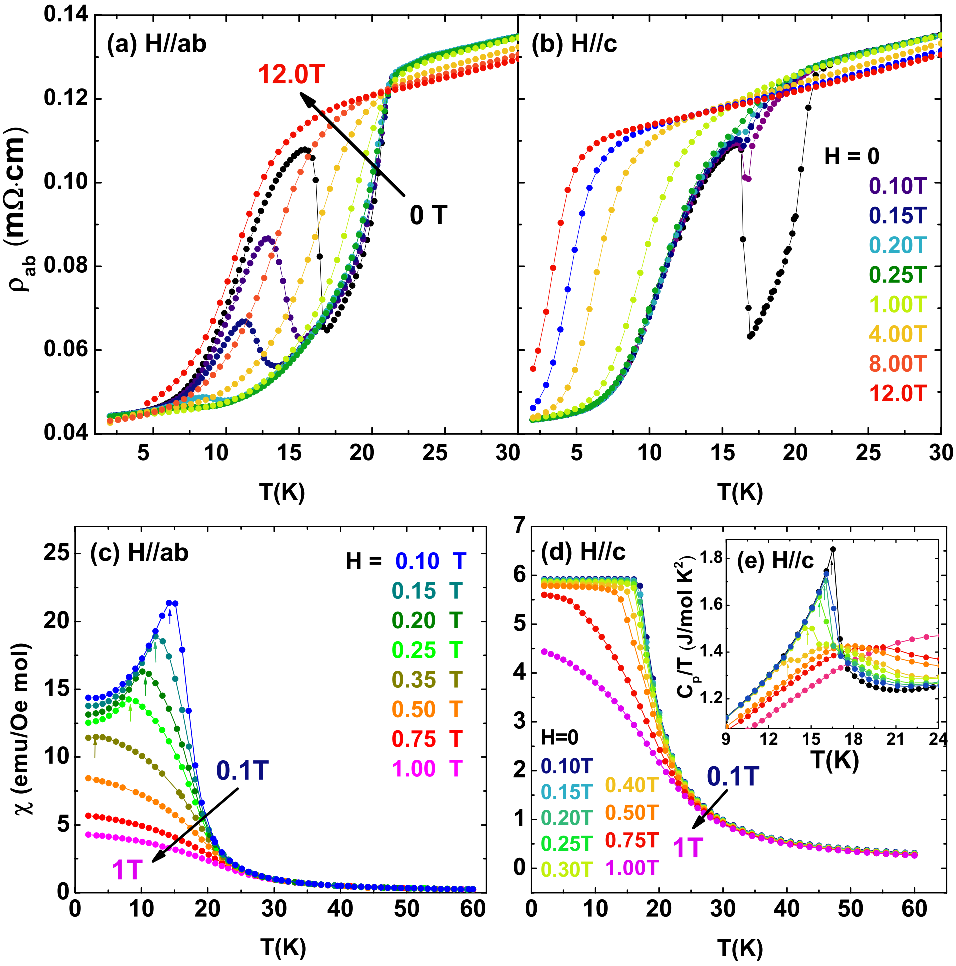

Figure 2. Temperature dependence of resistivity and susceptibility under different magnetic fields for optimal doped single-crystal $EuFe_{1.715}Co_{0.285}As_2$. (a) and (c) H applied within *ab* plane; (b) and (d) H applied along c axis; (e) specific heat under different H applied along c axis from 9 to 24 K.

be observed in figure 2(e). After the external field 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 [13]. These results give strong evidence that a resistivity reentrance arises from the AFM ordering of Eu^{2+} spins, indicating the competition between AFM and SC. It is striking that the resistivity reentrance can be completely suppressed by an external field due to the field-induced metamagnetic transition from AFM to FM for Eu^{2+} spins. All the above measurements on $EuFe_{2-y}Co_yAs_2$ have contributed to the picture that the spins of Eu^{2+} ions tend to establish an AFM order around 17 K, and are easily tuned to FM order with a small field. Such metamagnetism of Eu^{2+} ions provides a good system to study the intriguing interaction between AFM/FM and SC. AFM appears to have strongly counteracted SC, while FM could coexist rather at ease with the SC. It should be noticed that the superconductivity (zero resistivity and Meissner effect are both observed in further Sr-doped samples as will be demonstrated below) originates from the FeAs layer while FM order is established among Eu^{2+} ions. However, considering the Eu^{2+} ions' large local moment and vicinity to the FeAs layer, the molecular field from a classical viewpoint could still exert significant impact on conducting carriers in a ferromagnetic way, resembling the Ru ions' role in the $RuSr_2GdCu_2O_8$ system [6].

In order to make sure that it is AFM from the Eu^{2+} sublattice that destroys SC, we choose to partially substitute Eu^{2+} with nonmagnetic Ba^{2+}/Sr^{2+}. As expected, superconductivity shows up at 23 K as shown in figure 3(a) for the Ba-and Co-doped crystals, T_c being nearly the same as that in $BaFe_{1.8}Co_{0.2}As_2$ [17]. Figure 3(b) clearly demonstrates the evolution in resistivity in the $Eu_{1-x}Sr_xFe_{1.715}Co_{0.285}As_2$ 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 $x = 0.3$, the 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 a stable superconducting phase can be achieved for the crystal with $x = 0.5$. As a confirmation of the existence of metamagnetism in the Sr-doped system, figure 4(a) shows the overlapped FC and ZFC susceptibility in the Sr-doped sample $Eu_{0.75}Sr_{0.25}Fe_{1.715}Co_{0.285}As_2$, where the AFM peak behaves in an almost identical way to that in a non-Sr-doped system except for low Néel temperature. This behavior is similar to that observed in figure 2(c), indicating that

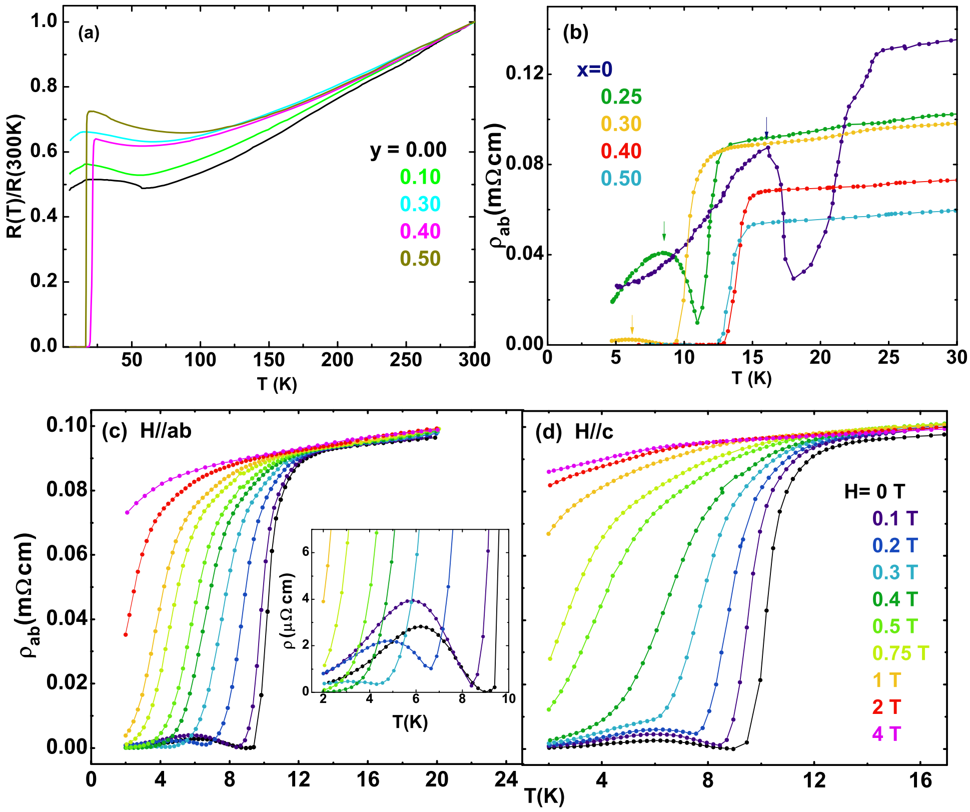

Figure 3. Temperature dependence of resistivity (a) for single-crystal $Eu_{1-x}Ba_xFe_{1.715}Co_{0.285}As_2$ and (b) for single-crystal $Eu_{1-x}Sr_xFe_{1.715}Co_{0.285}As_2$. Temperature dependence of resistivity under different magnetic fields for $Eu_{0.7}Sr_{0.3}Fe_{1.715}Co_{0.285}As_2$ single crystal, (c) *H* applied within *ab* plane; (d) *H* applied along *c* axis.

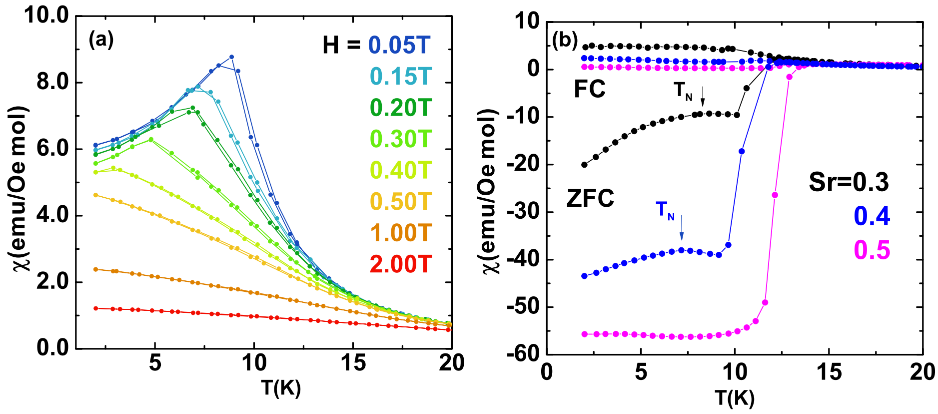

Figure 4. Temperature dependence of susceptibility with zero-field cooling and field-cooling, respectively, (a) for $Eu_{0.75}Sr_{0.25}Fe_{1.715}Co_{0.285}As_2$ under different magnetic fields. (b) For $Eu_{1-x}Sr_xFe_{1.715}Co_{0.285}As_2$ single crystals with $x = 0.3$, 0.4 and 0.5, respectively. It should be pointed out that in Meissner state the hump rightly corresponds to the resistivity reentrance shown in figures 3(c) and (d).

a metamagnetism transition occurs in Sr-doped samples. Figures 3(c) and (d) present resistivity behavior under different fields applied within the *ab* plane and along the *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 field applied within the *ab* plane. However, we didn't observe the same trend with magnetic field applied perpendicular to the *ab* plane. This could be understood in terms of the anisotropic magnetic structure and exchange integration within or between the Eu^{2+} layers. In the $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ system, the AFM appears to be more destructive to SC, while the FM can coexist with SC. It should be pointed out that such a metamagnetic transition from AFM to FM is induced by a small field. Therefore, the observed behavior here is intrinsic. Figure 4(b) further proves the bulk superconductivity emerging in further Sr-doped samples. It should be emphasized that in the Meissner state, there still exists the remnant of an AFM transition as shown in

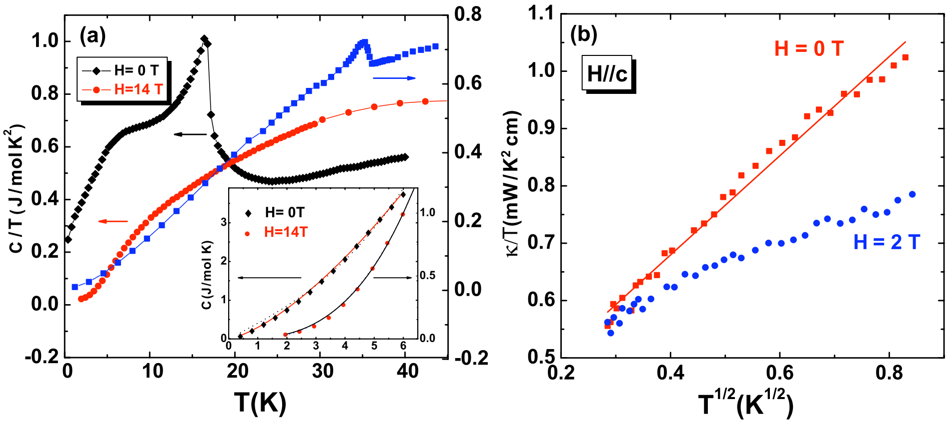

Figure 5. (a) Specific heat of $EuFe_{1.715}Co_{0.285}As_2$ (black diamond for $H = 0$ and red circle for $H = 14$ T) and $Ba_{0.65}K_{0.35}Co_2As_2$ (blue square for $H = 0$). Inset: low temperature fitting of $EuFe_{1.715}Co_{0.285}As_2$'s specific heat reveals evidence for strong magnon excitation. (b) Heat conductivity of $EuFe_{1.715}Co_{0.285}As_2$ which exhibits corresponding $T^{1.5}$-dependent behavior with specific heat measurement.

figure 4(b). Moreover, the humps in figure 4(b) are consistent with the resistivity reentrance due to an AFM transition shown in figure 3(c).

In the RNi_2B_2C system, a pronounced resistivity reentrance was observed when the AFM transition in RC layers happens below T_c [7], similar to the observation in the $Eu_{0.7}Sr_{0.3}Fe_{1.715}Co_{0.285}As_2$ system. If the AFM transition temperature (T_N) is lower than T_c, T_c could be negatively scaled by the de Gennes (DG) factor for the R^{3+} ions. This quantifies the strength of the local moment's influence on conducting carriers [19]. As suggested in s-wave superconductors, the presence of a local magnetic moment tends to destabilize the bonding of spin singlet Cooper pairs [7]. However, we find the linear DG scaling to be totally broken down in our double-doped $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ system. It resembles the result in Dy-doped $HoNi_2B_2C$ system [20], suggesting potential scattering from collective magnetic excitations (magnons) rather than conventional long range magnetic order in terms of the RKKY model.

Further experiments conducted on this system, including specific heat and heat conductivity shown in figures 5(a) and (b), suggest strong magnon excitation in the AFM ordering. In figure 5(a) and its inset, the specific heat is fitted by the formula $C_p = C_1T + C_{1.5}T^{1.5} + C_3T^3$ contributed from electrons, magnons and phonons (red curve) [22]. The fitting yields $C_1 = 0.048$ J mol^{-1} K^{-2}, $C_{1.5} = 0.348$ J mol^{-1} K$^{-2.5}$ and $C_3 = 0.000\,36$ J mol^{-1} K^{-4}, where $C_{1.5}$ is much larger than C_1 and C_3. The data are also fitted by $C_p = C_1T + C_3T^3$ without considering the magnon contribution (dotted curve). As shown in the inset of figure 5(a), the fitting with the magnon term is much better than that without considering the magnon contribution. It is more important that the fitting without the magnon term results in an unconventional large effective mass of the electron. It indicates there exists a strong FM magnon excitation at this temperature. Furthermore, figure 5(b) demonstrates the magnon excitation from thermal conductivity measurements. The thermal conductivity is also dominated by the $T^{1.5}$ term. Moreover, an external magnetic field suppresses the contribution of magnons to specific heat and thermal conductivity. As shown in figure 5(a), the magnitude of the specific heat for Co-doped Eu122 crystals is largely suppressed by an applied field of 14 T at low temperature, and is nearly the same as that for the nonmagnetic superconducting K-doped Ba122 system. As a matter of fact, the suppression of heat capacity rightly comes from the decrease in the contribution of magnons since spins can hardly flip under a high external field. In addition, the zero-field thermal conductivity follows the $T^{1.5}$ behavior as shown in figure 5(b), while the thermal conductivity under 2 T deviates from the $T^{1.5}$ behavior. It further indicates thermal conductivity is dominated by magnon excitation, which is suppressed by an external field. This once more braces the assumption that magnetism from Eu ions gives rise to that aforementioned exotic coexistence and competition between SC and FM/AFM. Provided that an external field can obviously suppress the magnetic elementary excitation which strongly destroys superconductivity, we reckon this to be one possible explanation for the coexistence of SC and FM in this system.

In this paper, we carefully measured the transport properties for $Eu_{1-x}Sr_xFe_{2-y}Co_yAs_2$ single crystals. Co doping suppresses the SDW transition and induces the superconducting transition. In contrast to the case of the $BaFe_{2-y}Co_yAs_2$ system, a resistivity reentrance is observed at the temperature corresponding to the antiferromagnetic ordering of Eu^{2+} ions and no zero resistivity is achieved in the $EuFe_{2-y}Co_yAs_2$ system. The resistivity reentrance can be completely suppressed by an external field due to a field-induced metamagnetic transition from antiferromagnetism to ferromagnetism for Eu^{2+} spins. These results suggest that the antiferromagnetism destroys the superconductivity, while the ferromagnetism can coexist with the superconductivity. Coexistence of SC from the FeAs layer and the inner field produced by the ferromagnetic Eu^{2+} layer suggests a possible p-wave component to the order parameter. Specific heat and thermal conductivity measurements provide evidence that magnon excitation can be suppressed by a magnetic field and the magnon excitation could be the reason to suppressed superconductivity.

Acknowledgments

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