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Phase diagram of superconductivity and antiferromagnetism in single crystals of Sr(Fe1–xCox)2As2 and Sr1–yEuy(Fe0.88Co0.12)2As2

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Abstract
We report magnetic susceptibility, resistivity, and heat capacity measurements on single crystals of the Sr(Fe1−xCox)2As2 and Sr1−yEuy(Fe0.88Co0.12)2As2 series. The optimal Co concentration for superconductivity in Sr(Fe1−xCox)2As2 is determined to be x~0.12. On the basis of this, we grew members of the Sr1−yEuy(Fe0.88Co0.12)2As2 series so as to examine the effects of well-defined, local magnetic moments on the superconducting state. We show that superconductivity is gradually suppressed by paramagnetic Eu2+ doping and coexists with antiferromagnetic ordering of Eu2+ as long as Tc>TN. For y ≥ 0.65, TN crosses Tc and the superconducting ground state (as manifested by zero resistivity) abruptly disappears with evidence for competition between superconductivity and local moment antiferromagnetism for y up to 0.72. It is speculated that the suppression of the antiferromagnetic fluctuations of the Fe sublattice by coupling to the long-range order of the Eu2+ sublattice destroys bulk superconductivity when TN>Tc.

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Phase diagram of superconductivity and antiferromagnetism in single crystals of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$

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We report magnetic susceptibility, resistivity, and heat capacity measurements on single crystals of the Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ series. The optimal Co concentration for superconductivity in Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is determined to be $x \sim 0.12$. On the basis of this, we grew members of the Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ series so as to examine the effects of well-defined, local magnetic moments on the superconducting state. We show that superconductivity is gradually suppressed by paramagnetic Eu$^{2+}$ doping and coexists with antiferromagnetic ordering of Eu$^{2+}$ as long as $T_c > T_N$. For $y > 0.65$, $T_N$ crosses $T_c$ and the superconducting ground state (as manifested by zero resistivity) abruptly disappears with evidence for competition between superconductivity and local moment antiferromagnetism for $y$ up to 0.72. It is speculated that the suppression of the antiferromagnetic fluctuations of the Fe sublattice by coupling to the long-range order of the Eu$^{2+}$ sublattice destroys bulk superconductivity when $T_N > T_c$.

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I. INTRODUCTION

The interplay between superconductivity (SC) and magnetism has been of a long-standing interest in condensed matter physics. SC and magnetism were originally considered to be mutually exclusive in conventional superconductors because magnetism breaks the time-reversal symmetry of the singlet Cooper pairs. The influence of paramagnetic impurities on SC was first studied theoretically by Abrikosov and Gor’kov (AG). It was shown that SC is drastically suppressed by dilute paramagnetic impurities. Early experimental investigations were limited to superconducting systems without long-range magnetic order. The coexistence of SC and long-range magnetism was realized in several families of ternary and quaternary rare-earth compounds discovered later, also referred to as magnetic superconductors, e.g., RMo$_6$S$_8$, RRh$_4$B$_4$, and RNi$_2$B$_2$C. In these compounds, the localized magnetic moment antiferromagnetism for Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ series so as to examine the effects of well-defined, local magnetic moments on the superconducting state. We show that superconductivity is gradually suppressed by paramagnetic Eu$^{2+}$ doping and coexists with antiferromagnetic ordering of Eu$^{2+}$ as long as $T_c > T_N$. For $y > 0.65$, $T_N$ crosses $T_c$ and the superconducting ground state (as manifested by zero resistivity) abruptly disappears with evidence for competition between superconductivity and local moment antiferromagnetism for $y$ up to 0.72. It is speculated that the suppression of the antiferromagnetic fluctuations of the Fe sublattice by coupling to the long-range order of the Eu$^{2+}$ sublattice destroys bulk superconductivity when $T_N > T_c$.

The parent compounds Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ series for which the magnetic and structural transition was suggested to disappear abruptly prior to the emergence of SC, the 122 series show a gradual suppression of the AF (ortho) transition, which coexists with SC for a range of dopings. For the Co-doped Ba-122 series, neutron scattering shows a suppression of the magnetic order parameter on entering the superconducting state, indicating strong coupling between AF and SC. In addition, both $\mu$ SR (Refs. 19 and 20) and $^{75}$As NMR (Ref. 21) measurements unambiguously indicate that AF order is present in all of the sample volumes when the sample is in the superconducting state, i.e., that the magnetic order and SC coexist homogeneously at the atomic scale. Whereas Fe-based AF coexists with SC and Fe-based AF fluctuations may well be vital to FeAs-based superconductors, a systematic study of effects of well-defined, local magnetic moments on this SC is lacking. Starting from optimally Co-doped SrFe$_2$As$_2$, we can have Eu$^{2+}$ substituting for Sr$^{2+}$ without introducing extra electrons and holes and assess the sensitivity of this SC to the large $J = S = 7/2$ local moment.

The Eu end compound EuFe$_2$As$_2$ exists as an isostructural member of the 122 series. Therefore, a continuous substitution can be expected between EuFe$_2$As$_2$ and EuFe$_2$As$_2$. EuFe$_2$As$_2$, in addition to the AF order of the iron sublattice at about 189 K, exhibits an $A$-type AF order of Eu$^{2+}$ ions at 19 K. On suppression of the AF order of iron with pressure or Co doping, the onset of SC was observed, which was then followed by a resistive re-entrance attributed to the magnetic order of Eu$^{2+}$. Given the sensitivity of the 122 compounds to strain and pressure, we choose SrFe$_2$As$_2$ as host, owing to the similar size of Sr$^{2+}$ (118 pm) and Eu$^{2+}$ (117 pm), so as to minimize the steric effects of the doping.

To perturb the SC of the Sr 122 phase by isoelectronic substitution of Eu and establish phase diagrams systematically using the same growth technique for Co and Eu doping, the phase diagram of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ as a function of Co substitution is constructed first. The optimal Co doping level of $x \sim 0.12$ is then kept the same across the whole range of...
Eu doping. We present the magnetic susceptibility, resistivity, and heat capacity measurements on Sr_{1−x}Eu_x(Fe_{1−y}Co_y)2As_2. Superconductivity of the optimally Co-doped SrFe_2As_2 is suppressed gradually by Eu doping (0 ≤ y < 0.43), and crosses over with coexistence of SC and Eu-based AF (0.43 ≤ y ≤ 0.60) with T_c increasing linearly with y. For y ≥ 0.65, T_c cuts across the T_N line and SC suddenly disappears, leaving just the Eu\(^{2+}\) AF-ordered state. An initial study of EuFe_2As_2 doped with both Sr and Co was recently published, but it used samples with nominal doping values and focused on the Eu-rich side.\(^{32}\) We will compare the results of our systematic study with Ref. \(^{32}\) in the discussion section.

II. EXPERIMENT

Single-crystal samples of both Sr(Fe\(^{1−x}\)Co\(^x\))\(_2\)As_2 and Sr\(_{1−y}\)Eu\(_y\)(Fe\(_{0.88}\)Co\(_{0.12}\))\(_2\)As_2 were grown via a self-flux method.\(^{16,33}\) The FeAs and CoAs precursors were first synthesized by solid-state reaction. Elemental Sr and Eu were mixed with FeAs and CoAs in the stoichiometry of 1:4 − 4x : 4x and 1 − y : y : 3.44 : 0.56, respectively, in an alumina crucible and sealed into an amorphous silica tube. The sealed ampoule was heated to 1180 °C and then cooled slowly to 1000 °C; finally, the excess liquid flux was decanted.\(^{33}\) The as-grown crystals were annealed under a static Ar atmosphere at 500 °C for 24 hours (as discussed below in Sec. III).\(^{34}\)

Powder x-ray diffraction (XRD), with Si standard, was performed using a Rigaku Miniflex x-ray diffractometer employing Cu Ka radiation (λ = 1.5418 Å). The lattice parameters were refined by RIETICA software.\(^{35}\) Chemical composition was determined by wavelength dispersive x-ray spectroscopy (WDS) in a JEOL JXA-8200 electron microscope. The actual composition of the single crystals was taken as the average of 10 spots measured on the crystal, and the error bar was taken as the standard deviation of the 10 values.

Magnetic susceptibility was measured in a Quantum Design MPMS, SQUID magnetometer. The in-plane ac resistivity was measured by a standard four-probe method using an LR-700 resistance bridge with an excitation of 60 μV on samples of typical size 3 mm × 2 mm × 0.2 mm. Electrical contacts were made using Dupont 4929N silver paint. Heat capacity data were collected using a Quantum Design PPMS.

All the samples were found to slowly degrade in air. Over a period of four months, a ferromagnetic background on the order of 10\(^−2\) emu/mol develops, although no obvious change in appearance of the crystal and no impurity phase in powder XRD pattern can be observed. Elemental analysis indicates significant presence of oxygen in the surface layer of the aged samples, implying the formation of oxides. In addition, the superconducting transition of the aged sample broadens and T_c decreases as measured by low-field magnetization. Therefore, all the measurements reported in this paper were performed shortly after the samples were prepared. It should be noted that, whereas the Sr-based 122 compounds are known to be susceptible to chemical changes as well as strain,\(^{27}\) we observed no sample quality change over time in the well-studied Ba(Fe\(_{1−x}\)Co\(_x\))\(_2\)As_2 samples.

III. RESULTS AND DISCUSSIONS

A. Sr(Fe\(_{1−x}\)Co\(_x\))\(_2\)As_2

The results of elemental analysis and lattice parameter determination from the powder x-ray measurements on Sr(Fe\(_{1−x}\)Co\(_x\))\(_2\)As_2 are shown in Fig. 1. The nearly linear dependence in Fig. 1(a), with a slope of 0.94, indicates good agreement between the actual Co concentration x\(_{WDS}\) and the nominal concentration x\(_{nominal}\). The compositional spread over a wide area on the sample surface for each concentration is less than 0.02. These results demonstrate the relative homogeneity of the Co doping in the single-crystal samples. Figures 1(b) and 1(c) show the lattice parameters a and c as well as the c/a ratio and unit-cell volume as a function of x\(_{WDS}\). The parameters c and c/a change linearly with x\(_{WDS}\) and the values are in good agreement with the previous report.\(^{17}\) By substitution of Co for Fe, the lattice is changed more along the c axis than in the ab plane. The lattice parameter c decreases by 0.6% (0.074 Å) for x = 0.17, whereas the lattice parameter a increases by only about 0.2% (0.007 Å). The random error of lattice parameter determined by our Miniflex x-ray diffractometer is about.
FIG. 2. (Color online) The annealing effect for pure SrFe$_2$As$_2$: as-grown sample (black squares), annealed sample (red circles).

0.02% (∼0.0008 Å for the $a$ lattice parameter), which is about the same order as the average deviation from a linear variation (∼0.0013 Å). Thus, lattice parameters can be regarded to vary linearly with $x_{\text{WDS}}$, within experimental errors, in accordance with Vegard’s law.

Annealing can have a clear effect on samples and has been shown to remove extrinsic effects associated with strain-induced defects.34 As shown in Fig. 2, the magnetic and structural transition of pure SrFe$_2$As$_2$ is increased from 192 K for the as-grown sample to 201 K for the annealed sample, which is very close to the previous reported values for polycrystalline (205 K) (Ref. 37) and Sn-flux-grown single crystalline (198 K) (Ref. 38) SrFe$_2$As$_2$. Based on these observations, our samples are heat treated under the conditions described in Sec. II.

The magnetic susceptibility for $H \parallel ab$ of the Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series was measured in a magnetic field of 10 kOe for $x \leq 0.07$ [Fig. 3(a)]. The parent compound SrFe$_2$As$_2$ manifests a sharp drop at 201 K in magnetic susceptibility, due to the magnetic and structural transition.37,38 With increasing Co doping, this transition is suppressed to lower temperature and becomes undetectable for $x > 0.07$. For $0.07 \leq x \leq 0.17$, SC is induced and is manifested in low-field ($H = 100$ Oe), zero-field-cooled (ZFC), and field-cooled (FC) measurements below 20 K [Fig. 3(b)]. The data are compared to $1/4\pi$ to give a rough estimate of the superconducting volume fraction. Although, as discussed in Ref. 39, the FC curves are routinely close to zero in these materials, due to pinning or surface barrier effects, the ZFC curves approaching $-1$ suggest bulk SC. The superconducting transitions remain very sharp for $x \geq 0.07$, except for $x = 0.17$, which becomes broad and barely visible, consistent with a $T_c$ reduced to a value close to our base temperature. The transition temperature increases from 7.4 K for $x = 0.07$, maximizes at 14.8 K for $x = 0.117$, and then diminishes to 5.7 K for $x = 0.17$.

Figure 4(a) shows the temperature dependence of the electrical resistivity of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$, normalized to the room-temperature values. Similar to the case of the Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series,16 the magnetic and structural transition of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ manifests itself as a sudden drop for $x = 0$, as an increase in resistivity for $x = 0.042 - 0.07$, and nearly disappears for $x = 0.087$. After the magnetic and structural transition is completely suppressed for $x \geq 0.092$, the series shows featureless, metallic temperature dependence.
FIG. 5. (Color online) Magnetic susceptibility, normalized resistivity, and the temperature derivatives, single peak associated with simultaneous magnetic and structural transition.

and this agrees with the bulk SC observed in magnetic measurements.

To establish the phase diagram for the Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ series, the transition temperatures were inferred in the same manner as used in Ref. 16. $T_c$ from magnetic susceptibility is determined from the intersection of the steepest slope and the linear extrapolation of normal magnetic susceptibility, as shown in Fig. 3(b). Resistive onset and offset of $T_c$ values are inferred from the intersects of the steepest slope with the normal state and zero resistance, respectively, shown in Fig. 4(b).

$T_M/S$ is inferred from the peak of $d(M/H)/dT$ and $d(\rho/\rho(300 \text{ K}))/dT$; data for $x = 0.056$ is shown in Fig. 5 as an example. It is argued by Gillett et al. 40 that only a single, first-order-like transition occurs in the heat capacity of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ with coincidence of magnetic and structural transitions. Our magnetization and resistance data also do not show a discernible splitting between $T_M$ and $T_S$; Fig. 5 further supports this observation.

Based on our magnetization and electrical resistance measurements, the phase diagram of Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ is mapped out in Fig. 6. A superconducting dome is found: SC is first stabilized for $x = 0.07$ at about 7.4 K, reaches a maximum $T_c$ of $\sim 14.5$ K for $x = 0.117$, then decreases to 5.7 K for $x = 0.17$. Our phase diagram is in good agreement with earlier ones. The phase diagram for polycrystalline Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ showed a complete suppression of magnetic and structural transition and appearance of SC at $x_{\text{nominal}} = 0.1$ with the highest $T_c$ of 19 K. 37 The difference between maximum $T_c$ of the polycrystalline and our single crystalline samples is probably due to strain effect. As it has been demonstrated, 25-30 strain can affect Sr-122 profoundly, especially when there is a high surface area to volume fraction (as in powders). Results consistent with our single-crystal Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ work, with highest $T_c$ $\sim 13$ K, were reported by Kasinathan et al. 41 The more recent one based on Sn grown single crystals, having larger density of data points, showed more clearly a coexistence of $T_M/S$ and SC transition for $x = 0.07 \sim 0.09$ and the superconducting dome with optimal $T_c$ of 16 K at $x = 0.10$. 40 The differences between our phase diagram and the published ones, in terms of transition temperature and optimal doping concentration, may be associated with differences in both sample preparation and uncertainties of concentration. For our self-flux grown samples, we can choose Co concentration $x \sim 0.12$, with the highest $T_c$ and suppressed
AF and ortho transition as the starting point for our study of the effects of local moments of FeAs-based superconductor via Eu substitution for Sr.

**B. Sr$_{1-x}$Eu$_x$(Fe$_{1-y}$Co$_y$)$_2$As$_2$**

For our Sr$_{1-x}$Eu$_x$(Fe$_{1-y}$Co$_y$)$_2$As$_2$ series, the Co concentration is kept at $x \sim 0.12$ and the series was doped by Eu for $0 \leq y \leq 1$. Figure 7(a) shows the elemental analysis results for the actual Eu and Co concentrations as a function of nominal Eu concentration. The actual concentration of Eu agrees well with the nominal, with a slope of 1.03, and the Co concentration is essentially constant. The lattice parameters agrees well with the nominal, with a slope of 1.03, and the Co concentration was kept at $0.4\%$ ($0$).

The actual concentration of Eu results for the actual Eu and Co concentrations as a function of nominal Eu concentration. The actual concentration of Eu agrees well with the nominal, with a slope of 1.03, and the Co concentration was kept at $0.4\%$ ($0$).

The in-plane magnetic susceptibility of Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{12}$)$_2$As$_2$ is shown in Fig. 8. Both ZFC and FC curves are measured in a magnetic field of 100 Oe. The data clearly indicate that there are three regions of low-temperature behavior across the series: (i) $0 \leq y \leq 0.34$, where SC is gradually suppressed by Eu doping but remains a simply identifiable transition. (ii) $0.43 \leq y \leq 0.60$, where, in this intermediate range, the Curie-Weiss paramagnetic background due to Eu$^{2+}$ moments gradually becomes large enough to shift the diamagnetic signal to positive values. In addition, a second feature appears and, as $y$ increases, it rises in temperature leading to a double-peak feature, which can be ascribed to the coexistence of SC and lower temperature AF associated with the Eu$^{2+}$ sublattice. The upper transition shows a splitting between ZFC and FC curves consistent with SC. The lower transition of AF origin is present on both ZFC and FC curves at the same temperature, indicated by arrows. These transitions are further confirmed by heat capacity measurements as shown below. (iii) $0.65 \leq y \leq 1$, where clear AF transitions manifest as cusps and $T_N$ continues to increase with Eu$^{2+}$ doping up to 17 K for $y = 1$. It is worth noting that FC and ZFC curves collapse on each other for these higher $y$ values, suggesting long-range antiferromagnetic order, similar to EuFe$_2$As$_2$, instead of other magnetic origins, e.g., spin glass or ferrimagnetic order.

Figure 9(a) shows $M(T)/H$ as a function of temperature measured in various fields for $y = 1$. The cusp initially shifts to lower temperature with higher field and then becomes saturated paramagnetic-like for fields above 4 kOe. Neutron scattering experiments on pure EuFe$_2$As$_2$ revealed that the long-range order of Eu$^{2+}$ is of A-type AF, namely, the Eu$^{2+}$ moments are parallel in the $ab$ plane and antiparallel along the $c$ axis with an ordering wave vector of $k = (0, 0, 1)$. Therefore, the metamagnetic transition for $y = 1$ is most likely due to the spin flip along the field direction between Eu$^{2+}$ layers, similar to EuFe$_2$As$_2$. Our results are in good agreement with the reported magnetic field dependence of $M/H$ for EuFe$_{1.71}$Co$_{0.28}$As$_2$, where metamagnetic transition occurs at a lower field of 3.5 kOe than that of pure EuFe$_2$As$_2$ (8.5 kOe). Because of this metamagnetic transition, the series for $y \geq 0.43$ all show similar field dependence [Fig. 9(b)], i.e., the slope of magnetization changes around 4 kOe and shows a saturation moment of $\sim 7\mu_B$/Eu$^{2+}$ in the high field. For $y = 0.43$ and 0.50, diamagnetic contribution of SC can be seen below 500 Oe. Given the metamagnetic transition, the AF transition temperature $T_N$ was inferred from the cusp of $d(\chi T)/dT$ measured in $H = 100$ Oe.

In Fig. 9(c), we examine the high-temperature behavior of the magnetic susceptibility. Since the Hund’s rule ground state for Eu$^{2+}$ is the same as Gd$^{3+}$ ($7/2$S), there is no spin-orbital coupling and, thus, the crystal field effect is absent and well-defined magnetic moments of Eu$^{2+}$ exhibiting Curie-Weiss law at high temperatures are expected. We are able to estimate the concentration of Eu$^{2+}$ from magnetic measurements by assuming each Eu$^{2+}$ carries an effective magnetic moment of 7.94 $\mu_B$. The magnetic background of Sr(Fe$_{0.88}$Co$_{0.117}$)$_2$As$_2$ in a magnetic field of 10 kOe is subtracted from all the datasets, and the inverse magnetic susceptibility normalized to a fitted Eu concentration $\gamma_M$ is plotted in Fig. 9(c) as a function of...
FIG. 9. (Color online) (a) Field dependence of the temperature-dependent magnetization divided by applied field for \( y = 1 \) for the \( H_{\|ab} \) plane; (b) \( M(H) \) normalized by \( y_{WDS} \); (c) inverse magnetic susceptibility \( H/M \) normalized by the actual Eu\(^{2+} \) concentration \( y_{WDS} \).

The magnetic susceptibility above 100 K is fitted by the Curie-Weiss law:

\[
\chi(T) = \frac{y_M N \mu(Eu^{2+})^2}{3k_B(T - \theta_{CW})} = \frac{7.94^2 y_M}{8(T - \theta_{CW})} \text{ (emu/mol),}
\]

where \( N \) is the Avogadro constant, \( k_B \) is the Boltzmann constant, and \( \theta_{CW} \) is Curie-Weiss temperature. As can be seen in Table I, \( y_{WDS} \) and \( y_M \) agree well with each other and follow the same trend with the nominal concentration. The positive Curie-Weiss temperature is consistent with an overall predisposition to ferromagnetic coupling between Eu\(^{2+} \) moments, at least in the magnetic field of 10 kOe.

The low-temperature (\( T < 20 \) K) heat capacity divided by temperature \( C_p/T \) versus \( T \) of \( \text{Sr}_{1-x}\text{Eu}_x(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2 \), where the solid line indicates \( C_p \sim T^2 \) for a FM magnon contribution. (b) An expanded view for \( 0 \leq y \leq 0.19 \) showing a SC jump, where \( T_c \) is indicated by arrows. Inset shows the isentropic reconstruction of the superconducting transition of \( C_p/T \).

FIG. 10.

and Table I. Results of elemental analysis for \( \text{Sr}_{1-x}\text{Eu}_x(\text{Fe}_{1-x}\text{Co}_x)_2\text{As}_2 \) and the Eu\(^{2+} \) concentration inferred from high-temperature magnetic susceptibility. The Curie-Weiss temperature \( \theta_{CW} \) is compared with the AFM transition temperature \( T_N \).

| \( y_{nominal} \) | 0.05 | 0.1 | 0.15 | 0.2 | 0.3 | 0.4 | 0.45 | 0.5 | 0.55 | 0.6 | 0.7 | 0.8 | 0.9 | 1 |
|------------------|------|-----|------|----|-----|----|------|----|------|----|----|----|----|---|
| \( y_{WDS} \)    | 0.04 | 0.15 | 0.16 | 0.19 | 0.34 | 0.43 | 0.5 | 0.54 | 0.6 | 0.65 | 0.72 | 0.85 | 0.96 | 1 |
| \( y_M \)        | 0.05 | 0.12 | 0.15 | 0.18 | 0.31 | 0.4 | 0.48 | 0.5 | 0.6 | 0.64 | 0.73 | 0.85 | 0.97 | 1 |
| \( \theta_{CW} \) (K) | 2.2  | 4.1  | 4.0  | 4.2  | 5.0  | 9.3  | 9.7 | 10.3 | 11.7 | 13.9 | 14.6 | 18.6 | 19.4 | 20.2 |
| \( T_N \) (K)    | 3.5  | 4.5  | 5.5  | 6.3  | 8.0  | 9.5  | 11.8 | 14.3 | 16.8 |
FIG. 11. (Color online) (a) Normalized electrical resistivity of \(\text{Sr}_{1-y}\text{Eu}_y(\text{Fe}_{0.88}\text{Co}_{0.12})_2\text{As}_2\); (b) low-temperature data showing the superconducting transition; (c) for \(0.85 \leq y \leq 1\), the loss of spin scattering around \(T_N\), where solid lines are the temperature derivatives.

worth noting that the low-temperature \(C_p/T\) below 5 K for \(0.65 \leq y \leq 1\) shows a linear dependence on \(T\), i.e., \(C \propto T^2\). This temperature dependence of heat capacity is consistent with the low-temperature AF magnon excitations of a two-dimensional magnetic lattice.46

Figure 10(b) shows that, starting from the low \(y\) side, SC can be identified as a weak jump for \(0 \leq y \leq 0.19\), but becomes hard to detect for \(y > 0.34\) because of the large background associated with the AF transition. Figure 10(b) inset shows a representative heat capacity jump for \(y = 0\). The SC transition temperature \(T_c\) is inferred by isentropic construction, i.e., the two shaded areas have the same size. For \(y = 0.34\), \(T_c\) is taken as the middle point of the jump.

Figure 11 shows the temperature dependence of the normalized resistivity of \(\text{Sr}_{1-y}\text{Eu}_y(\text{Fe}_{0.88}\text{Co}_{0.12})_2\text{As}_2\). Given the Co-doping level (near optimal), it is not surprising that the series remains metallic and featureless above 20 K. In Fig. 11(b), it can be seen that the superconducting transition temperature is gradually lowered by \(\text{Eu}^{2+}\) doping for \(0 \leq y \leq 0.60\). The transition becomes broader for \(y > 0.19\), e.g., \(\Delta T\) is 4 K for \(y = 0\) and 6 K for \(y = 0.5\). This wide transition is similarly observed in Ni-doped \(\text{SrFe}_2\text{As}_2\).34 For \(y = 0.65\) and 0.72, a resistivity re-entrance is observed as a broad peak below a local minimum in resistivity at 7.8 and 9.8 K, respectively. The minimum coincides with the AF order temperature measured by magnetic susceptibility and heat capacity, indicating that the bulk SC transition is interrupted by AF order. Such incomplete resistive transitions have been observed in \(\text{Sr}_{0.3}\text{Eu}_{0.7}(\text{Fe}_{0.86}\text{Co}_{0.14})_2\text{As}_2\) and \(\text{EuFe}_2\text{As}_2\) under pressure.23,32

The superconducting transition temperature \(T_c\) is inferred in the same way as in Fig. 4. For \(y = 0.65\) and 0.72, only \(T_{\text{onset}}\) is extracted. For \(0.85 \leq y \leq 1\) [Fig. 11(c)], the series remains a normal metal and manifests a very small change in slope at \(T_N\) (corresponding to the peak in \(d\rho/\rho(300 \text{ K})/dT\)), due to the loss of spin disorder scattering. We must note that the change in resistivity at \(T_N\) is very small, even smaller than that of \(\text{EuFe}_2\text{As}_2\).42 This implies very weak coupling between \(\text{Eu}^{2+}\) moments and conduction electrons. Recent detailed transport studies of \(\text{EuFe}_2\text{As}_2\) under high pressure showed that electron scattering due to \(\text{Eu}^{2+}\) has minor contribution to both resistivity and Hall effect, thus consistent with our conclusion.47

C. Analysis and discussion

Based on the transport and thermodynamic measurements, a phase diagram as a function of Eu doping can be constructed and is shown in Fig. 12. Starting from the Eu-rich side of the phase diagram, we can see that \(T_N\) decreases with decreasing Eu content and crosses through the \(T_c\) line, near \(y \sim 0.60\), without any resolvable change in slope (\(dT_n/dy\)). This is fairly standard behavior for an intermetallic compound with a local moment antiferromagnetic phase transition that is being reduced via site dilution with a nonmagnetic ion (i.e., \(\text{Sr}^{2+}\) for \(\text{Eu}^{2+}\)).11,48 Starting from the Sr-rich side of the phase diagram, we can see that, when \(\text{Eu}^{2+}\) is a paramagnetic
impurity, it suppresses SC monotonically but rather weakly. The weakness of the paramagnetic Eu$^{2+}$ as a pair breaker is not unexpected, given the rather weak coupling of the Eu$^{2+}$ moments to the conduction electrons, as most clearly manifested by the small loss of spin-disorder scattering seen in Fig. 11(c). The suppression of $T_c$ by magnetic impurities in a nonmagnetic superconductor has been discussed by Abrikosov and Gor’kov.\textsuperscript{1} The fit to AG theory for data 0 $\leq$ $y$ $\leq$ 0.34 gives a critical concentration $y_c$ = 1.08, implying SC could survive in Eu(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ if the Eu$^{2+}$ sublattice were to remain in the disordered paramagnetic state (which it does not). For 0.43 $\leq$ $y$ $\leq$ 0.60, both SC and AF states are clearly detected. As long as $T_c$ $>$ $T_N$, the advent of AF order does not lead to any re-entrance or other clear features in the $T$ $-$ $\eta_{WDS}$ phase diagram. This is in agreement with early findings that EuFe$_2$As$_2$ becomes a bulk superconductor with $T_c$ $\sim$ 30 K and SC coexists with AFM order with $T_N$ $\sim$ 20 K.\textsuperscript{40} The remarkable feature revealed in Fig. 12 is the sudden disappearance of bulk SC when the $T_N$ line intercepts the $T_c$ line. Superconductivity, as defined by a $\rho$ $=$ 0 state, suddenly disappears for $y$ $\gtrsim$ 0.65. This sudden truncation of the superconducting region is quite remarkable and demands further analysis.

As has been shown in this work and discussed before,\textsuperscript{27,34} the resistivity data associated with pure and doped SrFe$_2$As$_2$ samples is complicated, manifesting superconducting transition temperatures that appear to be higher than those determined by bulk, thermodynamic measurements such as magnetic susceptibility and specific heat. On the other hand, in both Figs. 6 and 12, the superconducting transition inferred from resistivity roughly tracks those inferred from magnetization and specific heat (in Fig. 12, even the $T_c$ data inferred from the onset criterion drop by a similar amount as the $T_c$ values inferred from thermodynamic data, just with an offset by a few degrees). This is consistent with the idea that a small portion of the sample has an enhanced $T_c$ associated with some strain and damage. Similar differences of $T_c$ between that inferred from resistivity and magnetic susceptibility, as well as transition width $\Delta T_c$, were observed in Fig. 6 and in Sr(Fe$_{1-x}$Ni$_x$)$_2$As$_2$\textsuperscript{34} This being said, the absence of any hint of superconducting drop in the $y$ $\geq$ 0.72 data [Fig. 11(c)] is a conclusive evidence that there is not even trace superconductivity in these samples. For the $y$ $>$ 0.65 and 0.72 samples, there appears to be an onset of filamentary superconductivity that is interrupted by the bulk AF.

In comparison to Ref. 32, whereas the above discussion and data show that AF appears to be very detrimental to the formation of the superconducting state when $T_N$ $>$ $T_c$, there is no evidence of the AF leading to dramatic re-entrance of the normal state when $T_N$ $<$ $T_c$ (i.e., for $y$ $\leq$ 0.60). The resistivity data as well as the susceptibility data do not show any feature that can be associated with the re-establishment of the normal state below the $T_N$ line as it cuts under the superconducting state.

These observations have several implications and also suggest several directions for future research. First, although dilute, paramagnetic, Eu$^{2+}$ only weakly suppresses SC, and antiferromagnetically ordered Eu$^{2+}$ appears to prevent its formation. As has been the case for other magnetic superconductors, specifically the RNi$_2$B$_2$C materials,\textsuperscript{11} a dramatic difference in the effects of local moments on SC can be observed when comparing disordered, single ions in paramagnetic state and an antiferromagnetically ordered sublattice. In the case of (Ho$_{1-x}$Dy$_x$)$_2$Ni$_3$B$_2$C,\textsuperscript{11,50} as $T_c$ crosses from above $T_N$ to below it, the cause of pair breaking changes from spin-flip scattering off of single impurities to interactions with magnetic excitations of the order state. In the case of Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$, the sudden loss of superconductivity as $T_N$ rises above $T_c$ implies that somehow long-range antiferromagnetic order of the Eu sublattice strongly suppresses (or removes) necessary ingredients for the establishment of the superconducting state. If antiferromagnetic fluctuations of the Fe sublattice [associated with the $k$ = (1,0,1) ordering\textsuperscript{51}] are associated with the pairing in the superconducting state, then long-range order of the large ($J = S = 7/2$) Eu sublattice with an ordering wave vector of $k$ = (1,0,0) could easily be related to a dramatic change in the Fe sublattice fluctuation spectrum. Such a dramatic change in the fluctuations could easily be the suppressed, missing ingredient for superconductivity invoked above. So, unlike DyNi$_2$B$_2$C, which apparently requires antiferromagnetic ordering of the Dy sublattice to suppress pair breaking of the individual Dy moments,\textsuperscript{11} Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ requires the Eu sublattice to remain in the disordered, paramagnetic state in order to establish the FeAs-based superconducting state.

Although this hypothesis readily explains the sudden loss of SC when $T_N$ $>$ $T_c$, it also would imply that the SC state below $T_N$, when $T_N$ $<$ $T_c$, should be modified; although Figs. 8, 11, and 12 show that there is no effect of $T_N$ on the low-field magnetization and zero-field resistivity when $T_N$ $<$ $T_c$, it is reasonable to anticipate that there will be changes in other superconducting parameters such as the superfluid density and penetration depth.

**IV. CONCLUSIONS**

Transport and thermodynamic measurements were performed on Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ and Sr$_{1-x}$Eu$_x$(Fe$_{0.88}$Co$_{0.12}$)$_2$As$_2$ single crystals. A superconducting dome is identified in Sr(Fe$_{1-x}$Co$_x$)$_2$As$_2$ as a function of Co doping and the optimal Co concentration is determined to be $x$ $=$ 0.12. The SC of the optimal Co doping is gradually suppressed by paramagnetic Eu$^{2+}$ following AG theory and found to coexist with AF of Eu$^{2+}$ for 0.43 $\leq$ $y$ $\leq$ 0.60. For higher Eu$^{2+}$ doping, bulk SC disappears suddenly when $T_N$ $>$ $T_c$. We speculate that the long-range order of the Eu$^{2+}$ sublattice is coupled to the AF fluctuations of the Fe sublattice and the suppression of the Fe fluctuations required for FeAs-based SC is what gives rise to the abrupt loss of bulk SC when $T_N$ surpasses $T_c$.

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