Journal of Physics: Conference Series

PAPER • **OPEN ACCESS**

Superconducting state in $(Eu_{1-x}Ca_x)RbFe_4As_4$ with 1144-type Structure

Superconducting state in $(Eu_{1-x}Ca_x)RbFe_4As_4$ with 1144-type Structure

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Abstract. We report the Ca substitution effect of the Fe-based superconductor $EuRbFe_4As_4$ with 1144-type structure (crystal system: tetragonal, space group: *P*4/mmm (No. 123)), in order to elucidate the relationship between the superconductivity and the magnetic order. The lattice constant systematically changed with the Ca concentration *x*. By Ca-substitution, the magnetic order is suppressed, while T_c is almost unchanged. The results indicate that the superconductivity of $EuRbFe_4As_4$ is not sensitive to the magnetic order.

1. Introduction

Since the superconductivity in La(O,F)FeAs has discovered[1)], variety of Fe-based superconductors have been reported. . Recently discovered $AeAFe_4As_4$ (*Ae* = Ca, Sr, Ba, Eu, *A* = K, Rb, Cs), which is so-called 1144-type compounds, is one of the Fe-based superconductors[2-5)]. $AeAFe_4As_4$ has a tetragonal structure (space group: *P*4/mmm (No. 123)) that consists of alternate stacking of two inequivalent $ThCr_2Si_2$ structures, namely, $AeFe_2As_2$ and AFe_2As_2 (inset of Fig. 1(a)) and shows the superconductivity with superconducting transition temperature: $T_c = 31 \sim 36$ K. The T_c values of 1144 compounds are closed to that of 122-type superconductors, $Ae_{1-x}A_xFe_2As_2$ (*Ae* = Ca, Sr, Ba, Eu, *A* = Na, K) [7-9)]. However, the *Ae* : *A* ratio in the 1144-type crystal structure is fixed to be 1 : 1 , because the *Ae* and the *A* ions do not occupy crystallographically equivalent sites in the 1144-type crystal structure. In the case of $EuAFe_4As_4$ (*A* = Rb, Cs), besides the bulk superconductivity with $T_c = {\sim}36$ K, the magnetic transition takes place at $T_m = {\sim}15$ K, indicating the coexistence of the superconductivity and the magnetic order. The magnetic order comes from the localized spin on Eu^{2+} [3-5)]. The magnetic transition was only confirmed in the magnetic susceptibility data. In the case of the related 122-type compound, $EuFe_2As_2$, it is known that the antiferromagnetic order shows up below 20 K arising from the Eu^{2+} ions[10)], and this magnetic order influences the superconductivity. By applying high pressure, zero resistivity was observed in a narrow pressure range of 2.6 GPa < *P* < 2.7 GPa[11)]. By substituting non-magnetic elements Ca^{2+} for Eu^{2+}, the magnetic order is suppressed and zero resistivity is observed in a wide pressure range of 1.2 GPa < *P* < 2.4 GPa compared to pure $EuFe_2As_2$[12)]. In $EuFe_2As_2$, the antiferromagnetic order likely competes with the superconductivity. In $EuAFe_4As_4$, the relationship between the magnetic order and the superconductivity remains has not been investigated so far.

In this paper, we investigate the physical properties of the Ca-substituted samples of $(Eu_{1-x}Ca_x)RbFe_4As_4$ to clarify the competition between the superconductivity and the magnetic order in the 1144 system.

2. Experimental details

Polycrystalline samples of $Eu_{1-x}Ca_xRbFe_4As_4$ (x = 0.0, 0.25, 0.5, 0.75, 1.0) were synthesized by a conventional solid state reaction using the stainless steel (SUS) pipe and cap method as described in Ref. 7. Details of the synthesis condition are given in Ref. 2. A powder x-ray diffraction (PXRD) pattern was measured at room temperature using CuKα radiation to evaluate the composition dependence of the lattice parameters. Intensity data were collected over a 2θ range from 5 to 80^o at 0.01^o step width. Magnetic susceptibility measurements were performed under a magnetic field H of 10 Oe using a magnetic-property measurement system (MPMS) (Quantum Design, MPMS-XL7). Data were collected during warming after zero-field cooling (ZFC) and then during field cooling (FC).

3. Results and discussions

Figure 1(a) shows the PXRD pattern of the $Eu_{0.5}Ca_{0.5}RbFe_4As_4$ sample. The main peaks can be indexed by employing a primitive tetragonal structure with space group $P4$/mmm (No. 123), indicating that $Eu_{0.5}Ca_{0.5}RbFe_4As_4$ possesses the 1144-type crystal structure. There are extra reflections due to 122-type $RbFe_2As_2$ and $(Eu,Ca)Fe_2As_2$ impurity phase[14,15)]. Similar characteristic diffraction patterns were also observed for other synthesized samples, ensuring that the samples have the 1144-type crystal structure. The calculated lattice constants a and c are plotted in Fig. 1(b), which decrease linearly with increasing x, as expected from the Vegard's low. No structural phase transition is recognized by substituting Ca for Eu.

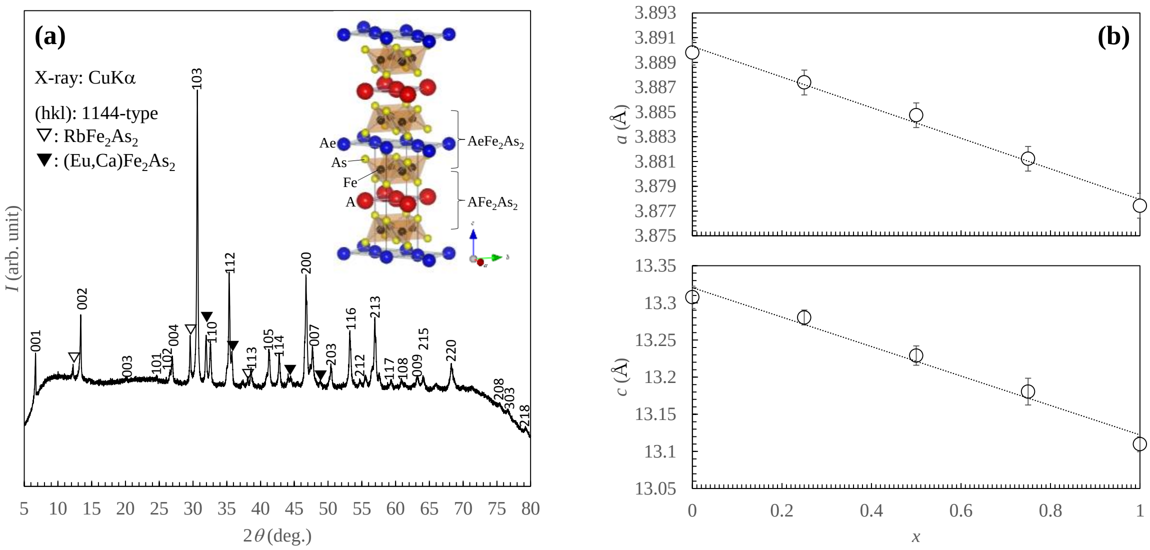

Fig. 1 (a) Powder X-ray diffraction pattern of $Eu_{0.5}Ca_{0.5}RbFe_4As_4$. Open and filled triangles show the impurity phases of $RbFe_2As_2$ and $(Eu,Ca)Fe_2As_2$, respectively. Inset shows the crystal structure of $AeAFe_4As_4$ (Program VESTA was used[13)]). (b) Ca concentration x dependence of the a- and c-axis lattice constants of $Eu_{1-x}Ca_xRbFe_4As_4$.

Figure 2(a) shows the temperature dependence of the magnetic susceptibility χ of $Eu_{1-x}Ca_xRbFe_4As_4$. The magnetic susceptibility data of all samples exhibits a large Meissner diamagnetic signal at approximately 36 K in both ZFC and ZC data, indicating the occurrence of bulk superconductivity. Because T_c of the $RbFe_2As_2$ and $(Eu,Ca)Fe_2As_2$ impurity phase is below 2.6 K under ambient pressure[14,15)], these impurity phases do not contribute for T_c as high as 36 K. The kink behaviour in the magnetic susceptibility is observed below T_c at T=15K for x = 0.0, T=12K for x =0.25, and T=7K for x

=0.5 , respectively. These features are the signatures of the magnetic phase transition and the transition temperature, T_m, decreases with increasing Ca concentration x (Fig. 2(b)). For the x = 0.25 sample, the magnetic susceptibility slightly increases near the lowest temperature. Observed T_m values of $Eu_{1-x}Ca_xRbFe_4As_4$ are different from those of $Eu_{1-x}Ca_xFe_2As_2$ ($x \leqq 0.5$) [12,15], indicating that the behaviour is not due to the $Eu_{1-x}Ca_xFe_2As_2$ impurity phase.

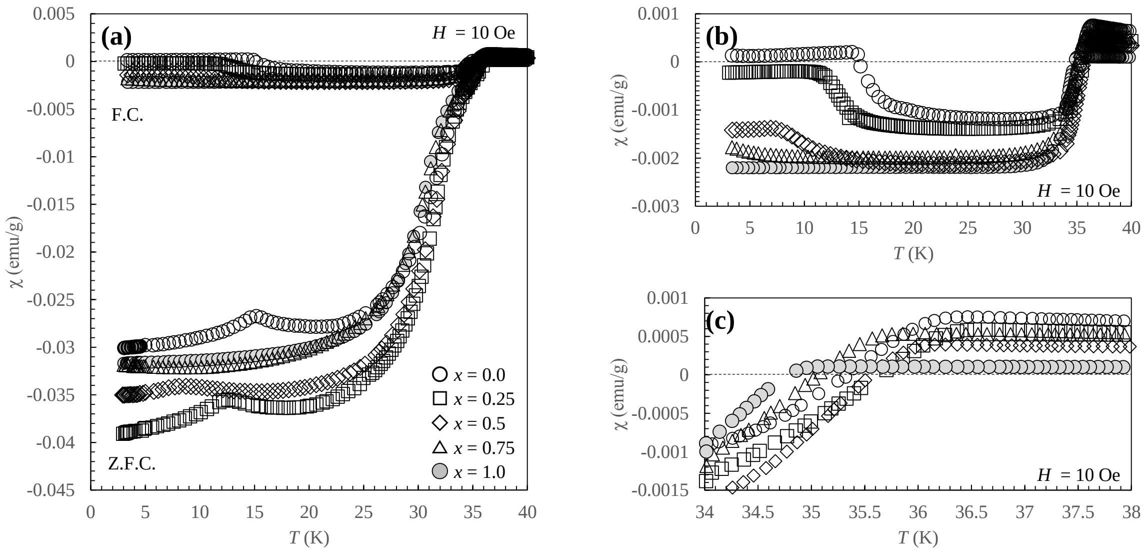

Fig. 2 (a) Magnetic susceptibility χ of $Eu_{1-x}Ca_xRbFe_4As_4$ as a function of temperature T. Enlarged view of the F.C. data and near T_c are shown in Fig. 2(b) and Fig. 2(c), respectively.

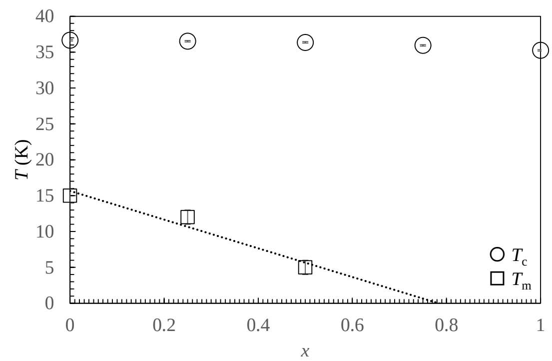

Fig. 3 T_c and T_m of $Eu_{1-x}Ca_xRbFe_4As_4$ as a function of Ca concentration x. T_c and T_m were defined from the χ -T data of Fig. 2(a).

Figure 3 shows T_c and T_m as a function of x. Although T_m monotonously decreases with increasing x, T_c is almost unchanged. This behaviour is in contrast to the case of $Eu_{1-x}Ca_xFe_2As_2$, in which pressure-induced superconductivity shows up at lower pressures in the Ca-substituted samples. It is noted that the c-axis length of $EuFe_2As_2$ sublattice in $EuRbFe_4As_4$ is larger compared to $EuFe_2As_2$ [4]. The longer distance between the Eu layer and the FeAs-layer in $EuRbFe_4As_4$,would weaken the influence of the Eu^{2+} magnetic order to the superconductivity compared to $EuFe_2As_2$.

4. Summary

We synthesized the polycrystalline sample of $Eu_{1-x}Ca_xRbFe_4As_4$ and characterized their properties. The lattice constants show the linear Ca concentration x dependence. Superconductivity was confirmed at around 36 K in all synthesized samples. The magnetic order shows up below 15K for $EuRbFe_4As_4$, which decreases with increasing x. On the other hand, T_c does not change with x. These behaviors suggest that the magnetic order in the Eu-layer have little influence on the superconductivity in the 1144system.

ACKNOWLEDGEMENT

This work was partially supported by JSPS KAKENHI Grant Number JP16H06439.

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