

Supporting information:

Single crystal growth and study of the ferromagnetic superconductor RbEuFe₄As₄

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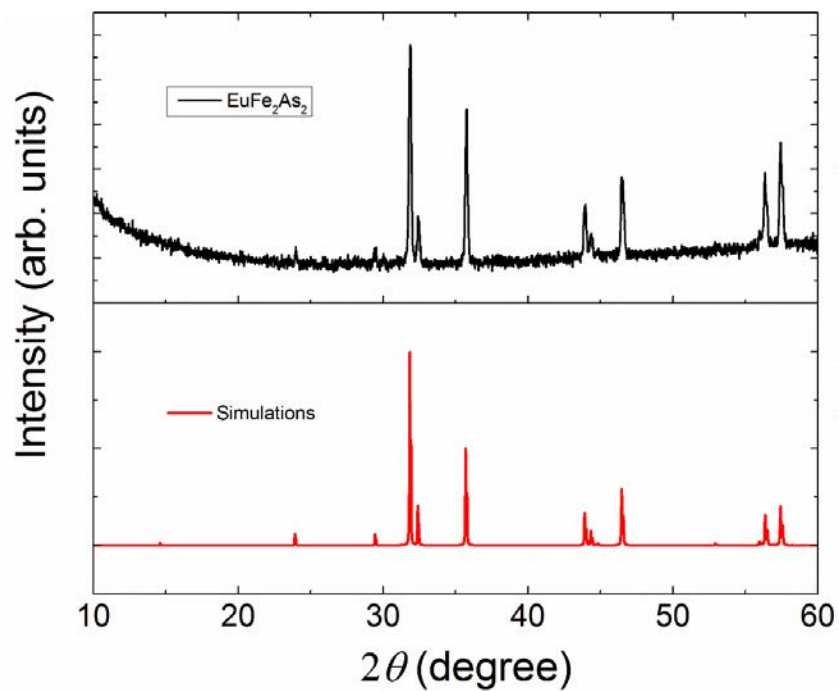


Figure S1 Powder x-ray diffraction data of EuFe_2As_2 and its simulated one.

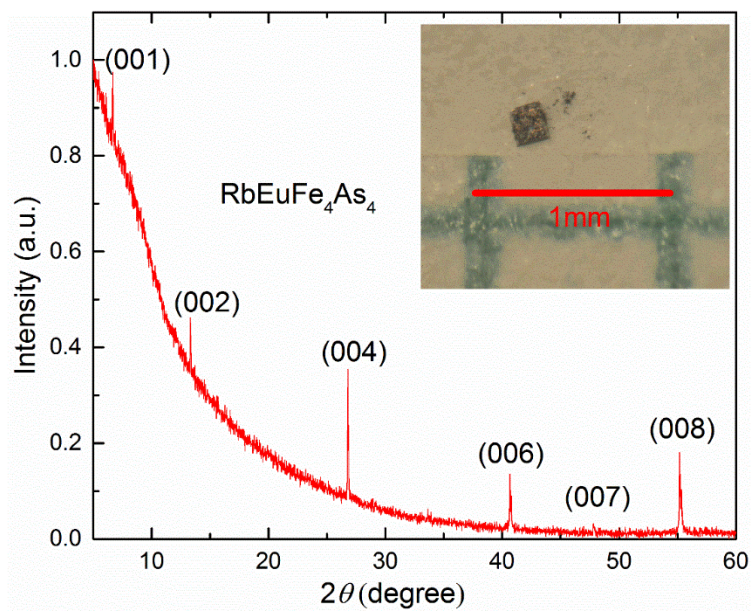


Figure S2 X-ray diffraction of one single crystal $\text{RbEuFe}_4\text{As}_4$ grown from the ratio $\text{RbAs} : \text{EuFe}_2\text{As}_2 = 5:1$. A photo of a tiny crystal with a length of ~ 0.2 mm under an optical microscope.

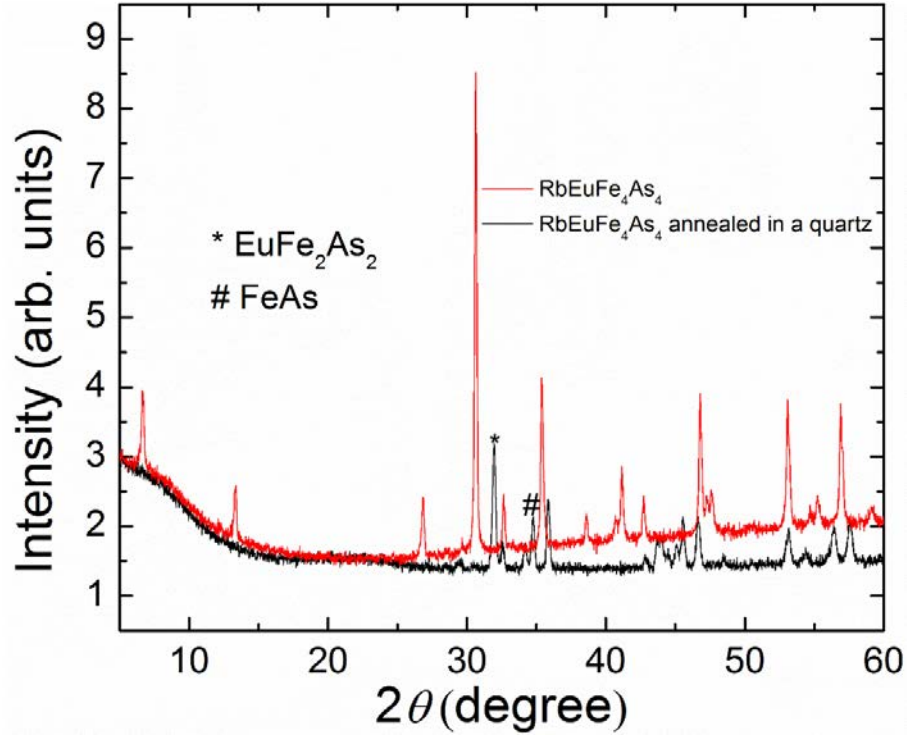


Figure S3 Powder x-ray diffraction of pure RbEuFe₄As₄ polycrystalline material before and after being annealed at 1173 K in a quartz tube. The main phase after annealing is EuFe₂As₂ and FeAs.

Table S1. Anisotropic displacement parameters U_{ij} ($\text{\AA}^2 \times 10^3$) for RbEuFe₄As₄ sample A1 at 293 K with estimated standard deviations in parentheses.

Label	U_{11}	U_{22}	U_{33}	U_{12}	U_{13}	U_{23}
Eu	8(1)	8(1)	8(1)	0	0	0
As1	9(1)	9(1)	6(1)	0	0	0
As2	8(1)	8(1)	5(1)	0	0	0
Fe	7(1)	7(1)	6(1)	0	0	0
Rb	15(1)	15(1)	11(1)	0	0	0

The anisotropic displacement factor exponent takes the form: $-2\pi^2[h^2a^{*2}U_{11} + \dots + 2hka^*b^*U_{12}]$.

Table S2. Bond lengths (Å) and bond angles (°) for RbEuFe₄As₄ sample A1 at 293 K with estimated standard deviations in parentheses.

Bond lengths (Å)	
Eu-As2	3.2125(8) × 8
Fe-As1	2.3904(11) × 4
Rb-As1	3.5112(9) × 8
Fe-As2	2.3844(12) × 4
Fe-Fe	2.7453(2) × 4
Bond angles (°)	
As2-Fe-As2	109.01(8)
As2-Fe-As1	109.81(2) × 4
As1-Fe-As1	108.60(7)
As2-Eu-As2	117.43(5) × 4
As2-Eu-As2	62.57(5) × 4
As2-Eu-As2	180.00 × 4
As2-Eu-As2	74.35(2) × 8
As2-Eu-As2	105.65(2) × 8
As1-Rb-As1	180.00 × 4
As1-Rb-As1	112.87(2) × 8
As1-Rb-As1	67.13(2) × 8
As1-Rb-As1	77.13(4) × 4
As1-Rb-As1	102.87(4) × 4

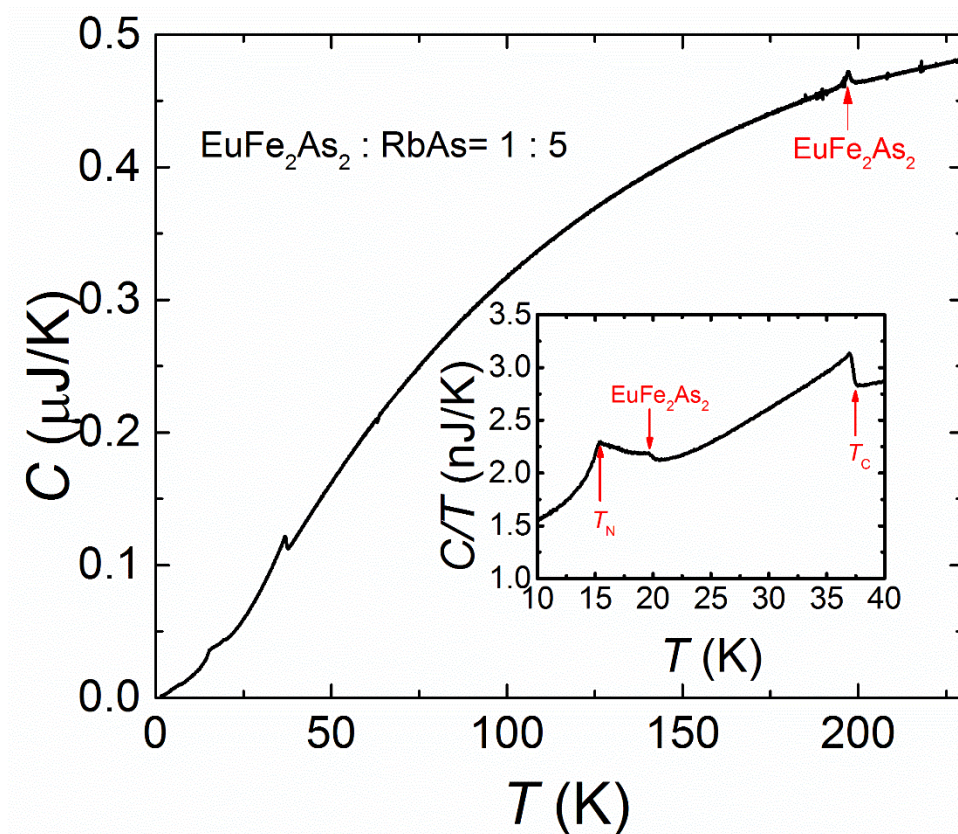


Figure S4 Temperature dependence of specific heat C from the crystals grown in the batch with the ratio $\text{RbAs} : \text{EuFe}_2\text{As}_2 = 5 : 1$. The inset shows C/T vs T from 10 to 40 K. There are two phase transitions at T_N and T_c from the $\text{RbEuFe}_4\text{As}_4$ phase. There are another two transitions at ~ 20 K and 195 K in the same crystal, demonstrating the existence of a tiny impurity phase of EuFe_2As_2 .

Sample label information:

Sample A1 and A2 are crystals from the same batch grown under the condition RbAs : Fe₂As : EuAs = 15 : 1 : 1. The single crystal refinements results in the article are from sample A1. Sample B1 and B2 are crystals from the same batch grown under the condition RbAs : EuFe₂As₂ = 5 : 1.

Table S3 As-Fe-As Bond angles in the FeAs₄ tetrahedron from four different samples.

Sample A1	
Bond angles (°)	
As2-Fe-As2	109.01(8)
As2-Fe-As1	109.81(2) × 4
As1-Fe-As1	108.60(7)
Sample A2	
Bond angles (°)	
As2-Fe-As2	108.93(8)
As2-Fe-As1	109.83(2) × 4
As1-Fe-As1	108.57(8)
Sample B1	
Bond angles (°)	
As2-Fe-As2	108.93(3)
As2-Fe-As1	109.834(11) × 4
As1-Fe-As1	108.56(3)
Sample B2	
Bond angles (°)	
As2-Fe-As2	109.03(5)
As2-Fe-As1	109.764(14) × 4
As1-Fe-As1	108.75(5)

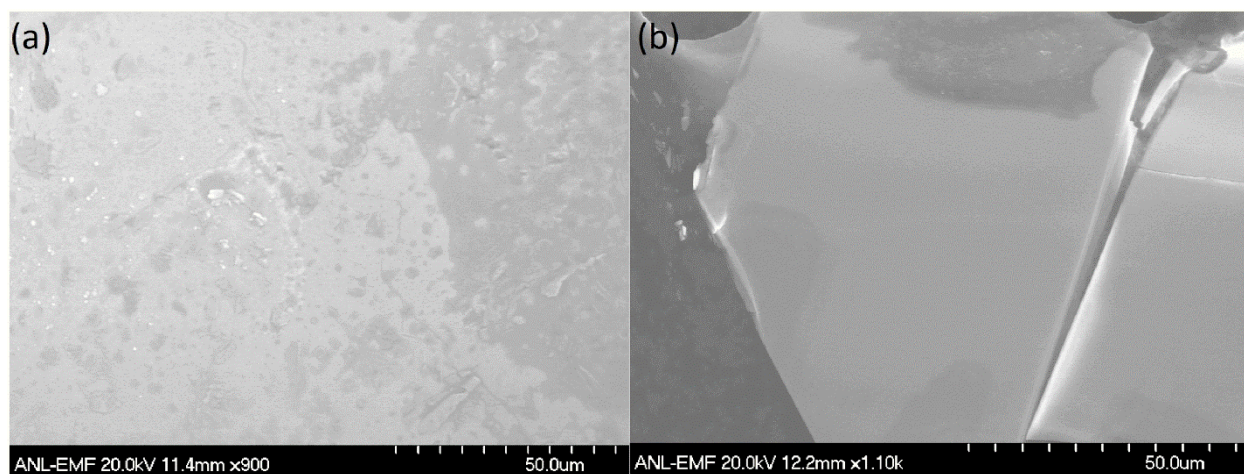


Figure S5 (a), (b) Scanning electron microscope images of one pristine crystal $\text{RbEuFe}_4\text{As}_4$ and another cleaved one, respectively.



Figure S6 Alumina crucible after the crystal growth

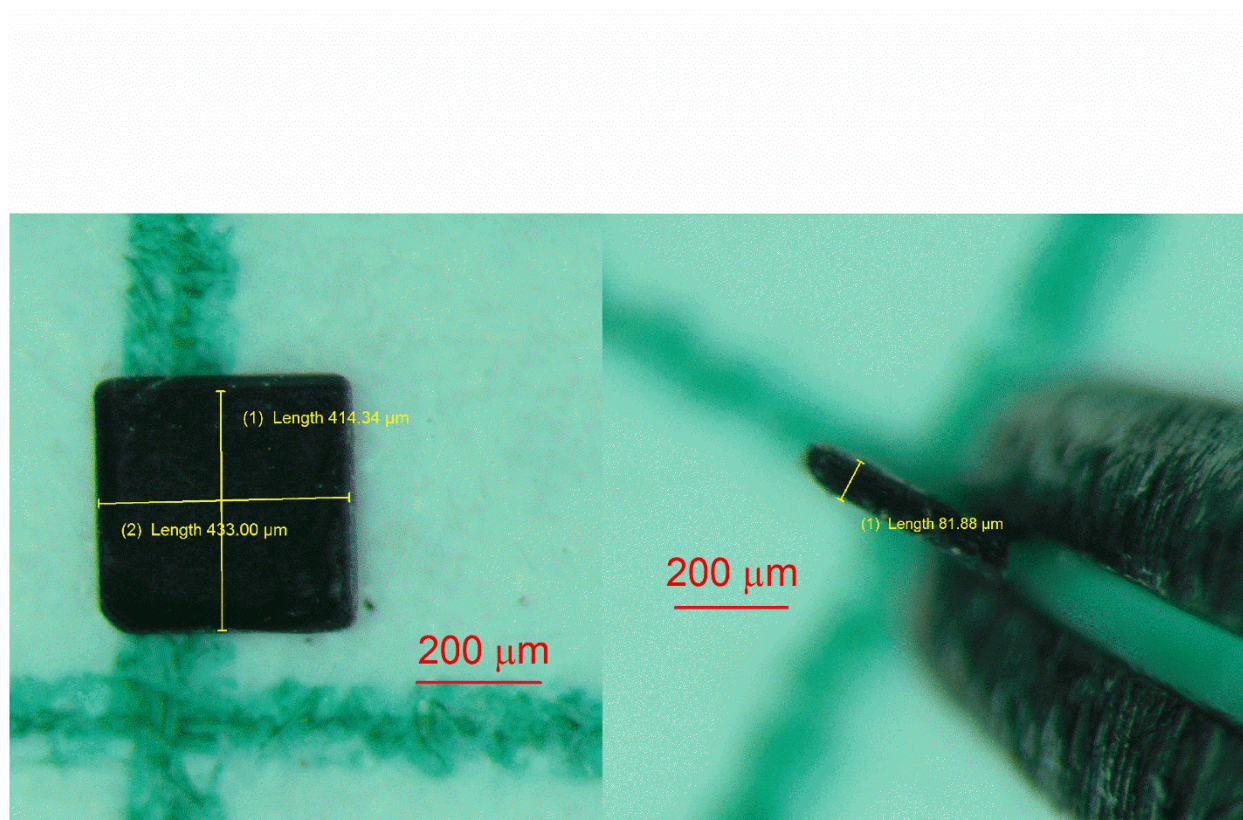


Figure S7 The dimension ($\sim 0.43\text{mm} \times 0.42\text{mm} \times 0.081\text{mm}$) of the $\text{RbEuFe}_4\text{As}_4$ crystal used for magnetic susceptibility measurements.