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Article

Anomalous AC Susceptibility Response and Paramagnetic Meissner Phase of $\text{EuRbFe}_4\text{As}_4$ Superconductor

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Abstract

Magnetic superconductor $\text{EuRbFe}_4\text{As}_4$ is a quite unique system in which macroscopic superconductivity and magnetic ordering coexist, with interesting interactions between Abrikosov vortices and Eu^{2+} spins that were investigated mostly by static (DC) magnetization measurements. Our aim is to study the dynamic interactions between the two sub-systems using AC susceptibility measurements in a wide range of temperatures and superimposed DC fields. In low DC fields, the magnetic transition at 15 K is clearly visible. We have observed very little difference between the AC susceptibility in different cooling regimes, but large difference for different field orientation. For field perpendicular to the superconducting planes, we have observed an anomalous dependence just below the critical temperature, which is absent in the parallel field orientation. We explained the anomaly by the interplay between the sample dimensions and the temperature dependence of the London penetration depth which may allow the paramagnetic Meissner phase to be detected in the susceptibility response.

Keywords: magnetic superconductor; $\text{EuRbFe}_4\text{As}_4$; AC susceptibility

1. Introduction

Soon after the discovery of iron-based superconductors (IBS) [1], the 122 family based on AEFe_2As_2 (AE being alkali-earth metal Ca, Sr, Ba) parent compound, became the most popular materials for both physical explorations and applications because of their critical temperature T_c as high as 38 K [2], very high upper critical fields $\mu_0 H_{c2}$ (> 70 T) [3,4] and low anisotropies ($\gamma < 2$) [4]. Superconductivity in AEFe_2As_2 is primarily induced by alkali metal ($A = \text{Na}, \text{K}, \text{Rb}, \text{Cs}$) substitution at AE sites with a concomitant suppression or elimination of the structural and magnetic ordering transition. The structure's crystallographic space group ($I4/mmm$) is not changed by this A substitution because AE and A randomly occupy crystallographically equivalent sites. Thus, $(\text{AE}_{1-x}\text{A}_x)\text{Fe}_2\text{As}_2$ (also noted as $(\text{AE}, A)\text{Fe}_2\text{As}_2$) are solid solutions between AEFe_2As_2 and AFe_2As_2 compounds with the same structural type. Later it was found that, if there is a large difference in the ionic radii Δr of AE and A , such solid solutions are not possible and a new type of IBSs has been reported [5], having a new structure, abbreviated as AEA1144 , namely $\text{CaAFe}_4\text{As}_4$ ($A = \text{K}, \text{Rb}, \text{Cs}$) and $\text{SrAFe}_4\text{As}_4$ ($A = \text{Rb}, \text{Cs}$). In these cases, because A does not mix with AE due to the large Δr , AEA1144 crystallizes through alternate stacking of the AE and A layers across the Fe_2As_2 layers, changing the space group from $I4/mmm$ to $P4/mmm$, the compounds being superconductors with T_c values between 31 and 36 K. One of the most studied superconductor (SC) in this new family is $\text{CaKFe}_4\text{As}_4$ (CaK1144) due to its excellent superconducting properties including very high critical current density and very high pinning potential [6]. Following the discovery of the 1144-type IBS, in 2016 two Eu-

containing materials in the family, $AEuFe_4As_4$ ($A = \text{Rb}, \text{Cs}$), were subsequently synthesized and characterized [7,8]. The two sibling compounds exhibit SC at $T_c = 36.5 \text{ K}$ ($A = \text{Rb}$) and 35 K ($A = \text{Cs}$), respectively, without extrinsic doping. The Eu^{2+} spins order at $T_m = 15 \text{ K}$ ($A = \text{Rb}$) or 15.5 K ($A = \text{Cs}$) [8]. The Mössbauer studies [9] indicate that the Eu^{2+} spins in $AEuFe_4As_4$ are ferromagnetically (FM) coupled and lie flat in the ab plane. Intuitively, the removal of every alternating magnetic Eu-layer in EuFe_2As_2 would give rise to Eu-spin FM. Nevertheless, recent neutron diffraction study [10] revealed a helical modulation with the magnetic propagation vector of $\mathbf{k} = (0, 0, 1/4)$ for the Eu-spin ordering. If a small magnetic field ($\sim 0.2 \text{ T}$ and $\sim 0.4 \text{ T}$, respectively, for $H \parallel ab$ and $H \parallel c$) is applied [11], this helically modulated magnetic structure easily changes into a genuine FM, the latter of which fully coexists with SC. Therefore, the new materials can be viewed as a natural atomic-thick superconductor–ferromagnet superlattice, as sketched in Figure 1. The parent compound EuFe_2As_2 is an antiferromagnet (Fig. 1a). Due to the large difference in atomic radii, a 50% substitution of Eu with Rb results not in $(\text{Eu}_{0.5}\text{Rb}_{0.5})\text{Fe}_2\text{As}_2$ solid solution, but in the new, layered 1144 structure, $\text{EuRbFe}_4\text{As}_4$, in which the Eu layers having helical magnetism sandwiches a thick superconducting layer composed of two Fe_2As_2 planes with a Rb plane between them (Fig. 1 b). By applying a small magnetic field, helical magnetism in the Eu planes is driven into a ferromagnetic state (Fig.1 c).

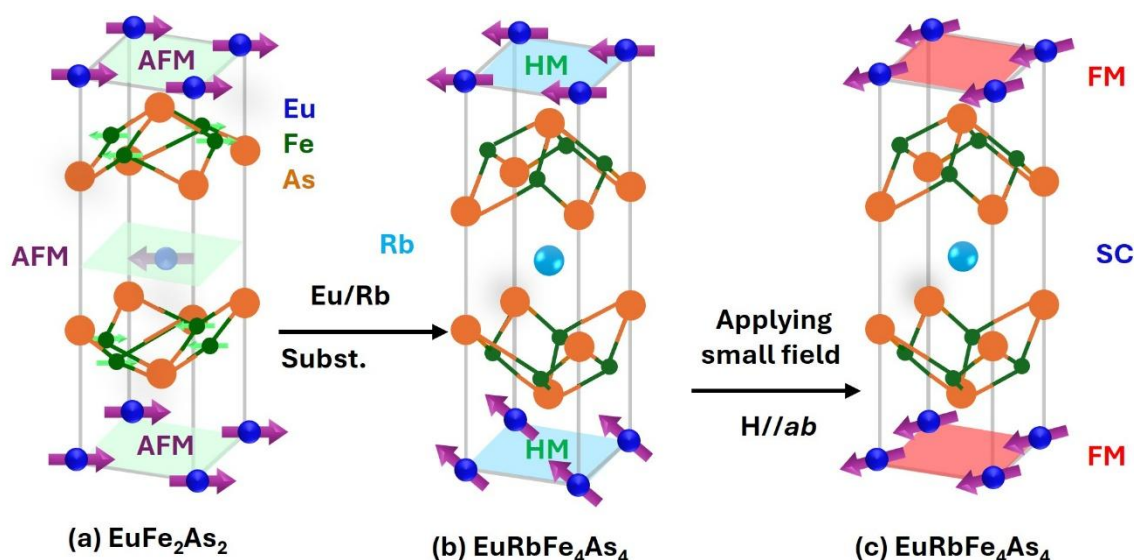


Figure 1. Schematic structures of: (a) EuFe_2As_2 ; (b) $\text{EuRbFe}_4\text{As}_4$ with helical magnetism of Eu^{2+} spins; and (c) $\text{EuRbFe}_4\text{As}_4$ with ferromagnetism of Eu^{2+} spins induced by magnetic field.

The new magnetic superconductor was characterized by many techniques to elucidate various aspects of superconductivity. After their discovery by Kawashima et al [7], the crystal structure was investigated by XRD and determined to be tetragonal with symmetry group $P4/mmm$ and by resistivity and DC magnetization that showed a critical temperature of about 36 K , an upper critical field (extrapolated at 0 K) of about 92 T and a coherence length of 1.8 nm [7]. More importantly, they discovered an anomaly in the magnetic response at 15 K which was correctly interpreted as coexistence of superconductivity and a magnetic ordered state created by Eu^{2+} ions. A few months later, Liu et al. [8] managed to replicate the material, and, in addition to resistivity and magnetization measurements that confirmed previous results, they performed also magnetization hysteresis measurements and specific heat measurements that revealed a very rare third-order type magnetic transition. In a seminal paper, Ishida et al. [12], by combining neutron diffraction and magnetization measurements, revealed that ferromagnetic alignment of Eu^{2+} moments is induced by superconducting vortices. They showed that the direction of the Eu^{2+} spins is dominated by the distribution of pinned vortices based on the critical state model, highlighting a unique interplay between magnetism and superconductivity. Vortex matter, dynamics and pinning are reflected

different in the case of AC susceptibility as compared to DC studies. For this reason, in this work we investigated the interplay between magnetic moments and vortex matter and dynamics in the case of AC fields superimposed on DC fields up to 9 T, for fields orientations perpendicular and, respectively, parallel to the superconducting planes, in both Zero-Field-Cooling (ZFC) and Field-Cooling (FC) procedures. The results showed the expected ferromagnetic signal at around 15 K superimposed on the diamagnetic screening at low DC fields (similar to DC susceptibility measurements), as well as a clear anomaly in the diamagnetic screening due to ferromagnetic ordering of the spins immediately after T_c at higher fields. The anomaly in the in-phase susceptibility is accompanied by a shoulder in the out-of-phase susceptibility (dissipation peak), in both ZFC and FC regimes, for perpendicular orientation. In the case of measurements with the thin sample parallel to the fields, such anomalies were not detected.

2. Materials and Methods

The $\text{EuRbFe}_4\text{As}_4$ single crystals were grown in AIST Tsukuba, Japan, by the RbAs-flux method [13]. EuAs, Fe_2As and RbAs precursors were prepared from Eu and As, Fe and As, and Rb and As, respectively, which were mixed at appropriate molar ratios. EuAs and Fe_2As mixtures were sealed in evacuated quartz tubes, while RbAs was sealed in a stainless-steel tube with an alumina crucible. The mixtures were sintered for 20 h at the following temperatures: 750 °C for EuAs, 900 °C for Fe_2As and 600 °C for RbAs, then were weighed at a ratio 1:1:15 to a total amount of 9g and placed in an alumina crucible which was sealed in a stainless-steel container. The thermal process leading to crystal growth consisted in heating in 5 h to 700 °C, maintaining this temperature for 5 h, then heated in 5 h to 970 °C and maintaining this temperature for 10 h. The final step was a very slow cooling for 350 h (1 °C/h) to 620 °C. After cleaving the surfaces of the single crystals, XRD patterns showed only the (00 l) peaks from $\text{EuRbFe}_4\text{As}_4$ [12]. The sample investigated in this work is a thin square with length $l = 0.9 \pm 0.1$ mm and thickness $t \approx 0.06 \pm 0.02$ mm.

AC magnetic susceptibility was measured using a *Quantum Design* PPMS-9 T with an ACMS option and the PPMS MultiVu software, which allows to pre-program the type of measurements, the temperature (fixed or variable, between 1.9 and 350 K), the field (fixed or variable, up to 9 T), the frequency of the AC field excitation (up to 10 kHz), and the amplitude of the AC field excitation (up to 16 Oe). For this work, AC susceptibility measurements were conducted as a function of temperature by applying an AC magnetic field perpendicular or parallel to the superconducting layers (a - b planes), with or without a superimposed DC magnetic field up to 9 T, in the Zero-Field Cooling (ZFC) conditions or Field Cooling (FC) conditions. The measurements were taken at a fixed AC field amplitude $h_{AC} = 1$ Oe and at the AC field frequency $\nu = 5686.4$ Hz. To prevent any potential effects of residual field trapped within the DC magnet on the sample response, before each measurement we warmed the magnet close to room temperature, applied 2 T and reduce the field to zero in the oscillation mode. This demagnetizing process was proven to reduce the trapped DC field below 1 Oe. After demagnetizing procedure, the sample was cooled to 5 K in ZFC or FC conditions. Susceptibility as function of temperature data were measured between 5 and 40 K with a sweep rate of 0.1 K/min, such small sweeping rate ensuring a large number of measured points for each curve.

3. Results

3.1. Fields Perpendicular to the Superconducting Planes

Figure 2 shows the temperature dependence of the in-phase fundamental susceptibility of the sample measured with an AC field with amplitude of 1 Oe and frequency of 5686.4 Hz superimposed on DC magnetic fields of 0.01, 0.1, 0.5, 1, 2, 3, 4, 5, 6, 7, 8 and 9 T.

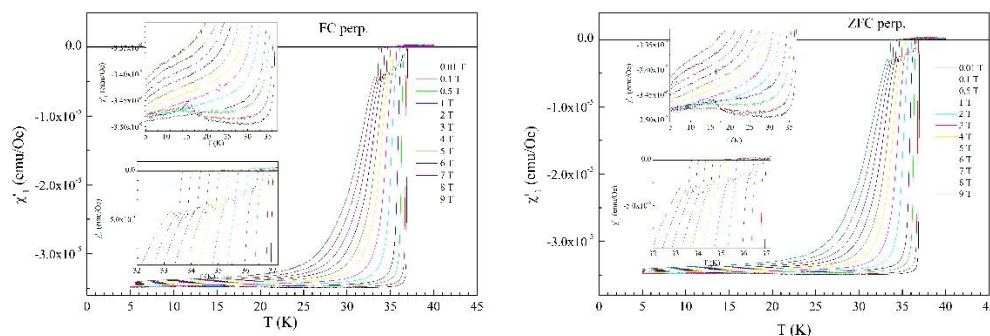


Figure 2. Temperature dependence of the in-phase susceptibility for the DC fields indicated in the Figure, for fields perpendicular to the superconducting planes, in ZFC (left-hand side) and FC (right-hand side) cooling protocols. Upper inserts show details of the diamagnetic signal around the temperature of the magnetic transition (15 K) while lower inserts show details of the diamagnetic signal near T_c .

The first thing we observed is the small signal at low temperature and very low DC fields (< 0.5 T) that indicates the magnetic transition, similar to the ones detected in DC magnetization measurements [7,8]. In higher fields the ferromagnetic signal from Eu^{2+} is masked by the decrease in diamagnetic screening due to the decrease of J_c with increasing DC field. Near T_c we observe a standard sharp superconducting transition in low DC fields, which, as expected, becomes wider and having smaller on-set temperature with increasing DC fields. However, for DC fields higher than 0.5 T the in-phase susceptibility shows an unexpected anomaly not far from T_c as can be seen in the main panels and, in detail, in the lower inserts. After the normal increase in diamagnetic screening with decreasing temperature, we can see a decrease in diamagnetic screening, which is consistent with the appearance of a small ferromagnetic signal from the Eu^{2+} spins. The anomaly starts as a “shoulder” for fields smaller than about 2 T, and develops as a clear local “maximum” for larger DC fields. Looking at both sets of graphs, we can see that there are small, but not significant differences between ZFC and FC cooling protocols.

Figure 3 show the temperature dependence of the out-of-phase susceptibility, in both ZFC and FC cooling protocols, for the same DC fields as in Figure 2. Figures show again very small, negligible, differences between ZFC and FC cooling protocols. For small fields, the dissipation signal has a very sharp peak and, as DC fields exceed 0.5 T, a “shoulder” is developing and becomes more visible with the increase of DC field, together with the expected broadening of the dissipation signal.

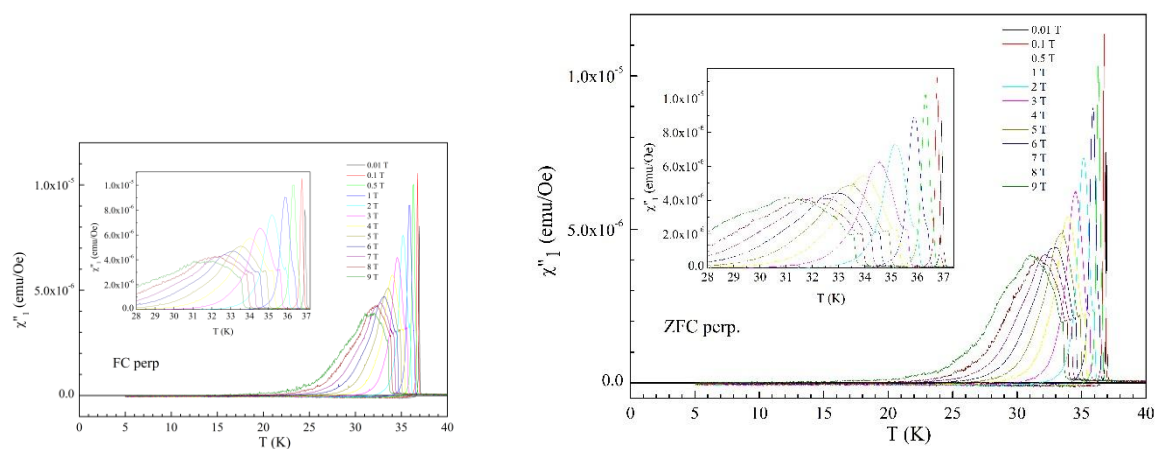


Figure 3. Temperature dependence of the out-of-phase susceptibility for the DC fields indicated in the Figure, for fields perpendicular to the superconducting planes, in ZFC (left-hand side) and FC (right-hand side) cooling protocols. Inserts show a more detailed picture of the dissipation signal.

The positions (temperatures) of the shoulders corresponds to those of the local “peaks” in the in-phase susceptibility as seen in Figure 2.

3.2. Fields Parallel with the Superconducting Planes

For this geometry, we have performed the measurements with two AC field amplitude, 1 and 10 Oe, and with two frequencies, 497 and 5686.4 Hz, in ZFC protocol. After looking at the results at DC fields 0 and 0.01 T, we have seen that the signal for 1 Oe and 497 Hz are very noisy, as can be seen in Figure 4.

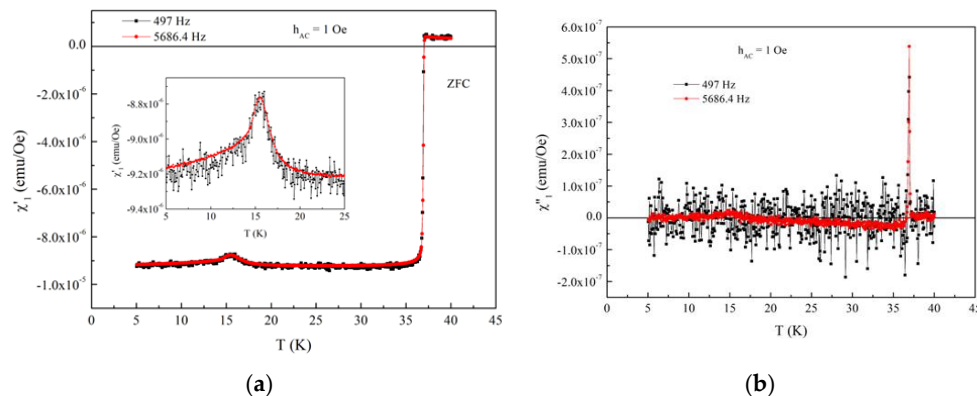


Figure 4. Temperature dependence of the in-phase (left-hand side) and out-of-phase (right-hand-side) susceptibility in zero DC field, with AC field amplitude of 1 Oe and the two AC frequencies indicated in the Figure. Insert shows the detail of the signal due to magnetic transition at 15 K.

Even for the low frequency, we can see a sharp superconducting transition in both diamagnetic screening and dissipation peak. The magnetic transition is clearly visible at 15 K, similar to the case of perpendicular orientation and DC susceptibility measurements.

For AC field amplitude of 10 Oe the measurements look similar, only without the noise at low frequency. For DC field of 0.01 T (100 Oe), both ZFC and FC measurements with AC field amplitudes of 1 and 10 Oe look similar to the zero DC field case (again with quite large noise at 1 Oe and 497 Hz). The data can be seen in Supplementary Material 1. For higher fields, the measurements in parallel configuration show standard in-phase susceptibility, without any anomaly just below T_c as was the case of perpendicular configuration. To summarize the results of the measurements in parallel configuration in zero or very small DC fields: the superconducting transitions are sharp, with the expected magnetic signal in the in-phase susceptibility at 15 K, very similar to measurements in perpendicular configuration and to low-field DC measurements reported in the literature. In higher fields, there is no anomaly in the diamagnetic signal as is the case for perpendicular fields configuration. The small positive signal in the normal state will be addressed in the next sub-section.

3.3. Comparison Between Perpendicular and Parallel Configurations

Since we have not observed any anomaly in the in-phase susceptibility in parallel orientation, it is interesting to have a comparison between $\chi'_1(T)$ for the two orientations, in the same DC field, measured with the same amplitude and frequency of the AC field. For such comparison to be meaningful, we need to take into account the huge difference in demagnetization factors for the two field orientations due to the plate-like shape of the sample with thickness one order of magnitude smaller than the larger dimensions, so we normalized the in-phase susceptibility curves to the largest values of the diamagnetic signal for the two orientations, which are about -3.5×10^{-5} emu/Oe in perpendicular configuration (see Figure 2), and, respectively, about -0.9×10^{-6} emu/Oe in parallel configuration (see left-hand-side of Figure 4), ensuring in this way that in both orientations the in-phase normalized susceptibility at 5 K is -1 (perfect diamagnetism). All the normalized susceptibility curves shown below were measured in various DC fields using ZFC protocol, with AC field amplitude of 1 Oe and frequency of 5686.4 Hz. FC protocols gave the same results, with small, minor differences. Figure 5 shows the results for zero and 100 Oe, below the transition between helical and ferromagnetic arrangements of the Eu^{2+} spins.

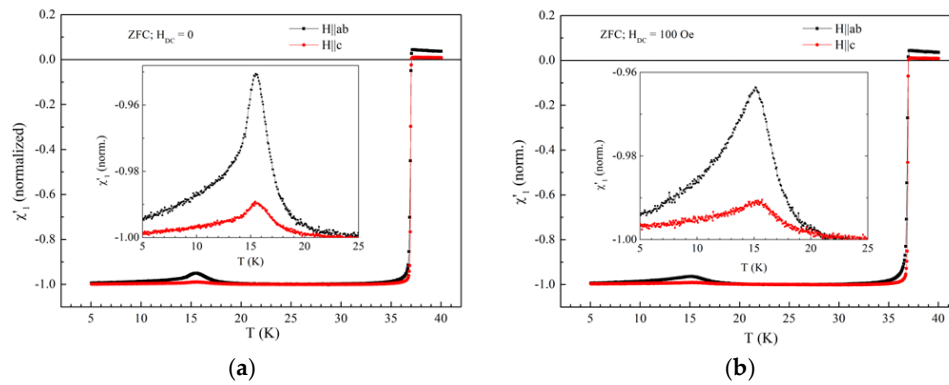


Figure 5. Temperature dependence of the in-phase susceptibility in zero DC field (left-hand side) and in DC field of 100 Oe (right-hand-side), measured with AC field amplitude of 1 Oe and AC frequency of 5686.4 Hz. Inserts show the details of the signal due to magnetic transition at 15 K.

It can be seen that, in zero and very low DC field, the superconducting transitions are sharp, with the same T_c . The signal due to magnetic transition is 5 times larger in the case of parallel orientation, which can be easily explained considering that is much easier for the Eu^{2+} spins to follow the direction of the AC field in the a - b planes for parallel orientation than to tilt away from the planes to follow the direction of perpendicular AC field. For the same reason there is a small, positive magnetic signal in the normal phase of the superconductors (for temperatures higher than T_c) in parallel configuration. Remarkably, the value of the positive susceptibility in the normal state is about the same as the height of the peak at 5 K, namely about 0.05 a.u. in the normalized scale. A larger signal for the parallel orientation at the magnetic transition at 15 K was observed also in DC magnetization measurements in a DC field of 30 Oe [14].

Figure 6 shows the temperature dependence of the in-phase fundamental susceptibility for the two orientations, for fields above the field of helical magnetism / ferromagnetism transition, but not very large, 0.1 T (left-hand side) and 0.5 T (right-hand side). It can be seen that, in such intermediate fields, there is a competition between the magnetic interaction of Eu^{2+} spins with the larger density of vortices induced by the DC field (similar to the so called “magnetic pinning”) and the tendency to follow the AC field orientation. In 0.1 T the signals due to magnetic transition are much smaller, and very noisy, while in 0.5 T the signal practically disappears.

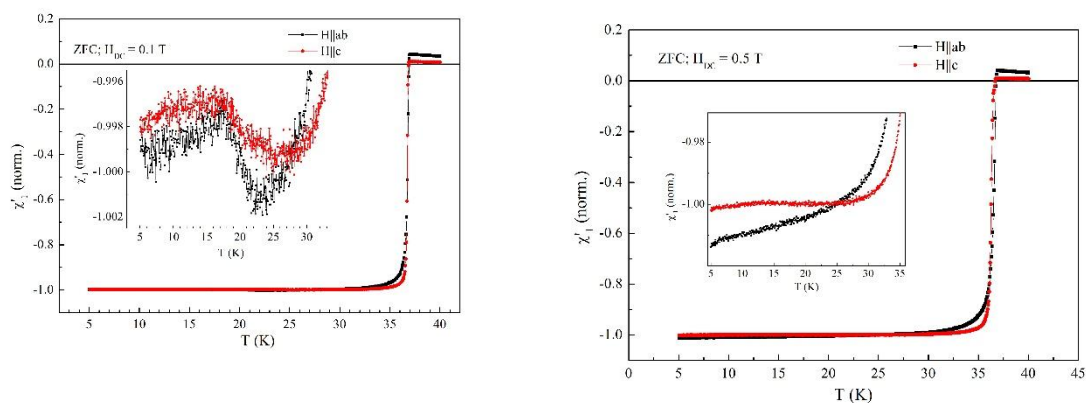


Figure 6. Temperature dependence of the in-phase susceptibility in DC field of 0.1 T (left-hand side) and in DC field of 0.5 T (right-hand-side), measured with AC field amplitude of 1 Oe and AC frequency of 5686.4 Hz. Inserts show the details of the signal due to magnetic transition at 15 K.

In higher fields there is no more visible signal at 15 K due to the much larger density of vortices which, through magnetic interaction, impede the Eu^{2+} spins from following the direction of AC field, but for such larger DC fields the anomaly just below T_c in perpendicular configuration appears.

Figure 7 shows the comparison between the temperature dependence of the normalized in-phase susceptibility for parallel and perpendicular configurations, in DC fields of 1 T (left-hand-side) and 3 T (right hand side). Inserts show details of curves just below T_c , where the anomaly starts to appear for perpendicular configuration. Apart from the anomaly, there are a few interesting features. The most visible is the appearance of a small difference in the critical temperatures for the two orientations, due to the large demagnetization factor in perpendicular configuration. Another feature is the decrease of the positive signal in the normal state with increasing DC field, again due to the magnetic interaction between Eu^{2+} spins and magnetic flux lines with an increasing density, preventing the spins to follow the direction of AC field. It is worth noting that we have kept the same normalizing factors as in zero DC field; this is the reason for in-phase susceptibility for parallel orientation at low temperature laying below -1 .

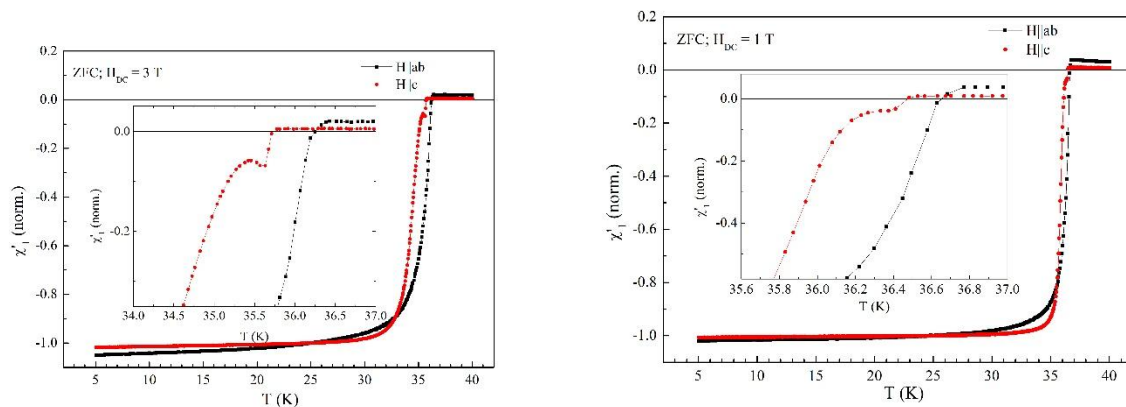


Figure 7. Temperature dependence of the in-phase susceptibility DC field of 1 T (left-hand side) and in DC field of 3 T (right-hand-side), measured with AC field amplitude of 1 Oe and AC frequency of 5686.4 Hz. Inserts show the details of the transition just below T_c .

4. Discussion

We will start our discussion with a comparison between the EuRb1144 magnetic superconductor and its more studied “relative” non-magnetic CaK1144. Quite unexpectedly, EuRb1144 shows a higher T_c of 37 K as compared to 35.8 K in CaK1144 [6,15] despite the presence of magnetism which usually suppress critical temperature. However, the interaction between Eu^{2+} spins and the vortex system leads to a much broader superconducting transition in high magnetic fields and a more pronounced reduction of T_c with increasing DC field. For example, in a DC field of 9 T, T_c decreases with 10% in EuRb1144 (see lower inserts of Figure 2) while in CaK1144 T_c decreases with only 8 % (see Supplementary Material 2). Another important aspect is the comparison between ZFC and FC cooling conditions. Similar to CaK1144, there is no significant difference between ZFC and FC AC susceptibility in EuRb1144, unlike the case of isovalently substituted $\text{BaFe}_2(\text{As}_{1-x}\text{P}_x)_2$ 122 compound which showed a very pronounced magnetic memory effect [16,17]. Of course, the main difference between the two 1144 materials is the signal around 15 K due to the magnetic transition, which was first detected through DC magnetization measurements and reported in the same papers that announced the new material [7,8]. The feature that was not observed previously in any studies is the anomaly in the susceptibility response to the AC field perturbation just below T_c in fields larger than approximately 0.5 T, *only with the fields perpendicular to the ab planes*. We explain this anomaly by the interplay between the temperature dependence of the London penetration depth, λ_{ab} and the dimensions of the sample, in the context of the very peculiar interaction between the vortex system and the subsystem of Eu^{2+} spins [18,19]. Vlasko-Vlasov *et al.* [18] performed a study of the magnetic-flux evolution in EuRb1144 using magneto-optical imaging and DC magnetization measurements. They showed that the interplay of magnetic susceptibility amplifying the magnetic induction and vortex pinning attenuating the magnetic flux entry results in a field- and temperature-dependent critical state that emulates a paramagnetic Meissner effect. They further concluded that the observed

vortex dynamics corresponds to a nontrivial spatial current distribution and yields a self-consistent inhomogeneous enhancement of the sample magnetization. Suppression of superconductivity by correlated magnetic fluctuations were also detected by high-resolution scanning Hall probe microscopy [19]. In our explanation of the origin of the anomalous susceptibility response in perpendicular configuration we are extending the concept of paramagnetic Meissner effect described in [18] to the case of penetration of AC perturbation of the mixed state stabilized by a large DC field. In this context, just below T_c the superimposed AC magnetic field disturbs the critical state inside the sample on a scale of London penetration depth, λ_{ab} , resulting in the AC susceptibility response of the sample. The region of the sample not perturbed by the AC excitation (in the center of the sample) we would describe as paramagnetic Meissner state following [18], although it contains Abrikosov vortices (not perturbed by AC magnetic field excitation) as well as oriented Eu^{2+} spins. For a clearer explanation of the anomaly just below T_c we are using the experimental data (detail near the superconducting transition) of the temperature dependence of the in-phase susceptibility χ' in a DC field of 9 T, showed in the left-hand-side of Figure 8. The temperatures indicated in the Figure are T_c (9 T) = 35.8 K, T_1 is a temperature in the first part of increasing diamagnetic signal, T_m is the temperature of the starting of anomalous susceptibility response, T_2 is a temperature where the diamagnetic response decrease with decreasing temperature due to Eu^{2+} spins, T_M is the temperature where the derivative $d\chi'/dT$ changes sign again and T_3 a temperature at which the circulating AC currents result in diamagnetic AC shielding overcoming the paramagnetic Eu^{2+} spins paramagnetic response due to much higher critical current density at this lower temperature. To check our scenario, we have calculated the London penetration depth, $\lambda_{ab}(T)$, shown in the right-hand side of Figure 8.

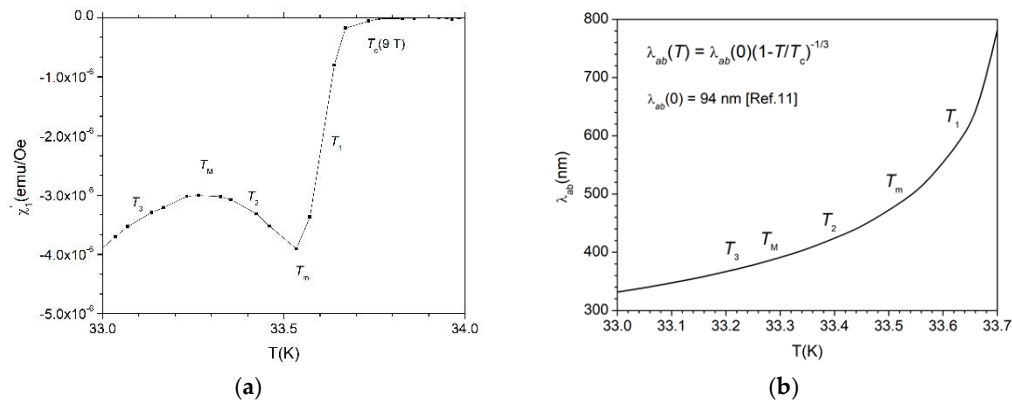


Figure 8. Details of temperature dependence of in-phase susceptibility just below T_c for $H_{DC} = 9\text{ T}$ (left-hand side), and of the calculated penetration depth λ_{ab} (right-hand side). Indicated temperatures are explained in the text.

For the value $\lambda_{ab}(0)$ we have taken the experimental value of 94 nm as determined in [11], while for the functional of temperature dependence of $\lambda_{ab}(T)$ we have used the dependence given by the 3D X-Y critical fluctuations model, $\lambda_{ab}(T) = \lambda_{ab}(0)(1 - T/T_c)^{-1/3}$ that gave the best fit compared with mean-field and two-fluid models, which predicts different dependencies, for both 1144 and 122 IBS systems [20]. The notations of temperatures are the same in the two panels. Please note that half-width of the sample in perpendicular configuration is about 450 nm, while in parallel configuration is about 30 nm, with the London penetration depth $\lambda_c(0) = 160\text{ nm}$ [11].

Figure 9 is a sketch that helps understanding our explanation for the appearance of the anomaly in the AC susceptibility in EuRb1144, only in the perpendicular configuration. Figure 9 (a) describe the penetration of the circulating AC supercurrents induced by the AC field excitation at temperature T_1 indicated in left-hand-side of Figure 8. At this temperature, $\lambda_{ab}(T_1) \approx 600\text{ nm}$ is larger than the half-width of the sample $d/2 \approx 450\text{ nm}$, so the sample is fully penetrated by the AC perturbation, with a small critical current density due to high temperature. Hence, there is the expected increase of the diamagnetic shielding due to the increase of critical current density with decreasing temperature, down to the temperature T_m where $\lambda_{ab}(T_m) \approx d/2 \approx 450\text{ nm}$.

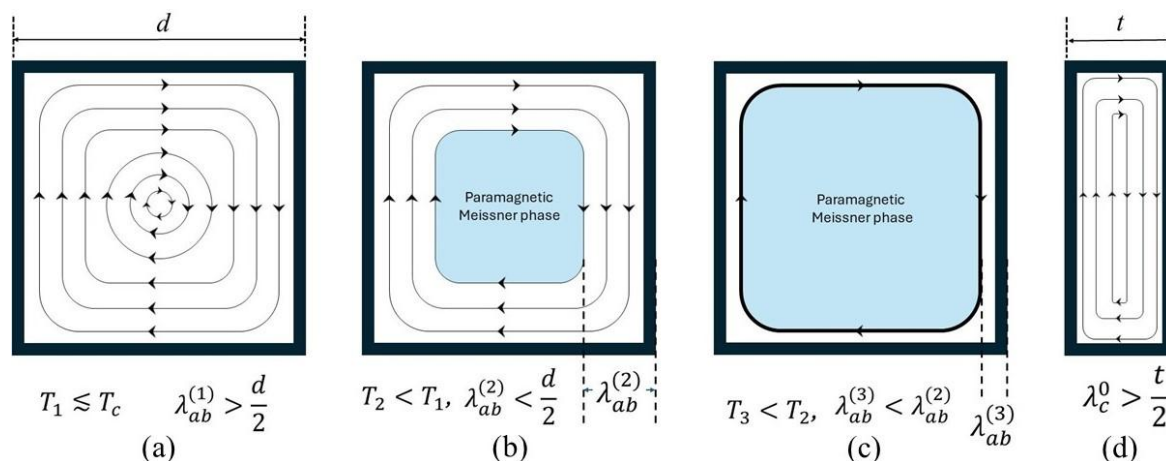


Figure 9. (a-c) Schematics of the circulating supercurrents in perpendicular orientation for three decreasing temperatures, same as those indicated in Figure 8; and (d) in parallel orientation.

With further decrease in temperature, as in Figure 9 (b), at T_2 , $\lambda_{ab}(T_2) \approx 400 \text{ nm} < d/2$ the AC excitation doesn't penetrate all the sample, so in the middle there is the so-called paramagnetic Meissner phase whose dimension expands due to the further decrease of London penetration depth with decreasing temperature. Since the circulating supercurrent is still small due to closeness to T_c , the paramagnetic signal from Eu^{2+} spins increases with decreasing $\lambda_{ab}(T)$ faster than the increase in critical current density, their combination leading to the anomalous decrease of the diamagnetic shielding between T_m and T_M as shown in Figure 8 (left-hand side). Again, please note that the paramagnetic Meissner phase in AC susceptibility is not a true Meissner phase as in the phase diagrams of type 2 superconductors, it indicates just the central region where the AC field perturbation doesn't penetrate. Further decrease in temperature below T_M , at T_3 (Figure 9 (c)) lead to a larger paramagnetic Meissner phase, but now the critical current density becomes large enough for the usual dependence of the diamagnetic shielding.

Figure 9 (d) explains the reason for the absence of the anomaly in the parallel configuration: even at the lowest temperature, the London penetration depth ($\lambda_c(T) > 160 \text{ nm}$) is much larger than the thickness of the crystal, so the AC field penetration in the parallel configuration is complete at all temperatures, and the AC response of the Eu^{2+} spins is not present in the susceptibility measurements.

The last noteworthy aspect resulted from our experiments is the small positive signal of fundamental in-plane AC susceptibility for parallel configuration in the normal phase ($T > T_c$), as seen in Figures 4-7. This is, to our knowledge, the first indication that the interaction of Eu^{2+} spins occurs not only with the Abrikosov vortices, as previously demonstrated, but also with the magnetic flux lines in the normal state. Clearly, the presence of a positive susceptibility signal in the normal phase for fields parallel to the ab planes indicates the fact that there is a plane of easy magnetization. For not too large DC fields in the parallel direction, Eu^{2+} spins can, to a small extent and with the help of thermal energy, follow the AC field excitation which occurs in the ab plane. When the DC field is large, the magnetic interaction between the Eu^{2+} spins and magnetic flux lines is larger and the tentative spins rotation to follow the variation of DC field is slowed down, like the movement of a particle in a medium with increased viscosity. For the perpendicular configuration, the absence of a noticeable positive in-phase susceptibility indicates that tilting of Eu^{2+} spins from the ab plane towards the c -axis is much more difficult. Since the interaction between the Eu^{2+} spins occurs not only with the vortices in the superconducting state but also with the magnetic flux lines in the normal state, a future research direction which will bring great benefits to a better understanding of the complexity of vortex matter, dynamics and pinning in magnetic superconductors will be to employ studies of third-harmonic susceptibility, which already proved great benefits for a better understanding on iron-based superconductors [17].

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Abbreviations

The following abbreviations are used in this manuscript:

MDPI	Multidisciplinary Digital Publishing Institute
IBS	Iron-based superconductors
AE	Alkali-earth metal
A	Alkali metal
AEA1144	AEAF ₄ As ₄
FM	Ferromagnet/Ferromagnetic
ZFC	Zero-field cooling
FC	Field cooling
DC	Direct current
AC	Alternate current
PPMS	Physical Properties Measurement System
ACMS	Alternate Current Measurement System

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