Muon spin rotation measurements on RbEuFe₄As₄ under pressure

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We report muon spin rotation and magnetization measurements on the magnetic superconductor RbEuFe₄As₄ under hydrostatic pressures up to 3.8 GPa. At ambient pressure, RbEuFe₄As₄ exhibits a superconducting transition at $T_c \approx 36.5 \, \mathrm{K}$ and a magnetic transition at $T_m \approx 15 \, \mathrm{K}$ below which the magnetic and the superconducting order coexist. With increasing pressure, T_c decreases while T_m and the ordered Eu magnetic moment increase. In contrast to iron-based superconductors with ordering Fe moments, the size of the ordered Eu moment is not proportional to T_m . The muon spin rotation signal is dominated by the magnetic response impeding the determination of the superconducting properties.

I. INTRODUCTION

In 2009, it was found that isovalent P substitution on the As site in $EuFe_2(As_{1-x}P_x)_2$ suppresses the spin density wave order of the Fe moments and changes the antiferromagnetic order of the Eu moments to ferromagnetic order which coexists with superconductivity for a small substitution range x [1, 2]. There has been a vivid debate how superconductivity and ferromagnetism can coexist in this so-called 122 system. One plausible theory states that the superconducting pairing and the ferromagnetic coupling of the Eu moments through the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction involve different Fe-3d orbitals. While it is mainly the $d_{x^2-y^2}$ and d_{z^2} orbitals, which provide the RKKY coupling, the super conducting pairing is dominated by the d_{yz} and d_{zx} orbitals [3]. In recent years, the intrinsically hole-doped iron-based superconductor RbEuFe₄As₄, an intergrowth of EuFe₂As₂ and RbFe₂As₂, has attracted a significant amount of attention due to its comparably high superconducting transition temperature $T_c \approx 36.5\,\mathrm{K}$ and the coexistence of superconducting and magnetic order below $T_m \approx 15 \,\mathrm{K}$ [4, 5]. The anion heights, i.e. the heights of the As above the Fe plane, are close to the empirical optimum of 1.38 Å [6, 7] to achieve highest T_c [4, 8]. It was shown that the in-plane ferromagnetic order in this compound is associated solely with the Eu magnetic moments that are aligned perpendicularly to the crystallographic c-axis [9]. The three dimensional magnetic structure is still under debate, however, with some studies arguing in favor of ferromagnetic order [4, 9] while a

recent study claims a helical antiferromagnetic structure [10]. The not so common coexistence of magnetism with superconductivity calls for microscopic investigations of RbEuFe₄As₄.

In this work we present a combination of local-probe muon spin rotation and relaxation (μ SR) measurements and magnetization measurements on RbEuFe₄As₄ under hydrostatic pressures up to 3.8 GPa. We find that T_c decreases with pressure while T_m increases, in agreement with data from literature [11, 12]. In addition, our local-probe μ SR measurements show that the ordered magnetic moment increases by about 4% at 2.4 GPa, while T_m increases by 24%. We do not find any signature of a significant coupling between the superconducting and the magnetic order.

II. EXPERIMENTAL METHODS

Polycrystalline RbEuFe₄As₄ was synthesized via a solid-state reaction method [4] and characterized using powder X-ray diffraction (PXRD). μ SR measurements were performed at the Swiss Muon Source (S μ S) using the General Purpose Surface-Muon (GPS) [13] and the General Purpose Decay-Channel (GPD) [14] spectrometers. The data were analyzed with the free software package Musrfit [15]. Magnetization measurements were performed using a commercial vibrating sample magnetometer (VSM) and a superconducting quantum interference device (SQUID) magnetometer. Hydrostatic pressure for the μSR measurements was applied using a double-wall piston cell made from MP35N alloy [14] with Daphne 7373 oil [16] as a pressure transmitting medium. A CuBe anvil-type cell with CuBe gaskets, self-aligning ZrO₂ anvils, and Daphne 7575 oil [17] as a pressure trans-

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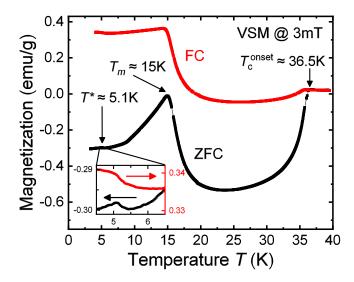


FIG. 1. Magnetization of RbEuFe₄As₄ at ambient pressure measured by vibrating sample magnetometry in 3 mT [cooled in zero field (ZFC) or in applied field (FC)] showing the superconducting transition at T_c and the magnetic transition at T_m . Inset: Magnetization in the temperature range around the impurity related anomaly at T^* for ZFC [black (dark), left axis] and FC [red (light), right axis] measurements.

mitting medium was used for magnetization measurements. Pressures were determined by either In (μ SR) or Pb (SQUID) manometers [18].

III. RESULTS

VSM measurements shown in Fig. 1 confirm a superconducting transition temperature $T_c \approx 36.5\,\mathrm{K}$ and a magnetic transition temperature $T_m \approx 15\,\mathrm{K}$. Further, there is a small anomaly at $T^* \approx 5.1\,\mathrm{K}$ (inset Fig. 1). This anomaly was previously observed by magnetization and heat capacity measurements [4] and was later realized to be likely due to very small amounts of $\mathrm{Eu_3O_4}$ impurities [19] which order antiferromagnetically at $T_N \approx 5\,\mathrm{K}$ [20].

uSR measurements, which require a comparably large amount of sample when performed under pressure, were carried out on 1.65 g of RbEuFe₄As₄ with 6.4% RbFe₂As₂ and 6.7% EuFe₂As₂ impurities. A small amount of $\mathrm{Eu_3O_4}$ impurity below the detection limit of the characterizing PXRD measurements was presumably present too, given the anomaly at T^* mentioned before. Representative zero-field (ZF) μ SR spectra recorded with no external magnetic field applied are shown in Fig. 2 for temperatures above and below $T_m \approx 15\,\mathrm{K}$. Below the magnetic transition temperature, spontaneous muon spin precession can be observed due to the static long range magnetic order. The data were analysed using two different models for the temperatures above and below T_m . Above T_m , a simple phenomenological model was applied:

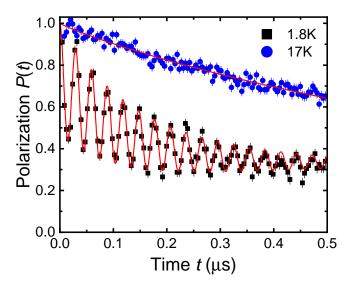


FIG. 2. Representative zero-field μSR spectra measured at ambient pressure above and below the magnetic transition temperature $T_m \approx 15\,\mathrm{K}$. The oscillations at low temperature indicate static long range magnetic order. The red lines are fits using the models introduced in Eqs. (1) and (2).

$$P_{\rm HT}(t) = (1 - f_1) \exp[-(\lambda_{\rm HT} t)^{\beta}] + f_1,$$
 (1)

where $\lambda_{\rm HT}$ is the relaxation rate and β is a stretching exponent. f_1 is a small nonrelaxing tail fraction due to the already present static magnetic order of the EuFe₂As₂ impurity [21–23]. The data below T_m were modelled by:

$$P_{\rm LT}(t) = \frac{2}{3} [f_{\rm osc} \cos(\gamma_{\mu} B_{\rm int} t) \exp(\lambda_T t) + (1 - f_{\rm osc}) \exp(\lambda_{\rm no} t)] + \frac{1}{3} \exp(\lambda_L t),$$
(2)

where the 2/3 (transverse) and 1/3 (longitudinal) components reflect the powder average of the internal fields with respect to the initial muon spin direction in a the polycrystalline sample. The transverse part consists of an oscillating fraction $f_{\rm osc} \approx 0.4$ and a nonoscillating fraction $f_{\rm no}=1-f_{\rm osc}\approx$ 0.6. $\lambda_T,~\lambda_{\rm no},~{\rm and}~\lambda_L$ are the corresponding relaxation rates. The oscillation frequency is given by $\gamma_{\mu}B_{\rm int}$, where $\gamma_{\mu}=2\pi\times135.5\,{\rm MHzT^{-1}}$ is the muon's gyromagnetic ratio and B_{int} is the magnetic field at the muon stopping site. The latter is proportional to the ordered magnetic moment and therefore a measure of the magnetic order parameter. The oscillating signal from the few percent of magnetically ordered EuFe₂As₂ and Eu₃O₄ impurities was not included in the analysis as it was too small to be resolved. Possible contributions from the impurity phases are absorbed by the last two terms of Eq. 2. An influence on the determination of $B_{\rm int}$ is very unlikely due to the significantly higher internal fields in EuFe₂As₂ [23, 24] and the very small amount

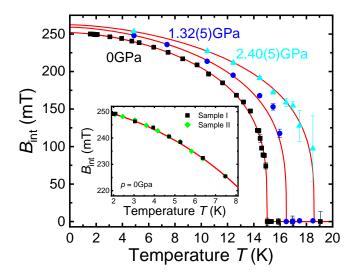


FIG. 3. Temperature dependence of the internal magnetic field $B_{\rm int}$ at the muon stopping site for different pressures. $B_{\rm int}$ is proportional to the ordered magnetic moment and therefore a measure of the magnetic order parameter. The red lines are fits using the model described in Eq. (3). Inset: No anomaly in $B_{\rm int}$ is observed around T^* .

of Eu_3O_4 . In the case of measurements under hydrostatic pressure, the signal from muons stopping in the pressure cell was treated in analogy to Ref. [14].

Fig. 3 shows the temperature dependence of B_{int} for different pressures. The red lines are fits using the phenomenological function [24]:

$$B_{\rm int} = B_{\rm int,0} (1 - (T/T_m)^{\alpha})^{\gamma}, \tag{3}$$

where $B_{\rm int,0}$ is the field at zero temperature. $\alpha =$ 1.63(4) and $\gamma = 0.29(1)$ were determined from the ambient pressure data and fixed for the fit of the pressure The magnetic transition temperature increases data. monotonically with pressure in agreement with literature data [11, 12]. Simultaneously, our ZF μ SR measurements show that the ordered magnetic moment is enhanced. At 2.4 GPa, the increase amounts to about 24% for the transition temperature T_m , but only about 4% for the magnetic moment. The inset of Fig. 3 includes data measured on a second batch of RbEuFe₄As₄ and focuses on $B_{\rm int}$ in the temperature region around the feature at $T^* \approx 5.1 \,\mathrm{K}$ observed by VSM (inset Fig. 1). The lack of an anomaly in the temperature dependencies of the internal field $B_{\rm int}$ and the transverse relaxation rate λ_T (Fig. 4) rules out a change in the magnetic structure like a spin reorientation and therefore supports the notion of impurities as a cause for this feature.

The fraction $f_{\rm no}=1-f_{\rm osc}$ in Eq. (2) describes those parts of the sample that are magnetic but too disordered to exhibit coherent muon spin oscillations (correlation length smaller than approximately 10 lattice constants [25]). The corresponding relaxation rate $\lambda_{\rm no}$ sharply increases to about $9\,\mu{\rm s}^{-1}$ within the first Kelvin below T_m

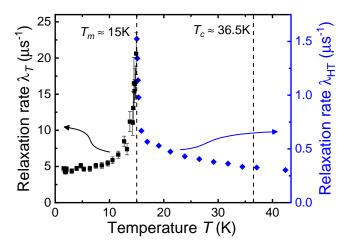


FIG. 4. Temperature dependence of the muon spin relaxation rates obtained by fitting the zero-field μ SR data measured at ambient pressure to Eqs. (1) and (2).

and stays roughly constant at this value for lower temperatures (not shown). The longitudinal relaxation rate λ_L (not shown), which can be nonzero only for dynamic systems, drops quickly to zero below T_m , indicating that the whole volume of the sample, including $f_{\rm no}$, is static below T_m . In the temperature region between T_m and T_c , magnetic fluctuations lead to a sizable and temperature dependent relaxation rate $\lambda_{\rm HT}$ (Fig. 4) which renders an investigation of the superconducting properties of RbEuFe₄As₄ by the means of μ SR unfeasible.

We therefore employed SQUID magnetometry to determine the pressure dependence of T_c and to further investigate the magnetic transition. Magnetization data

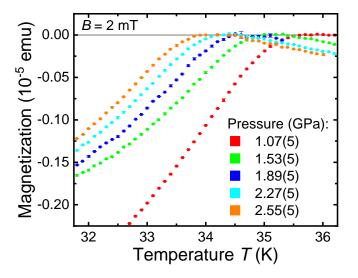


FIG. 5. Magnetization of a small grain of RbEuFe₄As₄ as a function of temperature for a representative series of pressures measured by SQUID magnetometery in a field of $2\,\mathrm{mT}$. For all measurements, the sample was cooled in zero field. The pressure cell background was subtracted and the data were shifted vertically to overlap around T_c .

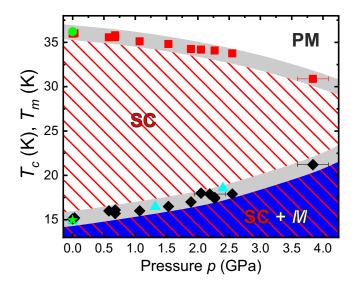


FIG. 6. Temperature-pressure phase diagram of RbEuFe₄As₄. Superconducting transition temperatures T_c were measured by SQUID magnetometry (\blacksquare) and vibrating sample magnetometry (\blacksquare). Magnetic transition temperatures T_m were measured by SQUID magnetometry (\spadesuit), vibrating sample magnetometry (\bigstar), and zero-field muon spin rotation measurements (\blacktriangle). With increasing hydrostatic pressure, T_c decreases while T_m increases. PM and SC denote the paramagnetic and the superconducting phase respectively. SC+M denotes the region of coexisting superconducting and magnetic order. The gray shaded areas are guides to the eye.

for the temperature range around T_c are shown in Fig. 5 for a representative series of pressures. The cell background was subtracted and the data were shifted vertically to overlap around the transition. T_c was determined by the intersection of two linear approximations of the data above and below the transition. The same method was used to determine T_m (not shown). The results are presented in the temperature-pressure phase diagram shown in Fig. 6, together with the transition temperatures obtained by ZF μ SR and VSM.

IV. DISCUSSION

Under hydrostatic pressure, the superconducting transition temperature T_c of RbEuFe₄As₄ decreases monotonically, while the magnetic transition temperature T_m increases (Fig. 6), in agreement with literature data [11, 12]. Additionally, our ZF μ SR measurements reveal sizable magnetic fluctuations already above T_m and show that the ordered magnetic moment also increases under hydrostatic pressure. At 2.4 GPa, T_m is enhanced by about 24% compared to ambient pressure, but the ordered magnetic moment increases by only about 4%. This nonproportional relation sets RbEuFe₄As₄ apart from magnetic members of the 122 and other families of iron-based superconductors where a proportional scaling of the two quantities was found [26, 27]. In these

systems, the magnetic order is usually associated with Fe moments, whereas in $RbEuFe_4As_4$ magnetism is due to the ordering of Eu moments [9], which might explain the different scalings.

Despite the seemingly antagonistic behavior of the superconducting and the magnetic state in RbEuFe₄As₄ our findings imply that there is no significant coupling between the two orders in agreement with Refs. [10, 12, 28, 29]. The rate of decrease in T_c is comparable to nonmagnetic CaKFe₄As₄ under pressure [30]. Therefore, the suppression of superconductivity is unlikely due to the enhanced magnetic order but rather caused by the pressure induced changes of the lattice parameters [11] which likely drive the anions away from the optimal height value. Further, substitution studies show that the superconducting and the magnetic order can be suppressed independent of each other [19, 28]. This is in agreement with various mechanisms proposed to explain the coupling among the Eu moments, namely the so-called d-f [31] interaction or As-Eu-As superexchange interactions as proposed in Ref. [19] or an RKKY interaction that involves different Fe-3d orbitals than the superconducting pairing as proposed in Refs. [3, 4]. Subtle effects like small influences of the onset of magnetic order on the vortex lattice like described in Refs. [32, 33] are not detectable by μSR however due to the dominance of the magnetic signal.

Neither the magnetic order parameter measured via the zero-field muon spin precession frequency nor the muon spin relaxation rates, which reflect the field distribution at the muon stopping sites, exhibit anomalies in the low temperature region (inset of Fig. 3, Fig. 4). The anomaly at $T^* \approx 5.1 \,\mathrm{K}$ in magnetization measurements (inset Fig. 1) is therefore clearly not related to the magnetic order in RbEuFe₄As₄. This supports the attribution to small Eu₃O₄ impurities mentioned in Ref. [19]. Eu₃O₄ represents the most likely impurity, not only due to the antiferromagnetic order below $T_N \approx 5 \,\mathrm{K}$ [20], but also due to the fact that the T^* anomaly is reported for the magnetic superconductor CsEuFe₄As₄ as well [34], but not for the Eu free members of the 1144 family. In contrast, a connection to the EuFe₂As₂ or the RbFe₂As₂ impurity phase seems unlikely since no features are reported around T^* in the literature for these compounds [21–23, 35, 36].

V. CONCLUSION

In conclusion, we have shown that the superconducting order in RbEuFe₄As₄ is suppressed by the application of hydrostatic pressure while the magnetic order is enhanced. The relation between the magnetic transition temperature T_m and the size of the ordered magnetic moment is not proportional, setting RbEuFe₄As₄ apart from magnetic members of the 122 and other families of iron-based superconductors, where a proportional scaling of the two quantities was found [26, 27]. No significant

coupling between the magnetic and the superconducting order was found in agreement with earlier reports [10, 12, 28, 29].

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