Anisotropy induced vortex lattice rearrangement in CaKFe$_4$As$_4$

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(Received 6 February 2019; published 19 April 2019)

The magnetic penetration depth anisotropy $\gamma_\lambda = \lambda_\parallel / \lambda_{ab}$ ($\lambda_{ab}$ and $\lambda_\parallel$ are the in-plane and the out-of-plane components of the magnetic penetration depth) in a CaKFe$_4$As$_4$ single crystal sample (the critical temperature $T_c \simeq 35$ K) was studied by means of muon-spin rotation ($\mu$SR). $\gamma_\lambda$ is almost temperature independent for $T \lesssim 20$ K ($\gamma_\lambda \simeq 1.9$) and it reaches $\simeq 3.0$ by approaching $T_c$. The change of $\gamma_\lambda$ induces the corresponding rearrangement of the flux line lattice (FLL), which is clearly detected via enhanced distortions of the FLL $\mu$SR response. A comparison of $\gamma_\lambda$ with the anisotropy of the upper critical field ($\gamma_{H_{c2}}$) studied by Meier et al. [Phys. Rev. B 94, 064501 (2016)] reveals that $\gamma_\lambda$ is systematically higher than $\gamma_{H_{c2}}$ at low temperatures and approaches $\gamma_{H_{c2}}$ for $T \rightarrow T_c$. The anisotropic properties of $\lambda$ are explained by the multigap nature of superconductivity in CaKFe$_4$As$_4$ and are caused by anisotropic contributions of various bands to the in-plane and the out-of-plane components of the superfluid density.

DOI: 10.1103/PhysRevB.99.140507

In the majority of superconducting compounds discovered so far, the crystal structure, as well as the electronic and phononic band structures, are all far from being isotropic. Anisotropic superconductors are usually treated within the phenomenological anisotropic Ginzburg-Landau (AGL) theory [1,2], which follows from the isotropic Ginzburg-Landau (GL) approach via replacement of the effective mass $m^*$ in the GL free energy functional by an effective mass tensor, with values $m^*_{ab}$, $m^*_{c}$, and $m^*_{\parallel}$ along the principal $a$, $b$, and $c$ axes [3,4]. In the most usual case of uniaxial anisotropy, all anisotropies are incorporated into the single parameter [5],

$$\gamma = \gamma_\lambda \equiv \frac{\lambda_c}{\lambda_{ab}} = \sqrt{\frac{m^*_{c}}{m^*_{ab}}} = \frac{H^*_{c2}}{H^*_{c2}} = \frac{\xi_{ab}}{\xi_c}, \quad (1)$$

Here, $\gamma_\lambda$ and $\gamma_{H_{c2}}$ are the anisotropy of the magnetic penetration depth ($\lambda$) and the upper critical field ($H_{c2}$), respectively, and $\xi$ is the coherence length. In AGL theory the same effective mass tensor determines the anisotropy of $\lambda$ and $H_{c2}$, thus making both $\gamma_\lambda$ and $\gamma_{H_{c2}}$ temperature and field independent [6].

Note, however, that the GL theory is strictly valid only for $T \rightarrow T_c$. Following Kogan [7], away from $T_c$, the theoretical approach for calculating $H_{c2}$ (the position of the second-order phase transition in high fields) has little in common with the evaluation of $\lambda$, so that the anisotropies $\gamma_\lambda$ and $\gamma_{H_{c2}}$ are not the same and can be substantially different. In MgB$_2$, e.g., both anisotropies approach a common value $\gamma_\lambda = \gamma_{H_{c2}} \simeq 1.75$ at $T_c$ and become $\gamma_\lambda \simeq 1.2$ and $\gamma_{H_{c2}} \simeq 6$ at low temperatures [8–10]. Different $T$ dependencies of $\gamma_\lambda$ and $\gamma_{H_{c2}}$ have also been reported for various cuprate and Fe-based superconductors [11–19].

It is worth mentioning here that the change of $\gamma_\lambda$ would necessarily lead to a corresponding flux line lattice (FLL) rearrangement. As follows from Fig. 1(a), for the external field $B_{ex}$ applied along the $ab$ plane ($B_{ex} \parallel ab$), the increase of $\gamma_\lambda$ shortens the distance between vortices within the $ab$ plane and enhances the intervortex distances in the $c$ direction. The displacement of vortex lines could be quite big. The simple estimate reveals that the increase of $\gamma_\lambda$ from 1.0 to 3.0 leads to a decrease/increase of the in-plane/out-of-plane intervortex distances by almost half of the isotropic FLL unit cell [see Fig. 1(a)]. This effect can be studied, e.g., by using techniques visualizing the vortex distribution in superconducting materials, such as small-angle neutron scattering (SANS), tunneling, or magnetic decoration. The important limitation comes, however, from the sample size and morphology. Most of the available good quality single crystals are very thin along the crystallographic $c$ direction, and a good surface preparation (needed for magnetic decoration and tunneling experiments) in $B_{ex} \perp c$ orientation is challenging.

In this Rapid Communication, we report on measurements of the magnetic penetration depth anisotropy $\gamma_\lambda = \lambda_c/\lambda_{ab}$ in a CaKFe$_4$As$_4$ single crystal sample ($T_c \simeq 35$ K) by means of muon-spin rotation ($\mu$SR). A comparison of $\gamma_\lambda$ with $\gamma_{H_{c2}}$ studied in Ref. [21] shows that $\gamma_\lambda$ is higher than $\gamma_{H_{c2}}$ all the way up to $T_c$. Only in the narrow region close $T_c$, $\gamma_\lambda = \gamma_{H_{c2}} \simeq 3.0$ [Fig. 1(b)]. The change of $\gamma_\lambda$ with temperature induces the corresponding rearrangement of the flux line order, which was detected via the enhanced distortions of the FLL $\mu$SR response. The temperature dependencies of the in-plane and the out-of-plane components of the superfluid density ($\lambda_{ab}^2$ and $\lambda_c^{-2}$) were found to be well described within the two-gap scenario, thus suggesting that the multiple-band nature of superconductivity in CaKFe$_4$As$_4$ is well pronounced for both $c$ and $ab$ directions.

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FIG. 1. (a) The contour plot of the field variation within the triangular vortex lattice of an anisotropic superconductor with $B_{\text{ex}} \parallel ab$ ($B_{\text{ex}} = 11 \text{ mT}$, $\gamma_\lambda = \lambda_c/\lambda_{ab} = 1.0$, 2.0, and 3.0; see the Supplemental Material [20]). $B_{\text{min}}$, $B_{\text{max}}$, and $B_{\text{sad}}$ are the minimum, maximum, and the saddle point fields. (b) The anisotropies $\gamma_\lambda$ and $\gamma_{H_{c2}}$ of CaKFe$_4$As$_4$. Solid circles are $\gamma_\lambda$ points from the present study. The solid and open squares correspond to $\gamma_{H_{c2}}$ ($T$) from Ref. [21], as obtained by using the “onset” and “offset” criteria of $T_c$ determination, respectively. The solid red line is the fit of the two-gap model [Eq. (2)] to $\lambda_{ab}^2(T)$ and $\lambda_{c}^2(T)$ data.

A CaKFe$_4$As$_4$ single crystal with dimensions of $\approx 4.0 \times 4.0 \times 0.1 \text{ mm}^3$ was grown from a high-temperature Fe-As rich melt [21,22], and it is characterized via magnetization measurements (see the Supplemental Material [20]). The $\mu$SR measurements were carried out at the $\pi M3$ beam line using the GPS spectrometer (Paul Scherrer Institute, Switzerland) [23]. The zero-field (ZF) and transverse-field (TF) $\mu$SR measurements were performed at temperatures from $\approx 1.5$ to 50 K. In two sets of TF-$\mu$SR experiments the external magnetic field ($B_{\text{ex}} \approx 10.8 \text{ mT}$) was applied parallel to the $ab$ plane and the crystallographic $c$ axis of the crystal, respectively. A fraction of the TF-$\mu$SR data for $B_{\text{ex}} \parallel c$ was previously reported in Ref. [24]. ZF-$\mu$SR data are discussed in the Supplemental Material [20]. A special sample holder designed to measure thin samples by means of $\mu$SR was used [25]. The experimental data were analyzed using the MUSRFIT package [26].

The homogeneity of the superconducting state in CaKFe$_4$As$_4$ was checked by performing a series of field-shift experiments. Figure 2 exhibits the fast Fourier transform of the TF-$\mu$SR time spectra, which reflects the internal field distribution $P(B)$. Figures 2(a)–2(c) correspond to $B_{\text{ex}} \parallel ab$ and Figs. 2(d)–2(f) to $B_{\text{ex}} \parallel c$ sets of measurements, respectively. The solid red symbols are $P(B)$’s obtained by following FCW and FCC protocols at $B_{\text{ex}} \approx 10.8 \text{ mT}$ (see text for details). The blue solid symbols are $P(B)$’s after the field shift down to $\approx 8 \text{ mT}$. Dashed lines correspond to the applied field ($10.8 \text{ mT}$) and to the “shifted” field (8.0 mT), respectively. From the data presented in Fig. 2, the following three important points emerge: (i) The main part of the signal, accounting for approximately 90% of the total signal amplitude, remains unchanged within the experimental error after a field shift. Only the sharp peak ($\approx 10\%$ of the signal amplitude) follows exactly the applied field. It is attributed, therefore, to the residual background signal from muons missing the sample (see also Refs. [25,28]). (ii) The asymmetric $P(B)$ distributions shown in Fig. 2 possess the basic features
expected for a well aligned vortex lattice, i.e., the cutoff at low fields \(B_{\text{min}}\), the peak arising from the saddle point midway between two adjacent vortices \(B_{\text{sad}}\), and a long tail towards high fields caused by regions around the vortex core \(B_{\text{max}}\) [24,29]. Note that the definition of a “well aligned” FLL is different, e.g., for \(\mu SR\) and SANS experiments. In the \(\mu SR\) case, the field at the muon stopping site is determined by the local flux line arrangement (within a couple of FLL unit cells). The SANS experiment collects reflections from flux-line planes, which requires a long-range arrangement of FLL. Following scanning tunneling microscopy (STM) results, the FLL in CaKFe\(_4\)As\(_4\) is well arranged on the short length scale, while the long-range arrangement is missing [see Figs. 3(d)–3(f) in Ref. [30]]. (iii) The \(P(B)\) curves obtained by following the FCC and FCW protocols coincide for both field orientations and within the full temperature range studied. To conclude, the above experiment demonstrates that the FLL in CaKFe\(_4\)As\(_4\) sample is well arranged and strongly pinned (at least at \(B_{\text{ex}}\simeq 11\) mT). The strong pinning in CaKFe\(_4\)As\(_4\) was also reported for \(B_{\text{ex}}\parallel c\) orientation in STM experiments [30], and for both \(B_{\text{ex}}\parallel ab\) and \(B_{\text{ex}}\parallel c\) sets of data in magnetization studies [31].

The TF-\(\mu SR\) data were analyzed by fitting a three-component expression to the time evolution of the muon-spin polarization (see the Supplemental Material [20]). The superconducting response of the CaKFe\(_4\)As\(_4\) sample was further obtained within the framework of the so-called momentum approach, which includes the calculations of the first moment \(\langle B \rangle\), and the second- \(\langle \Delta B^2 \rangle\) and third-central moments \(\langle \Delta B^3 \rangle\) of the magnetic field distribution function \(P(B)\) (see the Supplemental Material [20] for details). Following Ref. [32], the first moment (the mean field) scales with the sample magnetization. The second moment (the broadening of the signal), \(\langle \Delta B^2 \rangle = \langle B^2 \rangle + \sigma_{\text{rms}}^2\), contains contributions from the vortex lattice \(\langle B^2 \rangle\) and the nuclear dipole field \(\sigma_{\text{rms}}\), as is obtained from \(T > T_c\) measurements. In extreme type-II superconductors \((\lambda \gg \xi)\) and for fields \(B_{\text{ex}} \ll \mu_0 H_{\text{c2}}\), \(\langle \Delta B^2 \rangle \propto \lambda^{-4}\) [33,34]. The third moment accounts for the asymmetric shape of \(P(B)\), which is described via the skewness parameter \(\alpha_{\text{sk}} = \langle \Delta B^3 \rangle / \langle \Delta B^2 \rangle^{3/2}\). In the limit of \(\lambda \gg \xi\) and for realistic measurement conditions, \(\alpha_{\text{sk}} \simeq 1.2\), for a well arranged triangular vortex lattice [35]. It is very sensitive to structural changes of the vortex lattice which may occur as a function of temperature and/or magnetic field [35–40].

Figure 3 shows temperature dependencies of the skewness parameter \(\alpha_{\text{sk}}\) measured in \(B_{\text{ex}}\parallel c\) and \(B_{\text{ex}}\parallel ab\) sets of experiments. The temperature dependencies of the first moment \(\langle B \rangle\) and the second-central moment \(\langle \Delta B^2 \rangle\) are presented in the Supplemental Material [20]. Following Fig. 3, \(\alpha_{\text{sk}}\) stays temperature independent. Only in the close vicinity to \(T_c\), \(\alpha_{\text{sk}}\) decreases by approximately 8% (from \(\simeq 1.1\) to \(\simeq 1.03\)). This, together with the relatively high value of \(\alpha_{\text{sk}}\) \(\simeq 1.1\), suggests that in \(B_{\text{ex}}\parallel c\) orientation the FLL in CaKFe\(_4\)As\(_4\) is well developed and has little distortion. The presence of well arranged FLL is also confirmed in “field-shift” experiments [Figs. 2(d)–2(f)] showing asymmetric \(P(B)\)’s with all characteristic features attributed to FLL.

The temperature evolution of \(\alpha_{\text{sk}}\) is, however, different from \(\alpha^0_{\text{sk}}(T)\) (Fig. 3). By increasing temperature, \(\alpha^0_{\text{sk}}\) stays almost constant up to \(T \simeq 20\) K \((\alpha^0_{\text{sk}} \simeq 1.07)\) and it decreases down to \(\simeq 0.7\) by approaching \(T_c\). In general, the change of \(\alpha_{\text{sk}}\) as a function of magnetic field and temperature is associated to \(\gamma_\lambda\) for \(T \geq 20\) K is caused by a \(\gamma_\lambda\) induced rearrangement of the FLL. The change of \(\gamma_\lambda\), leading necessarily to the movement of the vortex lines [Fig. 1(a)], enhances the pinning caused FLL distortions and, as a consequence, reduces the value of the skewness parameter \(\alpha_{\text{sk}}\).

The \(T\) dependence of \(\gamma_\lambda\) was further studied via measurements of the temperature evolutions of the in-plane \(\langle \lambda^2 \rangle_\parallel\) and the out-of-plane \(\langle \lambda^2 \rangle_\perp\) superfluid density components (Fig. 4). The anisotropy parameter \(\gamma_\lambda = \lambda_\parallel / \lambda_\perp\) is presented in Fig. 1(b). Obviously, \(\gamma_\lambda\) in CaKFe\(_4\)As\(_4\) is temperature dependent. It is almost constant \((\gamma_\lambda \simeq 1.9)\) for \(T \lesssim 20\) K and increases up to \(\simeq 3.0\) by approaching \(T_c\). More remarkable is that \(\gamma_\lambda(T)\) follows the temperature evolution of the skewness parameter \(\alpha_{\text{sk}}\). Both \(\gamma_\lambda(T)\) and \(\alpha_{\text{sk}}\) are constant for \(T \lesssim 20\) K and change almost linearly in the \(25\) K \(\lesssim T < T_c\) region [Figs. 1(b) and 3]. These suggest that the enhancement of FLL distortions, seen via a decrease of \(\alpha_{\text{sk}}\) (Fig. 3), is caused by the vortex lattice rearrangement [Fig. 1(a)], which, in its turn, is initiated by the temperature dependent anisotropy coefficient \(\gamma_\lambda\) [Fig. 1(b)].
The comparison of \( \gamma \), obtained in our study with \( \gamma_{H_{2}} \) reported in Ref. [21] shows that these anisotropies coincide with each other (\( \gamma \approx \gamma_{H_{2}} \approx 3.0 \)) only for \( T \rightarrow T_{c} \) [Fig. 1(b)], thus resembling the situation in the famous two-gap superconductor MgB\(_{2}\) [8–10]. This may suggest that the range of applicability of AGL to CaKFe\(_{4}\)As\(_{4}\) is confined within a narrow range close to \( T_{c} \), in full analogy with MgB\(_{2}\), where the AGL description was found to be limited to temperatures less than 2\% away from \( T_{c} \) [42]. An alternative approach, based on microscopic Eilenberger calculations, reveals that in two-gap superconductors (as, e.g., MgB\(_{2}\)) \( \gamma \), and \( \gamma_{H_{2}} \) have different temperature dependencies and cross only at \( T_{c} \) [43]. Which of these approaches better describes the anisotropic properties of CaKFe\(_{4}\)As\(_{4}\) needs further investigation.

The temperature dependencies of \( \lambda_{ab}^{-2} \) and \( \lambda_{c}^{-2} \) (Fig. 4) were further analyzed within the framework of the phenomenological \( \alpha \) model by decomposing the \( \lambda^{-2}(T) \) into two components [12,41,44].

\[
\frac{\lambda^{-2}(T)}{\lambda^{-2}(0)} = \omega \frac{\lambda_{ab}^{-2}(T, \Delta_{1})}{\lambda_{ab}^{-2}(0, \Delta_{1})} + (1 - \omega) \frac{\lambda_{c}^{-2}(T, \Delta_{2})}{\lambda_{c}^{-2}(0, \Delta_{2})}
\]

(2)

Here, \( \lambda(0) \) is the value of the penetration depth at \( T = 0 \), \( \Delta_{i} \) is the zero-temperature value of the \( i \)th gap \((i = 1 \text{ or } 2)\), and \( \omega \) \((0 \leq \omega \leq 1)\) is the weight of the larger gap to \( \lambda^{-2} \). Each component in Eq. (2) was further evaluated by \( \lambda^{-2}(T, \Delta) / \lambda^{-2}(0, \Delta) = 1 + \beta_{i} f \int_{\Delta(T)}^{\infty} (\delta f / \delta E) E / \sqrt{E^{2} - \Delta(T)^{2}} dE \) [5]. Here, \( f = [1 + \exp(E / k_{B}T)]^{-1} \) is the Fermi function and \( \Delta(T) = \tanh[1.82(1/0.181(t - 1))^{0.51}] \) [41]. Note that this equation is valid within the clean limit, which is indeed the case for CaKFe\(_{4}\)As\(_{4}\) [21]. The solid lines in Fig. 4 are fits of Eq. (2) to the \( \lambda_{ab}^{-2}(T) \) and \( \lambda_{c}^{-2}(T) \) data. The fit parameters are \( \lambda_{ab}^{-2}(0) = 23.4 (\mu m^{-2}), \Delta_{1,ab} = 10.0 \text{ meV}, \Delta_{2,ab} = 2.5 \text{ meV}, \omega_{ab} = 0.64, \) and \( \lambda_{c}^{-2}(0) = 6.1 (\mu m^{-2}), \Delta_{1,c} = 6.0 \text{ meV}, \Delta_{2,c} = 3.5 \text{ meV}, \omega_{c} = 0.57 \) for \( \lambda_{ab}^{-2}(T) \) and \( \lambda_{c}^{-2}(T) \) data sets, respectively.

From the results of the two-gap model fit to \( \lambda_{ab}^{-2}(T) \) and \( \lambda_{c}^{-2}(T) \), three important points emerge: (i) The “theoretical” two-gap \( \gamma(T) \) curve stays in good agreement with the experimental data [see Fig. 1(b)]. (ii) The density functional theory calculations suggest that in CaKFe\(_{4}\)As\(_{4}\), ten bands (six hole- and four electronlike bands) having different zero-temperature gap values cross the Fermi level [45,46]. Recent angle resolved photoemission spectroscopy (ARPES) [45], as well as the combination of ARPES and \( \mu \)SR experiments [24], confirm this statement. This suggests, on the one hand, that the two-gap approach described by Eq. (2) is oversimplified and contributions of all ten bands need to be included. Such an analysis requires, however, an exact knowledge of the electronic band structure [24,47]. On the other hand, similar two-gap approaches were previously used in Refs. [21,48,49] and allowed to describe satisfactorily the in-plane superfluid density data of CaKFe\(_{4}\)As\(_{4}\). (iii) The differences in the parameters obtained from the two-gap fit to \( \lambda_{ab}^{-2}(T) \) and \( \lambda_{c}^{-2}(T) \) are most probably caused by anisotropic contributions of various bands to the superfluid density. In such cases \( \Delta_{1} \) and \( \Delta_{2} \) refer to averaged gaps for a series of bands with “big” and “small” gap values, respectively (see, e.g., Ref. [46]), while \( \omega \) corresponds to the averaged weight of “big” gaps to \( \lambda^{-2}(0) \).

To conclude, the magnetic penetration depth anisotropy \( \gamma = \lambda_{c} / \lambda_{ab} \) in a CaKFe\(_{4}\)As\(_{4}\) single crystal sample was studied by means of the muon-spin rotation. \( \gamma_{H_{2}} \) is temperature independent for \( T < 20 \text{ K} \) (\( \gamma_{H_{2}} \approx 1.9 \)) and it increases up to \( \gamma_{H_{2}} \approx 3.0 \) for \( T \rightarrow T_{c} \). The change of \( \gamma_{H_{2}} \) induces the flux line lattice rearrangement, which is clearly detected via the enhanced distortions of the FLL, \( \mu \)SR response. The anisotropic properties of \( \lambda \) are explained by the multigap nature of superconductivity in CaKFe\(_{4}\)As\(_{4}\) and caused by anisotropic contributions of various bands to the in-plane and the out-of-plane components of the superfluid density.

The work was performed at the Swiss Muon Source (S\(_{\mu}\)S), Paul Scherrer Institute (PSI, Switzerland). The authors acknowledge helpful discussions with V. G. Kogan. W.R.M. was funded by the Gordon and Betty Moore Foundation EPiQS Initiative through Grant No. GBMF4411. Work at Ames Laboratory was supported by the U.S. Department of Energy, Office of Science, Basic Energy Sciences, Materials Science and Engineering Division. Ames Laboratory is operated for the U.S. DOE by Iowa State University under Contract No. DE-AC02-07CH11358.

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In $B_{ex} \parallel c$ experiments, the external field $B_{ex} = 10.8$ mT is applied perpendicular to the flat surface of the sample. Due to big demagnetization effects in such geometry (the demagnetization factor $N \approx 0.98$ [59]), the vortex lattice forms at fields higher than $\mu_0B_{c1,0} ^{lab} \approx 0.4$ mT ($\mu_0H_{c1,0} ^{lab} \approx 20.8$ mT [20]). In $B_{ex} \parallel ab$ experiments, $B_{ex} = 10.8$ mT is slightly smaller than $\mu_0B_{c1,0} ^{lab} \approx 12.2$ mT [20]. However, due to the presence of a strong pinning in CaKFe$_2$As$_2$ [30,31], the “rigid” FLL sets in slightly below $T_c$, where $B_{ex} \gg \mu_0H_{c1,0} ^{lab} (T \approx T_c) \approx 0$, and remains unchanged by further cooling down the sample.

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