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Inversion of the upper critical field anisotropy in FeTeS films

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Abstract

We present the complete superconducting upper critical field (H_{c2}) – temperature (T) diagram of FeTeS films measured at three crystalline orientations (**H**||c, 45° and ab). We find that H_{c2} is *almost* isotropic in magnetic field orientation with $\mu_0 H_{c2}(T=0)$~30T, and a transition temperature of T_c~7K. A small but clear H_{c2} angular anisotropy is observed, with a crossover around $T = 0.7\ T_c$, from $H_{c2}(||c)<H_{c2}(||ab)$ for $T>0.7T_c$ to $H_{c2}(||c)>H_{c2}(||ab)$ for $T<0.7T_c$. This change in the anisotropy is similar to that observed in FeTeS and FeTeSe single crystals but occurs at a higher T/T_c for our film. We analyze the $H_{c2}(T)$ in terms of pair-breaking mechanisms and two-band superconductor theory. Understanding the inversion of H_{c2}, opens the possibility to adjust the effective anisotropy of superconductors for different applications.

1. Introduction

Much effort has been put into studying the upper critical field (H_{c2}) of iron-based superconductors [1-5]. Their very high H_{c2} values lead research to focus into possible high field applications. The exploration of H_{c2} gave valuable information on the electronic band structure, suggesting from early on, by analogy with MgB_2, that pnictide superconductors have multiple bands contributing to superconductivity [2,6]. Single crystals and epitaxial thin films also allowed for measurements in different crystalline orientations and showed that (especially for the $BaFe_2As_2$ family) a low effective anisotropy [$\gamma_H = H_{c2}(||ab)/H_{c2}(||c)$] is possible, with values of γ_H~2 near T_c. It was also seen that γ_H decreases monotonically as a function of decreasing temperature (T), suggesting an isotropic superconductor at low temperatures [2-5]. Different scenarios have been discussed about the origin of this quasi-isotropic H_{c2}, such as the influence of multiple bands, Pauli limited H_{c2}, and Fulde–Ferrel–Larkin–Ovchinnikov inhomogeneous state at high fields and low temperatures [3-4, 7-12]. Recently the work by Kogan and Prozorov also indicates that a decreasing γ_H is also possible without the presence of multiple bands [13].

When analyzing H_{c2}(T) anisotropy one has to bear in mind that for a two-band superconductor γ_H is not the electronic mass anisotropy (γ) as it is in the case of a single-band superconductor such as $YBa_2Cu_3O_7$ [14-16]. In a single-band superconductor the angular anisotropic behavior of thermodynamic properties can be explained using a single scaling parameter γ that is temperature independent. In a multi-band superconductor, the contributions of different bands lead to a non-constant $\gamma_H(T)$, with each band contributing differently as T is changed. Moreover, the angular dependence of H_{c2} is also not fully described by a single γ value, but rather by the contribution of the bands in different angular regions [17]. This is particularly evident when the H_{c2} angular dependence in Co-doped $SrFe_2As_2$ film is analyzed [4]. Although at high temperatures the angular dependence of the $SrFe_2As_2$ film's H_{c2} can be fitted using a single band approximation; when this data is combined with the $H_{c2}(T)$ measured along the main crystallographic directions and fitted using a two band model; a double maxima in H_{c2} angular dependence at low temperatures emerges from the model and was also measured in the film [4]. Consequently, even with H_{c2} being identical for **H**||c and **H**||ab a lower value of H_{c2} was measured when **H** was applied along intermediate orientations. A decreasing γ_H(T) with decreasing T also indicates the possibility of finding materials with $H_{c2}(||c) > H_{c2}(||ab)$ with the consequently inversed H_{c2} anisotropy $\gamma_H<1$.

A natural candidate for observing $\gamma_H<1$ is the FeTe (11) family, the least anisotropic of all iron-based superconductors[17]. Indeed, different authors have found evidence of an H_{c2} inversion in single crystals of FeTeSe and FeTeS[7,8,10,18,19]. Initially a possible reversal of H_{c2} anisotropy was observed by Fang *et al* for FeTeSe, with $H_{c2}(||c)>H_{c2}(||ab)$ at the lowest temperature measured, however within the error bars of the experiment [8]. Also in FeTeSe Khim *et al* also observed a crossover at low temperatures (T~4K, $t=T/T_c$=0.3), with γ_H~0.95. They attributed this observation to a difference in the Landé *g* factor, of an otherwise isotropic H_{c2}, due to the Pauli-limited H_{c2}[7]. For FeTeS an almost isotropic upper critical field was observed with γ_H= 1.05 at t =0.65 [18]. Further experiments of the full H_{c2}-T diagram showed that in $Fe_{1+y}(Te_{1-x}S_x)_z$ γ_H< 1 for T<1 K in a single crystal with T_c=8 K [19]. Confirming the H_{c2} inversion for films will help understand the origin of this phenomenon with important impact on application-prone materials such as thin films; and could mean a big step forward for tailoring superconductor's performance.

In this letter we show the full H_{c2}-T measurements performed on FeTeS thin films grown by Pulsed Laser Deposition (PLD) on MgO single crystals. We find a temperature-dependent γ_H, with a crossover at T= 0.7 T_c from γ_H>1 at high T to γ_H~0.96 at low temperatures. This result indicates the possibility of Fe-based isotropic superconducting materials for high-field applications in the entire H-T phase diagram. The observation of similar behavior as that seen in single crystals confirms the robustness of this phenomenon.

2. Experimental

A Lambda Physik KrF excimer laser (λ = 248 nm) was used for the PLD of epitaxial $Fe(Te_{1-x}S_x)$ thin films on MgO (100) single crystal substrates following the procedures described in reference [20]. The film presented in this study has a T_c of ~7K with ΔT_c ~1 K (90-10%) [20]. The value of T_c is very close to that found in single crystals for x=0.11 and 0.09 [18,19] and is consistent with the EDS measurements of x=0.1, although the nominal content of S in the target was x=0.20 [20]. According to SEM/EDS the three elements are uniformly distributed without segregations or clusters. Indeed, all our Fe–Te–S films present a stoichiometric deficiency of Te and S (i.e., a Fe stoichiometric excess), respect to nominal content of 0.8 and 0.2, respectively. The average composition of films can be expressed as Fe:Te:S = 1:0:55:0:095. This behavior is probably connected to the fact that the stoichiometry of target deviates from the nominal FeTe0:8S0:2, being FeTe0:96S0:1 and that Te and S elements are lost during film deposition. However the depression of superconducting transition in thin films is less dramatic than in the case of corresponding Fe-rich bulk samples [21].

Electrical transport was measured using AC digital lock-in at 100KHz with a current density of 5 A/cm^2 in pulsed magnetic fields up to 35 T. Measurements were carried out in a rotating sample holder that allowed the sample to be rotated without removing it from the measuring system. The rotating stage has a pick-up coil parallel to the sample's surface to determine the sample orientation (Θ) between the c axis and the applied magnetic field (**H**) within ± 1°. The angle was measured before and after shots with no measurable difference. Linear (ρ) transport measurements were carried out using the maximum Lorentz force configuration ($\boldsymbol{J} \perp \mathbf{H}$). Pulsed magnetic field measurements were performed at the NHMFL - Pulsed Field Facility at Los Alamos. To ensure a linear response and exclude heating effects, current was doubled and halved without observing changes in the resistivity (ρ) values measured. Also, pulses of different magnitudes were performed, with no changes in the ρ(*H*) curve, confirming that the H_{c2} value obtained is independent of the rate of *H* change.

3. Results and discussion

In Figure 1 we show ρ(*H*) for **H**||c (Θ =0°) at different *T*. Similar to what Fang *et al.* have reported, we observe a small residual resistivity [8]. Nevertheless the determination of the H_{c2} values is not affected by this since we use a 95% of the normal state resistivity (ρ_n) criterion to determine H_{c2} (see figure 1). The origin of the small tail can be associated with small inhomogeneities or second phases that are affecting percolation. The first observation is that we do not see a significant magneto-resistance at any orientation, in contrast to Lei *et al*. [19]. The difference in magneto-resistance could be related to a higher degree of disorder in the film, or a slight difference in composition. A clear change in magneto-resistance with chemical composition has been observed in different iron-based superconductors such as NdBaFeAs by Riggs *et al* [22]. Also, a large magneto-resistance has been observed while entering the magnetic state, but is absent in the non-magnetic state [5,9].

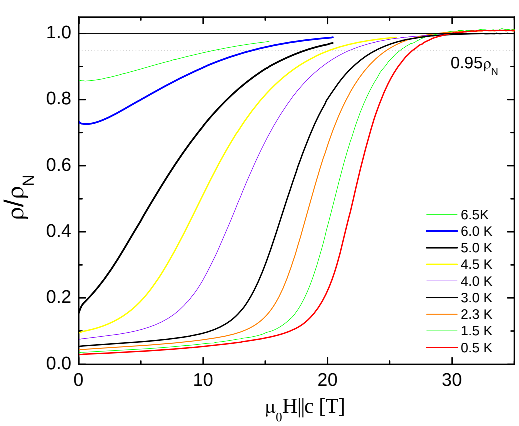

Figure 1. (Color online) ρ/ρ_N vs H for H||c at different temperatures, with ρ_N measured at T=10K and μ_0H = 0 T

In figure 2a we show H_{c2} –*T* measured at three orientations: **H**||c (0°), 45° and ab (90°). Given the lack of magneto-resistance, H_{c2} is determined using a $0.95\rho_N$ criterion; this also reduces the influences of possible artifacts related to the broadening of the transition. We also include the values of H_{c2}(T) for **H**||c of an identical sample reported by Mele *et al*.[20] At high temperatures a very steep H_{c2}(*T*) slope is observed (≥15 T/K), however as T is lowered, $H_{c2}(T)$ flattens out and $\mu_0 H_{c2}(0)$ ~ 27 T. This value of H_{c2} is almost twice the Pauli limit calculated as 1.84 T_c in the BCS approximation. Several authors have used the Werthamer-Helfand-Hohenberg (WHH) formalism to fit the H_{c2}(T) data for FeTeSe and FeTeS single crystals. In the case of FeTeS with T_c~8K and $\mu_0 H_{c2}(0)$~28T, good fits were obtained using the WHH parameters α~3-4 and λ_{so}~0.5-1, while for FeTeSe with T_c~15K and $\mu_0 H_{c2}(0)$ ~ 50 T the parameters were α~4-5 and λ_{so}~1 [7,10,19,23].

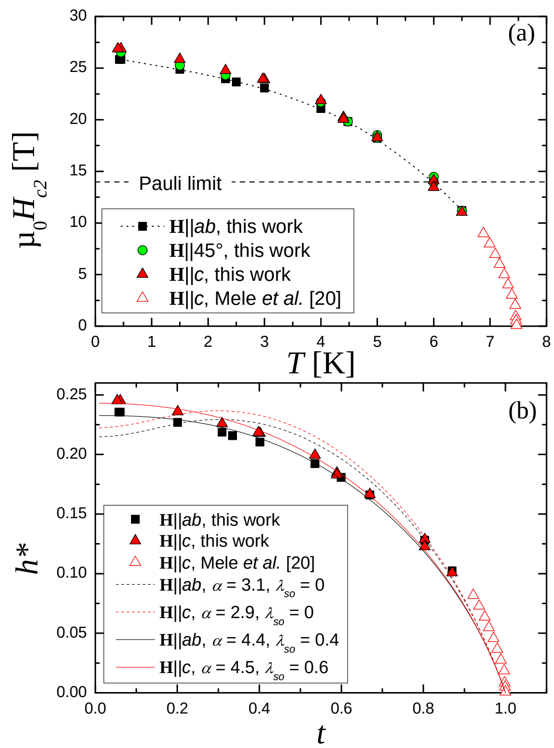

Figure 2 (Color online) **a)** Upper critical field H_{c2} – temperature phase diagram of this work (pulsed field) and DC fields [20]. **b)** $h^* = H_{c2}/H_{c2}'$ with $H_{c2}'=(-dH_{c2}/dt)_{t=1}$ versus t, for **H**||ab and ||c with fits using the WHH formulation [22], with H_{c2}'=15 T/K.

Given the fairly isotropic H_{c2} data any fits are expected to be similar for all directions. In figure 2b, we plot $h^* = H_{c2}/H_{c2}'$ with $H_{c2}'=(-dH_{c2}/dt)_{t=1}$ versus $t=T/T_c$. We start by keeping λ_{so}=0, but it is apparent in figure 2b that in order to fit our data with the WHH model it is necessary to have a non-zero value for λ_{so} since the shape of the fit obtained for λ_{so}=0 is drastically different from the data. Indeed, we can fit our data very well with α=4.4 and λ_{so}=0.4 for **H**||ab and α=4.5 and λ_{so} =0.6 for **H**||c. It needs to be pointed out, however, that this fit fails to capture H_{c2}(T) near T_c, partly because the values of h*(t) depend strongly on H_{c2}'(t=1). The h* values for both **H**||ab and **H**||c shown in figure 2b were calculated from the measured H_{c2} using a slope of H_{c2}'~15 T/K, obtained from a linear fit to the **H**||c dataset of the DC measurements [20]. As $H_{c2}(T)$ exhibits a clear curvature, a linear fit to the data points closest to T_c yields a significantly higher slope (>50 T/K) [20]. Thus, If one performs WHH fits with the resulting smaller $h^*(t)$, a much better fit is obtained at *higher T-lower H*, forcing higher values of α, namely α~30 – 40. However, α is not just a fitting parameter but is defined with $\alpha=(3e^2 \hbar\, \gamma_n \rho_n)/(2m\, \pi^2 k^2_B)$ where γ_n is the normal state electronic specific heat and ρ_n the normal state resistivity [23,24]. Given that ρ_n~ 1mΩ cm for both FeTeSe films [20,25,26] and single crystals [7], and there is no reason for γ_n to change from a single crystal to a film, there is no good explanation for an arbitrary bigger α several times higher than that found in single crystals. As a consequence of the strong dependence on $(dH_{c2}/dt)_{t=1}$, the values of α and λ_{so} need to be treated with a certain caution. This type of very high values for H_{c2}' near T_c have also been observed in $FeTe_{0.5}Se_{0.5}$ thin films by Tarantini *et al* [27] and fit with a $H_{c2}(T)$~ $(T_c\text{-}T)^{1/2}$, taken as in indication of complete suppression of orbital pair breaking. A closer look at the h*(T) diagram in Fig. 2(b) also points to $h^*(T=0)$ similar to what Lei *et al* observed in FeTeSe and FeTeS crystals [10,19].

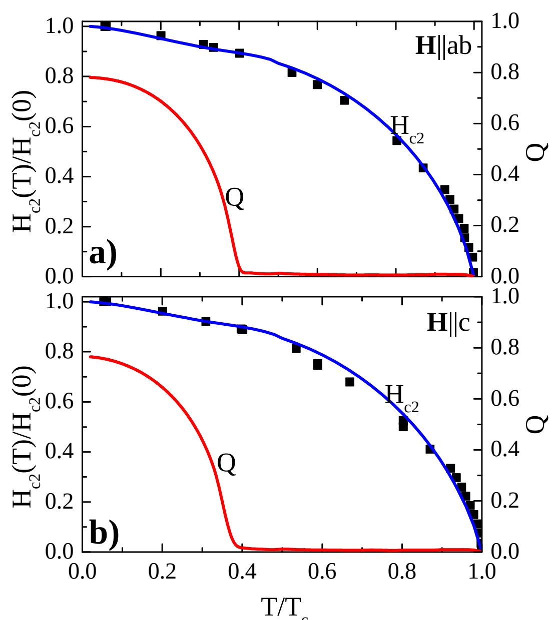

Figure 3 (Color online) **a)** Blue line is the fit to the upper critical field H_{c2} for **H**||ab with fit using Ref. [11], in red we show the resulting Q vector from the fit. **b)** *Idem* for **H**||c.

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Given the unsatisfactory results at *high T-low H* using the WHH model; in figure 3 we turn to a more refined model that takes into account the effects of multibands [11]. Note that this model is different to that used in ref. [4] since the model we apply here is taken in the clean limit, a choice more justifiable in this material with such small superconducting coherence length. By taking the *Fulde-Ferrel-Larkin-Ovchinnikov* (FFLO) instability with a modulated superconducting order parameter along the c axis with a wave vector Q into account, the upper critical field H_{c2} for **H**||*c* orientation is given by [11,27]

$$a_1G_1 + a_2G_2 + G_1G_2 = 0,$$
$$G_1 = \ln t + 2\exp(q^2)\,\mathrm{Re}\sum_{n=0}^{\infty}\int_q^{\infty}\exp(-u^2)\left[\frac{u}{n+1} - \frac{t}{\sqrt{b}}\tan^{-1}\left(\frac{u\sqrt{b}}{t(n+1/2)+i\alpha b}\right)\right]du. \qquad (1)$$

G_2 is obtained by replacing $\sqrt{b} \to \sqrt{\eta b}$ and $q \to q\sqrt{s}$ in G_1. The wave vector $Q(T, H)$ corresponds to the case when $H_{c2}(T, Q)$ is maximal. The other parameters are

$$a_1 = \frac{(\lambda_0 + \lambda_-)}{2w}, a_2 = \frac{(\lambda_0 - \lambda_-)}{2w}, \lambda_- = \lambda_{11} - \lambda_{22}, \lambda_0 = \sqrt{\lambda_-^2 + 4\lambda_{12}\lambda_{21}}, w = \lambda_{11}\lambda_{22} - \lambda_{12}\lambda_{21},$$
$$t = T/T_c, b = \frac{\hbar^2 v_1^2 H}{8\pi\Phi_0 k_B^2 T_c^2}, \alpha = \frac{4\mu\Phi_0 k_B T_c}{\hbar^2 v_1^2}, q^2 = \frac{Q^2\Phi_0\varepsilon_1}{2\pi H}, \qquad (2)$$

where $s = \varepsilon_2/\varepsilon_1$ and $\eta = (v_2/v_1)^2$ with v_i the in-plane Fermi velocity for the *i*-th band and $\varepsilon_i = m_i^{ab}/m_i^c$ the mass anisotropy ratio. Here λ_{11} and λ_{22} are the intraband coupling strength, and λ_{12} and λ_{21} are the interband coupling strength. $\Phi_0 = hc/(2e)$ is the flux quantum and μ is the magnetic moment of a quasiparticle. In the fits we made, we have assumed $\varepsilon_1 = \varepsilon_2 = \varepsilon$ for simplicity.

For **H**||*ab*, H_{c2} is given by the following replacement for G_1 and G_2 in Eqs. (1-2)

$$q \to q\varepsilon^{-3/4}, \alpha \to \alpha\varepsilon^{-1/2}, \sqrt{b} \to \sqrt{b}\varepsilon^{1/4} \text{ for } G_1 \text{ and } \sqrt{\eta b} \to \sqrt{\eta b}\varepsilon^{1/4} \text{ for } G_2.$$

Using the typical coupling strength for iron-based superconductors $\lambda_{11} = \lambda_{22} = 0$ and $\lambda_{12}\lambda_{21} = 0.25$, [11] we find that the fitting parameters are $\alpha = 3.0$, $\eta = 0.9$, $\varepsilon = 0.8$. The results of our fits also indicate the existence of a FFLO phase, for $T/T_c<0.4$ characterized by the vector Q shown in red in figure 3. We also find a linear H_{c2}(T) at low temperatures, consistent with an FFLO phase as suggested in ref. [11]. Nevertheless, thermodynamic measurements are needed to establish the creation of the FFLO instability at low *T*- high *H*.

We now turn our attention to the temperature where the H_{c2} crossover takes place as it appears to be related to the existence of the FFLO phase.

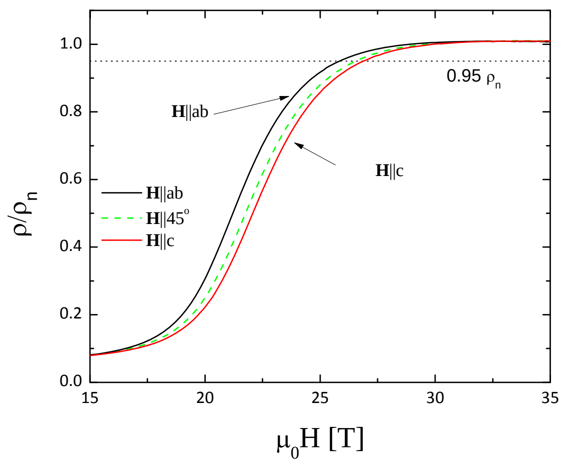

Figure 4 (Color online) ρ/ρ_N vs H for H||c, ab and 45° for T=0.50 K. Samples were immersed in liquid ^{3}He to avoid heating.

Although overall H_{c2} is almost isotropic in the three orientations measured with only a 5% anisotropy, our measurements allow us to ascertain a clear trend in H_{c2} at different orientations. At low temperatures we find that $H_{c2}(||c)>H_{c2}(||45°)>H_{c2}(||ab)$, see Figs. 2 and 4. This anisotropy is higher than any systematic error as can be observed in figure 4, where ρ(H) is shown at T=0.5 K. We can clearly see how $H_{c2}(||c) >H_{c2}(||ab)$ by one Tesla. Since the bigger source of uncertainty comes from the reproducibility of the angular position we repeated this measurement for both field orientations with the same result. As seen in figure 4, this is not affected by the width of the transition

In figure 5 we plot $\mu_0\Delta H_{c2}=\mu_0 H_{c2}(||ab)- \mu_0 H_{c2}(||c)$. It can be observed how ΔH_{c2} decreases and crosses to $\Delta H_{c2} < 0$ at $T=0.7T_c$ as the temperature decreases. Similarly, as plotted in the inset of figure 5, γ_H exhibits a monotonous T dependence with $\gamma_H < 1$ below the crossover. The value of γ_H=0.96 obtained at low temperatures is similar to that reported by Khim *et al.* [7] for FeTeSe crystals and very close of that of Lei *et al* [19] in FeTeS crystals. This could be an indication that indeed the small anisotropy and the crossover are due to a difference in the Landé *g* factor. However, the apparent difference in the H_{c2} ratio between the data from a single crystal [19] and our data, shown in the Inset of figure 5, might be indicating that disorder can be playing a larger role, noting that T_cs are only slightly different (7.5 K and 8.5 K for the films and crystal respectively), and doping is also very close. A note of attention is required

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concerning the results at higher temperatures; these could be affected by the width of the superconducting transition, but as explained previously this can only affect the crossover temperature, not the fact that at low temperatures the H_{c2} has an inverted anisotropy. Indeed, in the samples where this crossover has been observed (FeTeSe and FeTeS), it was only seen at much lower temperatures, and with a much bigger starting γ_H, indicating that manipulation of H_{c2} can be obtained with high and isotropic H_{c2}.

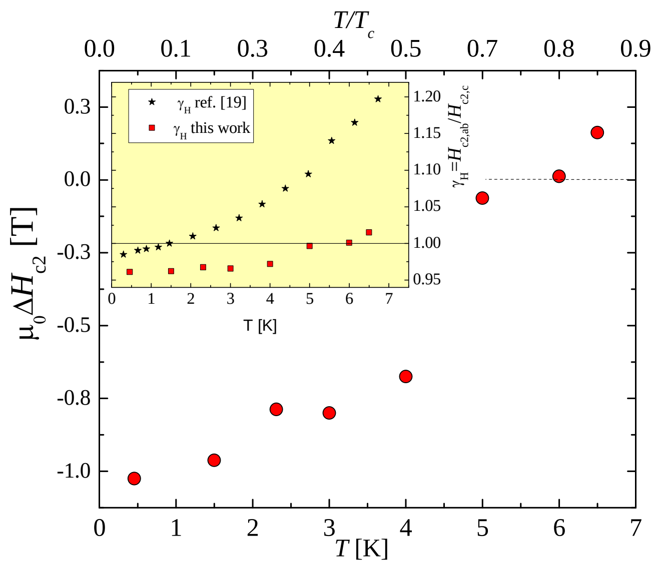

Figure 5(Color online) $\mu_0\Delta H_{c2}$=$\mu_0 H_{c2}$(||ab) -$\mu_0 H_{c2}$(||c). Inset: $\gamma_H = H_{c2}(||ab) / H_{c2}(||c)$ from the film (x=0.10) from this work and a FeTeS single crystal from ref. [19] with x=0.09.

CONCLUSIONS

In summary, we report on the full H_{c2}-T diagram for FeTeS films, measured at different orientations. We find an almost isotropic H_{c2} with clear evidence of the inversed H_{c2} anisotropy (γ_H<1) for $T<0.7T_c$. The H_{c2} ration γ_H is a strong indication of the role of multiple bands in H_{c2} of iron-based superconductors. We also analyze the data using a WHH formulation and obtain a very good fit, but we find that the fitting parameters depend strongly on the way the slope of H_{c2} near T_c was obtained with unrealistically high values for WHH fits that accommodate the $H_{c2}(T)$ curvature near T_c. Results using a two band superconductor in the clean limit led us to a fit that indicates the presence of a FFLO at low temperatures and more reasonable values for α and λ. The value for H_{c2}(||ab)/H_{c2}(||c) = 0.96 at low temperatures is very similar to that observed in single crystals, indicating that this property is robust against disorder. Further experiments are needed in order to elucidate how to control the H_{c2} crossover

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temperature and the appearance of a FFLO phase. The possibility to tune the effective anisotropy of superconductors is extremely appealing for different applications and a step closer to obtaining superconductors by design.

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