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Upper critical field of KFe_2As_2 under pressure: A test for the change in the superconducting gap structure

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We report measurements of electrical resistivity under pressure to 5.8 GPa, magnetization to 6.7 GPa, and ac susceptibility to 7.1 GPa in KFe_2As_2. The previously reported change of slope in the pressure dependence of the superconducting transition temperature $T_c(p)$ at a pressure $p^* \sim 1.8$ GPa is confirmed, and $T_c(p)$ is found to be nearly constant above p^* up to 7.1 GPa. The T-p phase diagram is very sensitive to the pressure conditions as a consequence of the anisotropic uniaxial pressure dependence of T_c. Across p^*, a change in the behavior of the upper critical field is revealed through a scaling analysis of the slope of H_{c_2} with the effective mass as determined from the A coefficient of the T^2 term of the temperature-dependent resistivity. We show that this scaling provides a quantitative test for the changes of the superconducting gap structure and suggests the development of a k_z modulation of the superconducting gap above p^* as a most likely explanation.

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Since the discovery of superconductivity in $LaFeAs(O_{1-x}F_x)$ [1], the iron-based superconductors have been the focus of numerous experimental and theoretical studies. Taking advantage of the lessons learned from the cuprate high-temperature superconductors, the investigation of the symmetry of the superconducting state has been given priority [2]. However, unlike the cuprates, the gap structure of the iron-based superconductors is not universal and several gap symmetries have been proposed both experimentally and theoretically [3]. The stoichiometric compound KFe_2As_2, which has a superconducting transition temperature $T_c \approx 3.5$ K, is one of the cleanest examples where different gap structures appear likely [4–15]. Recently, it has been suggested that a change of pairing symmetry from d wave to s wave occurs upon applying pressure to KFe_2As_2 [9]. The argument was based on the experimental observation of a change in the pressure dependence of T_c from negative to positive at $p^* \approx 1.8$ GPa. Following this study, ac magnetic susceptibility and de Haas–van Alphen (dHvA) oscillations under pressure confirmed the change of slope in $T_c(p)$ at p^* and supported the earlier inference that this change is not due to drastic modifications of the Fermi surface [16]. A similar change of slope of $T_c(p)$ was also observed in $CsFe_2As_2$ [17]. Although there have been theoretical predictions that the d-wave and s-wave states are very close in energy [14,18,19], the experimental data available so far do not provide information about the changes in the superconducting gap function at p^*.

In this study we significantly extend the pressure range of previous studies (~2.5 GPa [9,16]) to 7.1 GPa. We confirm the observation of a change of slope in $T_c(p)$ but find that the phase diagram is very sensitive to the hydrostaticity of the pressure medium in this high-pressure range. In addition, we report on the temperature dependence of the upper critical field H_{c_2} with the magnetic field applied along the c axis under pressure. By scaling $(-d\mu_0 H_{c_2}/dT|_{T_c})/T_c$ versus the A coefficient of the T^2 term of the resistivity, we find a change at p^* which allows for a quantitative test of the modification of the superconducting gap structure. The present data are not able to test whether a change in symmetry between d wave and s wave actually takes place at 1.8 GPa. We find, however, that such a change alone would not be sufficient to account for our results. We suggest that a k_z modulation of the superconducting gap is involved in the slope change at p^*.

The Fermi surface of KFe_2As_2 has been investigated experimentally by dHvA oscillation [16,20] and angle-resolved photoemission spectroscopy (ARPES) [4,21,22] experiments. The Fermi surface consists of three hole cylinders at the Γ point [α (inner), ζ (middle), and β (outer) bands], and four small hole cylinders near the X point (ϵ band). ARPES experiments down to 2 K indicate that the gap is nodeless on the α and β bands, and nodal with octet line nodes on the ζ (middle) band [4]. The nodes have also been detected by thermal conductivity [5,6], penetration depth [7], and nuclear quadrupole resonance [8]. The question of whether those nodes are accidental with an s-wave state [4] or imposed by symmetry in a d-wave state [6,9] is still under debate [10]. Other possibilities include a time-reversal symmetry breaking $s + id$ state [11–14], or an $s + is$ state between two kinds of $s\pm$ states which has been proposed upon Ba doping in $Ba_{1-x}K_xFe_2As_2$ [15] in the vicinity of $x \sim 0.7$ where deviations in the jump in specific heat have been observed [23,24]. In this context, the evidence for a change of gap function under pressure in KFe_2As_2 illustrates the near degeneracy of these states and the possibility of studying the interplay between different superconducting states. Our results suggest that, in addition to the considered possible in-plane symmetry of the gap functions, a k_z modulation of the superconducting gap is involved in the slope change at p^*.

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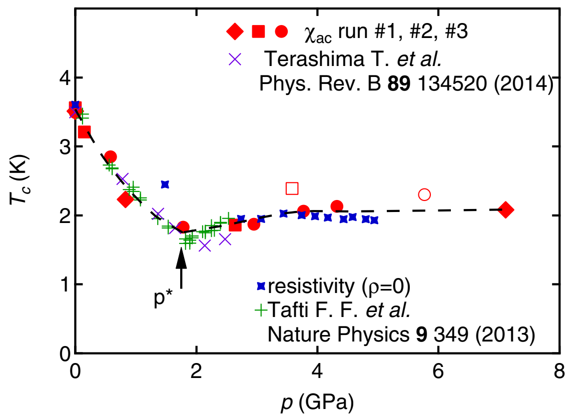

FIG. 1. (Color online) Superconducting phase diagram for KFe_2As_2 determined from our resistivity and ac susceptibility measurements [28]. For the latter, filled symbols are used when pressure was applied on liquid helium, whereas open symbols are used when pressure was applied on solid helium (at 10 K). The dashed line is a guide to the eye which does not use the two data points where the pressure was applied on solid helium. The data from Refs. [9,16] are also shown.

In this study, several high-quality single crystals of KFe_2As_2 were grown from KAs flux as detailed in Ref. [25]. For the electrical resistivity measurements, pressure was applied at room temperature using a modified Bridgman cell [26] with a 1:1 mixture of n-pentane:isopentane as a pressure medium. The ac susceptibility measurements to hydrostatic pressures as high as 7.1 GPa were carried out in a membrane-loaded diamond-anvil cell [27]. Helium was used as pressure medium. To promote hydrostaticity, pressure was increased at temperatures well above the melting curve of helium, unless stated otherwise. Further experimental details are given in the Supplemental Material [28] (see also Refs. [9,16,26,27,29–49]) together with other measurements using less hydrostatic pressure media.

The superconducting phase diagram obtained from ac susceptibility and resistivity measurements is shown in Fig. 1. The previously reported change of slope in $T_c(p)$ at $p^* \approx$ 1.8 GPa [9,16] is confirmed. T_c increases very slowly above p^* up to 7.1 GPa. A remarkable property of this phase diagram is its strong sensitivity on the pressure conditions. As shown by the open symbols in Fig. 1, T_c is increased when the pressure is applied on solid helium by comparison with liquid helium. As expected, the effect is even more dramatic with less hydrostatic media. When using a 1:1 mixture of Fluorinert FC70:FC77, T_c is only slowly reduced with pressure and $T_c \approx 3.19$ K at our pressure limit of 5.8 GPa [28]. In our dc magnetization measurements using Daphne 7474, a second superconducting dome is even obtained with a maximum T_c as high as 3.8 K at 5.5 GPa, which is above the room temperature solidification point of this medium [28]. Such a large sensitivity to the hydrostaticity is most likely a consequence of the anisotropic uniaxial pressure dependence of T_c. In KFe_2As_2, $\partial T_c/\partial p_a|_0 \approx -1.9$ K GPa^{-1} along the a axis, whereas $\partial T_c/\partial p_c|_0 \approx +2.1$ K GPa^{-1} along the c axis [49]. Under hydrostatic conditions, the three axes will contribute equally to give rise to the phase diagram presented in Fig. 1. However, under nonhydrostatic conditions, as already explained in Refs. [32,42], the pressure will be larger along the c axis and smaller in the ab plane. This results in larger values of T_c and a modification of the superconducting phase diagram [28].

Not only do we confirm the kink in $T_c(p)$ previously reported [9,16], but we observe that T_c remains roughly constant up to 7.1 GPa. A Lifshitz transition can produce such a kink in $T_c(p)$. In that case, the observed increase of T_c just above p^* is consistent with the formation of a new Fermi-surface pocket [50]. However, no anomaly was observed in the Hall coefficient to support this mechanism [9], and dHvA oscillations indicate no drastic change in the Fermi surface up to ∼2.5 GPa [16].

We note that the change in slope at the characteristic pressure of 1.8 GPa could be a simple consequence of the fact that the uniaxial pressure dependencies of T_c are of opposite sign and large. At ambient pressure, $\partial T_c/\partial p_a|_0 \approx -1.9$ K GPa^{-1} and $\partial T_c/\partial p_c|_0 \approx +2.1$ K GPa^{-1} in Ref. [49] or $\partial T_c/\partial p_c|_0 \approx$ $+1.1$ K GPa^{-1} in Ref. [42]. These partial derivatives cancel each other to a considerable degree, yielding a hydrostatic pressure derivative that is negative. Any nonlinearity in $T_c(p_a)$ or $T_c(p_c)$ would generate a much larger relative nonlinearity in the dependence of T_c on hydrostatic pressure $T_c(p)$. For example, were the magnitude of $\partial T_c/\partial p_a$ to gradually decrease by a factor of ~ 3 under 3 GPa hydrostatic pressure, $\partial T_c/\partial p_c$ remaining constant, the hydrostatic pressure dependence $T_c(p)$ would be forced to pass through a minimum. We also note that, even though the modulus of elasticity is almost identical along the a and c axis, the first derivative of the modulus is over an order of magnitude smaller along the c axis [17]. This implies a larger compression along the c axis, so that the effect of pressure on T_c may become dominated by the p_c component. In such a scenario, a theoretical explanation of the uniaxial pressure dependencies of T_c would be the key to understanding the slope change at p^*.

Another possibility that may even induce the previous idea is a transition to a superconducting phase of a different symmetry. In such a case, changes in other thermodynamic quantities, such as the thermal expansion or the specific heat, are also expected. However, the combination of high pressures and low temperatures makes the experimental investigations of these quantities challenging. Figure 2 shows the temperature dependence of H_{c_2} for the magnetic field applied along the c axis at different pressures. At ambient pressure, the upper critical field along the c axis is known to be due to the orbital limit with negligible effect due to the Pauli limit [49,52]. With increasing pressure, as T_c decreases, H_{c_2} is also decreasing. Interestingly, above p^* where T_c remains roughly constant or increases very slowly, H_{c_2} continues to decrease (see inset of Fig. 2). In the following, we will relate this decrease with a commensurate decrease of the electrons' effective mass.

Figure 3 shows the low-temperature dependence of the resistivity as a function of T^2 at various pressures (full lines). For each pressure, we performed fits with a Fermi liquid behavior $\rho = \rho_0 + AT^2$ up to 8 K (dashed lines). The pressure dependence of the A coefficient is shown in the inset. At ambient pressure, $A \approx 0.02$ $\mu\Omega$ cm K^{-2}, in agreement with previous reports [5,52–54]. Under pressure, A decreases smoothly, which is consistent with the decreasing trend in effective mass observed in dHvA oscillations [16].

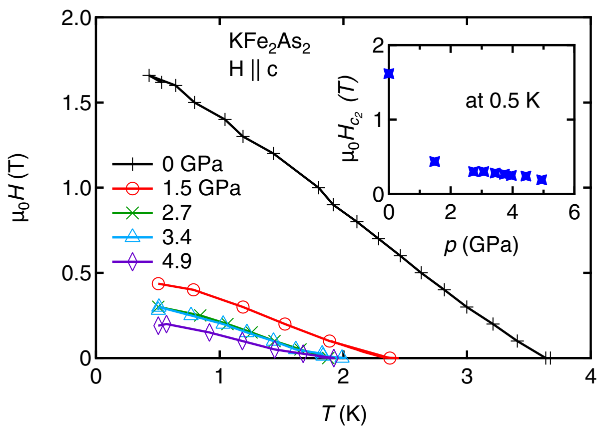

FIG. 2. (Color online) Temperature dependence of the upper critical field H_{c_2} at different pressures. The data were determined using a resistive transition offset criteria ($\rho = 0$, see Ref. [25]). We note that, unlike the use of the transition midpoint criteria, the chosen offset criteria agree more closely with ambient pressure specific heat measurements [51]. The inset shows the pressure dependence of H_{c_2} at 0.5 K.

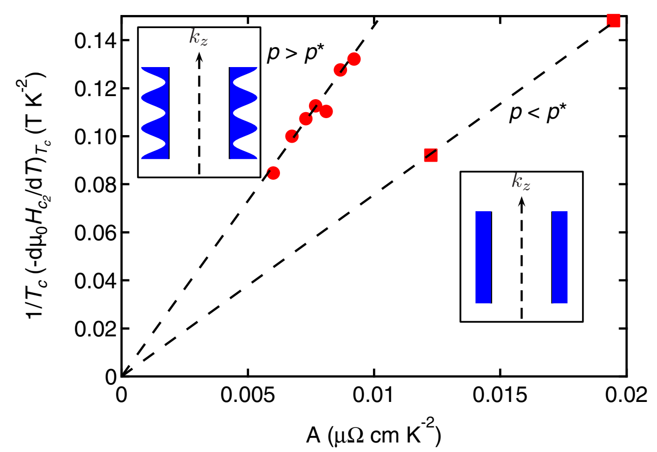

FIG. 4. (Color online) Plot of $(-d\mu_0 H_{c_2}/dT|_{T_c})/T_c$ vs the A coefficient of resistivity. Each point corresponds to a different pressure. All measured points fall onto two lines of different slope corresponding to $p < p*$ and $p > p*$. The two insets show a schematic gap structure with the appearance of a modulation along k_z above p^*.

In Ref. [55], the Helfand-Werthamer theory is examined for the case of uniaxial anisotropy with an anisotropic superconducting gap. For $H||c$ (one-band case, clean limit):

$$\frac{1}{T_c}\left(-\left.\frac{d\mu_0 H_{c_2}^{\text{orb}}}{dT}\right|_{T_c}\right) = \frac{8}{7\zeta(3)\langle\Omega^2\mu_c\rangle}\frac{\phi_0 2\pi k_B{}^2}{\hbar^2 v_0{}^2}. \quad (1)$$

The function $\Omega(\mathbf{k_F})$ which determines the $\mathbf{k_F}$ dependence of the superconducting gap $\Delta = \Psi(\mathbf{r},T)\Omega(\mathbf{k_F})$ is normalized so that $\langle\Omega^2\rangle = 1$. The averages over the Fermi surface are shown as $\langle\cdots\rangle$.

$$\mu_c = \frac{v_x{}^2 + v_y{}^2}{v_0{}^2} \quad \text{and} \quad v_0{}^3 = \frac{2E_F{}^2}{\pi^2\hbar^3 N(0)},$$

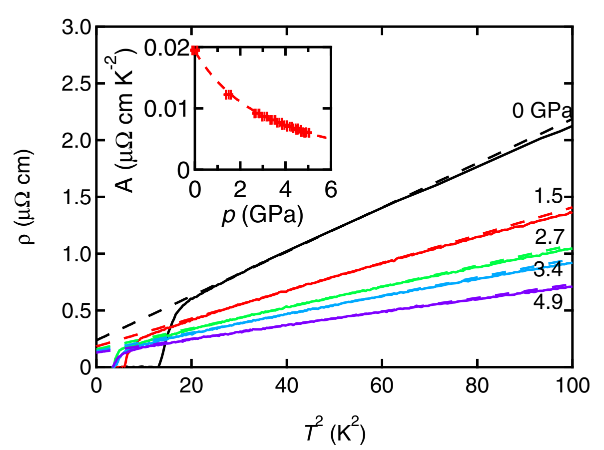

FIG. 3. (Color online) Electrical resistivity ρ of KFe_2As_2 at different pressures at low temperatures as a function of T^2. The dashed lines show fits of the resistivity to a Fermi liquid behavior $\rho = \rho_0 + AT^2$. The pressure dependence of the A coefficient is shown in the inset. The dashed line represents $A(0)/(1+\beta p)^2$ where $\beta = 0.16(1)\ \text{GPa}^{-1}$.

where $N(0)$ is the total DOS at the Fermi level E_F per spin. Assuming that the A coefficient of the T^2 term of the resistivity, when resistivity is measured along x, obeys $A \propto n/\langle v_x{}^2\rangle$, the A coefficient in the tetragonal case will reflect the dependence on $1/(v_0{}^2\langle\mu_c\rangle)$:

$$\frac{1}{T_c}\left(-\left.\frac{d\mu_0 H_{c_2}^{\text{orb}}}{dT}\right|_{T_c}\right) \propto \frac{\langle\mu_c\rangle}{\langle\Omega^2\mu_c\rangle}\frac{A}{n}, \quad (2)$$

where the carrier density n can be estimated from Hall measurements. A more detailed expression for A can be found in various publications [56,57] and would lead to a more complicated expression than that given in Eq. (2). A refinement to the case of several bands would certainly be of interest. In the present form, Eq. (2) shows a proportionality between the slope of H_{c_2} at T_c and the A coefficient. This result is known for heavy fermions, both quantities being proportional to the square of the effective mass [58,59].

Figure 4 shows the plot of $(-d\mu_0 H_{c_2}/dT|_{T_c})/T_c$ versus A. All measured points fall onto two straight lines of different slope corresponding to $p < p*$ and $p > p*$. It is remarkable that both lines go through the origin as expected from the proportionality relation in Eq. (2). Equation (2) indicates that a change of slope when plotting $(-d\mu_0 H_{c_2}^{\text{orb}}/dT|_{T_c})/T_c$ versus A implies a change in either n, Ω, or μ_c. In KFe_2As_2, the carrier density does not change significantly with pressure as inferred from Hall resistivity measurements [9] and from the smooth pressure variation of the A coefficient. Therefore, the observed change of slope is more likely due to a change in $\langle\Omega^2\mu_c\rangle$. We note that a change of the superconducting gap symmetry is not the only possible explanation, since a change of the Fermi surface will also modify the value of $\langle\Omega^2\mu_c\rangle$. However, dHvA oscillations experiments indicate that the global structure of the Fermi surface hardly changes up to $p \sim 2.5$ GPa. We mention that $\langle\mu_c\rangle/\langle\Omega^2\mu_c\rangle = 1$ for any Ω that does not depend on k_z. Therefore, a simple change between s wave and d wave would not be able to explain the change of slope of nearly a factor of two observed in Fig. 4.

On the other hand, the appearance of a k_z modulation of the superconducting gap at p^* can explain an increase of nearly a factor two in $\langle\mu_c\rangle/\langle\Omega^2\mu_c\rangle$. Let us assume a superconducting gap with a modulation along k_z [55,60,61]:

$$\Delta = \Delta_0[1 + \eta\cos(k_z c^*)], \tag{3}$$

where $c^* = \hbar v_0/(2E_F)$ is the length scale [55]. Let us also assume a prolate ellipsoidal Fermi surface ($\epsilon = 0.1$ in the notations of Ref. [55]). We find that $\langle\mu_c\rangle/\langle\Omega^2\mu_c\rangle$ changes from 1 to $\sim$1.9 if η changes from 0 to -0.8. Therefore, if we assume that the Fermi surface does not change at p^*, the appearance of a k_z modulation of the superconducting gap at p^* can be a possible explanation of our experimental observations. A k_z dependence of the superconducting gap has been observed by ARPES in $Ba_{0.6}K_{0.4}Fe_2As_2$ [61], in agreement with a theoretical prediction for the pairing strength [62]. The pairing between the layers is predominantly responsible for the gap dispersion with k_z. In contrast, in $Ba_{0.1}K_{0.9}Fe_2As_2$, the superconducting gap size on all the Γ-centered hole Fermi surfaces does not vary much along k_z [63]. This is consistent with the near two-dimensionality of KFe_2As_2 by comparison with the other members of the 122 family [22,64–67]. It is possible that, by applying pressure on KFe_2As_2, the pairing between the layers induces a k_z modulation of the superconducting gap. This is also consistent with dHvA oscillations measurements showing that three-dimensionality increases with pressure [16].

In conclusion we have shown that there is very likely a change in the k_z modulation of the SC gap at $p^* \sim 1.8$ GPa. We base this conclusion on a change in the scaling of $(-d\mu_0 H_{c_2}/dT|_{T_c})/T_c$ with the A coefficient of the T^2 term of the resistivity. We have shown that this indicates either a change of the Fermi surface, of the carrier density, and/or of the superconducting gap symmetry. In addition, we significantly extended the pressure temperature phase diagram from $\sim$2.5 to 7.1 GPa. For $p > 2.5$ GPa we found that T_c increases only slightly up to 7.1 GPa. By using various pressure cells and several different pressure-transmitting media, we have demonstrated the extreme sensitivity of KFe_2As_2 to nonhydrostaticity [28] and propose that it is due to the anisotropic dependence of T_c on strain.

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