Multiorbital Effects on the Transport and the Superconducting Fluctuations in LiFeAs

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The resistivity, Hall effect, and transverse magnetoresistance have been measured in low residual resistivity single crystals of LiFeAs. A comparison with angle resolved photoemission spectroscopy and quantum oscillation data implies that four carrier bands unevenly contribute to the transport. However the scattering rates of the carriers all display the T^2 behavior expected for a Fermi liquid. Near T_c low field deviations of the magnetoresistance with respect to a H^2 variation permit us to extract the superconducting fluctuation contribution to the conductivity. Though below T_c the anisotropy of superconductivity is rather small, the superconducting fluctuation displays a quasi-ideal two-dimensional behavior which persists up to 1.4 T_c. These results call for a refined theoretical understanding of the multiband behavior of superconductivity in this pnictide.

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Introduction.—The superconductivity (SC) and normal state of the iron-based materials are governed by their electronic structure involving the five iron $3d$ orbitals, as established by band structure calculations and angle resolved photoemission spectroscopy (ARPES) studies [1]. A spin fluctuation exchange mechanism for SC, with a $s^{\pm}$ symmetry, has been suggested early on, based on the observation of a good nesting between hole and electron Fermi surfaces [2]. This multiband character also highly influences the normal state transport, and experimental studies, performed mainly in the $BaFe_2As_2$(122) family, show that the electron mobility usually overcomes that of holes [3,4]. But the conductivity σ and Hall constant R_H do not permit one to disentangle the two carrier contributions to the transport.

Unlike other iron-based SC, LiFeAs is a stoichiometric compound with a relatively high $T_c \simeq 18$ K without any chemical doping [5,6], so that it is a nearly compensated semimetal, rather free from defects. Cleaved single crystals display no surface states [7], and clean ARPES data have evidenced [8,9] a dominant large holelike Fermi surface (FS), a much smaller one centered at the Γ, and two electronlike FSs at the corners of the Brillouin zone. The absence of matching between the sizes of the electron and hole FSs being a major difference with other pnictide families, it has been claimed [8] that the poor nesting prevents SC driven by spin fluctuations, while an orbital fluctuation mechanism [10] could be more appropriate.

In this Letter, we benefit from the reduced defect content in LiFeAs to take accurate magnetoresistance (MR) data, which together with σ and R_H should permit us to determine unambiguously the T dependences of the carrier contents and mobilities. While electron bands are found to dominate the transport as in undoped $BaFe_2As_2$, the detailed quantitative comparison with ARPES [8,9] and de Haas–van-Alphen (dHvA) data [11] shows that the holes involved in the largest hole band have the weakest mobility in LiFeAs. Furthermore, the MR data permit a precise determination of the contribution of superconducting fluctuations (SCFs) to the conductivity using the method we recently established for the cuprates [12,13]. These SCFs have been so far poorly studied in multiband SC, even in MgB_2 [14], and usually give information on the microscopic properties of the SC state [15]. Here, we find that the SCF paraconductivity can be very well fitted by the Gaussian Ginzburg Landau expectation for two dimensional single-band SC systems. These results should trigger theoretical studies of SCFs taking into account both the multiband aspect and the microscopic origin of SC, such as that initiated in Ref. [16].

Resistivity and Hall effect.—Six samples grown by a self-flux technique as detailed in Ref. [17] were studied, three with a four-probe configuration (labeled FP1,2,3) and three with a van der Pauw configuration [18] (labeled VDP1,2,3). The reproducibility of the data for the in-plane $\rho(T)$ is displayed in Fig. 1(a), the small differences (20% at most at room T) being ascribed to errors in the sample geometrical factors. The SC transition curves in the inset of Fig. 1(a) evidence increasing T_c values (15.5 to 18.2 K) for decreasing $\rho(T_c)$, except for FP1. As shown below, the data can be fitted with $\rho(T) = \rho_0 + AT^2$ below $\simeq 30$ K. For our best samples FP1 and FP2, $\rho_0 \sim 1.3\ \mu\Omega \cdot$ cm corresponds to residual resistivity ratios RRR $= \rho(300\text{ K})/\rho_0 \simeq 250$, that is 5 times larger than previously reported [19–22]. Comparing all those data [17], we find that both ρ_0 and A increase with decreasing T_c; i.e., Matthiessen's rule does not apply and $\rho(T)$ cannot be analyzed in a single-band model in LiFeAs.

For any temperature, we found that the Hall voltage is linear in field up to 14 T. The similar negative Hall coefficients R_H, reported in Fig. 1(b) for three samples with slightly different T_c, show that electrons dominate the

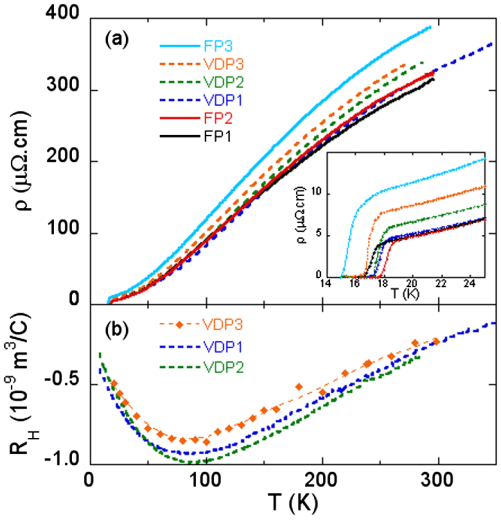

FIG. 1 (color online). (a) In-plane resistivity versus temperature for the six samples of LiFeAs measured. An expanded view of the superconducting transitions is reported in the inset. Except for one sample, one can see a correlation between the values of T_c and those of the residual resistivity. (b) Hall coefficient versus temperature for the three samples mounted in the van der Pauw configuration. The plotted curves correspond to the different samples ordered for decreasing room temperature resistivity or Hall effect.

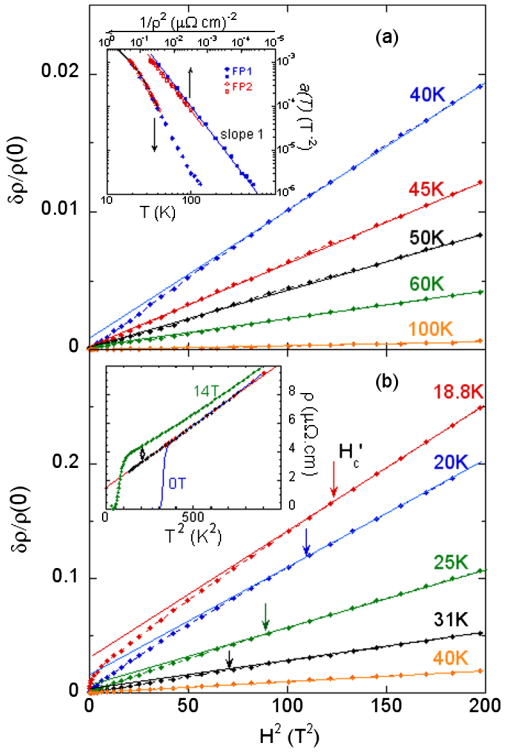

FIG. 2 (color online). The transverse magnetoresistance measured in FP1 is plotted as a function of H^2 for $T > 40$ K in (a) and below 40 K in (b). At high T, the MR is linear in H^2 for any H while it recovers this variation only above a given threshold field H_c' (indicated by arrows) for $T \lesssim 50$ K. In the inset of (a), the MR coefficient $a(T)$ of Eq. (1) is plotted versus T and $1/\rho^2$ for FP1 and FP2. The full lines of slope 1 indicate that Kohler's rule is very well obeyed (see text). In the inset of (b) the resistivity measured in a 14 T field down to 10 K is plotted versus T^2 for FP1, together with the extrapolated zero field data.

transport in this nearly compensated compound as in the nonmagnetic state of $BaFe_2As_2$ [3]. The broad minimum of $R_H(T)$, seen around 100 K, coincides with the observed change of curvature in the $\rho(T)$ curves. $R_H(T)$ tends towards zero with increasing T, which signals that the mobilities of the holes and electrons become similar. This behavior bears some resemblance to that found in overdoped Co-doped $BaFe_2As_2$ samples which also exhibit a minimum around 100 K, albeit less pronounced, and a similar increase towards room T [3].

Transverse magnetoresistance.—The small values of the residual resistivity ρ_0 permitted us to perform accurate measurements of the transverse MR above T_c up to 160 K. As shown in Fig. 2(a), for $T \gtrsim 45$ K, the MR increases as H^2 in the whole field range investigated here ($H \leq 14$ T). At lower T [see Fig. 2(b)], the low H increase of conductivity detected when approaching T_c is reminiscent of the contribution of superconducting fluctuations as evidenced in YBCO [13]. These SCFs will be analyzed later, but we shall focus first on the normal state which is fully restored beyond a threshold field $H_c'(T)$, allowing us to define the MR coefficient $a(T)$ as

$$\delta\rho/\rho(T,0) = [\rho(T,H) - \rho(T,0)]/\rho(T,0) = a(T)H^2. \quad (1)$$

As shown in the inset of Fig. 2(a), $a(T)$ is found to be identical in FP1 and FP2 and to decrease by about 3 orders of magnitude from T_c to 160 K.

Compensated two-band model.—The most natural approach to analyze these transport properties is within a two-band model. The transport coefficients are related to the respective conductivities σ_h, σ_e and mobilities μ_h, μ_e of the holes and electrons by

$$\rho^{-1} = \sigma = \sigma_e + \sigma_h, \quad (2)$$

$$R_H = (\sigma_h\mu_h - \sigma_e\mu_e)/\sigma^2, \quad (3)$$

$$\delta\rho/\rho(T,0) = \sigma_e\sigma_h(\mu_h + \mu_e)^2H^2/\sigma^2. \quad (4)$$

In undoped pnictides such as LiFeAs one expects an equal number of electrons and holes $n_e = n_h = n$ [8,9] and the two last equations condense into $\sigma R_H = \mu_h - \mu_e$ and $a(T) = \mu_e\mu_h$, so that the data permit us to deduce the T variations of μ_h, μ_e, and n. In particular, with $R = 1/ne$, these equations can be combined into

$$a(T) = (R^2 - R_H^2)/4\rho^2(T,0). \quad (5)$$

We can see in Fig. 2(a) that $a(T)$ scales as ρ^{-2}; that is, Kohler's rule [23] is well obeyed in LiFeAs, which indicates that $R^2 - R_H^2$ has a weak T dependence. We show

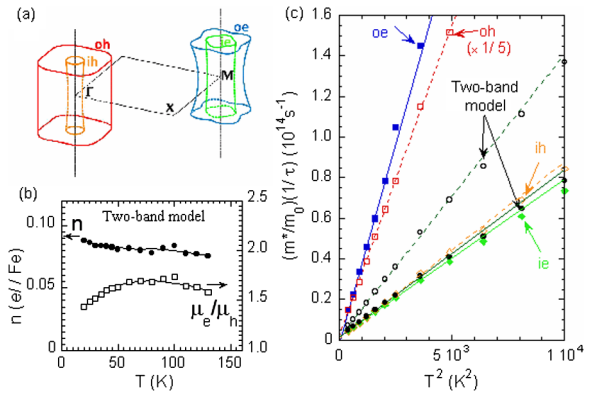

FIG. 3 (color online). (a) Schematic view of the Fermi surface sheets of LiFeAs. (b) Number of carriers and mobility ratio extracted from the compensated two-band model. (c) Scattering rates deduced in a two-band analysis (circles) and for the four-band solution given in the text, plotted versus T^2. Full (empty) symbols and full (dashed) linear fits are for the electrons (holes). Notice that the data for the oh band have been divided by 5.

in Fig. 3(c) that both scattering rates display a T^2 variation up to ~70 K [17], with $\mu_e/\mu_h \sim 1.5$ [see Fig. 3(b)] [24], and that the number of carriers is nearly T independent below 120 K. At higher T, thermal population of narrow bands might induce an increase of $n(T)$ [21], in analogy with the proposal we have made to explain the transport properties of $Ba(Fe_{1-x}Co_x)_2As_2$ [3].

Comparison with ARPES and dHvA: beyond the two-band model.—However, the deduced carrier content $n \sim 0.09$ el/Fe being 2 times smaller than that given by ARPES or by dHvA [9,11] implies that the two-band model only allows us to determine separate average parameters for the holes and electrons. Indeed, we do know from ARPES data [8,9] that hole carriers are located in a small inner (ih) band included in a large outer (oh) band. Similarly, it has been suggested [25] that, due to strong spin-orbit coupling, the electron bands should not be considered as two crossed degenerate elliptic pockets. As found in dHvA experiments in both LiFeP and LiFeAs [11], they split into an inner (ie) band included in an outer one (oe). The carrier mobilities are expected to differ substantially for these four bands, and the MR coefficient has now a more complicated expression than given in Eq. (4) [26]:

$$H^{-2}\Delta\rho/\rho(T,0) = \sigma_e\sigma_h(\mu_e + \mu_h)^2/\sigma^2 + \sigma_h A_h/\sigma + \sigma_e A_e/\sigma \qquad (6)$$

with $A_h = \sigma_{ih}\sigma_{oh}(\mu_{ih} - \mu_{oh})^2/\sigma_h^2$, where μ_h is now an effective hole band mobility given by $\sigma_h\mu_h = \sigma_{ih}\mu_{ih} + \sigma_{oh}\mu_{oh}$ and an effective number of holes is defined by $\sigma_h = n_h^{\rm eff} e\mu_h$. Similar expressions hold for the electron bands.

To approach a solution we may use for the T independent carrier contents those obtained from ARPES and dHvA experiments $n_{oh} \sim 0.16$ h/Fe, $n_{ih} \sim 0.03$ h/Fe, $n_{oe} \sim 0.11$ el/Fe, and $n_{ie} \sim 0.08$ el/Fe for the outer and inner hole (electron) bands, respectively. As we are still lacking sufficient experimental information, a unique solution cannot be acquired. However the dHvA results imply that the mobilities for the two electron bands are comparable. Matching the data with all these assumptions yields a strong differentiation of the two hole bands with a surprisingly much lower mobility for the outer band compared to the inner one. Also, imposing $\mu_{oe} \simeq \mu_{ie}$ puts some constraints on the value of $n_h^{\rm eff}$ that cannot be larger than ~0.06 h/Fe (see the Supplemental Material Ref. [17]). A solution with $n_h^{\rm eff} = 0.05$ h/Fe for any T is illustrated in Fig. 3(c) and gives a ratio between the electron (hole) mobilities, respectively, of $\mu_{ie}/\mu_{oe} = 3 - 6$ and $\mu_{ih}/\mu_{oh} \sim 17$. It corresponds to $n_e^{\rm eff} \sim 0.13$ el/Fe and it results in similar values for the effective electron and hole mobilities (see the Supplemental Material Ref. [17]), which would justify why Kohler's rule is obeyed in this compound. Whatever the value taken for $n_h^{\rm eff}$ in the range considered, we always find that the scattering rates for the different carriers increase as T^2 up to ~70 K, as seen in Fig. 3(c). This confirms a Fermi liquid behavior for both holes and electrons in agreement with density-functional theory and dynamical mean-field theory (DFT + DMFT) calculations [25].

The $T = 0$ extrapolated values correspond to mobilities of ~1000 $cm^2/(V \cdot s)$ for the smallest hole and electron bands, that is, using a Fermi velocity $v_F \sim 1 \cdot 10^5$ m/s [27] and an effective mass $m^* \simeq 4m_0$ [11], to mean free paths ≈ 2000 Å, in agreement with the high purity of our FP1 sample. Much faster relaxation is observed for the outer hole band which is exclusively constructed from the d_{xy} orbital, for which a stronger incidence of correlations on m^* and the scattering rates is expected from DFT + DMFT calculations [25,28].

Superconducting fluctuation contribution to conductivity.—We detail in the Supplemental Material [17] why the low field deviations from a H^2 behavior of the MR cannot be associated with a saturation of the normal state MR. This is confirmed experimentally, as such deviations were not detected in LiFeP, which has a lower $T_c = 7$ K than LiFeAs, with a similar band structure and residual resistivity as our samples [22]. Following our extensive study of cuprates [13], the SCF contribution to the conductivity is given by $\Delta\sigma_{\rm SF}(T) = \rho^{-1}(T) - \rho_n^{-1}(T)$, where $\rho_n(T)$ is the $H = 0$ extrapolation of the H^2 variation of the normal state MR. Let us notice here that $\Delta\sigma_{\rm SF}(T)$, which does not exceed 3% of the normal state values [see Fig. 2(b)], would be extremely difficult to extract directly from the $\rho(T)$ curves.

For our samples with the highest T_{c0}, the $\Delta\sigma_{\rm SF}$ data reported in a log-log scale in Fig. 4 as a function of the reduced temperature $\epsilon = \ln(T_c/T_{c0})$ resemble those found in cuprates [13]. After an initial power law behavior,

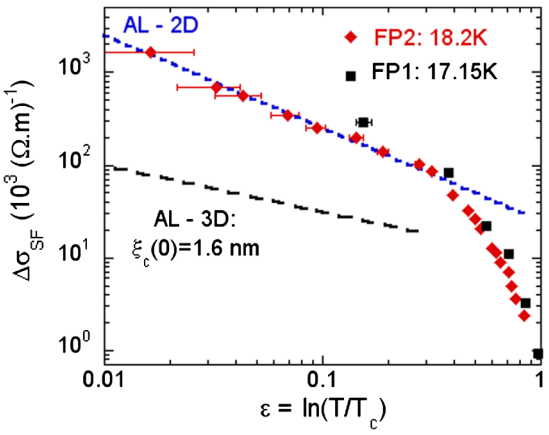

FIG. 4 (color online). Superconducting fluctuation conductivity $\Delta\sigma_{SF}$ for samples FP1 and FP2 as a function of $\epsilon = \ln(T/T_{c0})$, where T_{c0} is taken at the midpoint of the transition. The error bars for ϵ were determined using T_c values at 25 and 75% of the resistive transition. Estimates of the AL contribution in two and three dimensions are displayed as dashed lines for comparison.

$\Delta\sigma_{SF}(T)$ displays a cutoff, and only becomes negligible for $\epsilon \sim 1$. This larger SCF regime $\sim 2.5T_c$ than we find in cuprates points towards a two-dimensional (2D) character. This is better seen from the good fit with the 2D Aslamazov-Larkin (AL) formula

$$\Delta\sigma^{AL}(T) = (e^2/16\hbar s)\epsilon^{-1}, \quad (7)$$

for $0.017 \lesssim \epsilon \lesssim 0.3$, corresponding to $T = 1.02$ to $1.4T_c$, without any fitting parameter except the interlayer distance s taken here as the lattice parameter value $c = 6.36$ Å. The value of T_c is taken here at the midpoint of the transition but our conclusion remains valid for slightly different T_c determinations as shown by the error bars for ϵ in Fig. 4.

These 2D SCFs appear at first sight difficult to reconcile with the three-dimensional (3D) character of the normal and SC states in LiFeAs suggested by the weak anisotropies of the resistivity and upper-critical field H_{c2} [20,29,30] (see also the Supplemental Material Ref. [17]). Using, for instance, a single-band scheme to deduce the coherence length along the c axis from H_{c2}, we find $\xi_c \sim 16$ Å, which is more than twice the interlayer spacing. So one would expect for 3D fluctuations the much smaller contribution to the conductivity shown in Fig. 4. All this suggests that the present result is specific to multiband effects and that, above T_c, the fluctuating pairs are driven by a single 2D band. In view of our discussion above on the normal state, only the outer hole pocket that originates from in-plane d_{xy} orbitals is purely 2D [25,28,31].

It is worth comparing our results in LiFeAs with the case of MgB_2, a presumably much simpler multiband superconductor which also displays a rather low H_{c2} anisotropy factor ~ 2 at T_c. It has been proposed [14] that the SCFs are governed by a unique critical mode, which is dominated for $T >> T_c$ by the quasi-2D σ bands with the larger SC gap. However, near T_c, the critical mode should recover a 3D character due to both band contributions and the paraconductivity should diverge slightly slower than $\sim 1/\sqrt{\epsilon}$. Preliminary data [32] have been analyzed as suggesting 2D SCFs but, to our knowledge, no more reliable experimental work has been performed since.

Concerning the properties of the SC gaps in LiFeAs, they are found weakly anisotropic by ARPES, the largest $\Delta = 5$–6 meV for the small 3D hole pocket, while $\Delta = 3$–4 meV for the other bands [9,31], including the large 2D hole band. In that context one should rather expect a pronounced 3D character for the SCFs in LiFeAs [33].

Discussion.—In the specific case of pnictides, it has been underlined recently [16] that the SCFs are indeed expected to behave differently than for MgB_2 [14], as the pairing should be dominated by interband spin fluctuation interactions [34]. These authors have shown that, in such a case, the critical mode controlling the SCFs should have a simpler relation to the various bands, so that the AL formula known for single-band SC should remain valid either in two or three dimensions. Further experimental investigations of the SCFs along the same lines in other pnictide families would be necessary to get a more complete understanding of SCFs in these multiband compounds. For instance, one needs to ascertain whether the SCFs are dominated by a 3D contribution in the hole doped $BaKFe_2As_2$, as has been suggested from an investigation of the diamagnetism above T_c [35].

To conclude, we emphasized here the relevance of magnetoresistance data to unveil new physical phenomena. Our data provide indications which should help to investigate the incidence of spin fluctuations on the carrier scattering in the normal state and on the SC pairing in this specific LiFeAs system. On the other hand, it would be interesting to study whether other proposals such as orbital fluctuations or p-wave SC [10,36] might provide a natural explanation for these 2D SCFs. Unveiling the origin of these 2D SCFs should therefore give useful hints to clarify the mechanism of SC in LiFeAs.

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