Possible Kondo effect in the iron arsenides

Jianhui Dai,1 Guanghan Cao,1 Hai-Hu Wen,2 and Zhuan Xu1
1Department of Physics, Zhejiang University, Hangzhou 310027, China
2National Laboratory for Superconductivity, Institute of Physics, Chinese Academy of Science, Beijing 100190, China

The normal state of the iron arsenides shows the poor metallic behavior mixed with strong magnetic fluctuations. In particular, some FeAs-1111 and FeAs-122 compounds show the linear-T dependence of susceptibility above the spin-density wave (SDW) transition and the logarithmic upturn of resistivity at low temperatures. We suggest that this is due to the spin-flip scattering between the charge carriers and the local moments in the undoped FeAs layer where Kondo effect coexists with the SDW. This scenario is also accounted for the change of the magnetoresistance from positive to negative in the Sr3Sc2O5Fe2As2 compound.

PACS numbers: 71.10.Ay, 75.20.Hr, 71.27.+a, 74.20.Mn

Introduction. The recent discovery of superconductivity in the iron-arsenide materials with transition temperatures up to 56 K poses a new intellectual challenge in the correlated electron systems 1, 2, 3, 4, 5, 6. These materials consist of a unique two-dimensional conducting FeAs layer. The parent quaternary compounds (the 1111 series), such as LaFeAsO, show a structure distortion and a collinear spin-density wave (SDW) transition. They are suppressed by electron doping, giving rise to the superconductivity 7. This is remarkably similar to the cuprates is that the parent compound of the former is a multi-orbital metal. Yet the normal state properties of the FeAs-based materials are unusual. One of the most striking feature is the linear temperature dependence of static susceptibility above the SDW transition temperature. This behavior has been recently suggested as due to the strong exchange interaction among the so-called "preformed SDW moments" 8 or to the peculiarity of a two dimensional Fermi liquid 9. Another silent feature, perhaps of equal significance, is the resistivity upturn at low temperatures in some 1111 series with 1, 2, 3, 10 or without oxygens 10. This feature is hindered by the emergent SDW instability or superconductivity and is hidden in some ternary compounds (the 122 series) 11, 12.

In our point of view, the above two features are characteristic of the normal state of a single FeAs layer. The experimental evidence for the resistivity upturn being robust and intrinsic to the FeAs layer is accumulating. The transport measurement on a sizable single crystal of the BaFe2As2 sample exhibits this behavior both in the zero and finite magnetic field 13. Recently, a noticeable resistivity upturn behavior was found to pertain above \( T_c \) in the superconducting LaFe1-xNixAsO sample 14. Furthermore, a new 32522 compound, i.e., Sr3Sc2O5Fe2As2, was found to show a significant crossover from the metallic behavior at high temperatures to the upturn feature at low temperatures 13. The inter-layer spacing in this compound is much larger than those in the 1111 and 122 series. So it serves as an ideal system to study the physics of a single FeAs layer.

In this paper, we suggest that the above two characteristic features can be understood as due to the coexistence of Kondo effect and the SDW in the undoped FeAs layer. This Kondo effect may be regarded as a manifestation of the 3d multi-orbital correlations and it can be suppressed by electron doping and enhanced by small applied field. We note that the resistivity upturn at low temperatures is usually attributed to the charge localization or the Jahn-Teller effect. The purpose of the present paper is to provide qualitatively evidence that the Kondo scenario is more likely. Our interpretation of the resistivity upturn assumes the existence of "local moments" 10, while the latter is evidenced in LaFeAsO by recent electron spin resonance experiment 15.

Susceptibility and resistivity. Let us first briefly discuss some experimental results. The linear temperature dependence of the susceptibility above the SDW ordering temperature is universal in the iron arsenides including the F-doped 1111 13 and 122 series 12, 13. This behavior is an indication of strong antiferromagnetic interaction among the local moments, described by the \( J_1-J_2 \) Heisenberg model 20, 21. If one chooses \( S = 1 \) and assumes \( J_1 = J_2 (\equiv J_d) \), the susceptibility can be fitted by \( \chi = \chi_0 (1 + a T) \) (for \( T < 0.3 J_d \)) 8. For LaFeAsO1-xF_x or LaFe1-xNixAsO, however, while the coefficient of the linear-\( T \) term is nearly independent on \( x \), the Pauli part, \( \chi_0 \), decreases with increasing \( x \) 14.

For the resistivity, a noticeable upturn just before the SDW transition could be seen in LaFeAsO [the upper inset in Fig. 1(a)]. This behavior can be identified even in the normal state of the optimally-doped superconducting samples of LaFe1-xNixAsO [the upper inset in Fig. 1(b)]. The upturn after the minimum can be fitted by \( R \propto c_1 - c_2 \log T \) [Fig. 1(a,b)]. Moreover, the resistivity crossovers to \( R(T) \propto R(0) [1 - c T^2] \) as \( T \) decreases further [the lower inset in Fig. 1(b)]. Such kind of resis-
tivity evolution is a benchmark of Kondo effect [22].

\[ \rho(T) \text{ as a function of } T \text{ (Color online) Temperature dependence of the resistivity in the LaFeAsO and LaFeO}_{0.96}\text{Ni}_{0.04}\text{AsO samples.} \]

The spin-flip scattering and the local Fermi liquid. We now argue that the linear-\( T \) susceptibility and the Kondo-like resistivity can be understood as due to the coexistence of the local moments (\( S_{\alpha \beta} \)) and itinerant carriers (\( c_{k\alpha \sigma} \)) in the FeAs layer [10]. In the LDA language [23], they correspond to the coherent excitations in the vicinity of the Fermi-level and the incoherent excitations away from the Fermi-level, as illustrated in Fig. 2(a). The incoherent part is modeled by the \( J_1-J_2 \) Heisenberg model:

\[ H_J = J_J \sum_{(n.n.,\alpha)} S_{\alpha \beta} + J_J \sum_{(n.n.,\alpha)} S_{\alpha \beta} \cdot S_{\gamma \delta} + J_H \sum_{j,\alpha,\beta} S_{\alpha \beta} \cdot S_{\gamma \delta} \cdot \cdot \cdot \]

The coherent part is modeled by non-interacting fermions with hole and electron pockets near the \( \Gamma \) and \( M \) points respectively. The interaction between them is generally described by

\[ \sum_{kq\alpha\beta\sigma'\gamma} \sqrt{w_\alpha w_\beta} G_{k,q,\alpha,\beta,\gamma} c_{k+q,\alpha,\sigma}^\dagger \frac{\sigma_{\sigma'\gamma}}{2} c_{k,\beta,\sigma'} \cdot \cdot \cdot \]

where \( w_\alpha \) is the weight of the coherent excitations in the \( \alpha \)-th orbital [24]. So the system may be regarded as a generic momentum resolved Kondo lattice. Some components of \( G_{k,q,\alpha,\beta,\gamma} \) may be antiferromagnetic due to the strong orbital-mixing and the moderate 3d-Coulomb interaction, surpassing the Hund’s coupling. Though all five 3d-orbitals contribute to \( w \), some of \( w_\alpha \) may be vanishingly small due to possible orbital-selective Mott transition [Note that \( w = \sum_\alpha w_\alpha \) is small for a bad metal]. With these in mind, we consider a simplified model with a single conduction-electron band:

\[ H = w \sum_{k\sigma} (E_k - \mu) c_{k\sigma}^\dagger c_{k\sigma} + w J_K \sum_{j,\alpha} s_{j,c} \cdot s_{j,\alpha}^\gamma + H_J, \]

where \( E_k \) is the band energy with the width 2D [as illustrated in Fig. 2(a)], \( \mu \) is the chemical potential, \( s_{j,c} \) is the spin operator of the charge carrier, \( J_K \) the Kondo coupling and \( \mu \) the chemical potential.

FIG. 1: (Color online) Temperature dependence of the resistivity in the LaFeAsO and LaFeO\(_{0.96}\)Ni\(_{0.04}\)AsO samples. The temperature axis is scaled in log \( T \), while it is scaled in \( T^2 \) in the lower inset in Fig. 1(b). The straight red lines are guide to the eyes. The experiment data are taken from Ref. [14].

For the simplest case with \( \alpha = 1 \), i.e., the \( S = 1/2 \) Kondo lattice model, the coexistence of Kondo effect and antiferromagnetic ordering is possible for suitable \( J_K/D \) and \( J_\delta/(wD) \). A more plausible case is for \( \alpha = 1,2 \). This case is similar to the underscreened \( S = 1 \) Kondo lattice with an additional Hund’s coupling. It can be solved by the mean field (MF) approximation [27], and the coexistence of Kondo effect and magnetic ordering arises naturally in the MF solution. The ground state can be intuitively understood by distinguishing the hybridization and non-hybridization components [see in Fig. 2(b)], defined by \( \bar{S}_A = \bar{S}_1 + \bar{S}_2 \) and \( \bar{S}_B = \bar{S}_2 - \bar{S}_1 \) respectively. The distinction between the two components becomes more apparent if there is a weak Ising anisotropy. So the local moments are partially screened by the charge carriers, leading to the residual effective spins at each site. The magnitude of each of them decreases with increasing \( J_K \) and arrives at the minimal value 1/2 if the hybridization sector is in the Kondo phase. The local magnetic entropy is suppressed further by the \( J_1-J_2 \) frustrations, leading to a collinear SDW with a reduced ordered moment \( m_d \) observed experimentally.

In the Kondo phase of both cases, the low energy physics is determined by the Kondo temperature, \( T_K \), and the coherent temperature, \( T_{coh} \). The ratio of them can be approximated by

\[ T_{coh}/T_K \sim e^{-f(\mu)} \]

where \( f(\mu) = \int_{-D-\mu}^{D-\mu} \frac{1}{2} \rho(\omega) d\omega \).

FIG. 2: (Color online) (a) Schematic picture for coherent and incoherent excitations. (b) The hybridization (red) and non-hybridization (green) components distinguished in the MF approximation.
liquid. Here the bad metal behavior is also featured by comparably small ratio of the saturated resistivity to the one at room temperature (say at $T = 300$ K) due to small $w$. In the high temperature regime, however, the transport may deviate from the Fermi liquid behavior due to fluctuations beyond the MF approximation or other neglected multi-orbital effects.

Implications of Kondo effect for the 1111 and 122 series. The local Fermi liquid solution provides a scenario for the coexistence of Kondo effect and collinear SDW. The true long range ordering can be established at finite temperatures with small inter-layer coupling $J_z$. The ordering temperature $T_{SDW} \approx 4\pi\Delta_{SDW}/\ln(\Delta_{SDW}/J_z)$, with $\Delta_{SDW} = J_d m_d^{\Delta}$ [32].

In the absence of $J_K$, the static susceptibility is contributed from both types of excitations: $\chi = \chi_{inc} + \chi_{coh}$. Above $T_{SDW}$, $\chi_{inc}$ is given by the $J_1$-$J_2$ model, $\chi_{inc} = \chi_0(1+aT)$ for $T < 0.3J_z$ [38]. $\chi_{coh}$ is contributed from the coherent excitations, which is of the Pauli form. This part is proportional to the DOS at the Fermi level, $\rho_F$. So we have $\chi = \chi_0 + \alpha T$, where $\chi_0 = \chi_{coh} + \chi_0$, $\alpha = a\chi_0$. According to the LDA calculation, the electron doping (either via F or Ni) will shift $\mu$ to the right, leading to a decrease in $\rho_F$, as illustrated in Fig. 2(b). Thus the Pauli part $\chi_0$ shows an overall decreasing tendency, in agreement with the Ni-doping experiment [14]. With the finite $J_K$, as $w$ is very small for a bad metal, the effective coupling $wJ_K$ does not essentially change the linear behavior above $T_{SDW}$ but may slightly enhance the Pauli susceptibility below $T_K$. As a peculiar feature of the coexistence of the Kondo effect and the $J_1$-$J_2$ coupling, the local moments do not show a significant Curie-like behavior for $T > T_K$ as in the conventional Kondo effect.

The low temperature upturn behavior of electronic resistivity is usually attributed to the localization effect driven by doping-induced disorders. But this is less likely in the iron arsenides, as the upturn appears most significantly in the parent compounds and decreases with increasing Ni-doping. We emphasize that the electron correlation could be responsible to the upturn behavior in a translational invariant system. In the underdoped cuprates, the strong Coulomb repulsion results in a gauge interaction between the spinon and the holon, leading to a metal-insulator transition in the pseudo-gap phase [31]. In the iron arsenides, the Coulomb repulsion is moderate so that the normal state is a bad metal which was suggested to be in proximity to the Mott transition [20]. Here we just interpret the incoherent transport as due to the coexistence of Kondo effect and the SDW. Because $T_{coh} \approx 0$, the normal state is characterized by the Kondo temperature $T_K \sim D e^{-1/(w J_F \rho_F)}$. For $T \leq T_K$, the resistivity follows the well-known logarithmic behavior: $R \approx c_1 - c_2 \log T$, with the coefficient $c_2 \sim w J_K \rho_F$. This explains why $c_2$ and $\chi_0$ show the same overall doping dependence in the LaFe$_{1-x}$Ni$_x$AsO compounds [14].

When $T$ goes down to $T_{SDW}$, the SDW ordering gap $\Delta_{SDW}$ opens, triggering the SDW instability of the charge carriers. So $\rho_F$ has a drop across $T_{SDW}$. As a result, $\chi_0$ has a drop too as observed in some 122 series [13]. Accordingly, there are two different Kondo temperatures, $T_K^{(1)}$ and $T_K^{(2)}$, associated with $\rho_F$ for $T > T_{SDW}$ and $T < T_{SDW}$, respectively. The resistivity has also a drop due to a drastic increase in the scattering rate $1/\tau$ at the nested Fermi surfaces. So if $T_K^{(1)} \sim T_{SDW}$, there is a resistivity upturn above the SDW transition (as in many 1111 series). Otherwise, for $T_K^{(1)} \ll T_{SDW}$, no upturn appears above $T_{SDW}$ (as in some 122 series). These two cases are in accordance with the fact that $J_z$ decreases from the 122 to 1111 series. In the SDW phase, the upturn shows up again when $T \lesssim T_K^{(2)}$ [as in Fig. 1(a)]. The feature is most significant if $T_K^{(1)} \gtrsim T_{SDW}$ or $\Delta_{SDW} \rightarrow 0$. This is the case for the 32522 sample (see the inset in Fig. 3) where $J_z$ should be very small.

![FIG. 3: (Color online) Temperature dependence of the resistivity for the 32522 sample. The temperature axis is scaled in log $T$. The straight red lines are guide to the eyes. The experiment data are taken from Ref. [19].](image-url)

The magnetoresistance and the possible ordered moment in the 32522 sample. At zero temperature the magnetic field dependence of the resistivity, the magnetoresistance (MR), can be deduced from the Friedel sum rule [22]. In the conventional Kondo problem, the impurity spin is completely screened so that the MR is negative. This is because the spin-flip scattering and spin compensated ground state are suppressed in the presence of magnetic field.

However, the situation for the present Kondo effect is different. Now the local moments are only partially compensated. The residual ordered moment plays the role of an intrinsic magnetic field. It is in general a function of the external magnetic field $h$, denoted by $m_d^{(h)}$. Note that $m_d^{(h)}$ will be always suppressed by $h$ for $h < h_1$, with $h_1$ being defined by $m_d^{(h_1)} = 0$. For $h > h_1$, the local moment polarizes gradually along $h$ and saturates at the original quantized magnitude (e.g., 0.5 in the unit of $g\mu_B$) as $h$ approaches $J_d$. Applying the sum rule
gives contribution of the local moment to the resistivity: \( R_{loc}(m_d) = R_{loc}(0) \cos^2(\pi m_d) \), with \( R_{loc}(0) \) being the zero temperature resistivity for \( h = 0 \) or \( m_d(0) = m_d \). So we have
\[
\frac{\Delta R_{loc}(h)}{R_{loc}(m_d)} = \frac{\sin \pi (m_d - m_d(h)) \sin \pi (m_d + m_d(h))}{\cos^2(\pi m_d)}.
\]
When \( h > h_2 \), where \( m_d(h_2) = m_d \), the sign of the MR changes from positive to negative.

We find that the MR result depends only weakly on the details of \( m_d(h) \) if \( m_d, h_1, \) and \( J_d \) are properly fixed. Thus we can use a simplified molecular field approximation, i.e., \( m_d(h) = m_d(1 - h/h_1) \) for \( h < h_1 \) and \( m_d(h) = 0.5 \sqrt{\frac{h-h_1}{J_d-h_1}} \) for \( h_1 < h \leq J_d \). Eq.(2) is then compared with the \( T = 2 \) K data in the 32522 sample. Fig. 4 shows the MR result of this estimation with \( m_d \approx 0.014 \text{gB}, h_1 \approx 1.15 \text{ Tesla}, \) and \( J_d \approx 60 \text{ meV} \). We expect that the estimated MR curve could be more smooth if \( m_d(h) \) is accurately calculated from the \( J_1-J_2 \) or \( O(3) \) models in the presence of external field \( h \). It is interesting to note that the very small ordered moment obtained by fitting the MR data may be responsible to a tiny kink at zero field seen in Fig. 3 near \( T \approx 20 \text{ K} \). The kink disappears at \( h = 9 \text{ Tesla} \) when the SDW gap \( (\approx 0.8 \text{ meV}) \) is completely suppressed. Our observation also suggests that both the positive and negative parts of the MR should be robust as \( T \to 0 \).

![FIG. 4: (Color online) The magnetoresistance at \( T = 2 \) K for the 32522 sample. The experimental data (open squares) are taken from Ref. 13. The red dashed line is a theoretical estimation at zero temperature based on the Friedel sum rule.](image)

Finally, we remark that for the undoped 1111 and 122 compounds, \( \Delta_{SDW} \approx 15 - 25 \text{ meV} \), so \( h_1 \approx 150 - 250 \text{ Tesla} \). This means that the negative MR is hardly observable experimentally. Upon electron doping or chemical pressure, however, \( \Delta_{SDW} \) decreases dramatically, the negative MR could be observed if the superconductivity does not intervene the Kondo effect. The chemical pressure can be realized by P doping of As which effectively enhances \( w \) and suppresses \( m_d \) so that \( T_{coh} \) may increase slightly. All these could be possibly seen in the iron phosphides where the superconductivity is weak and can be completely suppressed by small magnetic field.

In summary, the logarithmic-\( T \) resistivity behavior, its relation with the doping dependence of the Pauli susceptibility, and the sign change in the MR, indicate a possible Kondo effect in the single FeAs layer. The coexistence of Kondo effect and the SDW is inherited from the multi-orbital 3d-electron correlations and plays a role in the rich complexities of the iron arsenides.

We thank W.Q. Chen, X.H. Chen, Y. Liu, Z.Y. Lu, Q. Si, N.L. Wang, Z.Y. Weng, H.Q. Yuan, F.C. Zhang, and J.X. Zhu for useful discussions. This work was supported by the NSF of China and PCSIRT (IRT-0754) of Education Ministry of China.

[1] Y. Kamihara et al., J. Am. Chem. Soc. 130, 3296(2008).
[2] X.H. Chen et al., Nature 453, 761(2008).
[3] G.F. Chen et al., Phys. Rev. Lett. 100, 247002(2008).
[4] Z.A. Ren et al., Europhys. Lett. 82, 17002(2008).
[5] H.H. Wen et al., Europhys. Lett. 82, 17009(2008).
[6] C. Wang et al., Europhys. Lett. 83, 67006(2008).
[7] Clarina de la Cruz et al., Nature 453, 899(2008).
[8] G.M. Zhang et al., arXiv:0809.3874.
[9] M.M. Korshunov et al., arXiv:0901.0238.
[10] X. Zhu et al., arXiv:0810.2531.
[11] M. Rotter, M. Tegel, and D. Johrendt, Phys. Rev. Lett. 101, 107006(2008).
[12] K. Sasmal et al., Phys. Rev. Lett. 101, 107007(2008).
[13] G.F. Chen et al., Chin. Phys. Lett. 25, 3403(2008).
[14] X.F. Wang et al., arXiv:0806.2452.
[15] G. Cao et al., arXiv:0807.4328.
[16] X. Zhu et al., arXiv:0811.2205.
[17] J. Dai et al., arXiv:0808.0305.
[18] T. Wu et al., arXiv:0811.2567.
[19] R. Klingeler et al., arXiv:0808.0708.
[20] G. Wu et al., J. Phys. Cond. Mat. 20, 422201(2008).
[21] Q. Si and E. Abrahams, Phys. Rev. Lett. 101, 076401(2008).
[22] F. Ma, Z.Y. Lu, and T. Xiang, arXiv:0804.3370.
[23] A.C. Hewson, The Kondo Problem to Heavy Fermions, Cambridge University Press, New York, 1997.
[24] K. Haule, J.H. Shim, and G. Kotliar, Phy. Rev. Lett. 100, 226402(2008); D.J. Singh and M.H. Du, Phy. Rev. Lett. 100, 237003(2008).
[25] Other approaches to the interplay of the itinerant carriers and local moments see: J. Wu, P. Phillips, and A.H. Castro, Phys. Rev. Lett. 101, 126401(2008); S.P. Kou, T. Li, and Z.Y. Weng, arXiv:0811.4111; A. Hackel and M. Vojta, arXiv:0812.2394.
[26] M. Vojta, Physica B378, 23(2006).
[27] M. Vojta, Phys. Rev. B78, 125109(2008).
[28] N.B. Perkins et al., Europhys. Lett. 79, 57006(2007).
[29] S. Burdin and V. Zlatic, arXiv:0812.1137.
[30] K. Haule and G. Kotliar, arXiv:0805.0722.
[31] C. Xu, M. Muller, and S. Sachdev, Phys. Rev. B78, 020501(R)(2008).
[32] P.A. Marchetti, Z.B. Su, and L. Yu, J. Phys. C19, 125212(2007).