Multiband effect and electron-hole asymmetry in the transport properties of iron pnictides

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## The different families of pnictides

| 1111 | SmFeAsO$_{1-x}$F$_x$ | Synthesis of single crystals difficult  
Few results  
Chemical composition |
<table>
<thead>
<tr>
<th></th>
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</thead>
<tbody>
<tr>
<td></td>
<td>Highest $T_c = 57$K</td>
<td></td>
</tr>
</tbody>
</table>

| 122  | BaFe$_2$As$_2$  
SrFe$_2$As$_2$  
CaFe$_2$As$_2$  | A very rich family  
Good single crystals |
|------|------------------|---------------------|

| 111  | LiFeAs            | Good single crystals  
Stoechiometric $\rightarrow$ low defect content  
Very sensitive to air |
|------|------------------|---------------------|

### Chalcogenides
The different families of pnictides

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<tr>
<th>Family</th>
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Chalcogenides
Outline

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   Necessity of a multiband description

Analysis of the resistivity and Hall effect measurements in a minimal two-band model: electron doped BaFe$_2$As$_2$ with Co and Ni substitution
   Strong electron-hole asymmetry in the scattering rates
Comparison with ARPES results

Isovalent substitution: Ru/Fe
Resistivity, Hall effect and magnetoresistance
   Evidence of a Fermi liquid behavior in a two band model

LiFeAs Resistivity, Hall effect and magnetoresistance
Analysis in relation with ARPES and quantum oscillations data
Beyond the two-band model
The 122 Phase $A\text{Fe}_2\text{As}_2$ ($A=$Ba, Sr, Ca, Eu)

Possibility to get large single crystals

$\text{Pr}^{3+}$, $\text{La}^{3+}$

Electron-doped

$T_{c}^{\text{max}} \sim 45\text{K}$ in CaFe$_2$As$_2$

$\text{Ba}^{2+}$, $\text{Sr}^{2+}$, $\text{Ca}^{2+}$, ..

$\text{K}^{+}$

Hole-doped

$T_{c}^{\text{max}} = 37\text{K}$

$\text{Ru}$ on Fe site

$\text{P}$ on As Site

Isovalent substitution

$\text{Co} : 3d^7$

$\text{Rh} : 4d^7$

$\text{Ir} : 5d^9$

$\text{Ni} : 3d^8$

$T_{c}^{\text{max}} = 25\text{K}$

$\text{Fe}$

$\text{Co}$

$\text{Ni}$

$\text{Ru}$

$26$

$27$

$28$

$44$
122 Phase BaFe$_2$As$_2$ : crystal growth

Self flux method

\[
\begin{align*}
\text{Fe} + \text{As} & \quad \xrightarrow{800^\circ\text{C}} \quad \text{FeAs} \\
\text{Fe} + \text{Co} & \quad \xrightarrow{700^\circ\text{C}} \quad \text{CoAs}
\end{align*}
\]

Ba + FeAs, with excess of CoAs

\[
\text{Ba(Fe}_{1-x}\text{Co}_x)_2\text{As}_2
\]

Synthesis in quartz tubes, sealed under vacuum
Kept at 1180°C for 4h and cooling at 5°C/h
1000°C for 6h
Cooled down to room T

Mechanical extraction of platelets single crystals,

Measurements of Transport properties
Thickness between 10 to 30 mm

Co content determined by wave length dispersive X-ray spectroscopy
Transport properties of Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$ single crystals

Structural and magnetic transitions

Coexistence of AF and SC states at local scale

NMR: Y. Laplace et al., PRB (2009)

FRA et al. PRL 2009

Ni et al. PRB (2008), J.H. Chu et al., PRB (2009), L. Fang et al., PRB 2009
Transport properties of $\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$ single crystals

No splitting between structural and AF transitions

$\rho_{ab}$ (m$\Omega$.cm)

$T$ (K)

$\text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2$

$\frac{d\rho_{ab}}{dT}$ (m$\Omega$.cm/K)

$T$(K)

$\text{Potassium content}$

B. Shen et al., PRB (2011)
Resistivity evolution by substitution in the BaFe$_2$As$_2$ family

Ba(Fe$_{1-x}$Co$_x$)$_2$As$_2$

Electron doped

F. R.A. et al., PRL 2009

BaFe$_2$(As$_{1-x}$P$_x$)$_2$

Isovalent

S. Kasahara et al., PRB 2010

Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$

Isovalent

F. R.A. et al., PRB 2010

Ba$_{1-x}$K$_x$Fe$_2$As$_2$

Hole doped

B. Shen et al., PRB (2011)
The CaFe$_2$As$_2$ family

Ca(Fe$_{1-x}$Co$_x$)$_2$As$_2$

Ca$_{1-x}$R$_x$Fe$_2$As$_2$

S.R. Saha et al., arXiv 1105.4798

L. Harnagea et al., arXiv 1011.2085
Hall effect: Large variation with temperature

\[ \text{Ba(Fe}_{1-x}\text{Co}_x)\text{As}_2 \]

Electron doped

\[ \text{BaFe}_2(\text{As}_{1-x}\text{P}_x)\text{As}_2 \]

Isovalent

\[ \text{Ba(Fe}_{1-x}\text{Ru}_x)\text{As}_2 \]

Isovalent

\[ \text{Ba}_{1-x}\text{K}_x\text{Fe}_2\text{As}_2 \]

Hole doped

F. R.A. et al., PRL 2009

S. Kasahara et al., PRB 2010

B. Shen et al., PRB (2011)
The Iron based superconductors: multiband structure

Electronic structure: band structure calculation

BaFe$_2$As$_2$

Compensated semimetal: $n_e = n_h$

Multiband effects
Modification by doping

Strong influence on transport properties
Resistivity, Hall effect, magnetoresistance, …
One-band versus multiband

Single band metal

- Drude formula
  \[ \rho = \frac{m^*}{ne^2 \tau} \]
- Hall effect
  \[ R_H = \frac{1}{ne} \]

Iron-based superconductors

Minimal two-band model

- Drude formula
  \[ \sigma = \sigma_e + \sigma_h \]
- Hall effect
  \[ 1/\rho = 1/\rho_e + 1/\rho_h \quad \rho_{e,h} = \frac{m_{e,h}^*}{n_{e,h} e^2 \tau_{e,h}} \]
- Hall coefficient
  \[ R_H = \frac{1}{e} \frac{(-n_e \mu_e^2 + n_h \mu_h^2)}{(n_e \mu_e + n_h \mu_h)^2} \]
- Hall coefficient
  \[ \frac{\delta \rho}{\rho(0)} = \frac{\sigma_e \sigma_h (\mu_e + \mu_h)^2}{(\sigma_e + \sigma_h)^2} H^2 \]
- Magnetic field
  \[ m_{e,h} = \frac{e \tau_{e,h}}{m_{e,h}^*} \]

\[ \mu_{e,h} = \frac{e \tau_{e,h}}{m_{e,h}^*} \]
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Hall effect: $\text{Ba(Fe}_{1-x}\text{Co}_x\text{)}_2\text{As}_2$ and $\text{Ba(Fe}_{1-x}\text{Ni}_x\text{)}_2\text{As}_2$

Transport dominated by the electrons all over the phase diagram

Drastic change of the Hall coefficient in the AFM state near $x \sim 0.02$

Seen also by thermopower measurements

E.D. Mun et al. PRB (2009)
Hall number: $\text{Ba(Fe}_{1-x}\text{Co}_x\text{)}_2\text{As}_2$ and $\text{Ba(Fe}_{1-x}\text{Ni}_x\text{)}_2\text{As}_2$

In a two-band model:

$$\frac{1}{n_H e} = |R_H| = \frac{-R_e \sigma_e^2 + R_h \sigma_h^2}{(\sigma_e + \sigma_h)^2}$$

- $R_e = \frac{1}{n_e e}$
- $R_h = \frac{1}{n_h e}$

$n_H$ upper bound for $n_e$
Analysis in a two-band model

One electron band and one hole band: 4 unknown quantities: \( n_e, n_h, \tau_e, \tau_h \)

Experimentally: 3 equations

Conductivity: \( \sigma = \sigma_e + \sigma_h \)

Hall effect: \( \frac{1}{n_he} = |R_H| = \frac{\left| -R_e\sigma_e^2 + R_h\sigma_h^2 \right|}{(\sigma_e + \sigma_h)^2} \)

Charge conservation: \( n_e = n_h + x_{Co} \)

Estimate of the electron density: ARPES measurements
Using ARPES measurements to determine $n_e$ and $n_h$

V. Brouet et al., PRB (2009)

Hole bands: two 2D, one 3D

2 degenerate electron bands: 2D

4% Co

7% Co

Hall number at low T

$n_e - n_h = x_{Co}$

Good agreement between ARPES and transport data

Holes not directly visible in the transport properties
Electronic scattering rates

\[ n_e(T) = n_H(T) \]

\[ \rho(T) = \frac{m_e}{n_e(T)e^2\tau(T)} \]

\[ \cot(\Theta_H) = \rho \left| R_H \right| = \frac{m_e}{e\tau} \]

Up to 150K: \( 1/\tau = 1/\tau_0 + BT^2 \)

Electron-electron interaction

Fermi liquid behavior

\[ 1/\tau_0 \text{ independent on Co doping for } x \leq 20\% \]

No contribution of phonons nor spin fluctuations to the electronic scattering rates
Electron and hole mobilities in $\text{Ba(Fe}_{1-x}\text{Co}_x\text{)}_2\text{As}_2$

Holes more strongly scattered than electrons

Optical measurements
E. Van Heumen et al., arXiv 0912.0636

Electronic Raman scattering
R. Hackl et al.

Superconducting state for $x=7\%$

The holes are 15 times more scattered than the electrons

Interband scattering more efficient for the holes than for the electrons
Anisotropy of the scattering rates along the FS sheets

Multiorbital composition of each Fermi pocket

A.F. Kemper et al. PRB 2011

d_{xz}, d_{yz}, d_{xy}

Anisotropy of the effective mass and Fermi velocity

Scattering by spin fluctuations
One particle scattering rates affected by
- the orbital character of the initial and final states
- momentum dependence of the spin susceptibility

Strong anisotropy of the scattering rates

Transport dominated by small parts of the electron FS sheets with d_{xy} character with long lifetimes and large v_F
Anisotropy of the scattering rates along the FS sheets

ARPES: in Ba(Fe$_{1.2}$Co$_{0.8}$)$_2$As$_2$ : V. Brouet et al., arXiv 1105.5604

 lifetimes about twice longer on the $d_{xy}$ parts of the SF sheets both for the hole and electrons pockets

$$1/l = 1/v_F \tau$$

<table>
<thead>
<tr>
<th>State</th>
<th>$k_P (\pi/\alpha$ units)</th>
<th>$v_P$ (eV.Å)</th>
<th>$m^*/m_b$</th>
<th>$\delta k(E_F) (\pi/\alpha)$</th>
<th>$n$ (carr/Fe)</th>
<th>$\hbar/\tau$ (meV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>hole $d_{xz}/d_{yz}$</td>
<td>0.06</td>
<td>0.5</td>
<td>2.7</td>
<td>0.11</td>
<td>0.006</td>
<td>44</td>
</tr>
<tr>
<td>hole $d_{xy}$</td>
<td>0.22</td>
<td>0.4</td>
<td>2.3</td>
<td>0.07</td>
<td>0.038</td>
<td>22</td>
</tr>
<tr>
<td>electron $d_{xz}/d_{yz}$</td>
<td>0.25</td>
<td>0.6</td>
<td>2.4</td>
<td>0.09</td>
<td>0.12</td>
<td>43</td>
</tr>
<tr>
<td>electron $d_{xy}$</td>
<td>0.30</td>
<td>0.7/1.2</td>
<td>5/2.9</td>
<td>0.04</td>
<td>22/38</td>
<td></td>
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Seems difficult to reconcile with the Hall effect dominated by the electrons
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Transport properties of Ru-substituted BaFe\textsubscript{2}As\textsubscript{2}

In a two band model:
mobility of holes overcomes that of electrons
at low T for x > 0.25
Ba(Fe$_{1-x}$Ru$_x$)$_2$As$_2$ : $\rho$, $R_H$, Magnetoresistance

For a compensated semi-metal: $n_e = n_h = n$

$$\frac{\delta \rho}{\rho(0)} = a(T)H^2$$

$R_H / \rho = \mu_h - \mu_e$

$$\frac{\delta \rho}{\rho(0)} = \frac{\mu_e \mu_h H^2}{n}$$

$1 / \rho = (\mu_h + \mu_e)ne$

$T^2$ behavior for both holes and electrons

Fermi liquid
Comparaison with ARPES measurements

V. Brouet et al, PRL (2010)

For the hole pockets:
FS significantly warped along $k_z$

Ru : isovalent substitution

$\frac{n_e}{n_h} = n \approx 0.11/Fe$

But other ARPES studies show no change of the carrier concentration

Electronic correlations strongly reduced $v_F$ nearly three times larger

Might explain the increase of the hole mobility
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LiFeAs

$T_c \sim 17-18\text{K}$

Stoechiometric

Nearly compensated semi-metal

Defect free

Small upper-critical fields

Quantum oscillation measurements

Non polar surface : ARPES

Drawback: Very sensitive to air

Mechanism of superconductivity?

- Absence of nesting: Spin fluctuations?
- Orbital fluctuations : $s_{++}$

$a=b=3.7715(2)\text{Å}, \ c=6.3574(3)\text{Å}$
ARPES results on LiFeAs

Good agreement with calculations if correlations are taken into account

Borisenko et al. PRL (2010)

Two hole bands
Two electron bands

\( n_e \approx n_h \approx 0.2 \) carriers/Fe

Yin et al., Nature materials
De Haas van Alphen experiments on LiFeAs

C. Putzke et al., PRL (2011)

3 different frequencies
area of the FS sheets
Electron bands

Inner band: ~ 0.08 el/Fe
Outer band: ~ 0.11 el/Fe
Transport properties of LiFeAs: resistivity

Compared to previous reports

Lower residual resistivity
larger RRR = $\rho(300\text{K})/\rho_0 \sim 250$
LiFeAs: a Fermi liquid compound?

\[ \rho(T) = \rho_0 + AT^2 \]

\[ A \approx 9 \, n\Omega \cdot \text{cm}/K^2 \]

\[ \rho_0 = 1.5 \, \mu\Omega \cdot \text{cm} \]

Matthiessen’s rule is not obeyed

Multiband material

<table>
<thead>
<tr>
<th>Sample</th>
<th>( T_c ) (K)</th>
<th>( \rho_0 ) (( \mu\Omega \cdot \text{cm} ))</th>
<th>RRR</th>
<th>( A ) (( n\Omega \cdot \text{cm}/K^2 ))</th>
</tr>
</thead>
<tbody>
<tr>
<td>VDP1</td>
<td>17.75</td>
<td>4.15</td>
<td>80</td>
<td>9.7</td>
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<tr>
<td>VDP2</td>
<td>17.6</td>
<td>2.93</td>
<td>119</td>
<td>9.3</td>
</tr>
<tr>
<td>VDP3</td>
<td>17</td>
<td>4.55</td>
<td>79</td>
<td>9.5</td>
</tr>
<tr>
<td>FP1</td>
<td>18.2</td>
<td>1.47</td>
<td>225</td>
<td>8.8</td>
</tr>
<tr>
<td>FP2</td>
<td>17.15</td>
<td>1.21</td>
<td>263</td>
<td>9.3</td>
</tr>
<tr>
<td>FP3</td>
<td>15.6</td>
<td>6.34</td>
<td>62</td>
<td>12.7</td>
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<tr>
<td>Ref.[2]</td>
<td>17.75</td>
<td>15.2</td>
<td>38</td>
<td>22</td>
</tr>
<tr>
<td>Ref.[3]</td>
<td>17.3</td>
<td>13</td>
<td>53</td>
<td>20</td>
</tr>
</tbody>
</table>
Transport properties of LiFeAs: Hall effect

The Hall coefficient is negative

Transport dominated by the electrons as in BaFe$_2$As$_2$

Small minimum in $R_H$: corresponds to the change in curvature of $\rho(T)$
Transport properties of LiFeAs: Magnetoresistance

\[ \frac{\delta \rho}{\rho(0)} = \frac{\rho(H) - \rho(0)}{\rho(0)} = a(T)H^2 \]

In a single band metal: 
\[ \frac{\delta \rho}{\rho(0)} \propto \left( \frac{H}{\rho} \right)^2 \]

Kohler’s rule
Interpretation of the transport data in a two-band model

Ratio $\mu_e / \mu_h \approx 1.5$

Scattering rates $1/\tau \propto T^2$

Fermi liquid behavior for both carriers

BUT : $n \approx 0.08$ carriers/Fe

More than twice less than given by ARPES or Quantum oscillations experiments
Beyond the two band-model - 1

4 electronic bands with 4 different mobilities

Number of carriers taken from ARPES and dHvA data

\[ \frac{1}{H^2} \frac{\partial \rho}{\rho(0)} = a(T) = \frac{\sigma_e \sigma_h (\mu_e + \mu_h)^2}{(\sigma_e + \sigma_h)^2} + \frac{\sigma_h A_h}{(\sigma_e + \sigma_h)} + \frac{\sigma_e}{(\sigma_e + \sigma_h)} A_e \]

\[ A_h = \frac{\sigma_{ih} \sigma_{oh} (\mu_{ih} - \mu_{oh})^2}{\sigma_h^2} \]

Effective mobility

Effective hole band

Effective number of carriers

\[ \sigma_h = n_h^{\text{eff}} e \mu_h \]

Can vary with temperature

Effective electron band

Two-band model with a Hall coefficient

\[ R_H = \frac{(-n_e^{\text{eff}} \mu_e^2 + n_h^{\text{eff}} \mu_h^2) e}{(\sigma_e + \sigma_h)^2} \]
Beyond the two band-model - 2

Four different unknown parameters and only three experimental equations

A unique solution cannot be acquired

Good solution must be such as \( \mu_{ie} \geq \mu_{oe} \) and \( 0.02 \ h / Fe \leq n_h^{\text{eff}} \leq 0.06 \ h / Fe \)

For instance:

\[ n_h^{\text{eff}} = 0.05 \ h / Fe \]

independent of \( T \)

\[ \mu_{ie} \cong \mu_{ih} \]

For all the carriers:

\[ 1/\tau \propto T^2 \]

For the outer hole band

ARPES measurements give a MDC half width: \( \delta k = 18 \ \text{meV} \) \quad (A.A. Kordyuk et al. PRB 2011)

\( (m^*/m_0)(1/\tau) \sim 8 \times 10^{13} \ \text{s}^{-1} \) comparable with \( \sim 2 \times 10^{13} \ \text{s}^{-1} \) found here by transport

Measurements of the lifetimes for the other pockets should be very interesting
Conclusion

- Multiband description of the transport properties

- For the compounds with isovalent substitution Ru/Fe
  - Description in a two band model seems reasonable
  - Number of carriers compatible with ARPES (Ru/Fe)
  - Fermi liquid behavior for both the holes and the electrons
  - Mobilities of holes and electrons comparable

- LiFeAs: low defect content
  - Comparison with ARPES and quantum oscillation measurements
  - Description more realistic of the transport properties by taking into account four different bands.

- Co substitution in BaFe$_2$As$_2$
  - Strong electron-hole asymmetry in the scattering rates: holes not visible in the transport properties
  - Difficult to reconcile with the ARPES data on the different lifetimes
  - Many theoretical questions….
Collaborators

**SPEC**  Dorothée Colson, Anne Forget
Aretha Olariu (post-doc)
P. Bonville (Mössbauer)

**LPS**  Véronique Brouet (ARPES)
H. Alloul
J. Bobroff, Y. Laplace, Y. Texier (RMN)

RMN: Y. Laplace et al., PRB **80** (2009).


**LiFeAs** :  Transport: FRA et al, submitted (2012).