Phase Diagram of Pnictide Superconductors

Interplay between magnetism, structure, and superconductivity



Leibniz-Institut für Festkörper- und Werkstoffforschung Dresden

B. Büchner



Institut für Festkörperforschung IFW Dresden



Leibniz-Institut für Festkörper- und Werkstoffforschung Dresden High Temperature Superconductivity

in FeAs Compounds



OUTLINE

- Introduction
- Phase Diagram of LaFeAsO_{1-x}F_x (and other pnictides)

Structural, magnetic, and superconducting transitions

Superconducting State

Gap, penetration depth, relaxation rate

Normal State Properties

Electronic Structure (XAS, PES, ARPES) "Pseudogap" (NMR, χ) Electronic transport and spin fluctuations Charge inhomogeneity (NQR)

Tuesday

Superconductivity in La(O_{1-x}F_x)FeAs: The initiation



Dec. 2008: More than 600 papers on pnictide superconductors

Superconductivity above 50 K



C. Hess et al., EPL (2009) similar data: e.g. Ren et al. Chin Phys. Lett. 2008

Superconductivity above 50 K

 $La \rightarrow \dots Sm \dots \rightarrow Gd$



S. Wurmehl et al, work in progress

Fe pnictides: A class of high T_c superconductors



1111 systems

La, Ce ... Gd-based F, O doped

F doped REOFeAs O defici, REOFeAs

Sm doped SrFFeAs



Ba(FeAs)₂

122 systems

Ba, Sr, Eu, Ca-based K, Na, **Co** doped

 T_c up to 39 K

Intermetallic comp. Single crystals

> M. Rotter, M. Tegel, and D. Johrendt **PRL 2008**

LaFeAsO





FeSe



of high T_c superconductors



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LaFeAsO





FeSe

Fe pnictides: A class of high T_c superconductors



Interplanar distance may differ very much!



H. Ogino et al., arXiv:0903.3314

Fe pnictides: A class of high T_c superconductors



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> M. Rotter, M. Tegel, and D. Johrendt PRL 2008





FeSe







Cuprates

Heavy fermions





Mathur, Julian, Lonzarich et al. 1998





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Some of the people @ IFW involved in the research on FeAs ...

Synthesis:	G. Behr, J. Werner, S. Singh, C. Nacke, S.Wurmehl et al		
M, Thermodynamics:	N. Leps, L. Wang, R. Klingeler et al.		
NMR, ESR:	H. Grafe, G. Lang. F. Hammerath, V. Kataev et al.		
Transport :	A. Kondrat, G. Friemel, C. Hess et al.		
X-ray diffraction:	J. Hamann-Borrero		
Spectroscopy:	S. Borysenko, D.V. Evtushinsky, A. Koitzsch, J. Geck A. Kordyuk, V.B. Zabolotnyy, T. Kroll, M. Knupfer et al.		

Cooperations

μSR, Mößbauer	H.H.Klauss, H. Maeter et al.	TU Dresden
μ S R	H. Luetkens, R. Khasanov et al.	PSI Villingen
Crystals	C.T. Lin, D. Inosov, V. Hinkov, B. Keimer et al.	MPI FKF Stuttgart
NMR	N. Curro	UC Davis
X-ray/neutron diffraction M. Braden et al.		U Köln
X-ray/neutron diffraction	HZ Berlin	

DFG-Research Unit FOR538

Doping Dependence of Phase Transitions and Ordering Phenomena in Copper-Oxygen Superconductors

Resistivity and Susceptibility of La(O_{1-x}F_x)FeAs



Preparation & Characterization



Lattice constants...



Preparation & Characterization



Resistivity of La(O_{1-x}F_x)FeAs



Two phase transitions in LaOFeAs



H.-H. Klauss et al., PRL 101, 077005 (2008)

LaOFeAs: Competing OP above T_s?





LaOFeAs

Thermal Expansion

• large fluctuation region at T>T_S

Magnetisation:

 linear T-dependence at high T, only weak changes due to fluctuations

Resistivity

 Peak at T_S enhanced scattering in the fluctuation regime

Strong link between electronic and structural degrees of freedom

Two phase transitions in (Pr,Sm)OFeAs





Structural Transition similar for all RE

• No transition in doped systems (x ~0.1)

REOFeAs: Structural changes



REOFeAs: Structural changes



Phase transitions in undoped 122 compounds







SDW magnetism in SrFe₂As₂



Krellner et al., PRL `08, arXiv:0806.1043

Resistivity of Ba_{1-x}K_xFe₂As₂



M. Rotter et al., Ang. Chem. 2008

Magnetic order of LaOFeAs



⁵⁷Fe Mössbauer spectroscopy

- → well defined magnetic hyperfine field
- → commensurate magnetic order below 138 K

B_{hyp} (T→ 0) ~ 4.86 T → ordered moment ~ 0.3 μ_B → itinerant magnet

H.-H. Klauss et al., PRL 101, 077005 (2008)

Magnetic order of LaOFeAs



C. de la Cruz, et al., Nature 453, 899 (2008)



S. Raghu et al., PRB 77 220503 (2008)

Magnetic order parameter in undoped LaOFeAs



H.-H. Klauss et al., PRL 101, 077005 (2008)










Resistivity of ReOFeAs



Resistivity

- Peak at T_s enhanced critical scattering
- Inflection point at T_N reduced scattering + gap opening at T<T_N
- Rare earth influence: induced in-gap states ?

$$ho \propto \frac{1}{n}, \frac{1}{\lambda}$$

C.Hess et al., arXiv:0811.1601 Kimber et al., PRB 08

ROFeAs (R = La, Pr, Ce, Sm)



- Zero Field Muon Spin Rotation
 - Static magnetic order below T_N
 - 100% magnetic volume fraction
 - Commensurate magnetic structure
- T-dependence of μ SR frequency
 - Second order transition at $T_N(Fe)$
 - $\Box T_{\rm N}(R)$
 - Why is CeOFeAs different ?
- Moessbauer spectroscopy
 - All compounds possess the same ordered Fe moment (~0.35 μ_B) !
- Neutron scattering: $0.25 0.8 \mu_B$?

Ce magnetization above T_N^{Ce}

S. Chi et al., arXiv:0807.4986 (2008).

CeOFeAs Ce magntic moment m_{z}^{Ce} (μ_{B}) CeOFeAs μSR data $f_{\mu}(T)$ 10 $f_{\alpha}(T)$ 80 60 100 120 140 20 40 0 Temperature (K) Quantum CEF (Fe-Ce) Brillouin (Fe-Ce + Ce-Ce) 0.0 20 60 80 100 120 40 140 0 Temperature (K) $f(T) = f_0 \left[1 - \left(\frac{T}{T_N}\right)^{\alpha} \right]^{\gamma} \cdot \left[1 + \frac{\tilde{C}}{T - \Theta} \right]$ Fe sublattice Curie-like Ce magnetization magnetization

H. Maeter, et al., arXiv:0904.1563 (2009).

- Magnetization in molecular field of the Fe sublattice
 - Contributes to the same magnetic Bragg peak as the Fe order
 - Creates a field at the muon site which is proportional to the Ce magnetization
 - This can be modeled by:
 - a Curie-like term to the μ SR frequency
 - a calculation of thermal population of crystal electric field (CEF) levels



Interplay of Rare Earth and FeAs electronic systems



L. Pourovskii et al., EPL 84 37006 (2008)

- Strong hybridization between Fe and Ce states
- CeOFeP is a moderate heavy fermion system

Pr states shifted 2 eV downwards



E. M. Brüning et al., Phys. Rev. Lett. 101, 117206 (2008)

Resistivity of La(O_{1-x}F_x)FeAs



Magnetism of lightly doped La(O_{1-x}F_x)FeAs



H. Luetkens et al., Nature Materials 2009

Magnetism of lightly doped La(O_{1-x}F_x)FeAs













Phase diagram of LaFeAsO_{1-x}F_x



Superconductivity of La(O_{1-x}F_x)FeAs



Phase Diagram of La(O_{1-x}F_x)FeAs





Phase Diagrams of CeFeAsO_{1-x}F_x and SmFeAsO_{1-x}F_x



Drew et al., Nature Materials 2009

- 100 % rare earth doping (5% lattice constant change!) does not change the structural distortion, but a few percent Flourine (electron doping) completely suppresses T_s !
- gradual decrease of FeAs Neel temperature to zero ?
- coexistence of magnetism and superconductivity? Yes

CeO_{1-x}Fe_xAs





Zero field µSR: CeO_{1-x}Fe_xAs

Determine $T_N(Fe)$ from magnetic volume:

- e.g.50% of signal shows static relaxation
- Strong reduction of T_N for x>0!

Appearance of **µSR frequency**

- → Long range coherent sublattice magnetization
- Strong reduction of sublattice magnetization for x>0
- No long range order for x=0.15
- but short range magnetic order present with f_µ(T)=0!
 → Mean sublattice magnetization = 0
 → not visible in neutron scattering



Frequency shift

- Additional diamagnetic frequency shift
- Bulk superconductivity \rightarrow T_C
- Bulk static magnetism for 15%!



Static line width

- Bulk superconductivity \rightarrow T_C
- Bulk static magnetism for 15%!

CeO_{1-x}F_xFeAs: coexistence of magnetism and SC?



Zhao et al., Nature Materials (2008), neutrons

Evidence for coexistence! XRD, ρ , μ SR,...

Phase Diagrams of doped BaFe₂As₂

BaFe_{2-x}Co_xAs₂





Rotter et al., Ang. Chem. 2008

Pressure induced Superconductivity in (Ba,Sr)Fe₂As₂



Colombier et al., arxiv.org/abs/0904.4488v1

Resistivity of La(O_{1-x}F_x)FeAs



Phase Diagram of Ba_{1-x}K_xFe₂As₂



Superconductivity and Crystal Structures of $(Ba_{1-x}K_x)Fe_2As_2$ (x = 0 - 1)

Marianne Rotter, Michael Pangerl, Marcus Tegel and Dirk Johrendt*

Ang. Chem. 2008

Resistivity of doped pnictides



Rotter et al., Ang. Chem. 2008



Resistivity of Co doped pnictides



Resistivity at the stuctural and SDW transistions





 $\rho \propto \frac{1}{n}, \frac{1}{\lambda}$

enhanced scattering at higher T

 Inflection point at T_N reduced scattering



Resistivity in Co doped systems

- Reduction of scattering at T_N smaller
- Mean free path limited by disorder due to Co atoms!!

Intrinsic sources of scattering masked by disorder in Co doped single crystals!!!



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Superconducting Order Parameter (Gap)

The fundamental parameter of a superconductor is the gap $\Delta({\bf k})$



How can we measure the superfluid density?

```
via the magnetic penetration depth \lambda n<sub>s</sub>/m<sup>*</sup> is proportional to 1/\lambda^2
```

• measure λ in vortex state of type II superconductor via field profile p(B) \rightarrow transverse field μ SR



In anisotropic powder superconductors P(B) shows Gaussian shape $\langle \Delta B^2 \rangle \propto \lambda_{ab}^{-4}$

Superconductivity in La(O_{1-x}F_x)FeAs

How can we measure the superfluid density?

via the magnetic penetration depth λ n_s/ m^* is proportional to 1/ λ^2

• measure λ in vortex state of type II superconductor via field profile p(B) \rightarrow transverse field μ SR



Penetration Depth of La(O_{1-x}F_x)FeAs



H. Luetkens et al., PRL 101, 097009 (2008)

Penetration Depth of La(O_{1-x}F_x)FeAs



T dependence of λ : consistent with BCS s-wave

B dependence of λ : not simple s-wave: d, ext. s, multiband

H. Luetkens et al., PRL 101, 097009 (2008)
Superconducting Properties of Co doped Ba122



Superconductivity of Co doped Ba122– TF-µSR



- Superfluid density (full line shape analysis) for field applied in different direction.
- Pronounced anisotropy of the penetration depth λ.
- Assuming $\lambda_a = \lambda_b$ one can calculate the low temperature values:
- λ_a(T=0) = 220 nm
- λ_c(T=0) = 1100 nm

H. Luetkens, C. Nacke et al., in preparation

Superconductivity of Co doped Ba122– TF-µSR



Superconductivity in Ba_{1-x}K_xFe₂As₂



Gap on the inner **F**-barrel, **F**M



D.V. Evtushinsky et al., PRB 2009

Anisotropy of Gap according to ARPES



D.V. Evtushinsky et al., PRB 2009 Similar results see eg. Ding et al EPL 2008

Penetration Depth of $Ba_{1-x}K_xFe_2As_2$, $T_c = 32 K$



Penetration Depth and Uemura Plot



The Fe-based superconductors are close to the Uemura line for hole doped cuprates \rightarrow hope for higher T_c in the future

Gap and Spin Lattice Relaxation Rate (NMR)





H.-J. Grafe et al., PRL 101, 047003 (2008)













Scenarios:

Transition from $s_{+/-}$ to conventional $s_{+/+}$ state due to impurities

As deficiencies cause mainly intraband scattering and further protect the unconventional $s_{+/-}$ state (higher T_c , higher H_{c2}) ("smart impurities")

F. Hammerath, H.-J. Grafe, ... S.L. Drechsler, I. Eremin et al.



Scenarios:

Temperature (K)

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SC transition in GdFeAsO_{0.85}F_{0.15}: Strange and Puzzling behavior



SC transition in GdFeAsO_{0.85}F_{0.15}



SC transition in GdFeAsO_{0.85}F_{0.15}



SC transition visible $\Delta c_{p, SC} = 2.0 \text{ J / (mol K)}$

BCS theory: $\gamma = \Delta c/(1.43 T_c)$ $\approx 70 \text{ mJ/(molK)}$

$$\frac{dT_c}{dp} = -TV \frac{\Delta \alpha}{\Delta c_p}$$
$$\approx 1.4 \frac{K}{GPa}$$

SC transition in GdFeAsO_{0.85}F_{0.15}



GdFeAsO_{0.85}F_{0.15}: Specific heat



GdO_{1-x}F_xFeAs - Gd HF-ESR: Width of the signal

Gd³⁺: 4f⁷, L = 0; J = **S =7/2** no coupling to lattice, **probes spin degrees of freedom**



Electron Spin Resonance of GdO_{1-x}F_xFeAs



Superconductor ($T_c = 21$ K): strong broadening and shift of the Gd-ESR response \Rightarrow onset of inhomogeneous quasi-static spin correlations at T < 80 K

SC transition in GdFeAsO_{0.85}F_{0.15}



GdFeAsO_{0.85}F_{0.15}: Magnetostriction



GdFeAsO_{0.85}F_{0.15}



SC transition in GdFeAsO_{0.85}**F**_{0.15}**:** Strange and Puzzling behavior



Spin lattice relaxation rate, T₁⁻¹, normal state





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