Short-range magnetic order in orbitally selective Ni-substituted $FeSe_{0.35}Te_{0.65}$ single crystals

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Abstract

Multi-orbital character of iron-based superconductors (IBS) results in various electronic phases evident in phase diagrams, including nematically or magnetically ordered phases. The role of magnetic or nematic fluctuations in superconducting (SC) pairing in IBS is a subject of intense discussions.

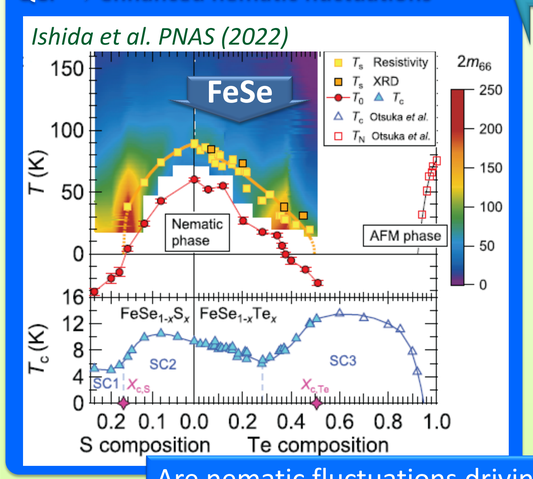
Here we study properties of $FeSe_{0.35}Te_{0.65}$ single crystals doped by Ni into Fe site. The crystals of this composition are located close to maximum T_c in heavily Te-doped SC dome. Transport properties combined with ARPES indicate that substitution of Ni dopes crystals with electrons, what eliminates some of the hole pockets from Fermi level, leaving only one, originating from the d_{xy} orbital. Anomalies in magnetization are observed, clearly linked to the localization of d_{xy} orbital on increasing temperature. Analysis of these anomalies suggests the presence of short-range magnetic orderings, both antiferromagnetic, and ferromagnetic.

This implies an importance of magnetic fluctuations in the studied system, confirming a possible link to pairing in heavily doped SC dome.

Motivation

Boudaries of nematic phase at x_c (elastoresistivity measurements) → QCP → enhanced nematic fluctuations

Ishida et al. PNAS (2022)

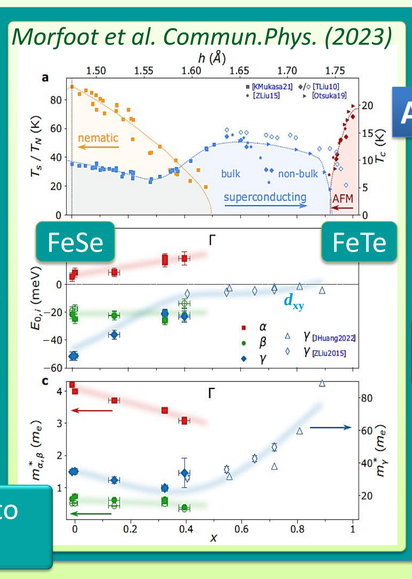


Are nematic fluctuations driving force for superconductivity?

Recent studies of FeSe isoelectronically substituted either by S or Te into Se-site have revealed one (in case of S) or two (in case of Te) SC domes, overlapping in some regions with nematic phase, suggesting a possible link between SC pairing and nematic fluctuations.

Another proposal in case of heavy Te-doping suggests an important role of the d_{xy} orbital, which approaches Fermi level with increasing Te content, possibly providing additional pairing channel via magnetic fluctuations.

Morfoot et al. Commun.Phys. (2023)

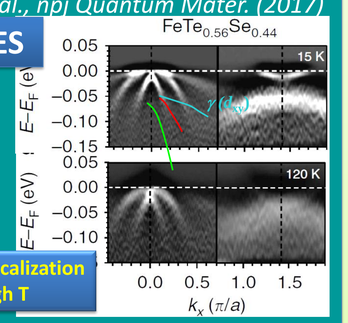


Additional pairing due to spin fluctuations ?

Orbital-selective Mott phase

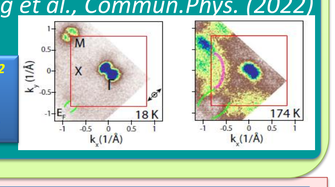
Yi et al., npj Quantum Mater. (2017)

ARPES



d_{xy} localization at high T

Huang et al., Commun.Phys. (2022)

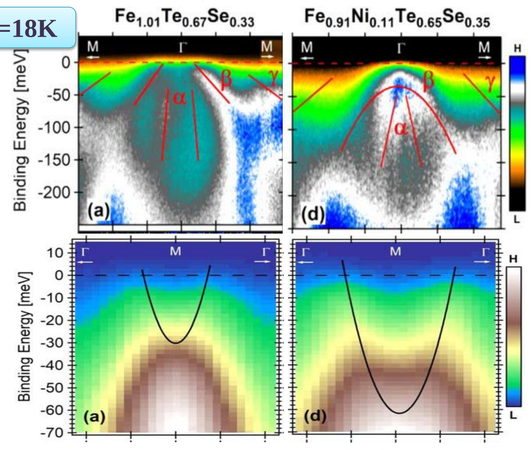


⇔ d_{z^2} at X point

ARPES

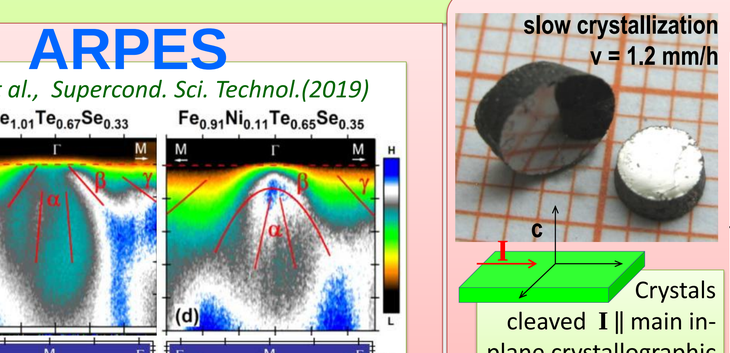
M. Rosmus et al., Supercond. Sci. Technol.(2019)

T=18K

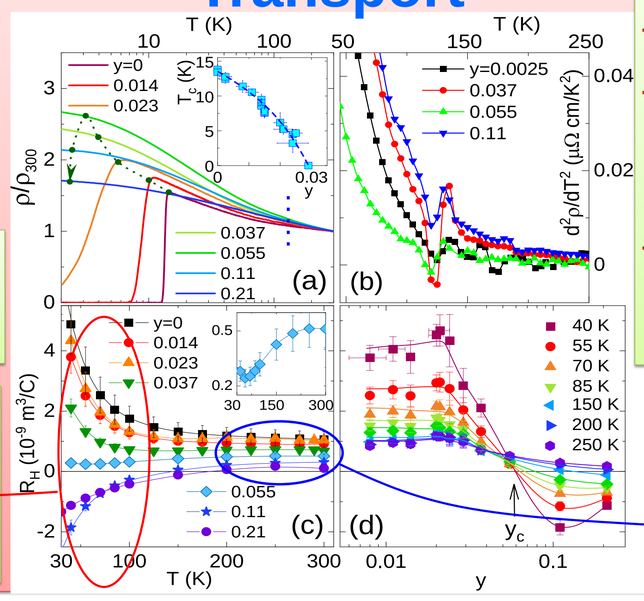


- Electron doping removes **some** hole pocket from Fermi surface – only d_{xy} remains
- electron pockets expand and become elliptical

Transport



Crystals cleaved **I** || main in-plane crystallographic axis



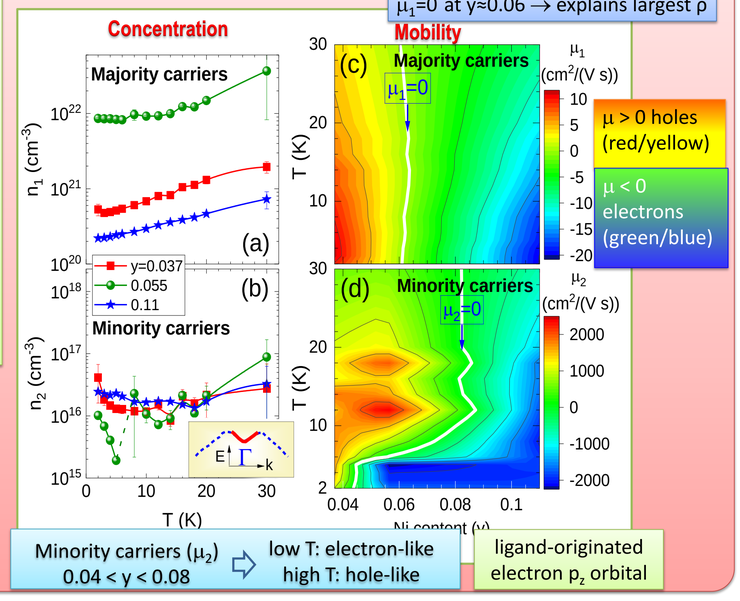
Hall effect
Low T:
- electron doping: R_H changes sign at $y_c \approx 0.06$
- 2-types of carriers

Hall effect
High T:
- R_H remains positive for all y

$Fe_{1-y}Ni_ySe_{0.35}Te_{0.65}$

- SC suppression at y ≈ 0.03
- non-monotonic evolution of ρ at low T: maximum ρ for $y_c \approx 0.055$
- change of slope of ρ at T≈125-130K (visible best as anomaly in $d^2\rho/dT_2$)

Majority carriers
μ_1=0 at y≈0.06 → explains largest ρ



μ > 0 holes (red/yellow)

μ < 0 electrons (green/blue)

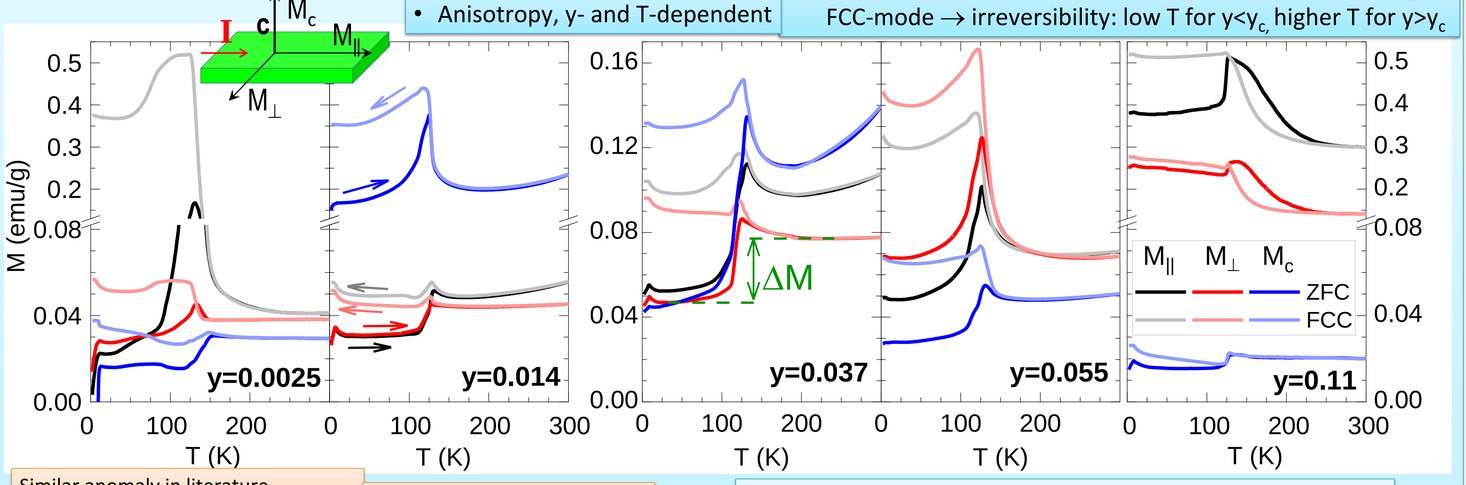
Minority carriers (μ_2) 0.04 < y < 0.08 ⇨ low T: electron-like, high T: hole-like

ligand-originated electron p_z orbital

Magnetization

- All components finite
- Anisotropy, y- and T-dependent

- y=0.0025: SC transition
- **Anomaly at 125-130 K – close to the anomaly in $d^2\rho/dT_2$**
  FCC-mode → irreversibility: low T for $y<y_c$, higher T for $y>y_c$

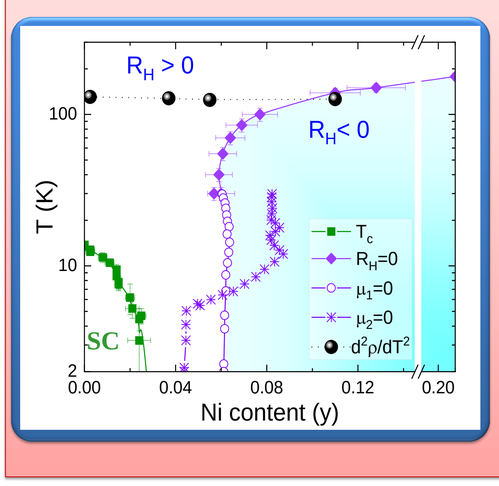


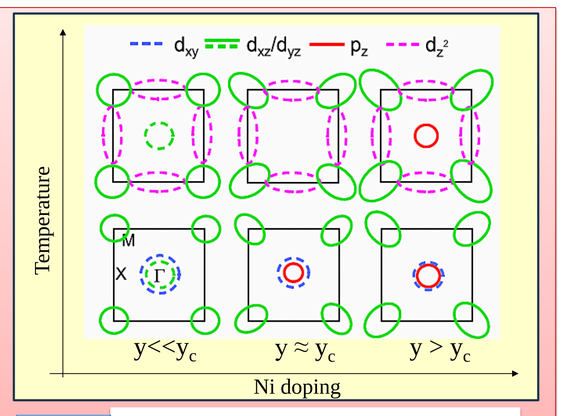
Similar anomaly in literature
Fang et al., PRB (2008) $Fe(Se_{1-x}Te_x)_{0.82}$
Rößler et al, PRB (2010) $Fe_{1+y}Te_{0.5}Se_{0.5}$
Wittlin et al., SST (2012) $FeSe_{0.35}Te_{0.65}$

Spin reorientation in hexagonal $Fe_7(Se,Te)_8$ phase or Verwey transition in Fe_3O_4

Present experiment: $Fe_7(Se,Te)_8$ – no trace; Fe_3O_4 < 0.7%; Fe excess < 0.03 → **intrinsic property**

Phase diagram

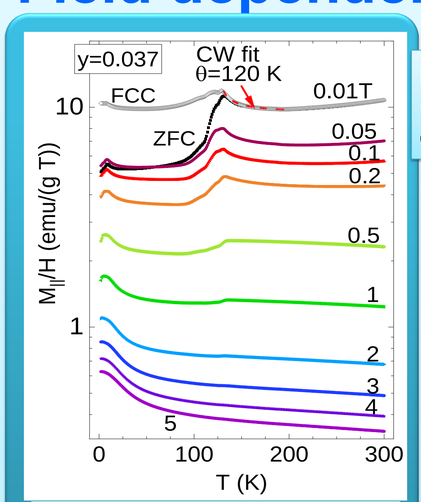




low T: Electron doping: only one d_{xy} hole pocket survives + small electron pocket of p_z origin

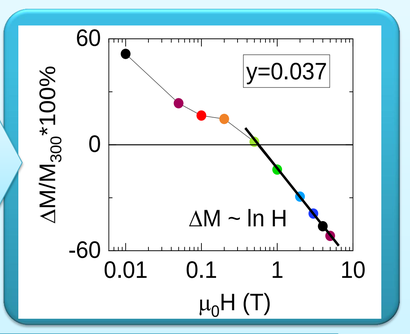
high T: OSMP: d_{xy} pocket at Γ disappears, and d_{z^2} hole pockets at X point appear → hole-dominated transport

Field dependence



Magnetic field suppresses peak, and reverses sign of ΔM

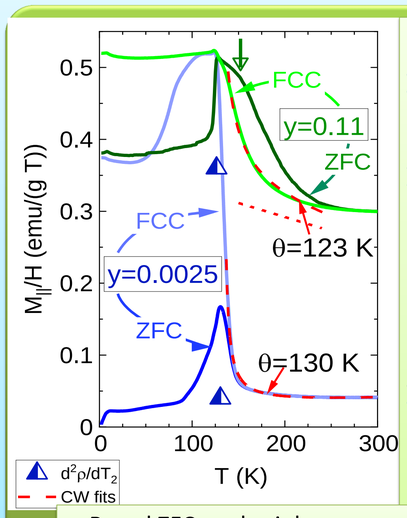
Negative ΔM ~ ln H →localization of carriers



- Both large magnetic field and large doping **reverse the sign of ΔM** from positive into negative (**localization**)
- Magnetic field suppresses the peak, but the doping does not → **ΔM and the peak are of different origin**

Peak: short-range magnetic ordering
→ magnetic field orients magnetic moments, Ni doping does not

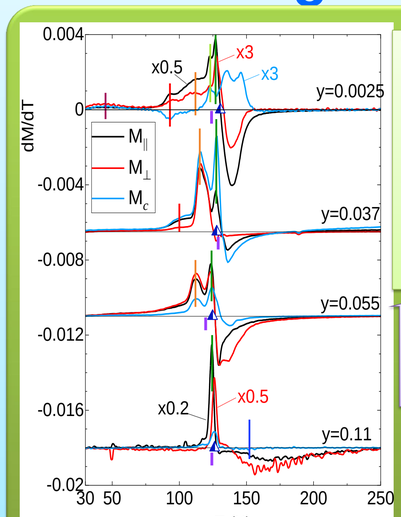
Inhomogeneous ordering



FCC vs ZFC

- CW fits to FCC: FM component
- Max FCC close to resistive anomaly, similar in both samples (FM y-independent)
- difference on decreasing T: related to localization

- Broad ZFC peaks: inhomogeneous ordering
- Broadening on low-T side (y=0.0025) or high-T side (y=0.11)

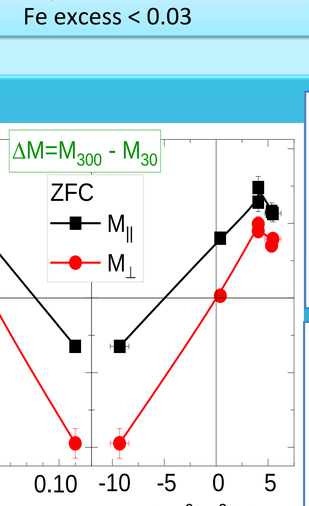


- y < y_c : precursors to the main peak
- y > y_c : no precursors, shoulder at higher T

AFM component y-dependent

AFM and FM clusters

close to the main peak FM component y-independent



Anisotropy ZFC, in-plane

◆ M_{30} small for $y<y_c$, large for $y>y_c$ → localization

★ M_{300} $y<y_c$ $M_{300} > M_{30}$; $y>y_c$ $M_{300} < M_{30}$ → transport and magnetism are linked

$y<y_c$ positive ΔM **delocalization of d_{xy}** holes on decreasing T → increase of density of itinerant carriers, decrease of M

$y>y_c$ negative ΔM **large Ni scattering**: on decreasing T carriers become incoherent → decrease of density of itinerant carriers, increase of M

Acknowledgements
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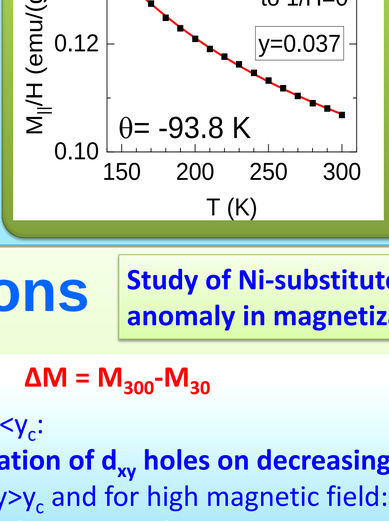
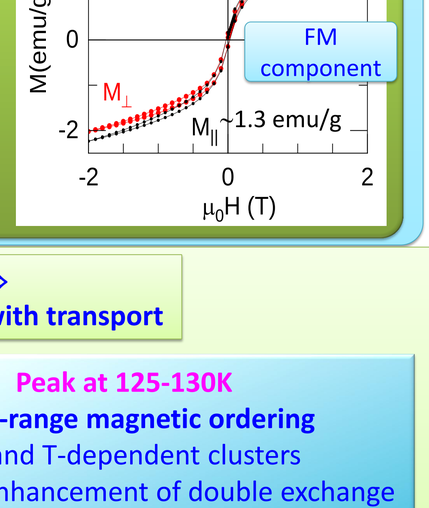


Curie-Weiss fits
$\chi = \chi_0 + C/(T-\theta)$
150-300 K, FCC

Low H: θ=120K
High H: θ= – 93.8K
AFM & FM



Conclusions

Study of Ni-substituted $FeSe_{0.35}Te_{0.65}$ ⇒ anomaly in magnetization correlated with transport

$\Delta M = M_{300}-M_{30}$

- positive for $y<y_c$: **delocalization of d_{xy} holes on decreasing T**
- negative for $y>y_c$ and for high magnetic field: **localization on decreasing T**

Peak at 125-130K
short-range magnetic ordering

- AFM : y-and T-dependent clusters
- FM – ? enhancement of double exchange (d_{xy} delocalization) or topological transition

More info: Marta Z. Cieplak et al., Phys. Rev. B 109, 144518 (2024)