IEEE/CSC & ESAS SUPERCONDUCTIVITY NEWS FORUM (global edition), October 2017.
Keynote presentation A3-K30-014 given at IUMRS-ICAM, 27 Aug. - 1 Sept. 2017, Kyoto, Japan.

IUMRS-ICAM 2017

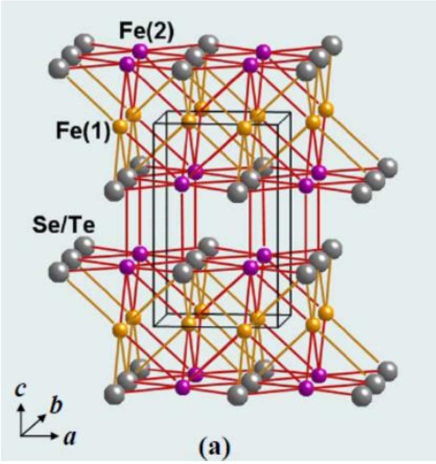
The 15th International Conference on Advanced Materials

Properties of $FeSe_{1-x}Te_x$ thin films

Carlo Ferdeghini
CNR-SPIN

Collaborators: E.Bellingeri, V.Braccini, A. Leveratto, A.Malagoli, C.Nappi, E.Sarnelli, G.Sylva
But also: M.Putti, R.Buzio, A.Gerbi, A.Martinelli, A.M.Massone, M.Piana….

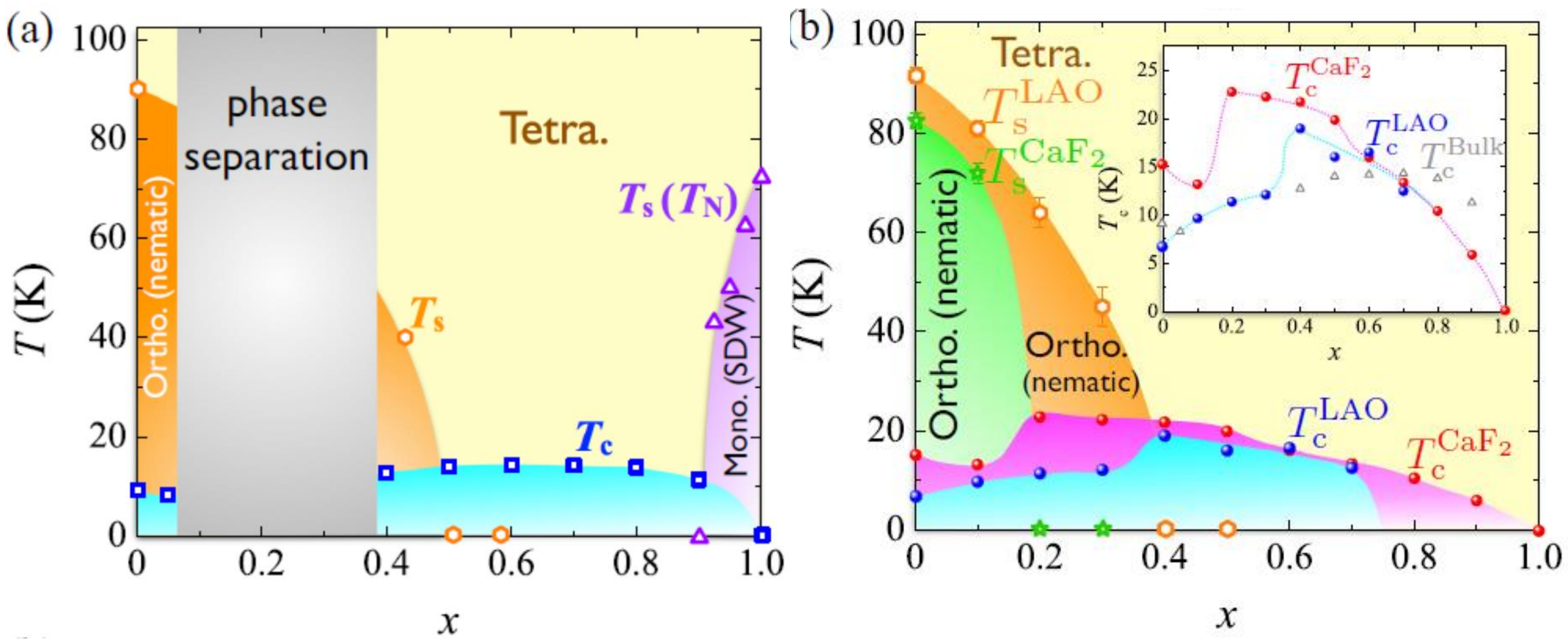
Why Fe(Se,Te) ?



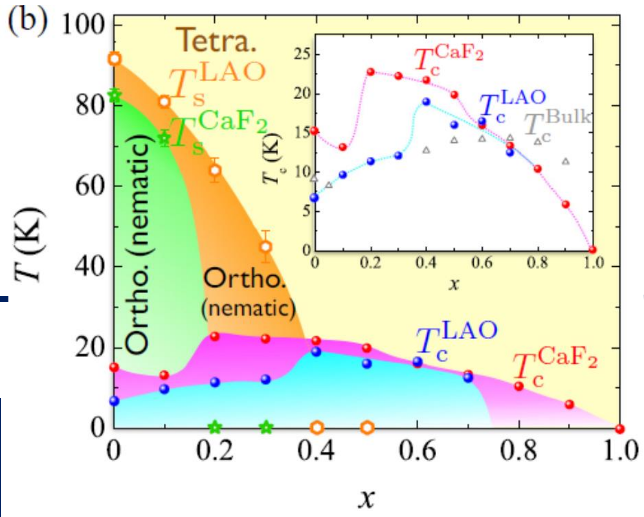
- **Simplest crystal structure**
- **Easy to grow *and to synthesize*** (Just 3 elements)
- ***Easy to handle stable in air*** *(not explosive)*
- **Do not contain As**
- T_c (~ 10-15 K) values, but increasable by straining the structure
- **Very high H_{C2}** values > 50 T
- **High J_c** (*~ 10^6 A/cm^2 in self-field) and* weakly depends on magnetic field.

Appealing from an applicative point of view

Simple system, complicated phase diagram



Y. Imai , F.Nabeshima, A.Maeda, Condens. Matter **2017**, 2, 25;



Fe Se

T_c up 100 K on monolayer!
Ge, J. F. et al. Nature Mater. 14, 285 (2015)

Electric-field-induced S/C in electrochemically etched ultrathin FeSe films on SrTiO3 and MgO (40K)
J. Shiogai et al., NATURE PHYSICS , 12 (2016)

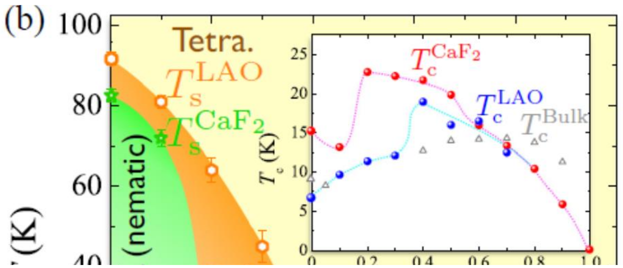
Fe Te

T_c up to 13K
Y. Han et.al. PRL 104, 017003 (2010)
Tensile stressed FeTe thin films (?)

T_c up to 8K
$Fe_{1.08}TeO_x$ thin films in controlled O_2 atmosphere, SC'ing at 8K.
W.Si et.al. Phy Rev B 81, 092506, 2010

$Fe\ Se_{1-x}Te_x$

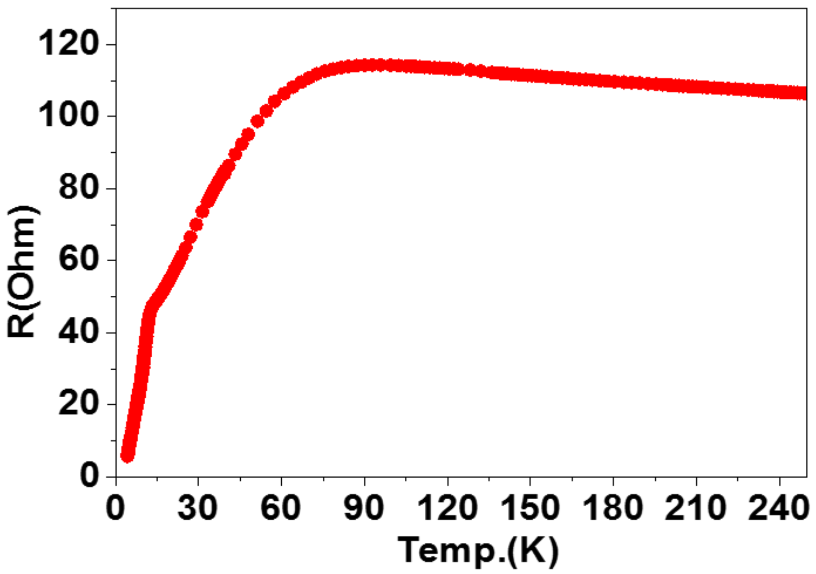
Wide tolerance in x values for SC: no phase separation.
T_c up to 25 K depending on the strain



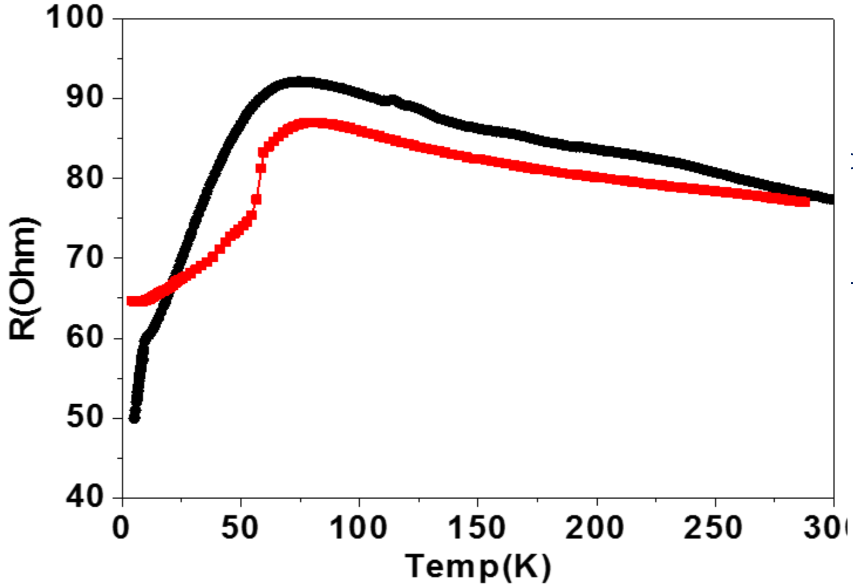
T_c u
Ge, J.

Elec
elec
FeS
J. Shic

Films deposited in O_2 atmosphere, ($P=2.4X10^{-5}$ mB)



Post deposition annealing: @150 ^0C in O_2 ($P=1X10^{-2}$mB)

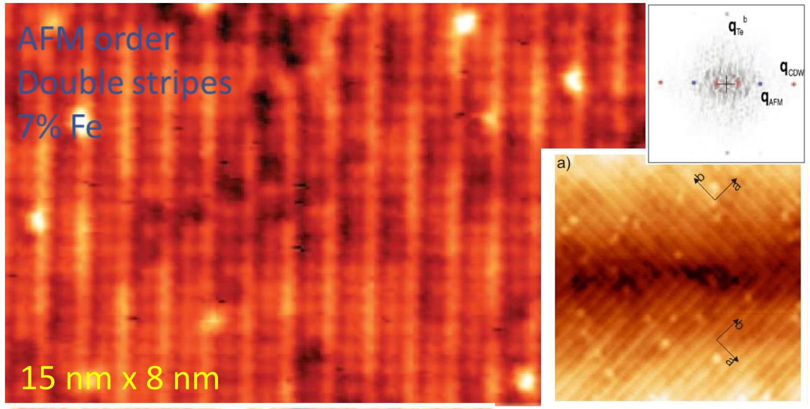


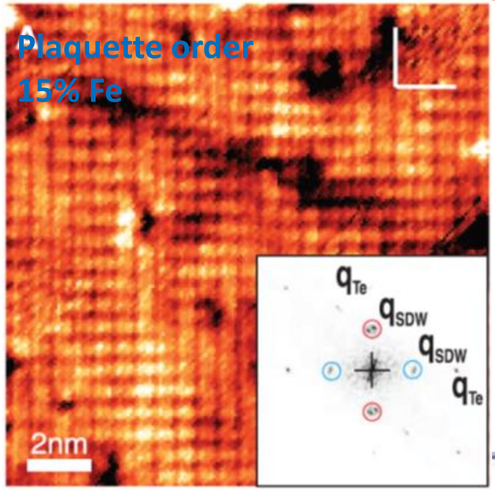
'ing at 8K.

$Fe_{1+y}Te$ single crystals: AFM ordering at the atomic scale (data from literature)

Fe Te

With STM tip Spin polarized **Fe(2)<12%**



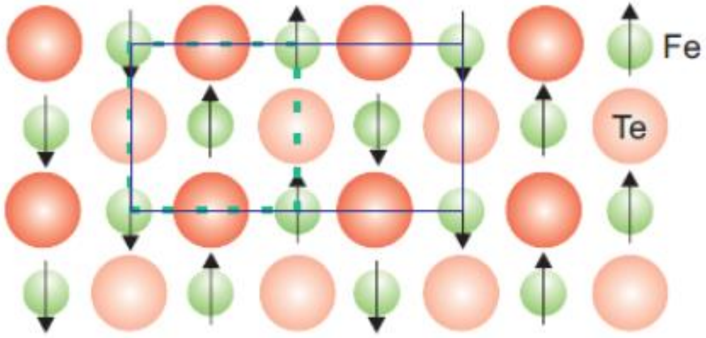


M.Enayat et al., Science 345, 653 (2014)

Only at grain boundaries between monoclinic domains the stirpes switch direction in region with low excess iron concentration (y<0.12)

A.Sugimoto et al., Phys. Rev. B 90 (2014)

T: Mizokawa et al,., J. Phys. Soc. Jpn. 81 (2012)



(π,0) order. Not superconductive

E-type symmetry

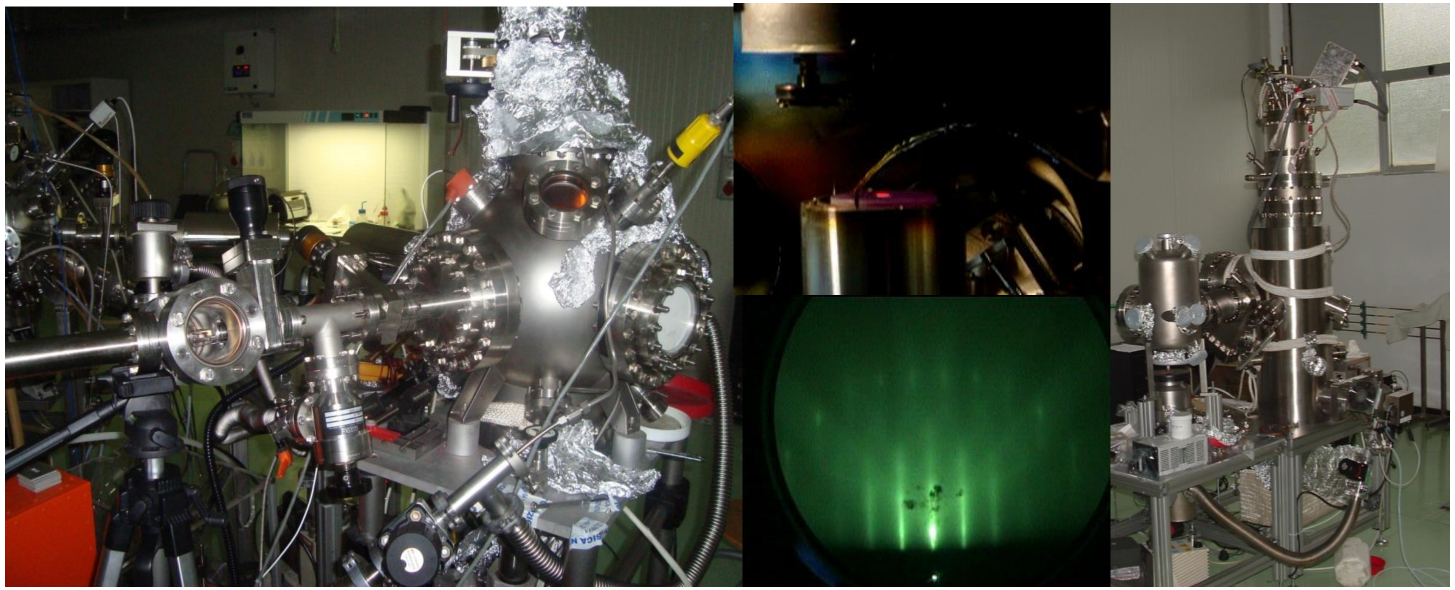
The parent compound FeTe exhibits a bicollinear AFM stripe order with a wave vector (π, 0) aligned along the Te square lattice

IUMRS-ICAM 2017 KYOTO

Pulsed laser deposition (PLD)

PLD system has been used for thin film deposition with following parameters-

- Ultra high vacuum (5 x 10^{-9} mbar) chamber.
- Deposition temperature ranged between 300 to 650°C.
- Repetition rates ranging from 3 to 10 pulse/s.
- Target-substrate distance was fixed at 5 cm.
- Laser beam KrF 248 nm fluency of 2 J/cm^2 and Nd-YAG @1064 nm.
- In-situ Reflection High Energy Electron Diffraction (RHEED) was been used to observe film growth.
- Facility to transfer sample to scanning tunnelling microscopy (STM) system under vacuum of 5 x10^{-8} mbar.



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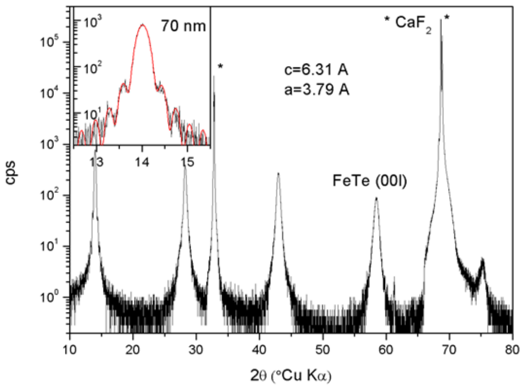
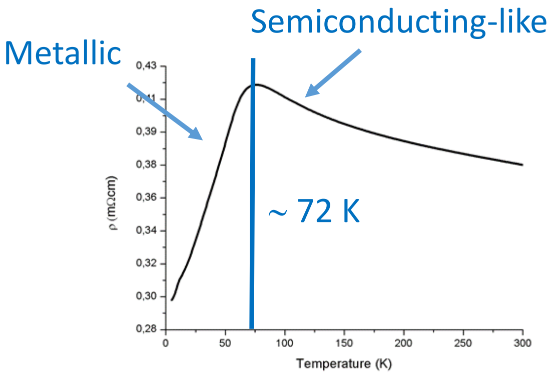
$Fe_{1+y}Te$ thin films: macroscopic characterization

Fe Te

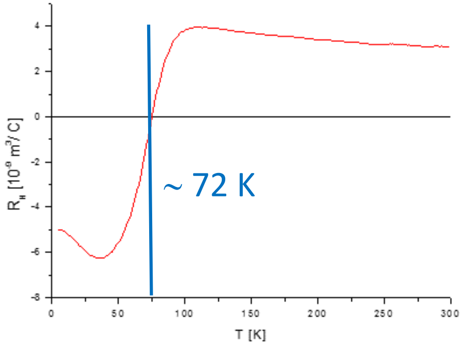
X-Ray Diffraction



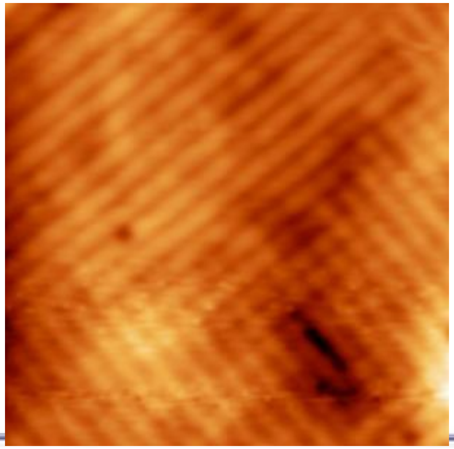
Resistivity vs Temperature



Hall Resistivity vs Temperature



a **=** ***b*** **= 3.79 Å**
c **= 6.31 Å**



$Fe_{1+y}Te$ thin film:
From STM:
Only at grain boundaries between monoclinic domains the stirpes switch direction in region with low excess iron concentration (y<0.12)

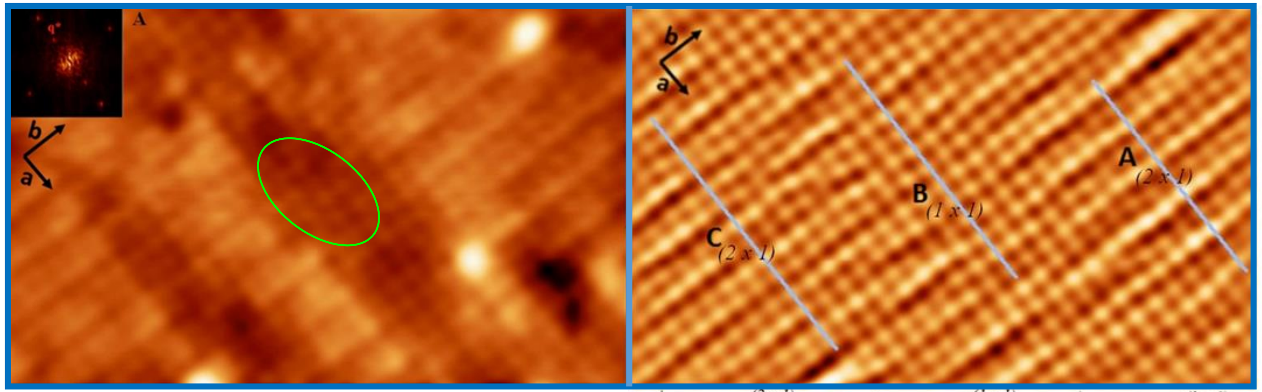
From Resistivity: y<11%

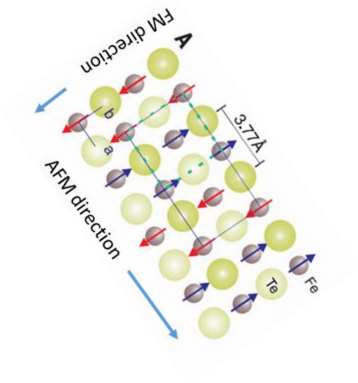
$Fe_{1+y}Te$ thin films LOW-BIAS

Appearance of (2 x 1) stripe structures (two extra spots in the FFT at q * = q (1 x 1) / 2 along the square diagonal direction.

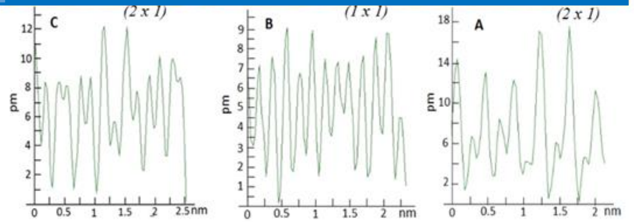
The AFM phase is stable only at a large z_{Te}, while reducing z_{Te} stabilizes the coupled stripe and the non-magnetic phases

Low-bias
< ± 50 mV



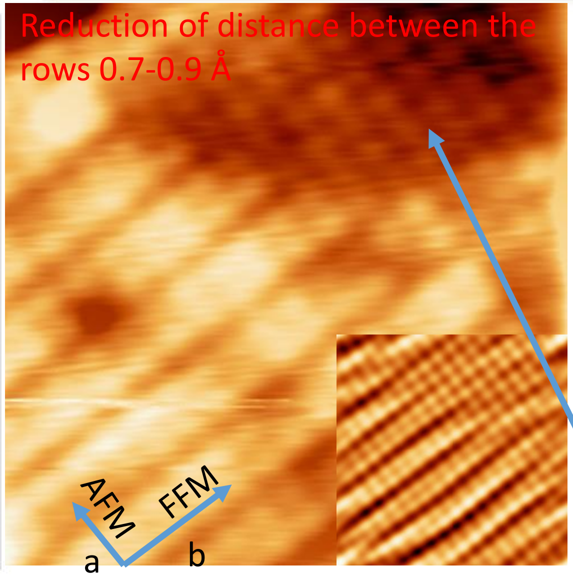


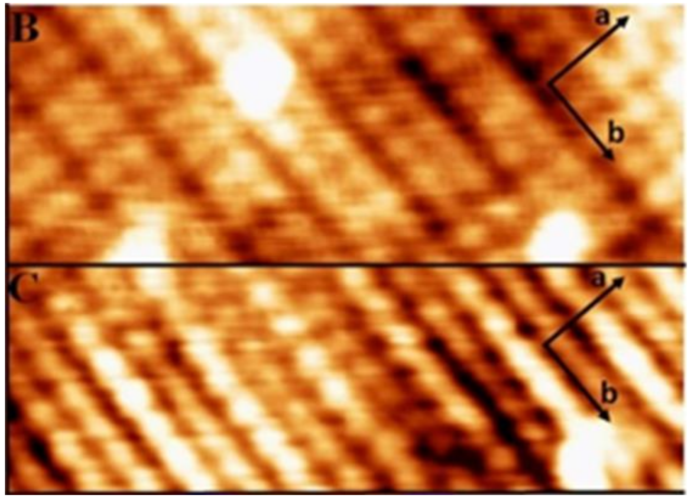
Region A Low Fe(2) concentration <6%
Region B High Fe(2) >11%-12%
Region C intermediate regime ~9%
helimagnet order compets with the bicollinear



The lattice distortion can be induced by the intercation between the excess Fe and the FeTe layer. The intercation is strong and extends a number of unit cells away from the interstitial Fe site. The strenght and the extend nature of the intercation suggests that this is the mechanism through which the excess Fe stabilizes the overall crystal structure

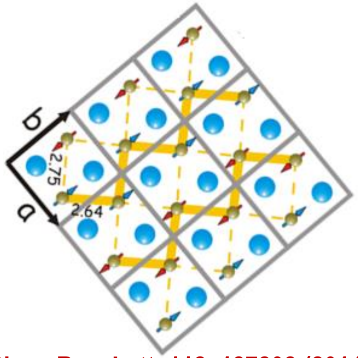
$Fe_{1+y}Te$ thin films LOW-BIAS



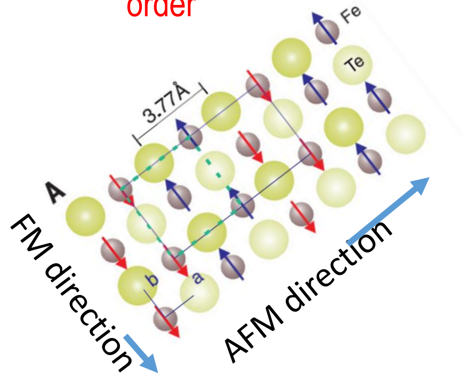


Intermediate excess Fe 9%-12%
helimagnetic order competes with the bicollinear commensurate magnetism

Low excess Fe
< 9%
AFM bicollinear commensurate order



Higher excess Iron
Incommensurate SDW
Or
Out of plane magnetism
Or
O + T phases
y> 11-12 %



Phys. Rev. Lett. 112, 187202 (2014

the local changing of hybridization leads to the formation of the ferromagnetic zigzag chains, which stabilize the bicollinear order with a reduced distance between the rows

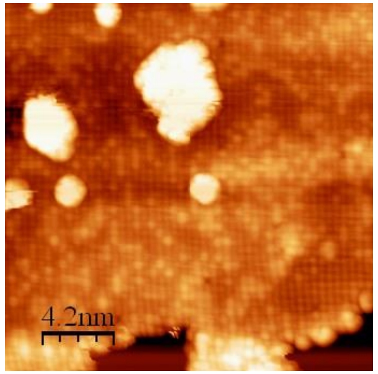
A. Gerbi *et al.* J. Phys.: Condens. Matter, submitted

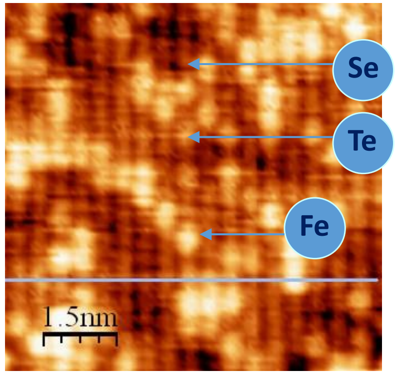
Local scale inhomogenities: a STM study

Fe $Se_{1-x}Te_x$

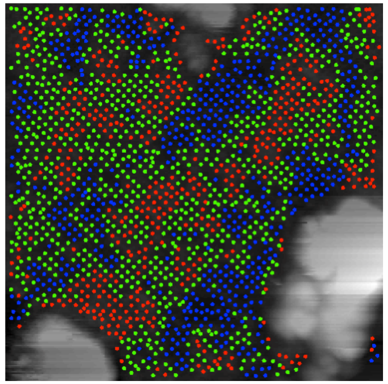
We exploited LT-STM (4.5K, UHV) to characterize the micrometric and atomic scale surface morphology of 150nm thick Fe(Se,Te) thin films with Tc=19K.

A. Gerbi *et al.* Supercond. Sci. Technol. **25**, 012001 (**2012**)

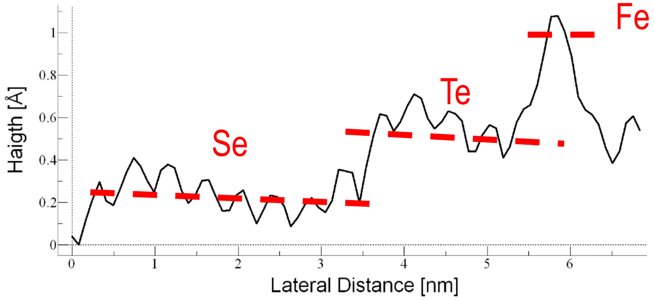




A. Perasso et al., *J. of Microscopy* (**2015**) 260

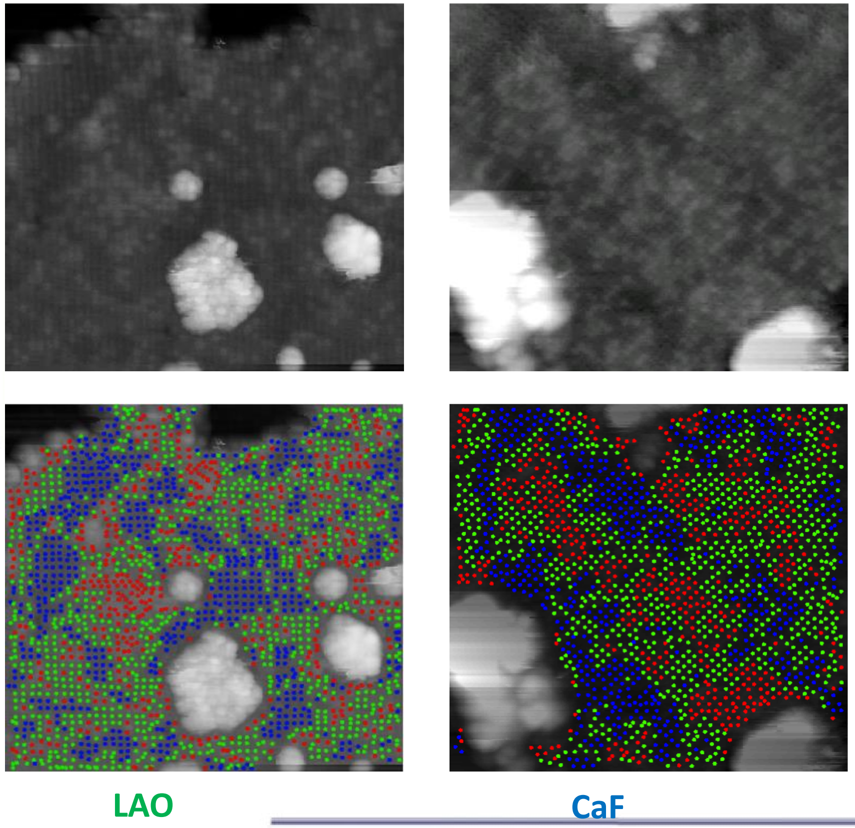


Se (Å)	Te (Å)	Fe (Å)
0 ± 0.11	0.28 ± 0.08	0.62 ± 0.14



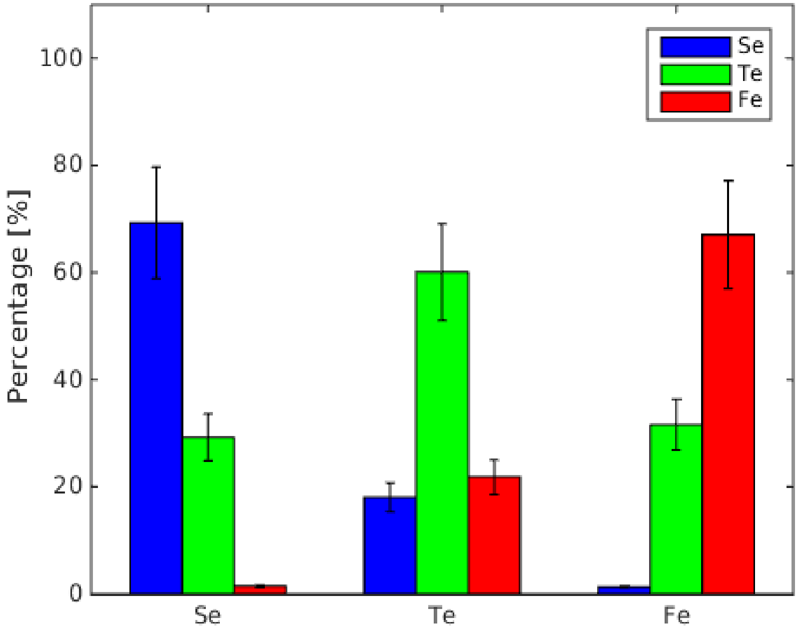
Surface stoichiometry for PLD thin films:
Se ~ 0.45
Te ~ 0.55
Fe ~ 0.20
Te diffuses from bulk towards surface (see also XPS and ARPES…)

Phase separation at nanometers length scale $Fe\ Se_{1-x}Te_x$

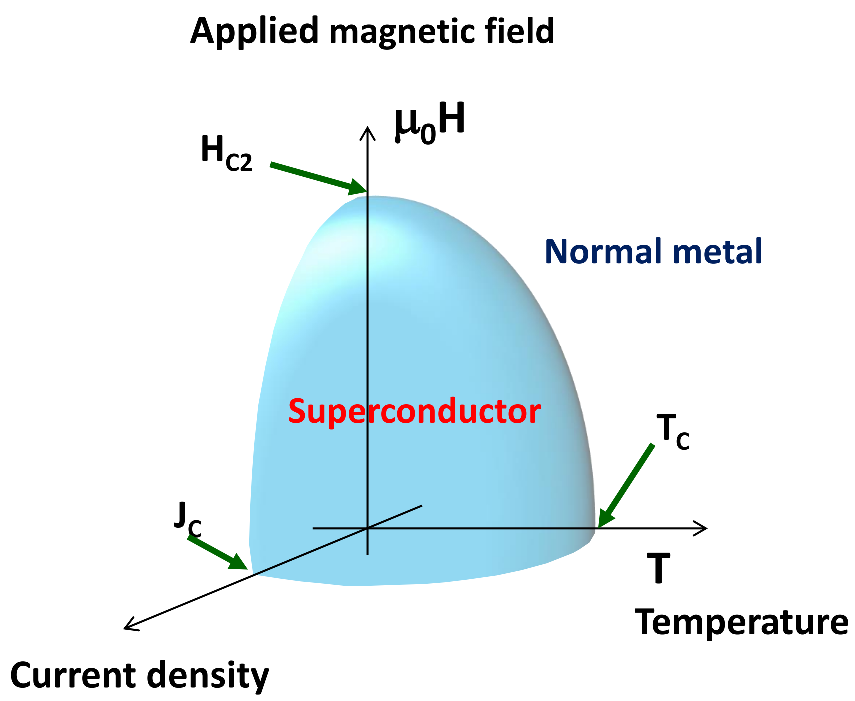


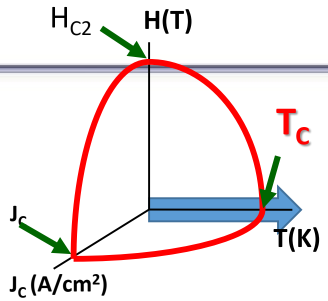
Atoms of the same species cluster together?

- histogram with three bins representing the probability of having a Se, Te or Fe neighbor, respectively. Under the hypothesis that atoms of the same species cluster, the histograms should not be uniform, but they should peak on the bin corresponding to the species itself.
- histograms indicates that our hypothesis is reasonable.



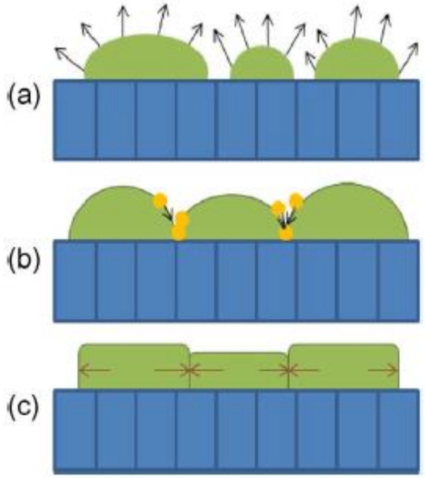
http://www.spin.cnr.it/index.php/software

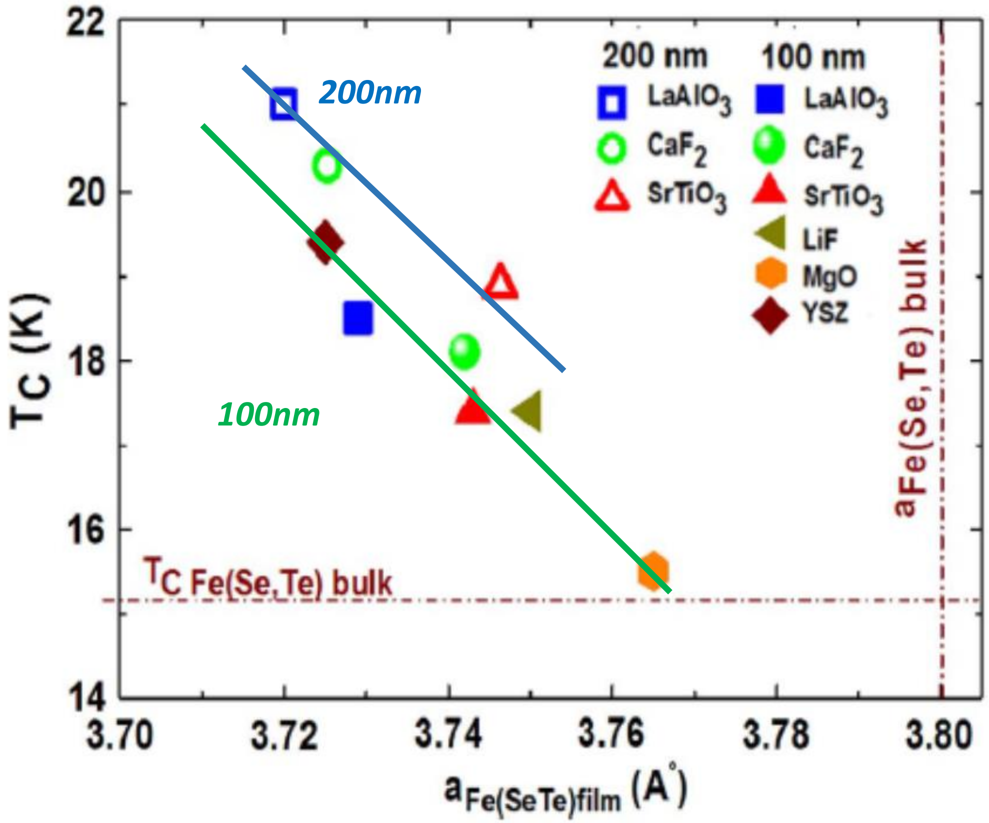

Applied magnetic field
μ_0H
H_{C2}
Normal metal
Superconductor
T_C
J_C
T
Temperature
Current density



T_C up to 23K reducing Se!

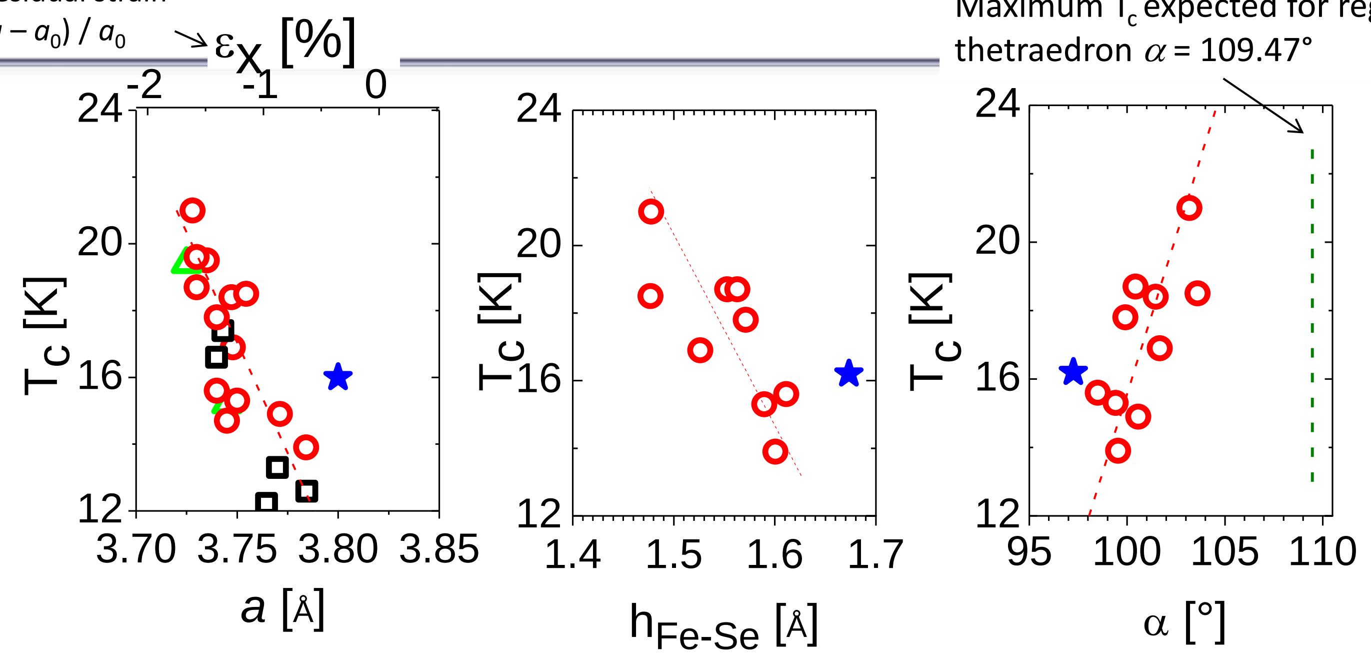
Y. Imai et al.PNAS 2015, 112



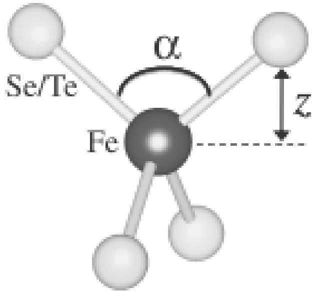


In bulk: up to **26K** by application of external pressure

S. Kavale et al. IEEE TRANS. APPL. SUPERCOND. 25, 2015



T_c **of several Fe(Se,Te) films as a function of i)the in-plane** ***a*** **cell parameter, ii)the Fe-(Se,Te) bond length and iii) the (Se,Te)-Fe-(Se,Te) bond angle.**
As an eyes guideline the linear fit of the data (dashed lines) is added in all the panels.

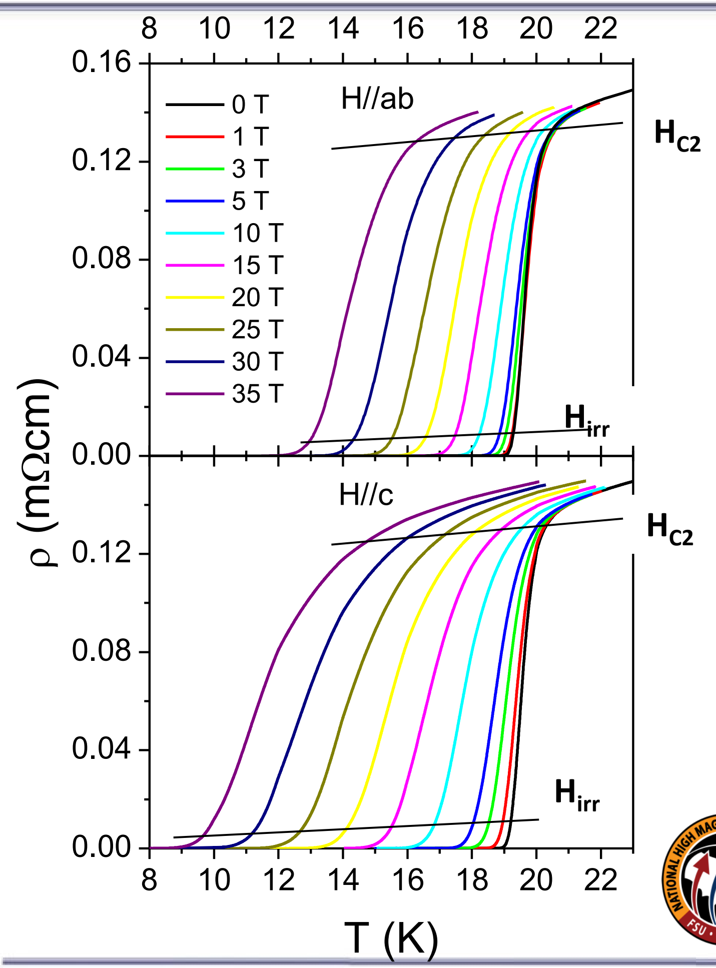


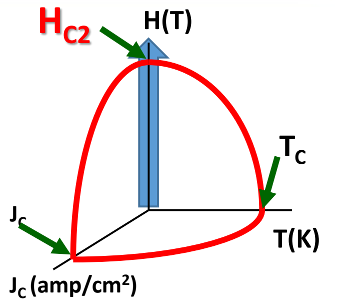
$LaAlO_3$ (O)
$SrTiO_3$ (□)
Y:ZrO (Δ)
the star represent the bulk value

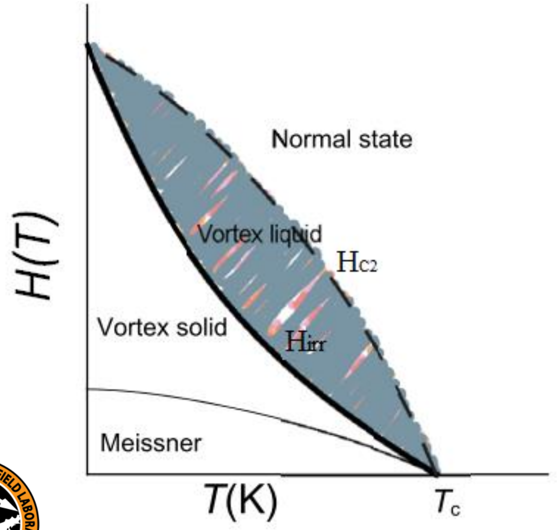
c axis is strongly reduced (5.85-5.90 Å), but NO T_C dependence was found

E.Bellingeri et al. Appl. Phys. Lett., 96, (2010)

Critical fields enhancement

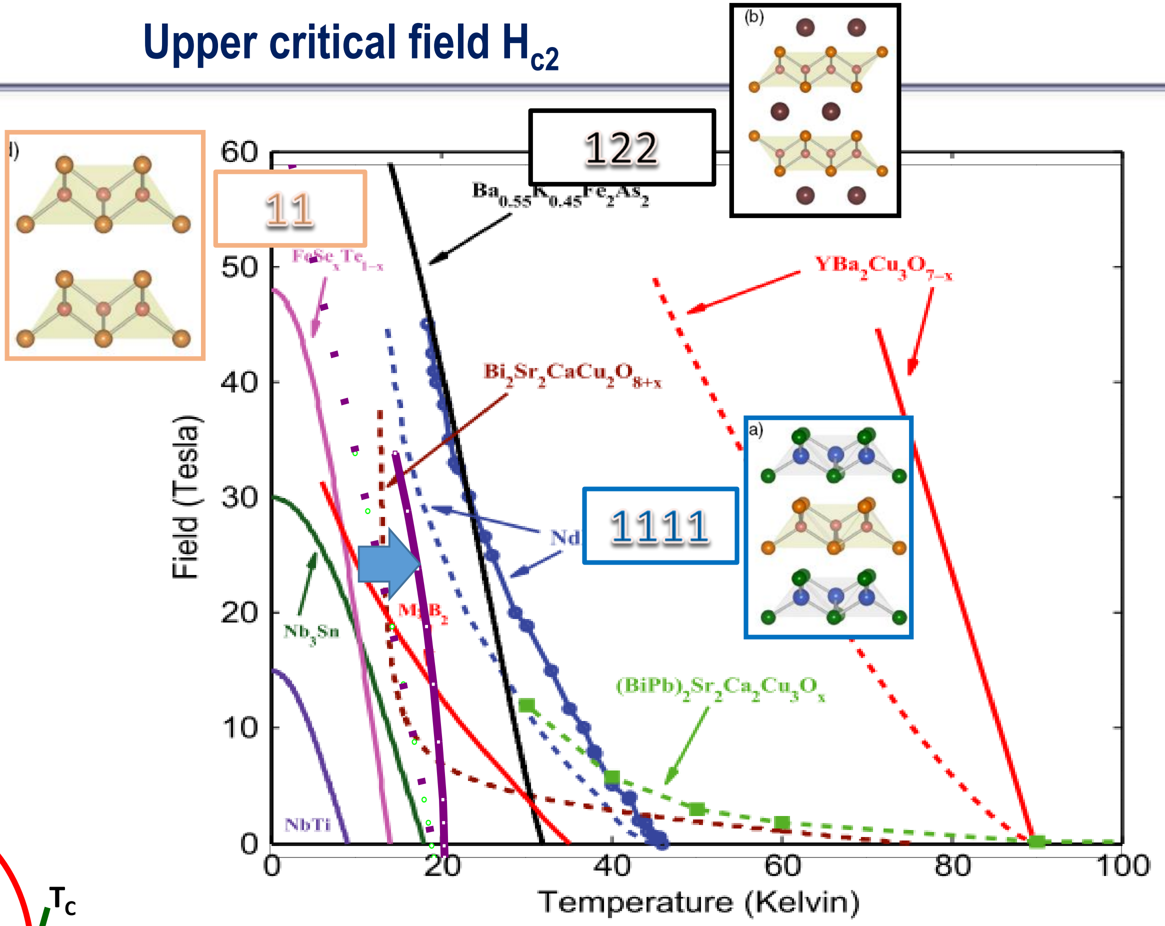


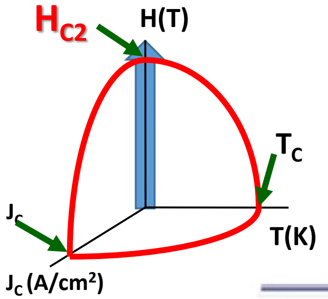




Up to 35T in DC field
Up to 80 T in Pulsed Field

Upper critical field H_{c2}

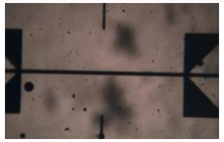


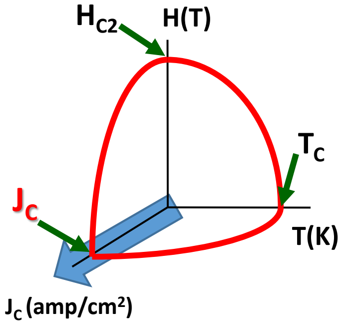


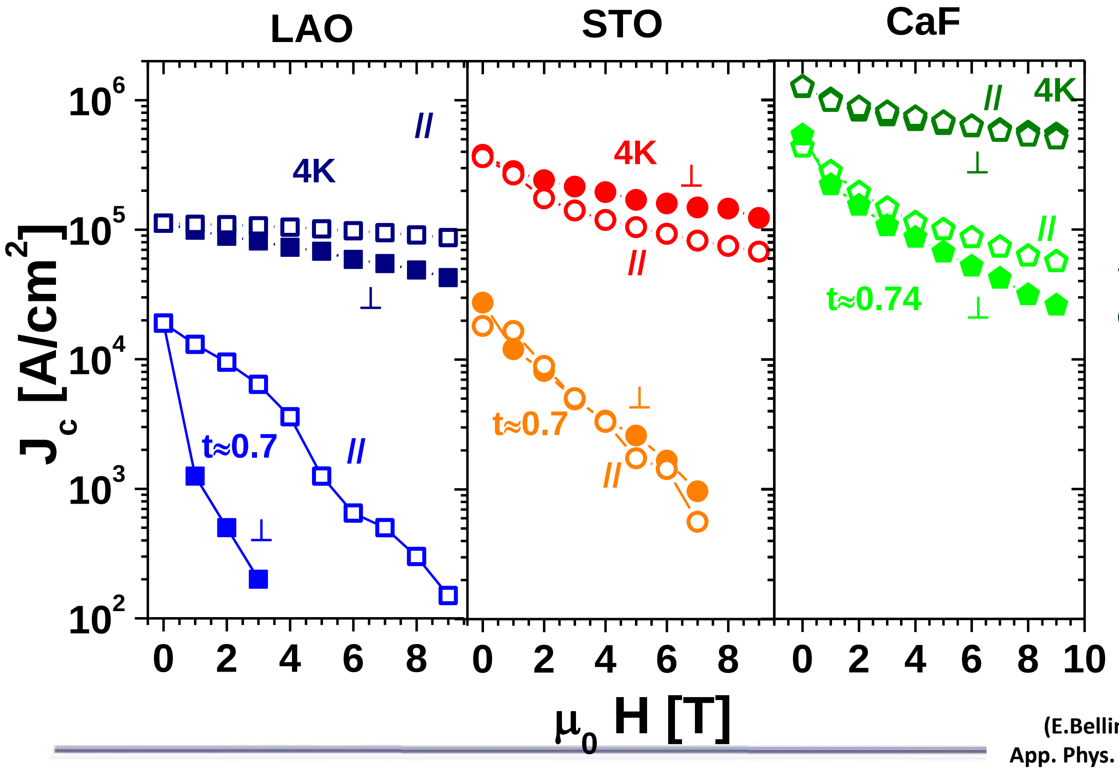
C. Tarantini et al., PRB 84, 184522 (2011)

Critical current densities

measured on thin (8–50 μm) strips patterned by photolitographic process + Ar ion milling etching





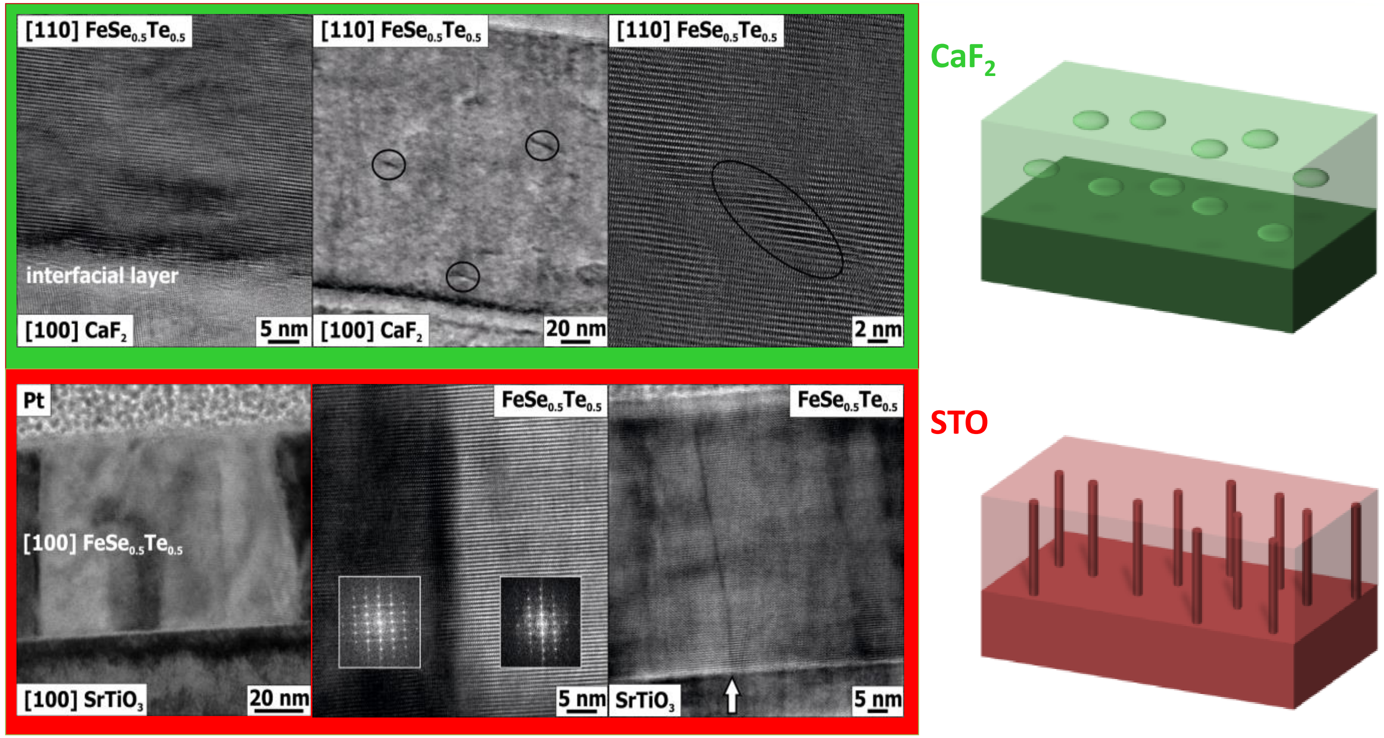


Very high J_c values + weak field dependence!

(E.Bellingeri, S.Kawale et.al.
App. Phys. Lett. 100,082601, 2012)

Leibniz Institute for Solid State and Materials Research Dresden

TEM analysis



STO: Single defect parallel to the *c*-axis, compatible with columnar defects shown by STM, which contribute to enhance pinning parallel to the *c*-axis as recorded by the highest J_c.

CaF_2 : No large defects or misorientations / grain structure. Small regions where the lattice seems to be disturbed on a very small local scale (5-20 nm). Such kind of defects might be the origin of the isotropic pinning present in the films grown on CaF_2.

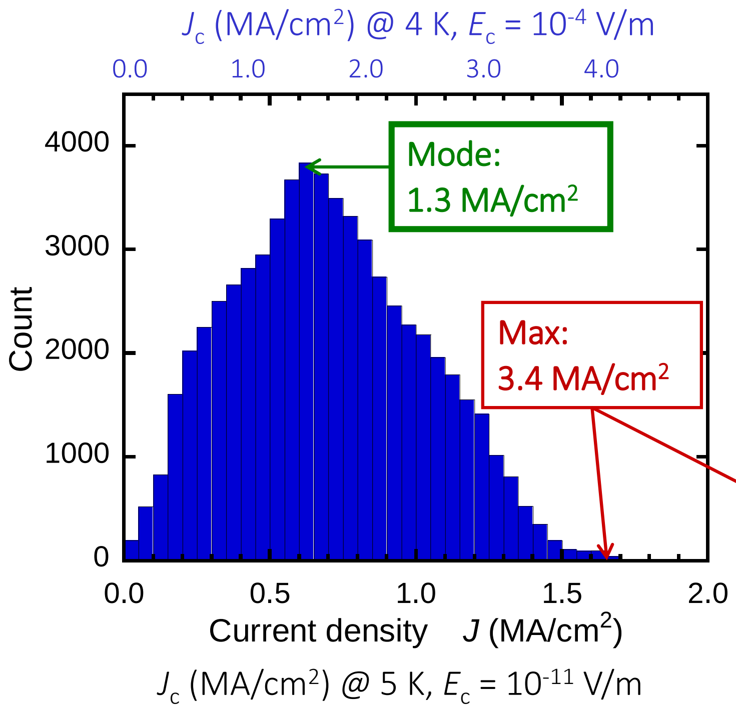
V. Braccini, et al. , Appl. Phys. Lett., 103, 172601 (2013)

Local Critical Current Density Distribution

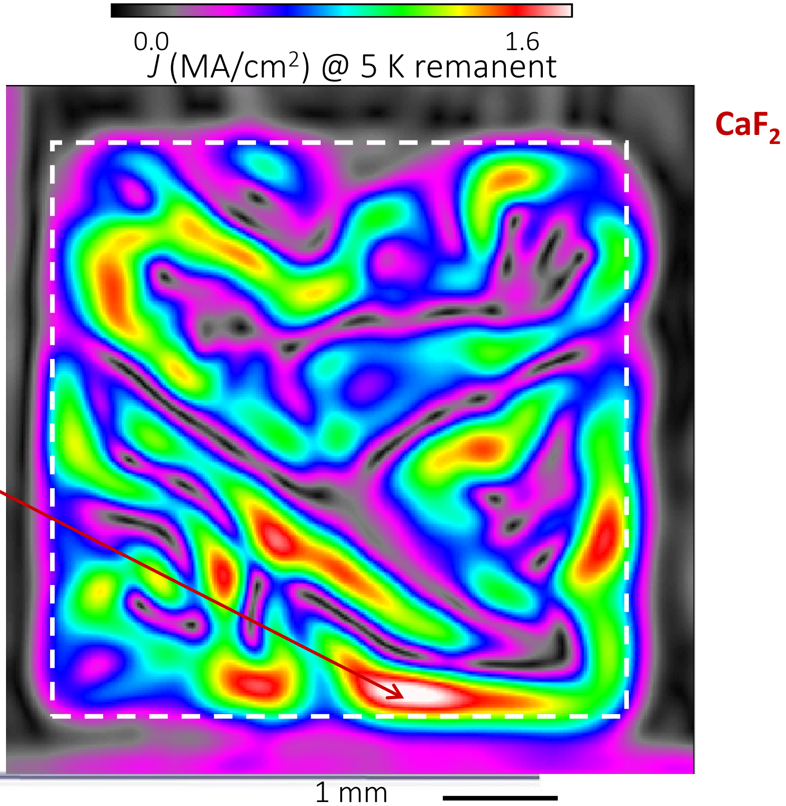
by LT-Scanning Hall Probe Microscopy (LT-SHPM)

KYUSHU UNIVERSITY

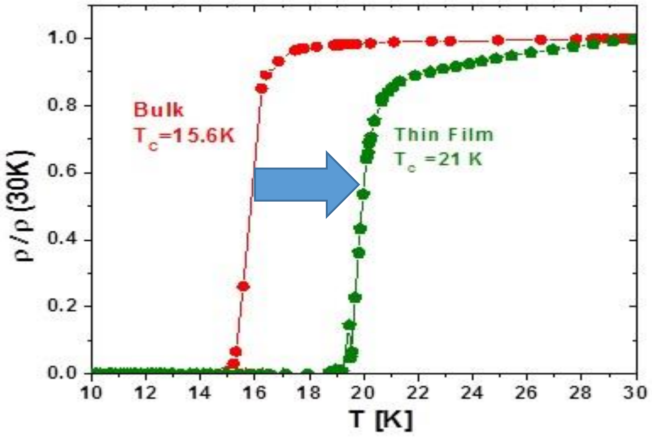
Mode value in agreement with transport J_c

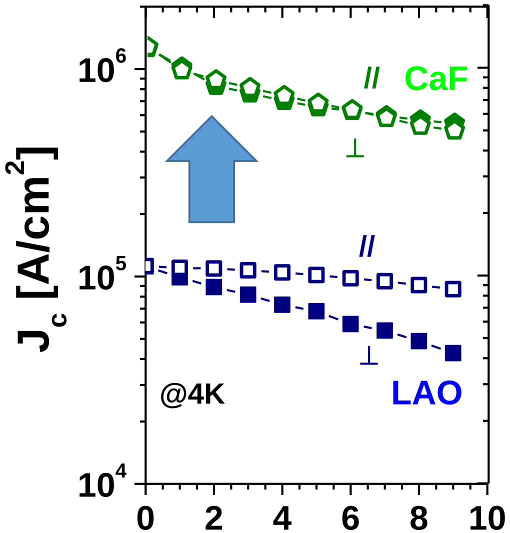


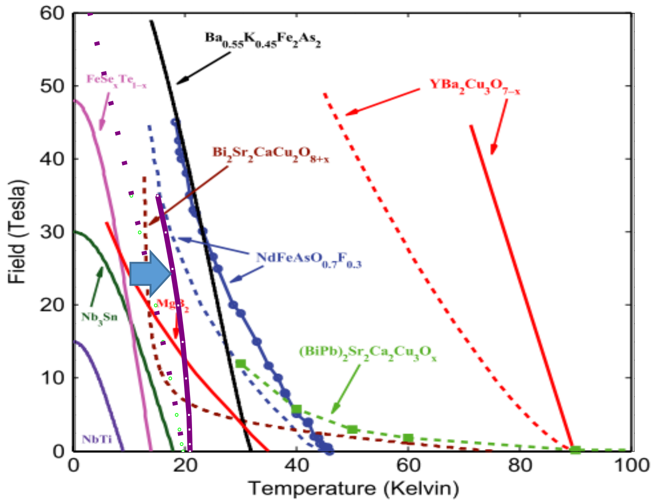
Histogram of the intensity of J_c distribution



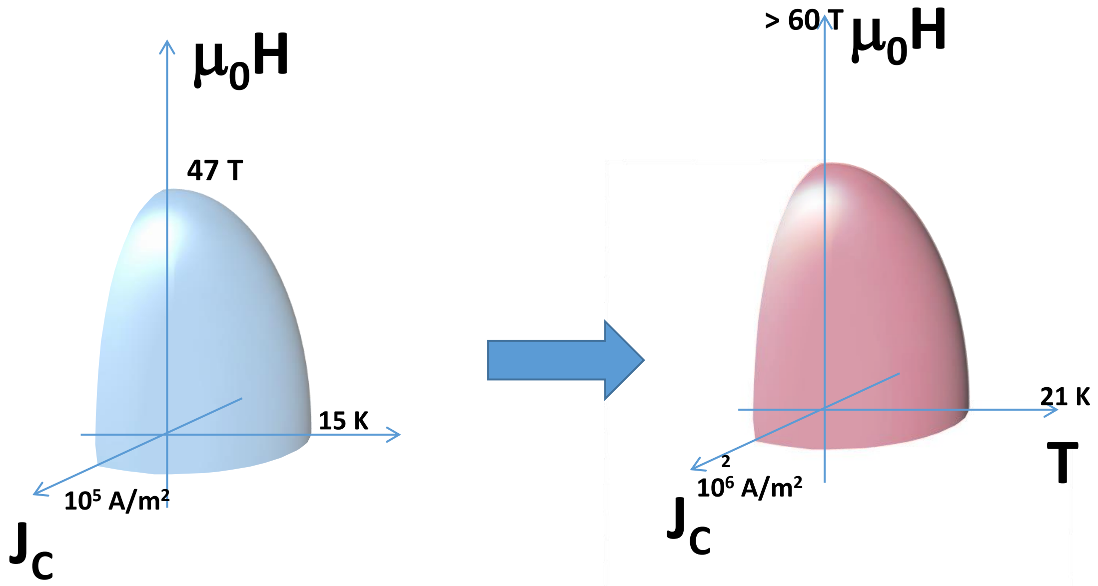
Summary of main achievements







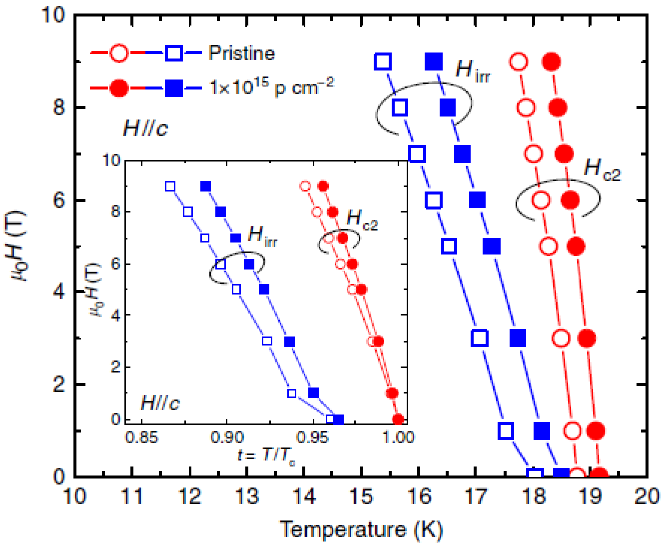
Summary of main achievements

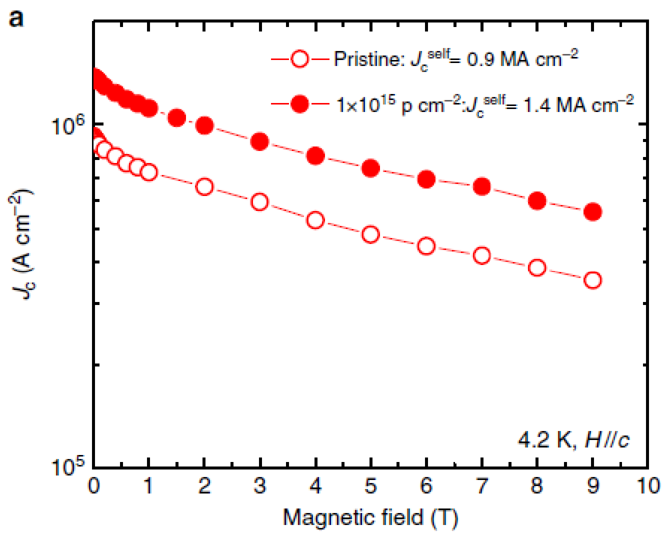


Effects of proton irradiation on the vortex dynamics of Fe(Se,Te) thin films

At BNL **irradiation with low energy (190 keV) protons** produced a simoultaneous increase in Tc and Jc.
Tc enhancement is due to the nanoscale compressive strain and proximity effect,
whereas Jc is increased through strong vortex pinning by the cascade defects and surrounding nanoscale strain.

T. Ozaki *et al.*, Nature Communication 7 13036 (2016)





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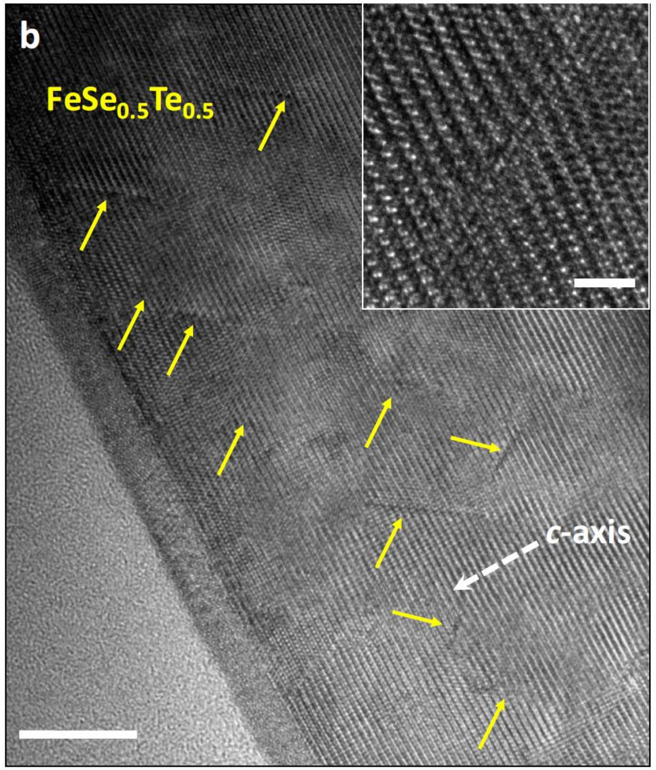
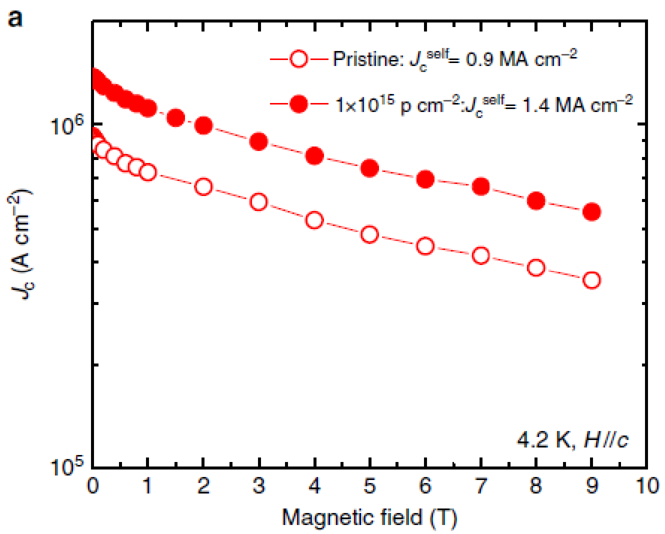


ature Communication 7 13036 (2016)



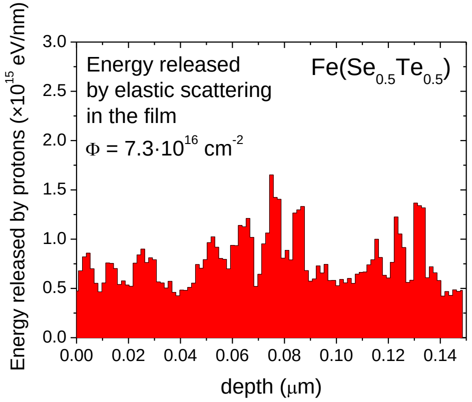
We tried irradiation with 3.5 MeV protons at the CN accelerator of INFN-LNL with fluences ranging from 0.7×10^{16} to 7.3×10^{16} cm^{-2}.

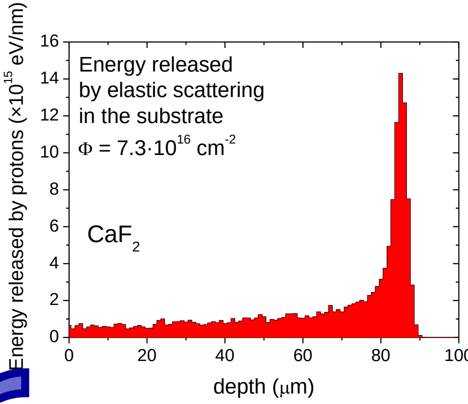
Laboratori Nazionali di Legnaro

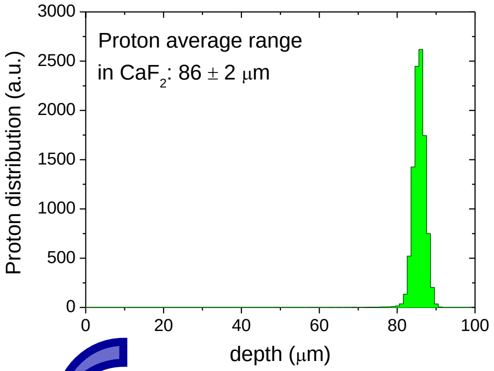
Collaboration with L. Gozzelino, G. Ghigo, C. Bennati, Department of Applied Science and Technology, Politecnico di Torino, and INFN-Torino, I.

Calculated damage and implanted atom distribution

Simulations of the energy loss of the protons (SRIM-2013 code) indicate that the protons cross the whole film thickness, creating a homogeneous defect distribution into the superconductor (calculations carried out using the monolayer collision step approach). Then protons implant into the substrate, where lost most of their energy.







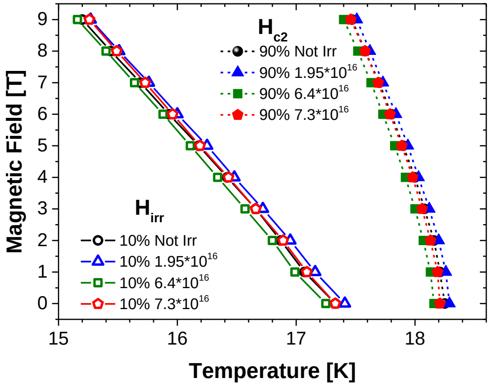
Energy release in the Fe(Se,Te) film due to the elastic scattering of the ions against the target nuclei.
Average displacement per atoms: 2.6×10^{-3} (Monolayer collision step mode)

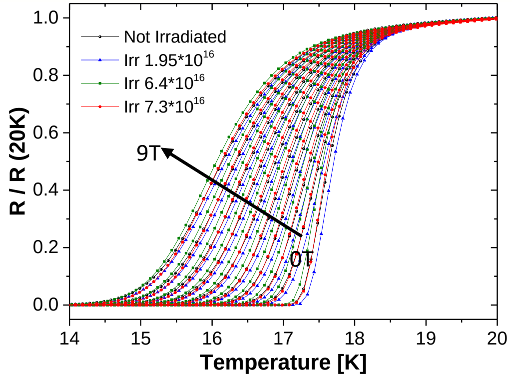
Energy release in the CaF_2 substrate due to the elastic scattering of the ions against the target nuclei

Spatial distribution of the implanted ions in the substrate

IUMRS-ICAM 2017 KYOTO

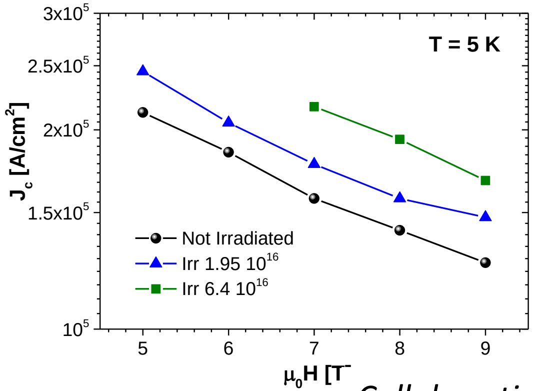
Critical temperature, critical fields and Critical current

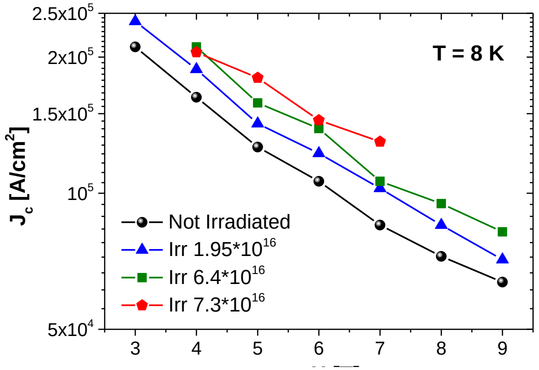


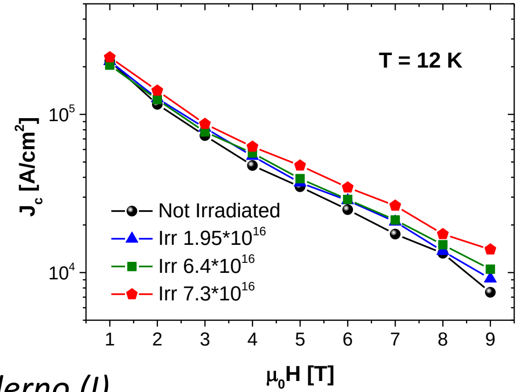


Even up to the highest fluences, T_c stays the same as well as the critical fields, while J_c monotonically increases with fluences at all the measured temperatures.

Increase of about 40% @ 4.2 K

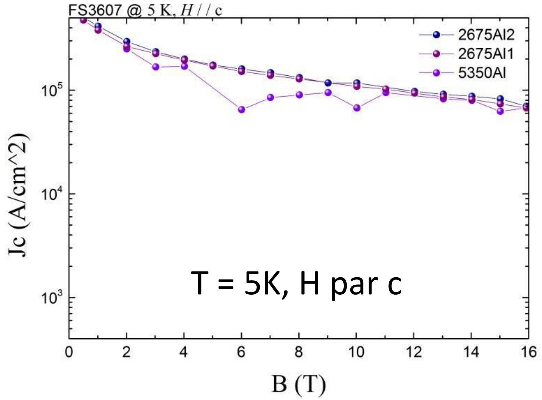


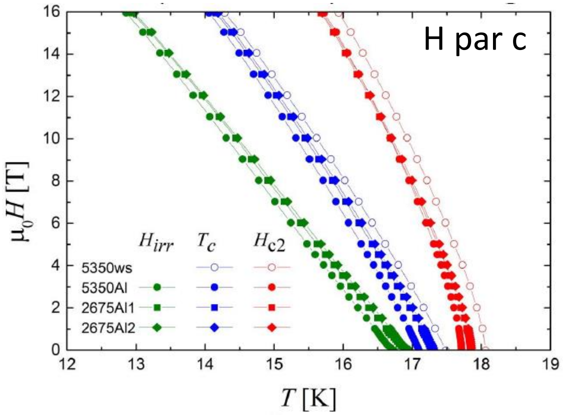


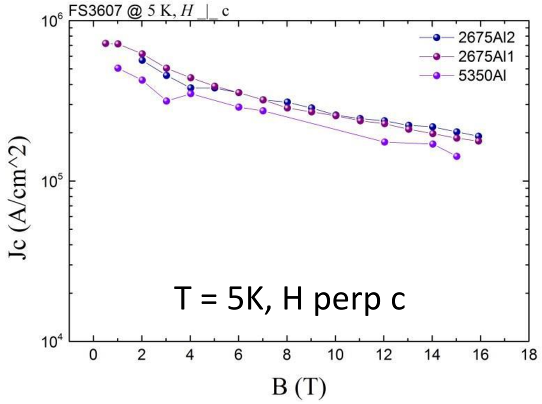


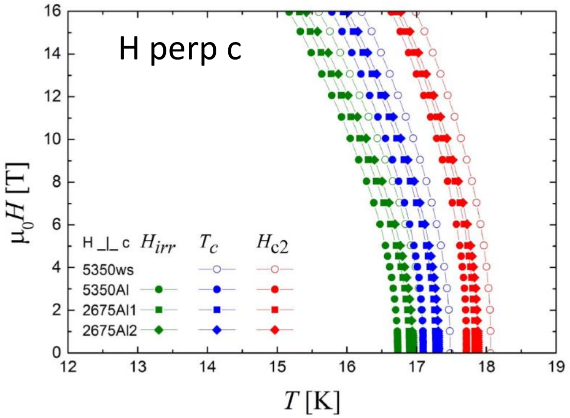
Collaboration with A. Leo and G. Grimaldi, CNR-SPIN Salerno (I)

Through simulations it is possible to demonstrate that using different proton energy the defects can be generate at different depth in film. The control of the implantation depth could be obtained by slowing down the particles through the positioning of an **Aluminium foil** on the top of the film. But we did not observe any relevant effect on Jc and Hc2.



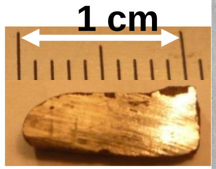


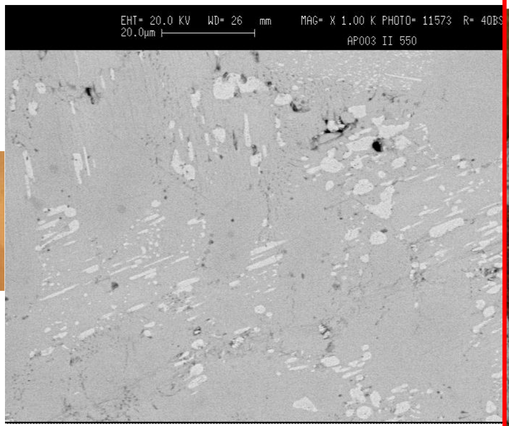


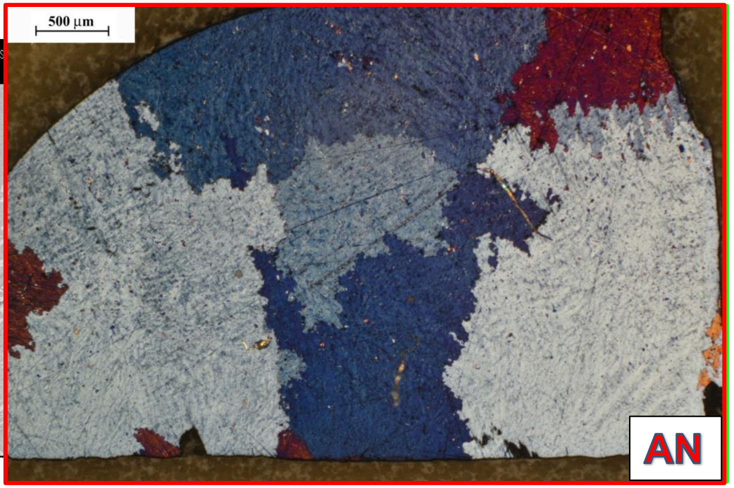


Collaboration with A. Leo and G. Grimaldi, CNR-SPIN Salerno (I)

Solid state synthesis (Fe, Se, Te → $Fe_y\ Se_{0.5}Te_{0.5}$) ⇨ **melting** ⇨ **annealing**







- Compact and dense

- Homogeneous matrix
- precipitates of Te-enriched phase like Fe2Ch3)

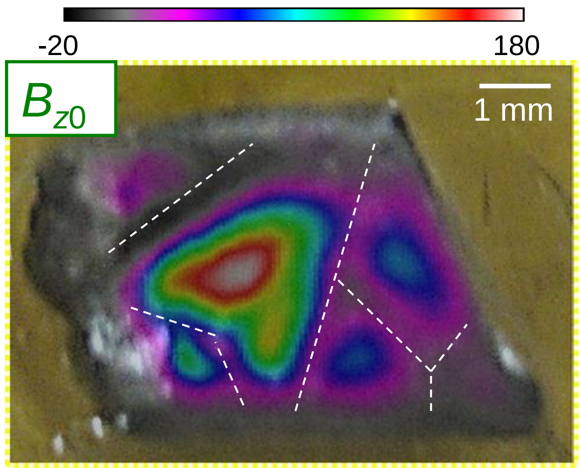
- Large grains
- No evidence of precipitation at grain boundaries

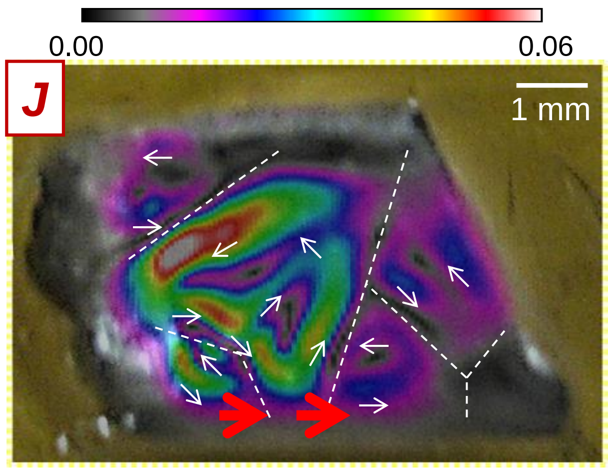
A. Palenzona et al., SUST 25 (2012) 115018

Scanning Hall Probe Microscopy

K. Higashikawa, T. Kiss

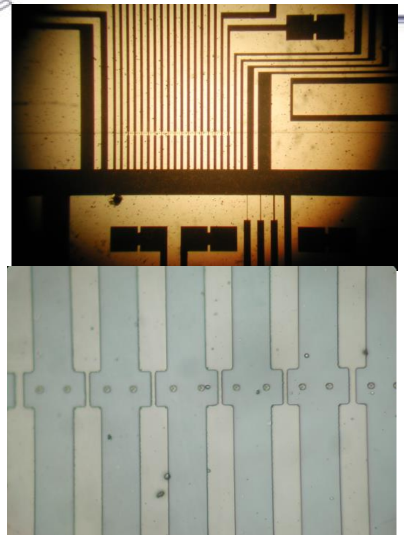
九州大学
KYUSHU UNIVERSITY

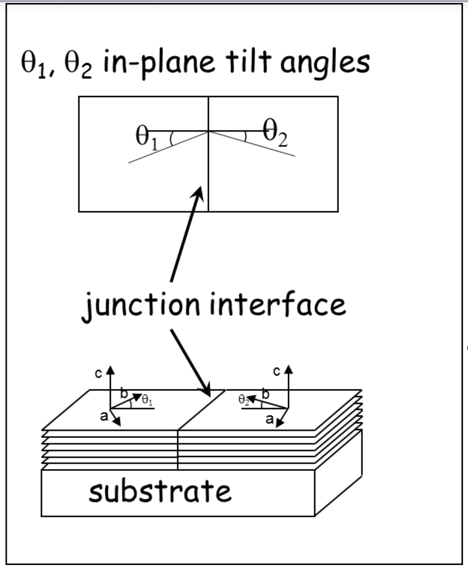


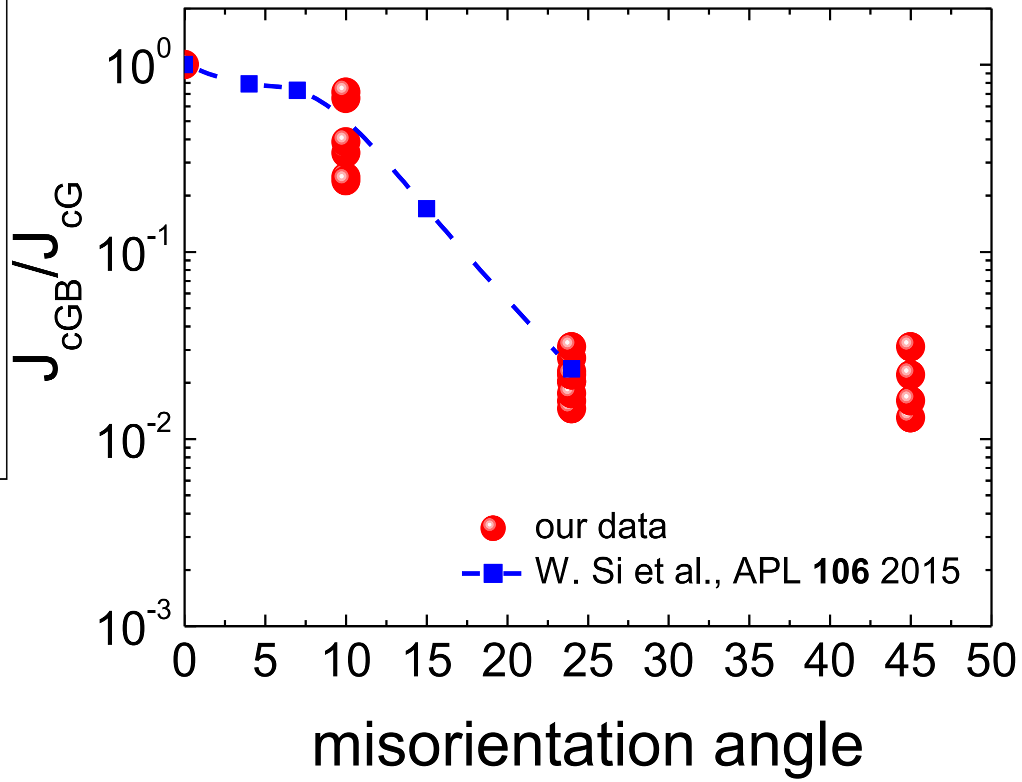


- **Magnetic field distribution in a remnant state at 5 K**
- **Dotted lines underline the GB, as confirmed by PLOM**
- **Intragrain J_c~7 x 10^4A/cm^2 @5 K**
- **Some inter-grain connections J_c~ 2 x 10^4A/cm^2**

Grain Boundaries behavior



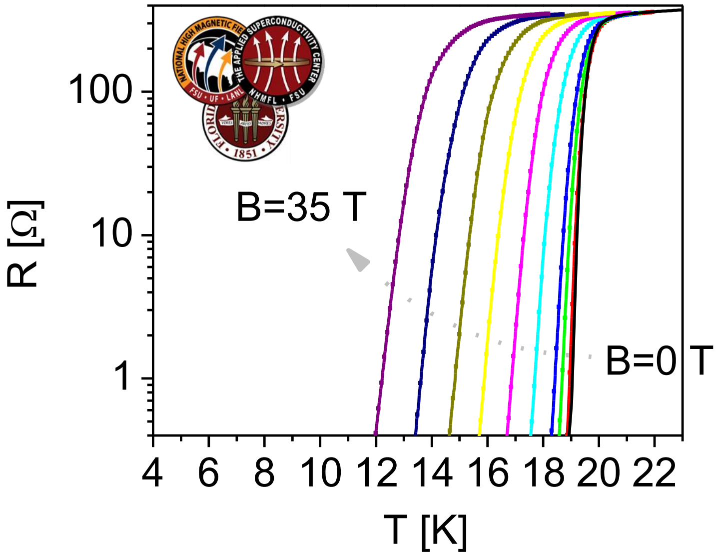




Si et al. state:
" A critical mis-orientation angle of around 9 degrees was identified that separates the strong coupling region from the weak link region. We found that the critical current densities across the grain boundary with a 24 degrees mis-orientation angle are modulated by the magnetic field, indicating a Josephson Effect".

S. Kawale et al., IEEE trans on Appl.Supercond. 25 (2015)
E. Sarnelli et al. IEEE trans on Appl.Supercond. 4 (2017)

Grain Boundaries behavior



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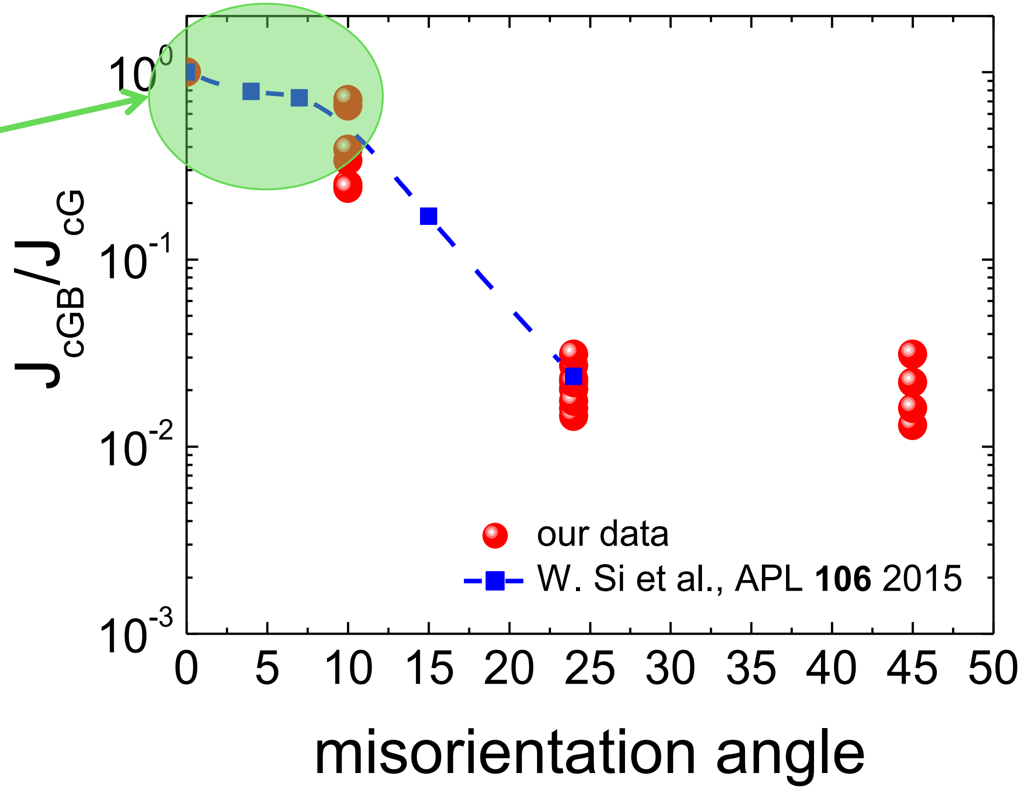
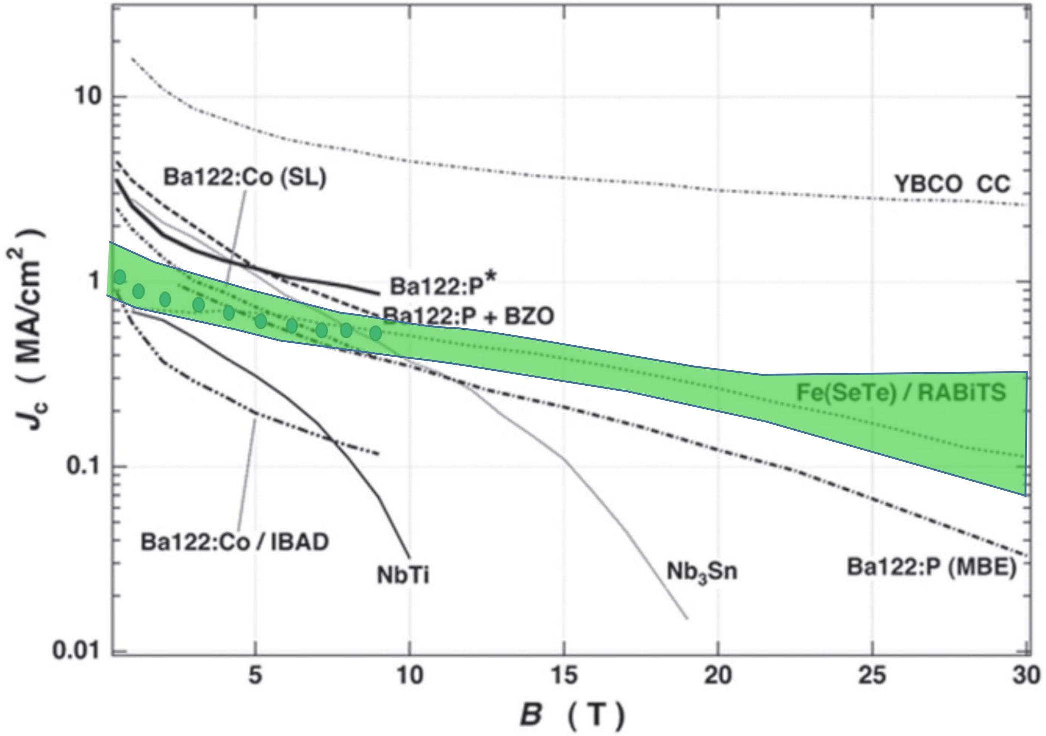
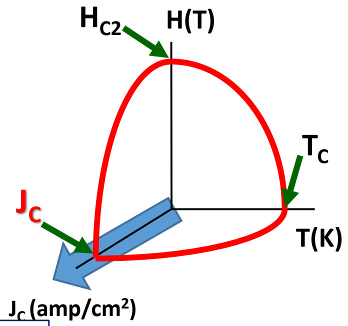


S. Kawale et al., IEEE trans on Appl.Supercond. 25 (2015)
E.Sarnelli et al. IEEE trans on Appl.Supercond. 4 (2017)

Critical Current density on technological substrates



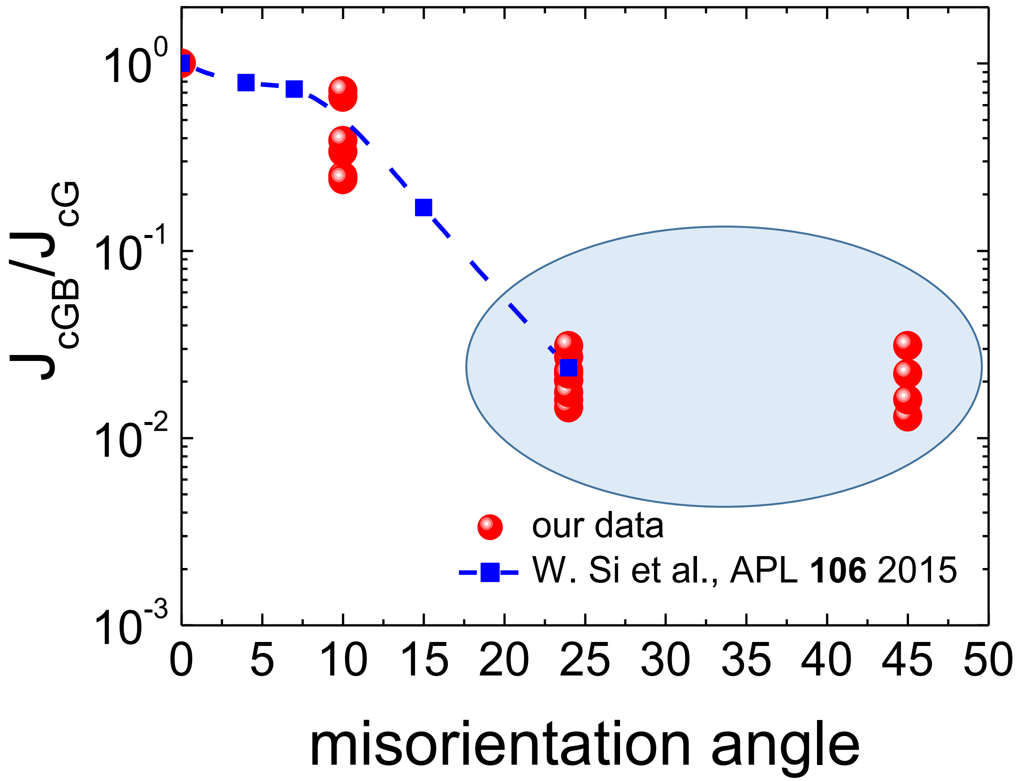
Jc of different Coated Conductors versus magnetic feld.
The BNL group shows on Fe-11 coated conductors, a critical current as high as 0.1MAcm-2 at 4.2K 30T!
Very nice results for applications.



A3-O30-017 V. BRACCINI et al , Fe(Se,Te) epitaxial thin films deposited on metallic alloys….
CERN: FCC-GOV-CC-0086 EDMS07 1750320/KE 35 Development of IBS conductors
Idea: Iron Alloys substrates (Fe/Ni) or Ni Alloys + buffer layer with low angle

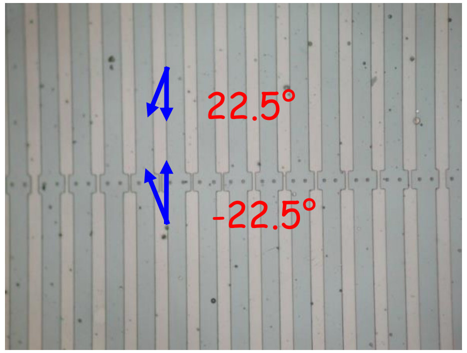
High Angle: electronics applications

Grain Boundaries behavior

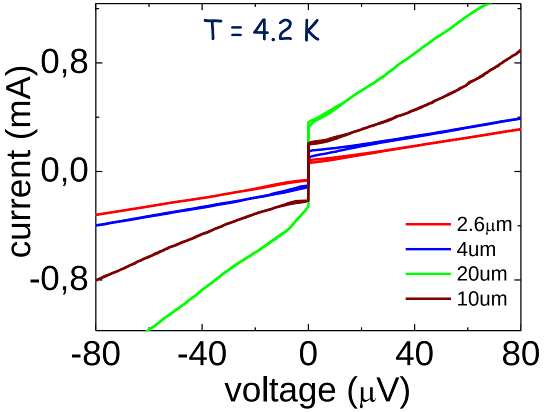


Fe(Se,Te) Josephson junctions

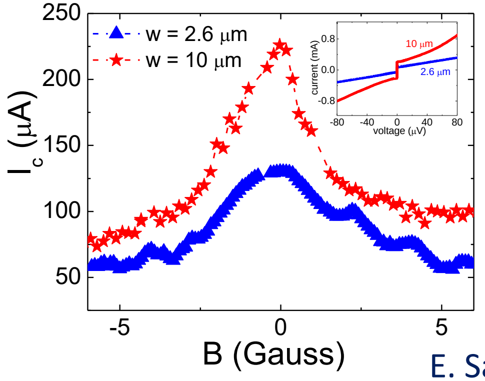
Symmetric 45° [001] tilt Fe(Se,Te) GBJs



IV characteristics

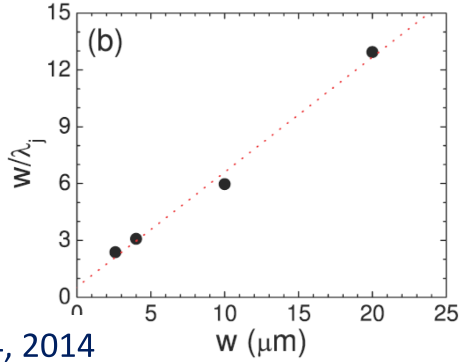


Magnetic field dependence of the critical current



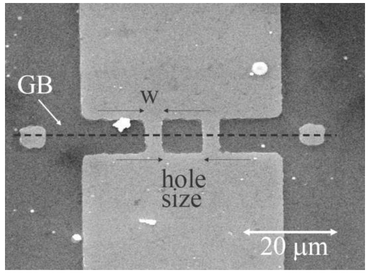
E. Sarnelli e al. APL 104, 2014

Scaling of the normalized junction width with the junction width

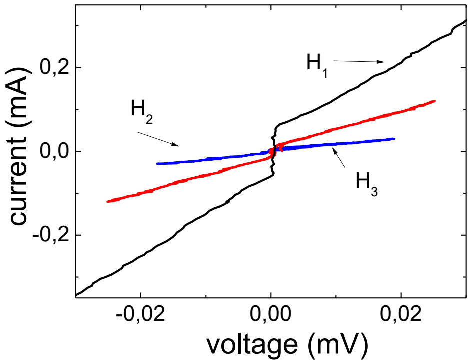


A linear scaling indicates that the critical current is uniformly distributed along the junction area

Fe(Se,Te) DC-SQUIDs



IV characteristics



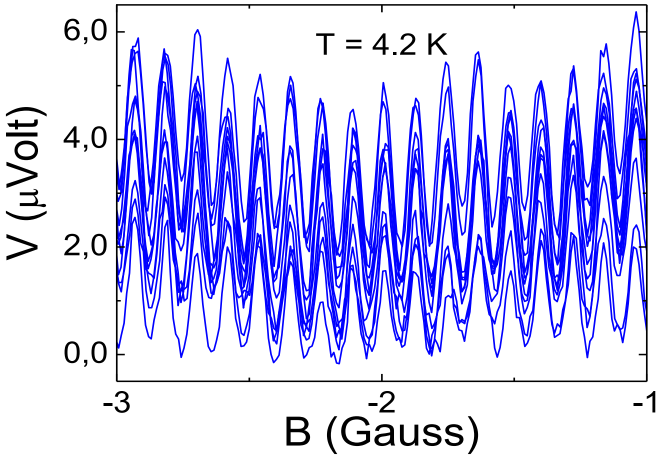


Table 2. Experimental electrical parameters of the fabricated dc-SQUIDs.

Sample	I_c (μA)	J_c (A cm^{-2})	R_N (Ω)	$\tilde{\rho}_N(\Omega \times \mathrm{cm}^2)$	I_cR_N (μV)
H1	45.0	3.2×10^3	0.08	1.1×10^{-9}	3.6
H2	17.7	3.2×10^3	0.25	1.4×10^{-9}	4.4
H3	10.7	3.9×10^3	0.63	1.8×10^{-9}	6.7

Sample	L (pH)	$\beta=\frac{2LI_c}{\Phi_0}$	ΔV_{Th} (μV)	ΔV_{exp} (μV))	$H_{exp}=\frac{\partial V}{\partial \Phi}\big\vert_{\Phi=\Phi_0/4}$ ($\mu V/\Phi_0$)
H1	10.3	0.23	2.0	2.5	3.2
H2	15.8	0.14	2.6	1.8	4.0
H3	22.5	0.12	4.0	3.8	9.4

$$\Delta V_{th} = \frac{7}{\pi^2}\frac{I_cR_N}{(1+\beta)}\left[1-3.57\frac{\sqrt{k_BTL}}{\phi_0}\right]$$

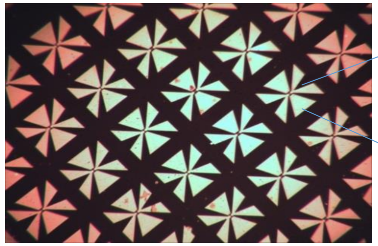
T. Ryhanen et al., J. Low Temp. Phys. 76, 287 (1989)
K. Enpuku et al., J. Appl. Phys. 73, 7929 (1993)

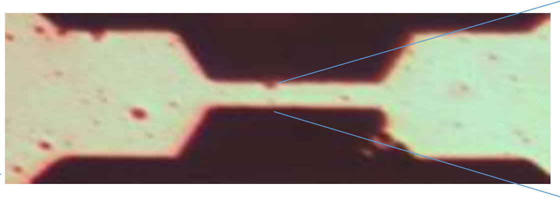
$$h = \frac{H_{\exp}}{I_cR_N}(1+\beta) = 2.4e^{\left[-3.5\pi^2\delta\Phi_n/\Phi_0^2\right]}$$

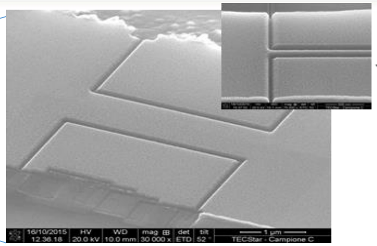
That is independent of electrical parameters I_c and R_n
Experimental values indicate high contribution of vortex motion to noise

E. Sarnelli et al. SUST 30, 065003 (2017)

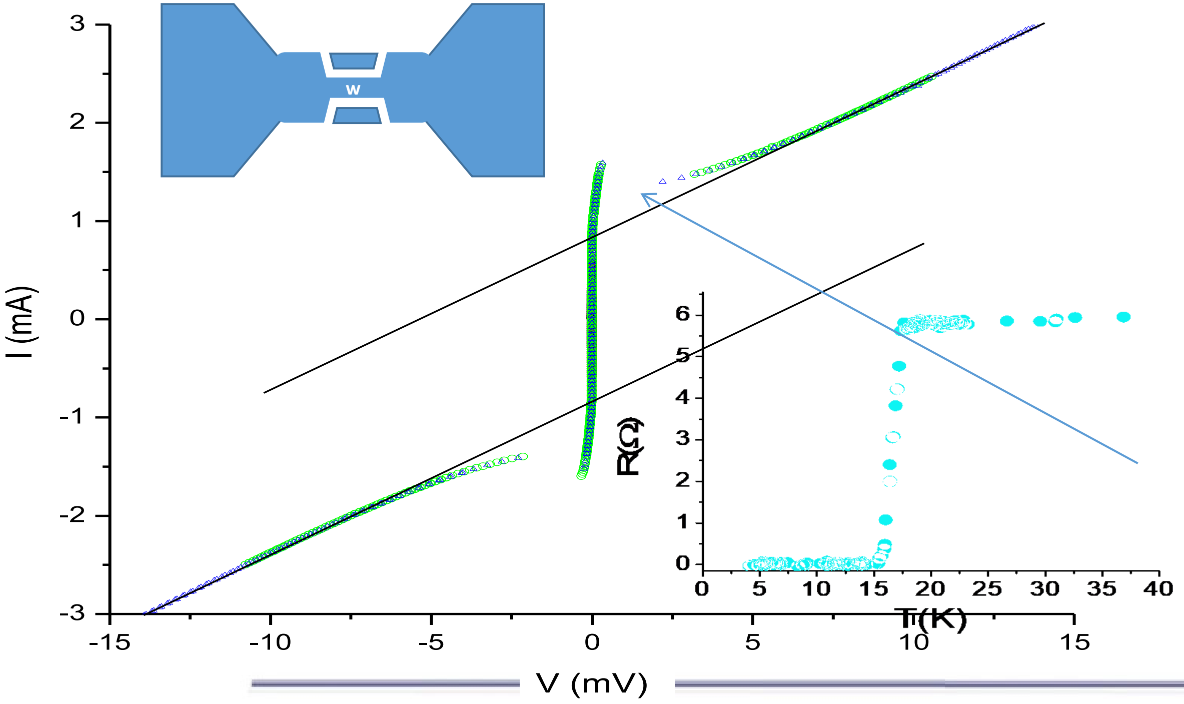
Submicrometric FIB patterning of Fe(Se,Te) films







50 nm wide
3 μm long nanostrip
By Focused Ion Beam



- Tc_0 = 15.5 K
- Jc =1.5 10^6 A/cm^2

No FIB damage

- Fast switch from 0 voltage to resistive branch Typical for S/C nanowire.
- IV useful as detector ?
- Flux line motion (displaced linear slope characteristic)?

C. Nappi et al., Sci. Rep. 7, 4115 (2017)

CONCLUSIONS

IUMRS-ICAM 2017

The 15th International Conference on Advanced Materials

- ✓**$FeSe_{1-x}Te_x$ in form of thin film presents important differences in respect to the bulk samples**
- ✓**$FeSe_{1-x}Te_x$ in form of thin film presents enhanced critical characteristics**
  - ✓Enhanced T_c beyond 20K
  - ✓Enhanced critical fields (60T)
  - ✓High J_c are sustained up to very high magnetic field
- ✓**Artificial GB shows a limit angle of 10° for strong SC coupling**
  - ✓Possibility for coated conductors on more simple oriented metallic substrates
  - ✓Good tolerance to irradiation
  - ✓Possibility for electronic devices and sensors