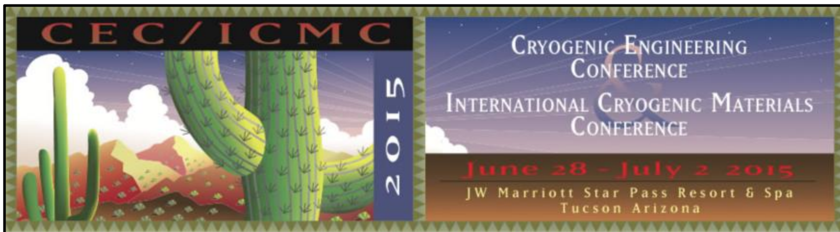

SPIN

LOW TEMPERATURE- HIGH FIELD PERFORMANCES OF IRON CALCHOGENIDES THIN FILMS

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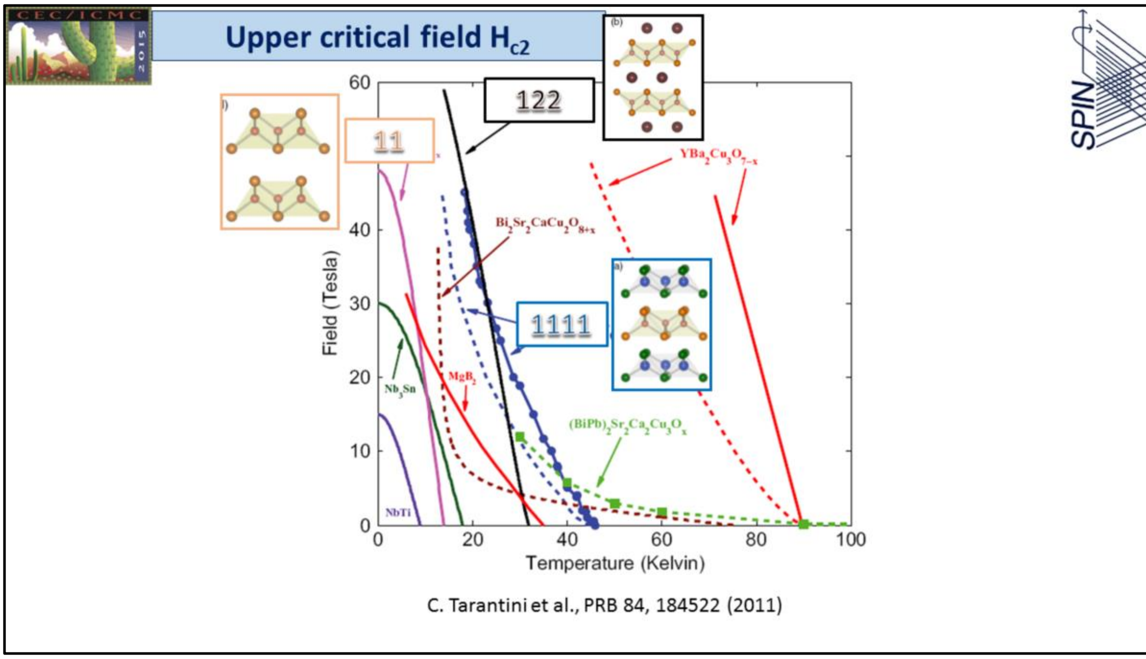
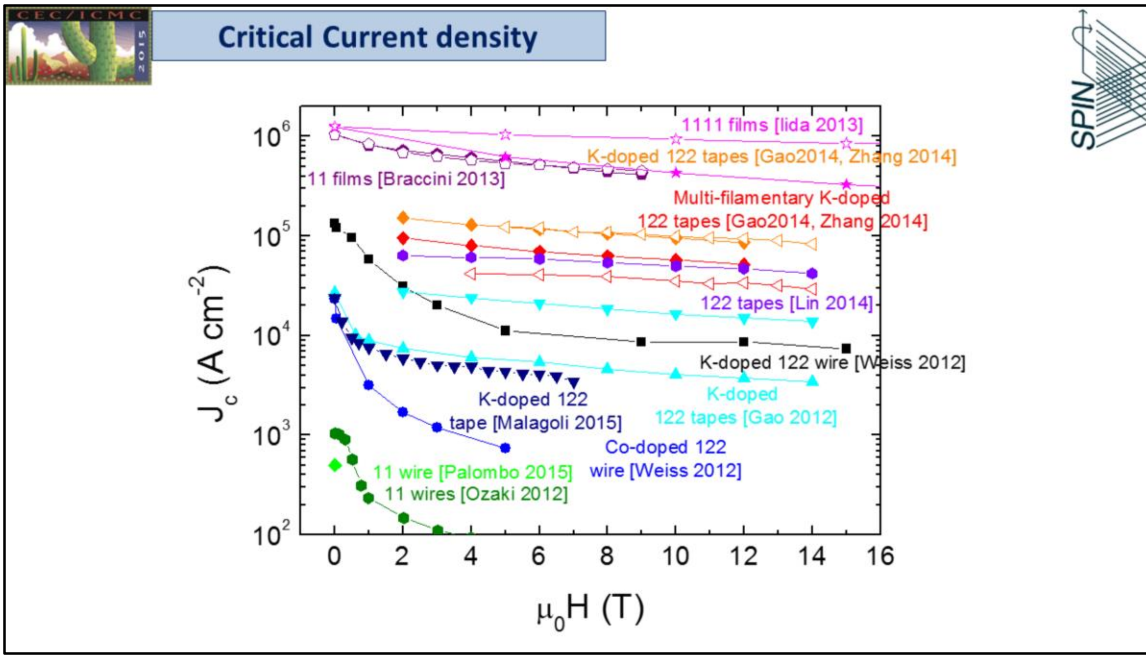
Super Iron
FP7-NMP-2011-Eu-Japan

The talk will treat the possible use of iron calchogenides at liquid He temperatures and high fields.



In fact, all families of Fe-based superconductors show very high critical fields. Despite the low critical temperature, also the 11 phase shows interesting critical fields.

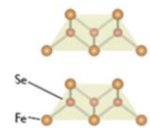


In the figure, the in-field critical current densities for the various families (literature values) are shown. The case of 122 is different in respect to the 11. The critical current density in the case of Fe- 11 wire/tapes is strongly depressed.

IEEE/CSC & ESAS SUPERCONDUCTIVITY NEWS FORUM (global edition), July 2015.
Invited presentation M3OrA-01 given at CEC-ICMC 2015, Tucson, USA, June 28 – July 2, 2015.

Why Fe(Se,Te) ?

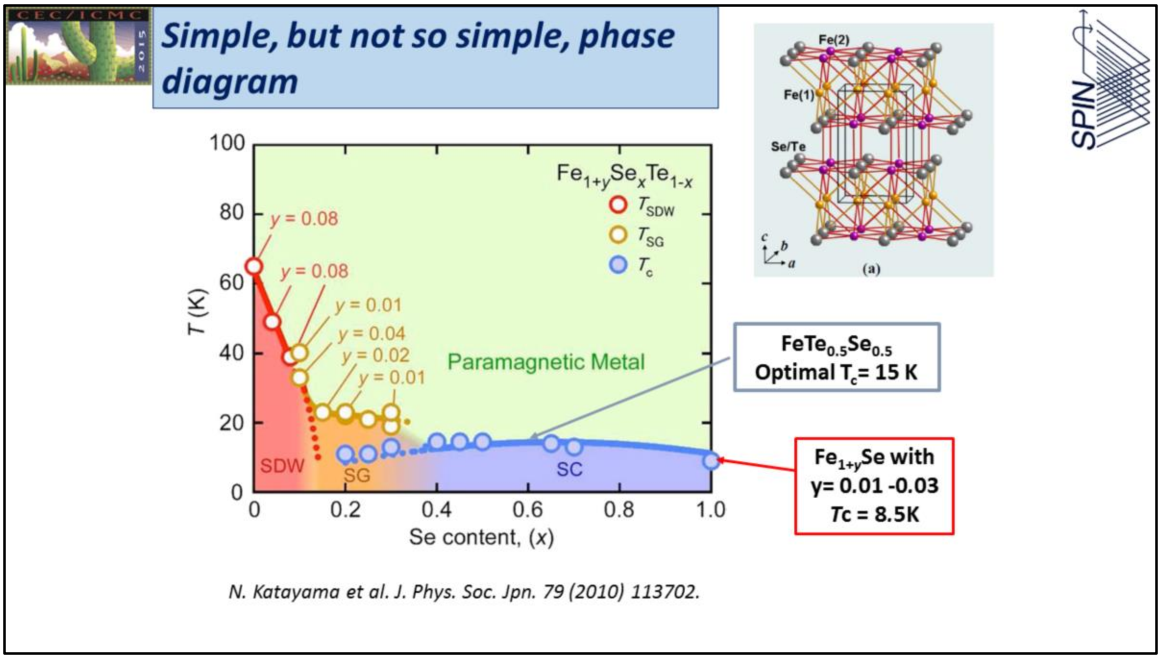
- T_c (~ 10-15 K) values, but tuneable with pressure
  In bulk: up to **26K** by application of external pressure
- **Very high H_{C2}** values > 50 T
- **High J_c** (*~ 10^6 A/cm^2 in self-field) and* weakly depends on magnetic field.
- **Simplest crystal structure**



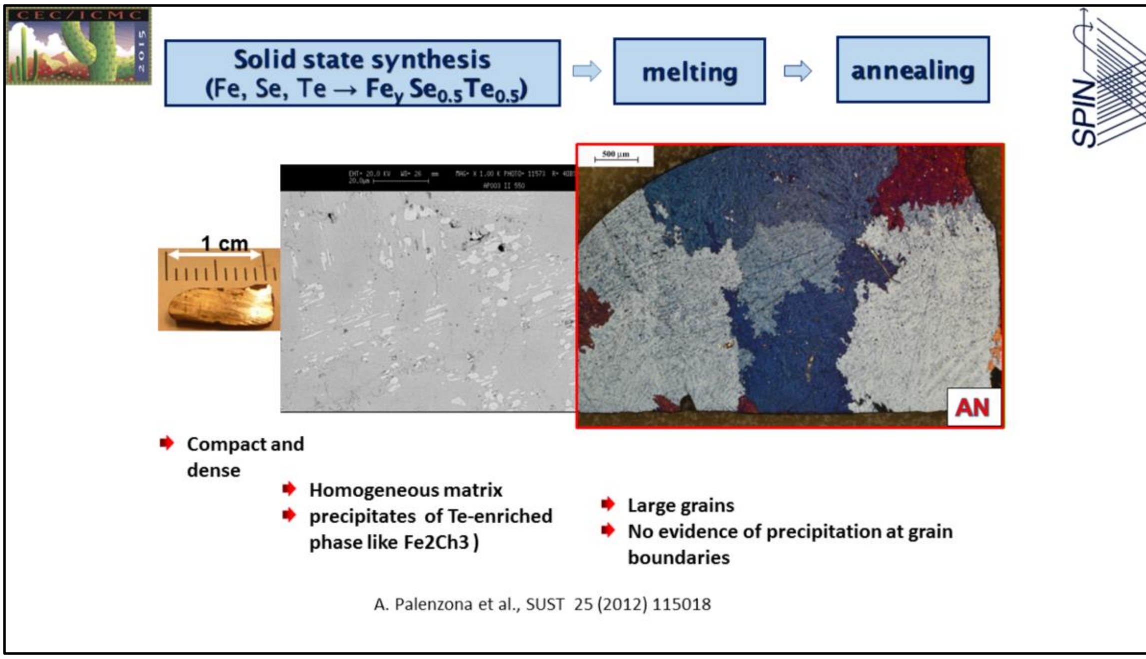
- **Easy to grow *and to synthesize*** (Just three components):
- ***Easy to handle stable in air*** *(not explosive)*
- **Do not contain As**

Appealing from an applicative point of view

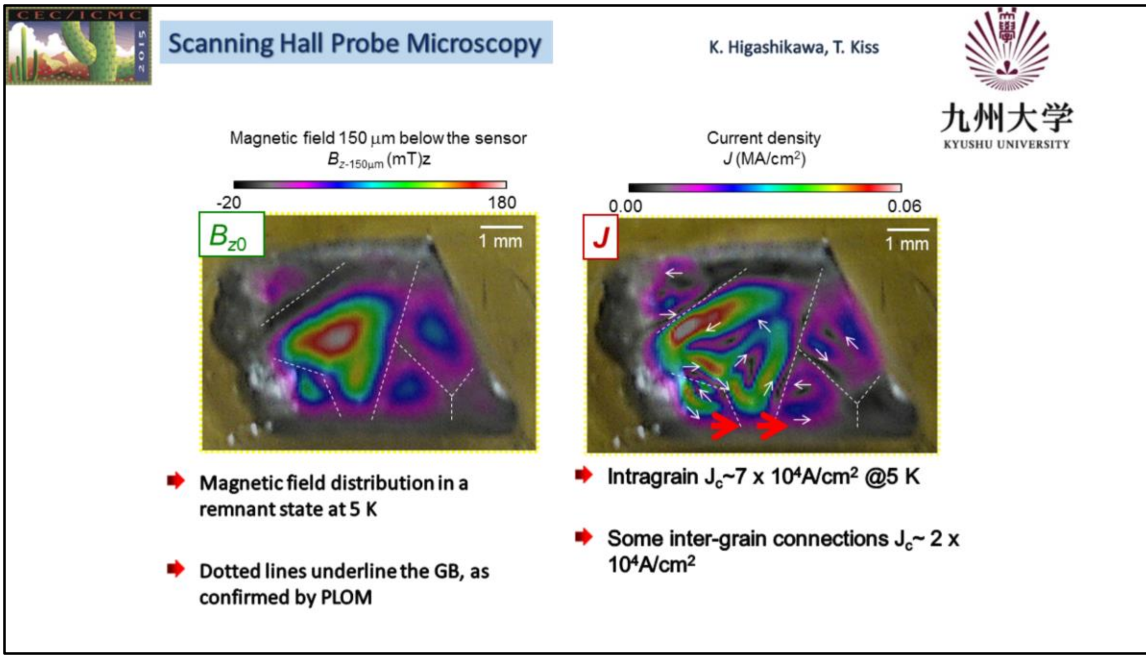
Here the favorable characteristics of the 11 phase.



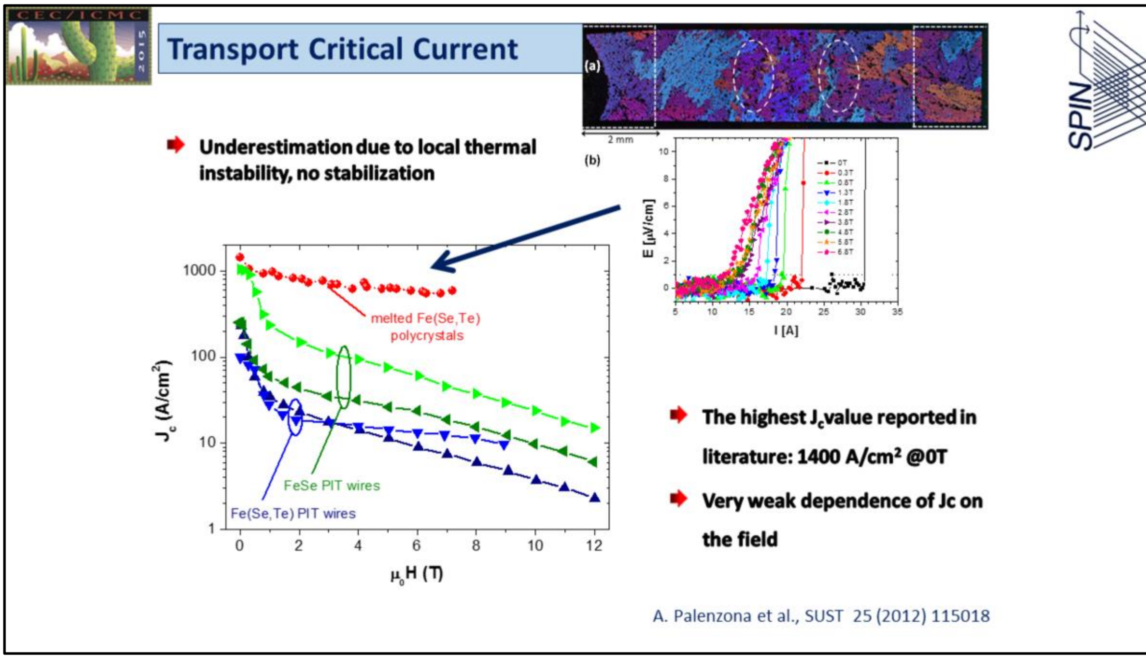
By replacing Te with Se, the structural transition temperature is gradually suppressed. Upon 6% Se doping, a trace of superconductivity starts to appear, and coexists with the antiferromagnetic order. With increasing Se content, the superconducting volume fraction improves, and Tc also becomes optimized for x ~0.5. Superconductivity extends all of the way to x = 1 in Fe(1+y)Se. All these samples showed nanoscale phase separation and chemical inhomogeneity of the Te/Se content. For x=1 it is found that the stable composition range of Fe(1+y)Se is between 0.01 and 0.03 with Tc = 8.5K. Therefore, all the work presented here is referred to a nominal Fe versus Te composition of 50%.



Here we present an original method to prepare bulk FeSe0.5Te0.5 samples, based on a melting process and a subsequent annealing treatment. With respect to the standard sintering technique, it produces much more homogeneous and denser samples, characterized by large and apparently well interconnected grains. In these samples we studied the effect of grain boundaries. In the photograph, realized by polarized light, different colors represent different crystallographic orientations.



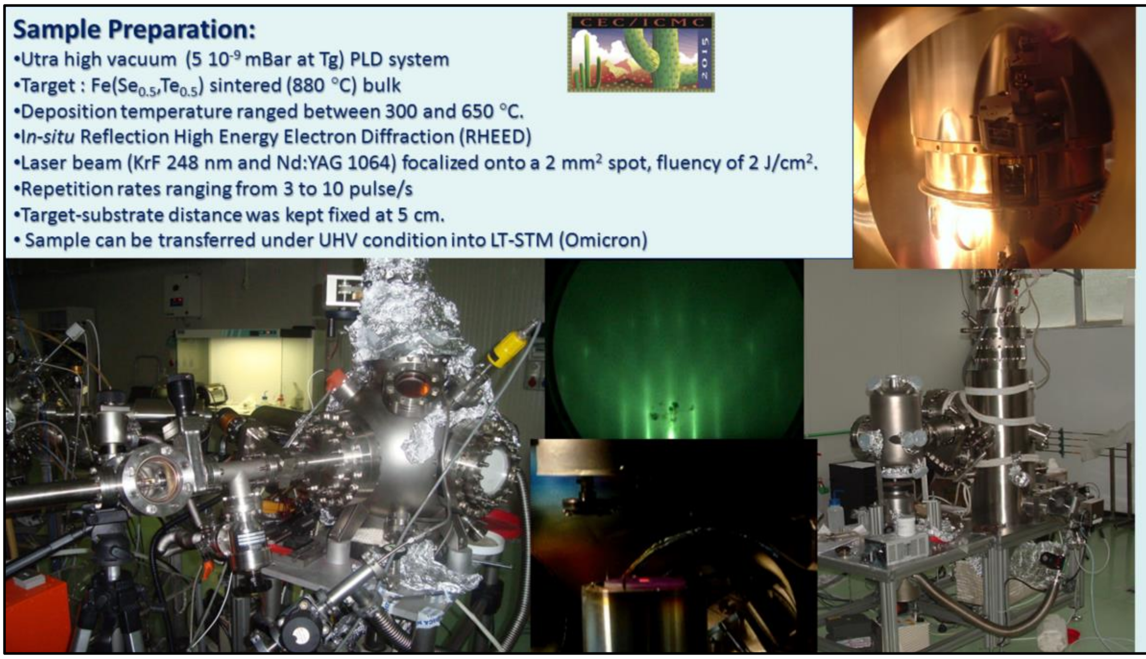
Scanning Hall probe microscopy measurements on these samples performed at Kyushu University. The harmful role of GBs is clearly visible. The sample seems to be not connected and divided in some differently magnetized regions. In these regions the intragrain Jc is of the order of 0.1 Ma/cm2.



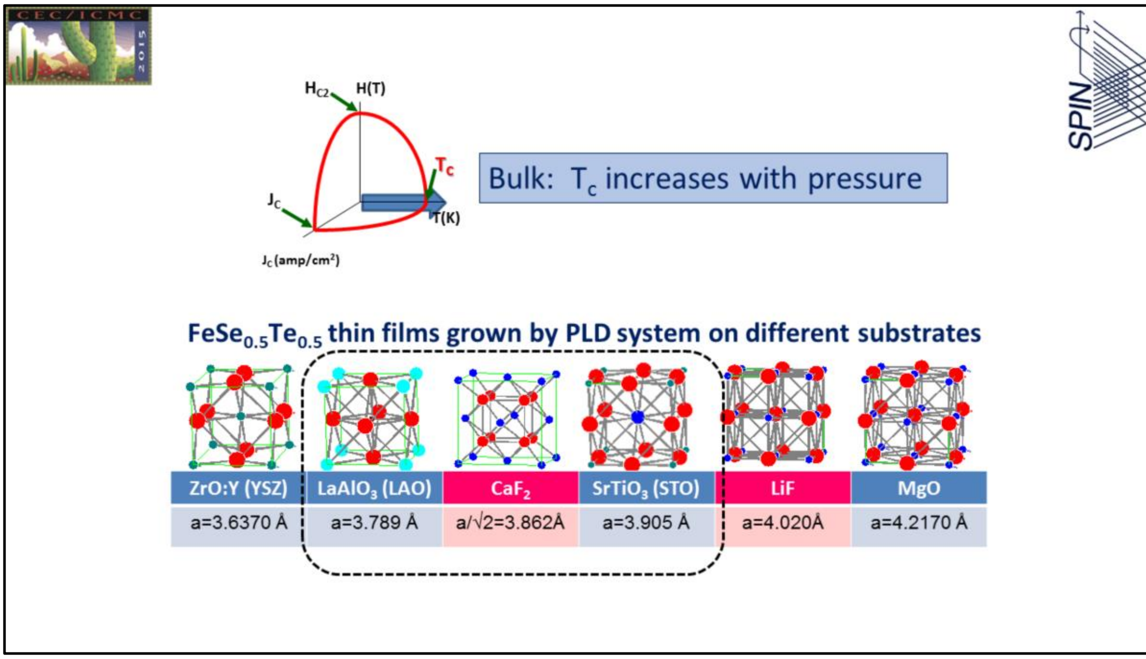
In the figure (on top, right side) we show the picture taken with a polarized light microscope on the relevant portion of the sample before measuring it by transport. A sketch of the voltage taps and of the position of the current leads is also shown, to better clarify that during this global measurement the current was flowing through portions of the sample presenting different orientations, therefore crossing several grain boundaries.

In figure below some selected I–V transitions are reported. We report also the transport Jc as a function of the magnetic field at 4.2 K extracted from the I–V curves. Despite the fact that transport Jc values are the highest ever reported for the 11 phase the results indicate that the connection between the grains is still critical.

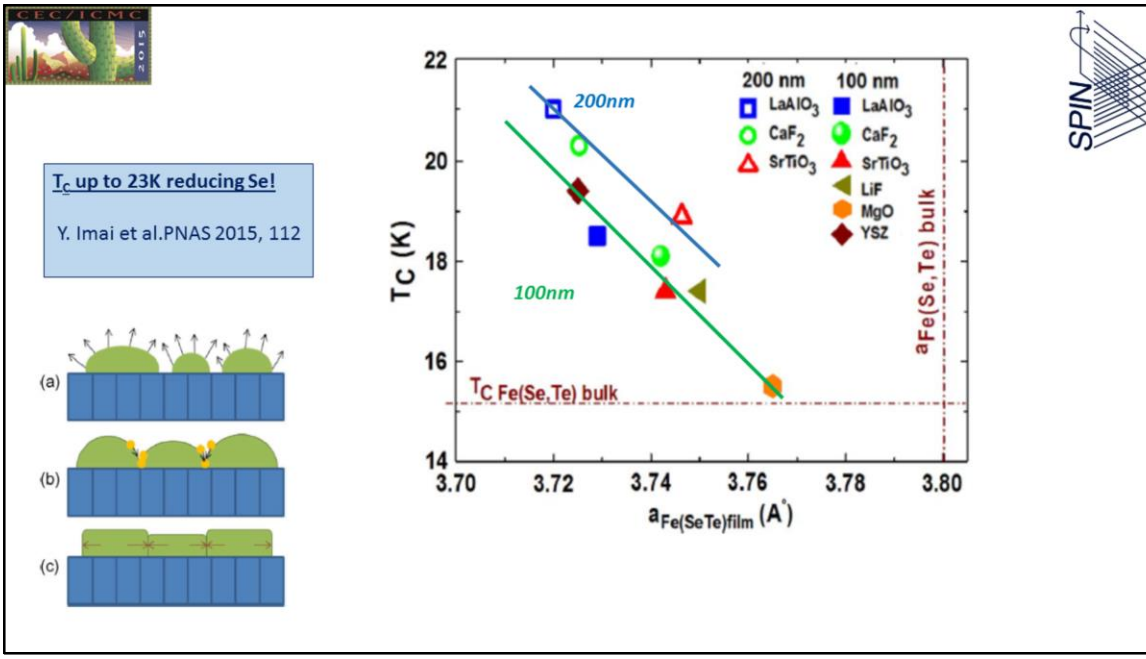
To study the effect of a single GB we considered 11 epitaxial thin films.



We grow thin films by pulsed laser ablation starting from bulk target of FeSe0.5Te0.5 (Tc= 16 K). Films were grown on single crystal substrates at temperatures between 300 and 650 °C with a deposition rate ranging from 0.06 up to 0.2 Ås−1. In the slide the laser characteristics are reported. The repetition rate was 3-10Hz. Films can be transferred to a cryogenic STM system under high vacuum for surface characterization, whereas the structural and transport properties were studied ex situ. In the slide, some other details are depicted.



FeSe0,5Te0,5 thin films were deposited on single crystal substrates having different cell parameters and different chemical compositions, namely magnesium oxide (MgO, a=4.217 Å), strontium titanate (SrTiO3, a= 3.905 Å), lanthanum aluminate (LaAlO3, d= 3:789 Å), yttria stabilized zirconia(YSZ, a= 3,637 Å) and calcium(CaF2, a=5.462 Å) and lithium fluoride (LiF2, a=4:020 Å). For all the substrates the (001) orientation was used. The substrates were glued by silver paint onto a stainless steel sample holder compatible with the used scanning tunneling microscope (Omicron Nanotechnology GmbH). The quality of the growth was in situ monitored by reflection high energy electron diffraction (RHEED). We will focus on thin films on the highlighted substrates.

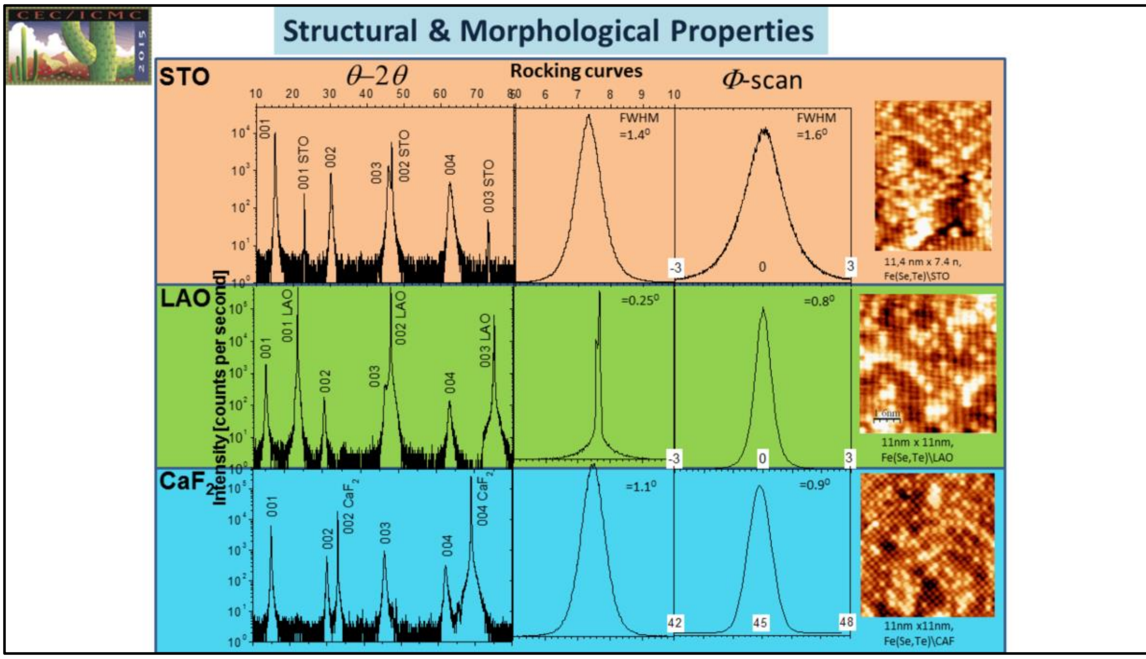


In figure the Tc values obtained for films with a thickness of 100 and 200 nm are reported as a function of the surface cell parameter of the substrate. Tc can reach the very high value of 21 K.

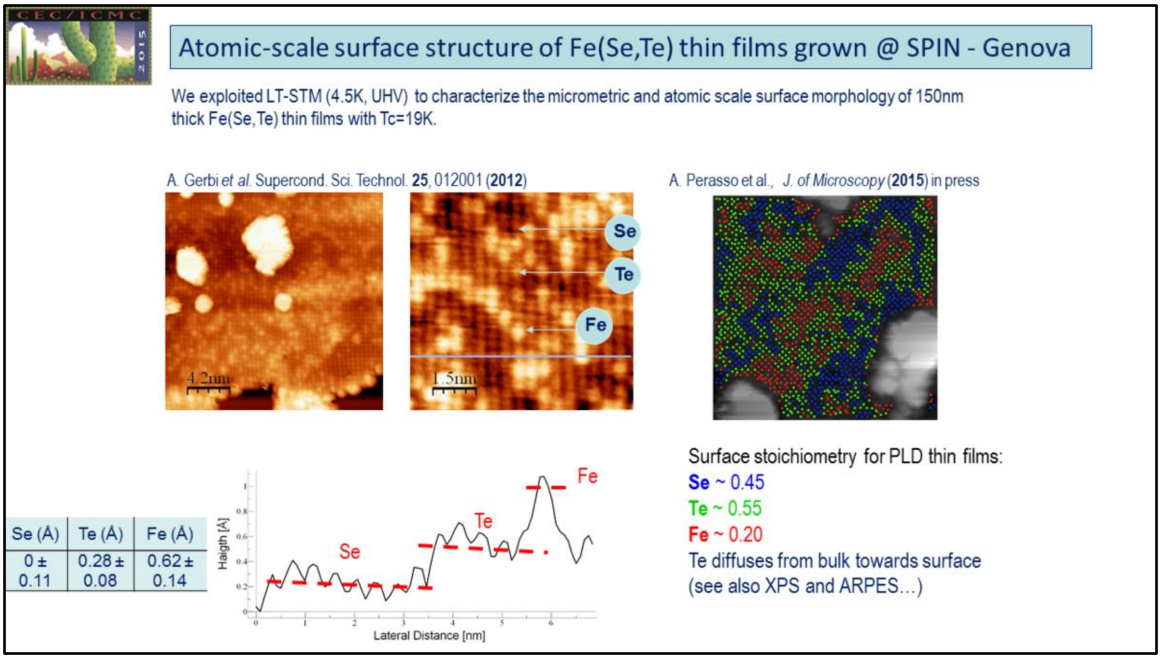
Films 200 nm thick have a higher Tc; however, the thickness is not the only parameter to determine the strain. Indeed, a non-negligible effect of the substrate is detectable as well. The measured in-plane cell parameter of the film plotted as a function of the substrate surface lattice parameter shows that the accumulated strain is affected by the substrate choice. In particular, we observe that on fluoride substrate the growth of the films proceeds in a very similar way as on oxide substrates.

For FeSe0:5Te0:5 thin films, the best Tc values—as high as 21 K—have been reported by our group. Very recently Y.Imai et al. were able, tuning the Se stoichiometry, to reach 23K.

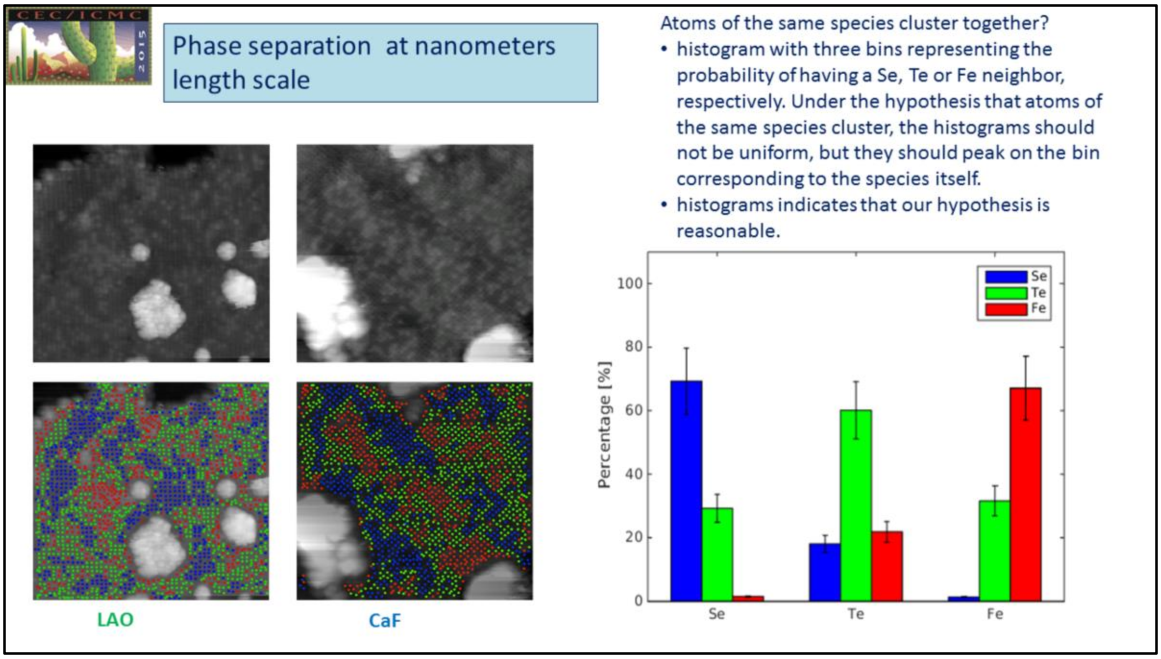
We attributed the high Tc values the compressive strain due to the Volmer–Weber growth mode which shrinks the c-axis, similarly to what happens through the application of an external pressure (see figure ref. Bellingeri APL 96 (2010)).



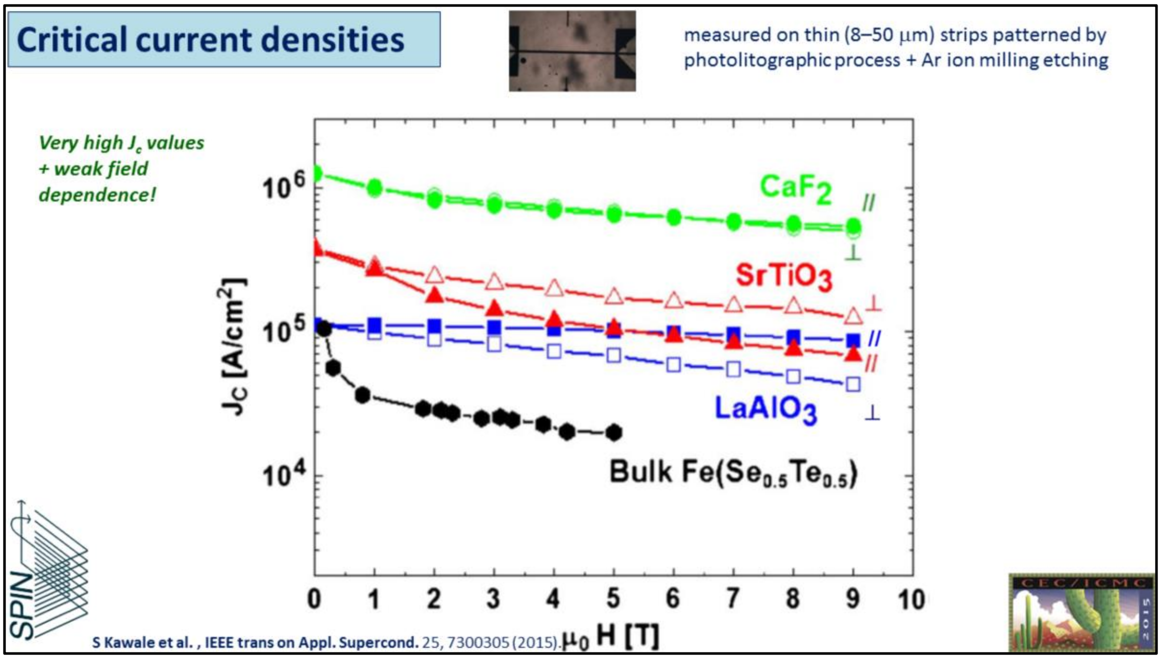
The high quality of the films on strontium titanate, lanthanum aluminate and calcium fluoride is confirmed by XRD analysis, showing that the FeSe0:5Te0:5 PbO-like tetragonal phase was obtained on all the substrates and no traces of elementary oxides or of the hexagonal phase were observed. In the teta–2teta scans (first column) only the (00l) reflections of the films and substrates are present, indicating the excellent purity of the phase and the optimum c-axis alignment of the growth. Typical omega scans on these reflections show rocking curves having a full width at half maximum (FWHM) in the range 0.2-2 degrees (second column). The c-axis values calculated by fitting the 00l peak positions are in the range 5.84–5.89 Å. These values are much reduced with respect to the bulk one(6.03 Å), but are in agreement with values previously reported. In order to establish the in-plane epitaxy of the growth, phi scans of the 101 reflection were performed. On all the substrates considered here, a single in-plane orientation is observed. On LaAlO3 and SrTiO3, the Fe(Se, Te) phase grows 'cube on cube' with the a axis parallel to the a axis of the substrate, whereas on CaF2, the film grows rotated by 45 degrees with respect to the a axis due to the good matching with half the diagonal (3.862). The FWHM of the phi scan peaks is approximately 0.8 degrees for samples deposited on LaAlO3 and CaF2 and 1.4 degrees for samples deposited on SrTiO3. In the 4th column STM images for the 3 samples are shown (the 45 degrees rotation in the case of CaF is confirmed). Despite the atomic resolution is reached the surface seems not uniform.



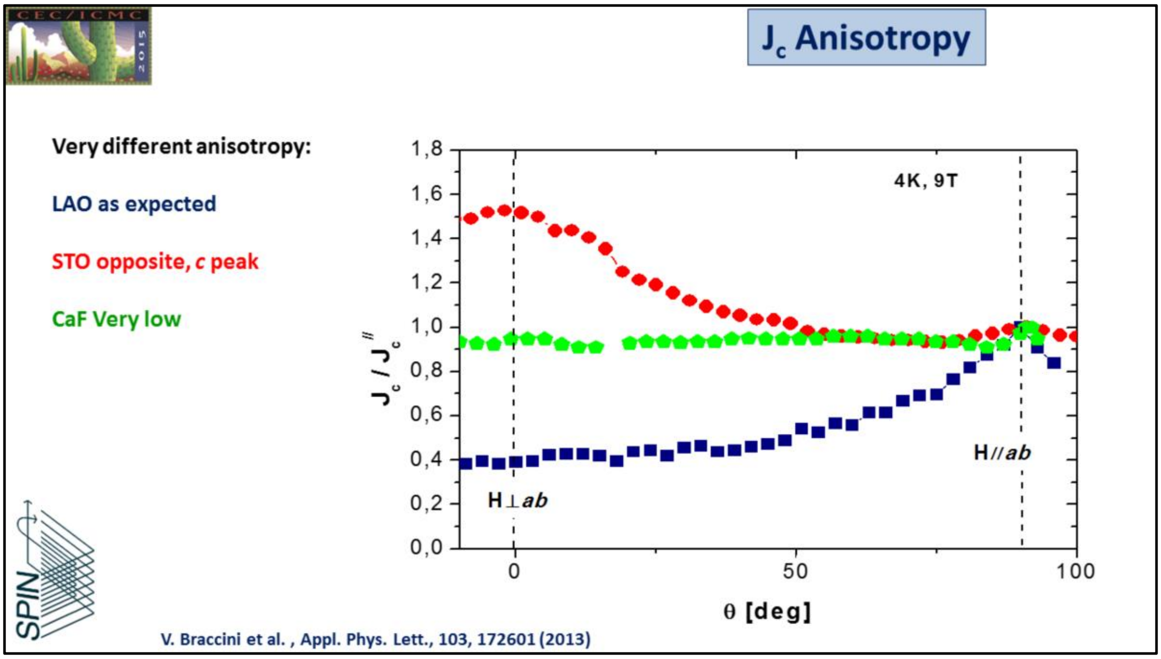
Atomic scale STM contrast, routinely achieved at low temperature, shows a squared lattice of ~3.8Å consistent with the arrangement of chalcogen atoms above the Fe plane in bulk single crystals. Small patches of bright and dark contrast highlight the occurrence of atomic scale surface disorder. In fact simple steric arguments, bulk structural refined data and direct STM imaging of cleaved single crystals, indicate that Se and Te atoms occupy distinct positions within the bulk unit cell, corresponding to a Se/Te splitting of 0.24Å along the z-coordinate axis. Moreover a small amount of excess Fe atoms is expected to be embedded in the chalcogen layer.
We estimate a Fe coverage of 6-7%, in reasonable agreement with the evaluation for excess interstitial Fe atoms from normal state magnetic measurements.
We developed an automatic method to recognize the different atomic species A.Perasso et al., J. of Microscopy (2015).



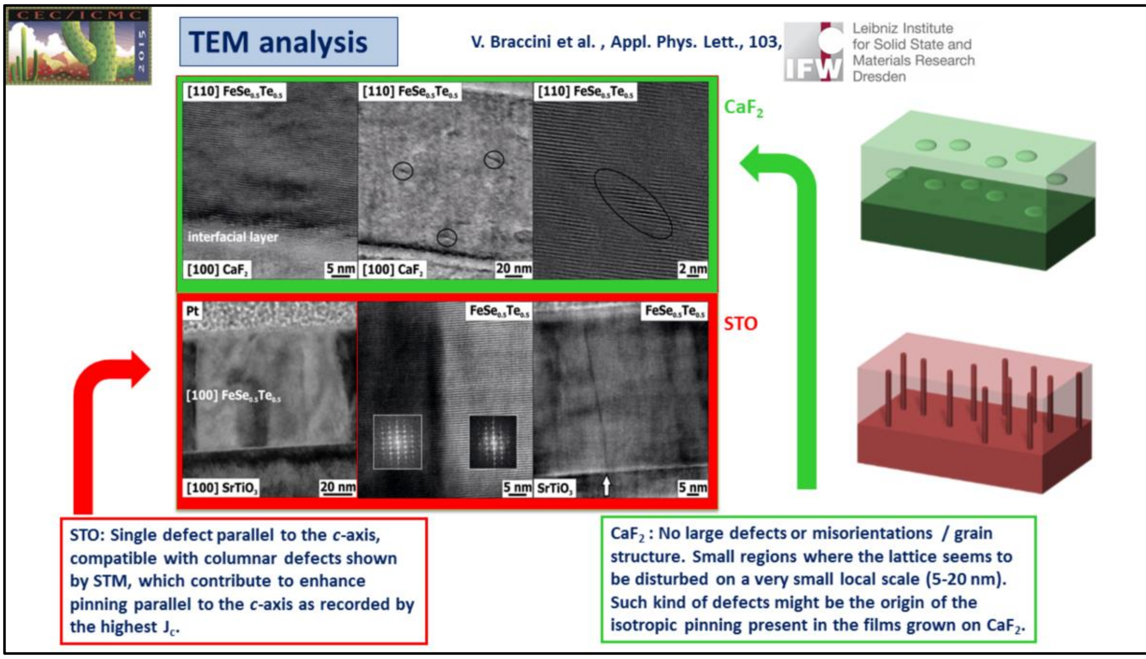
Atoms of the same species cluster together: histogram with three bins representing the probability of having a Se, Te or Fe neighbor, respectively. Under the hypothesis that atoms of the same species cluster, the histograms should not be uniform, but they should peak on the bin corresponding to the species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



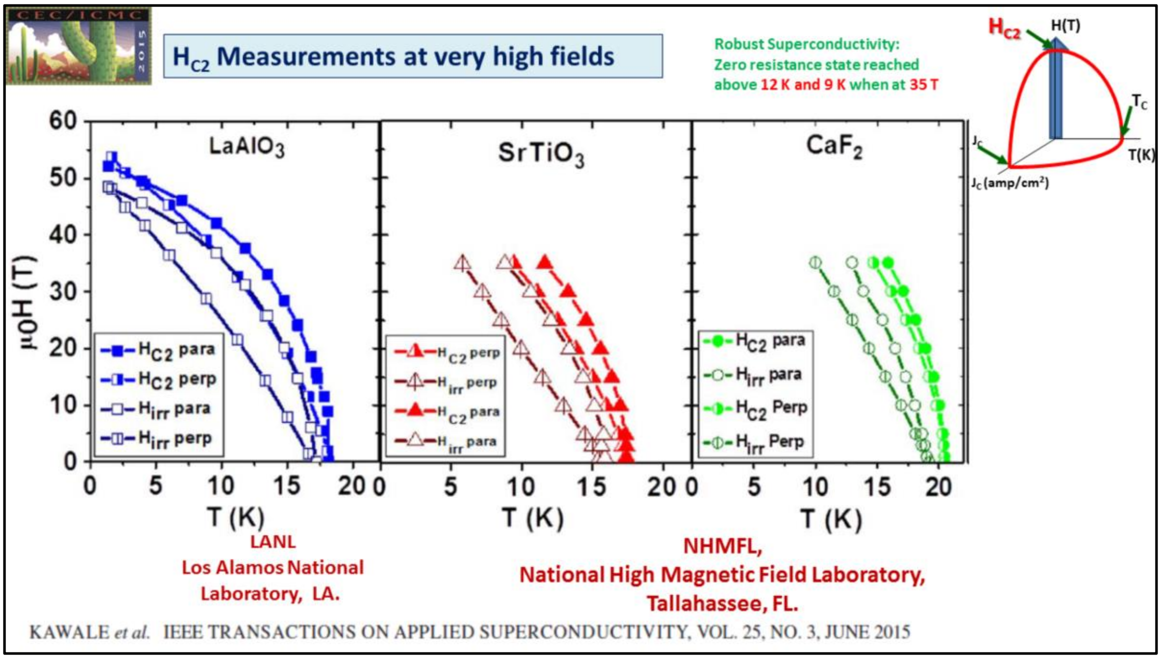
In the figure, critical current densities for thin films grown on STO, LAO and CAF. In the case of films on CAF, the anisotropy disappears and the Jc do not strongly depend on magnetic field applied. In the case of LAO and STO, the anisotropic behavior is different.



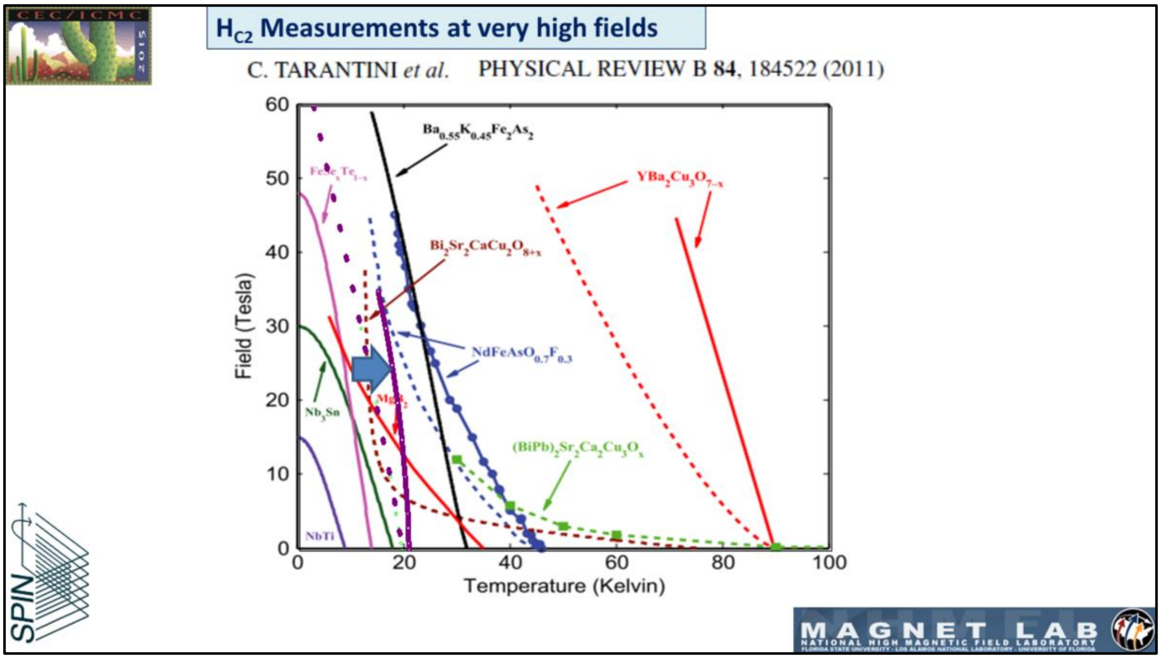
Comparison of the anisotropic behavior: in case of STO, a c-axis pinning is visible.



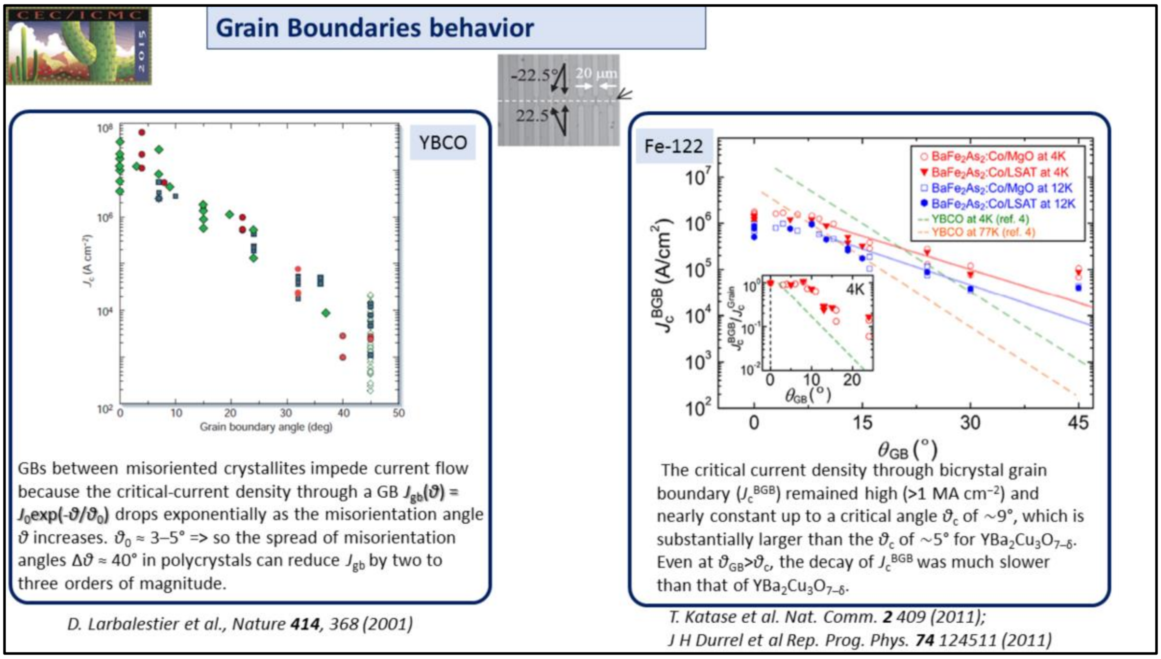
The figures show the microstructure of the sample deposited on CaF2 (green color). The microstructure of the film does not show any granular structure or large extended defects throughout the whole film; however, small regions appear where the lattice seems to be disturbed on a very local scale (5–20 nm) (second panel, marked in black). In the third panel of the figure, a higher resolution image is shown: at some point, the atoms start to deviate from the ideal plane while around the defects the lattice grows fine again. Such defects might originate from a different local stoichiometry between Se and Te – indeed FeTe and FeSe phases show quite different c axis values, 6.285 and 5.486Å, respectively. Given their size and distribution, it is likely that such kind of defects are the origin of the isotropic pinning present in films grown on CaF2. TEM analysis on a thin film deposited on SrTiO3 is reported (red color). Single defects parallel to the c-axis can be seen (third panel) which are compatible with the network of columnar defects parallel to the c-axis shown by STM previously hypothesized to contribute to enhance the pinning parallel to the c-axis.
In the cartoons a schematic view of the pinning centers in the two cases.



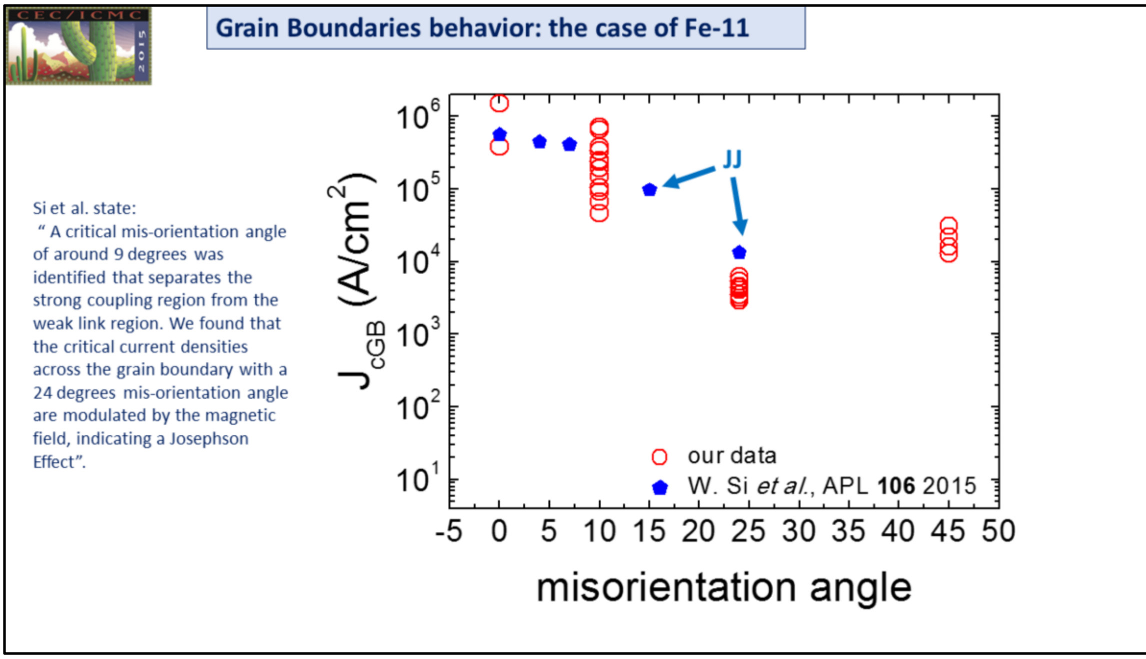
This set of measurements indicates the existence, at low temperature, of a large region in the H-T diagram where vortices are well pinned even at very large magnetic fields, hence the possibility of employing this superconductor at 4.2 K up to 40 T.



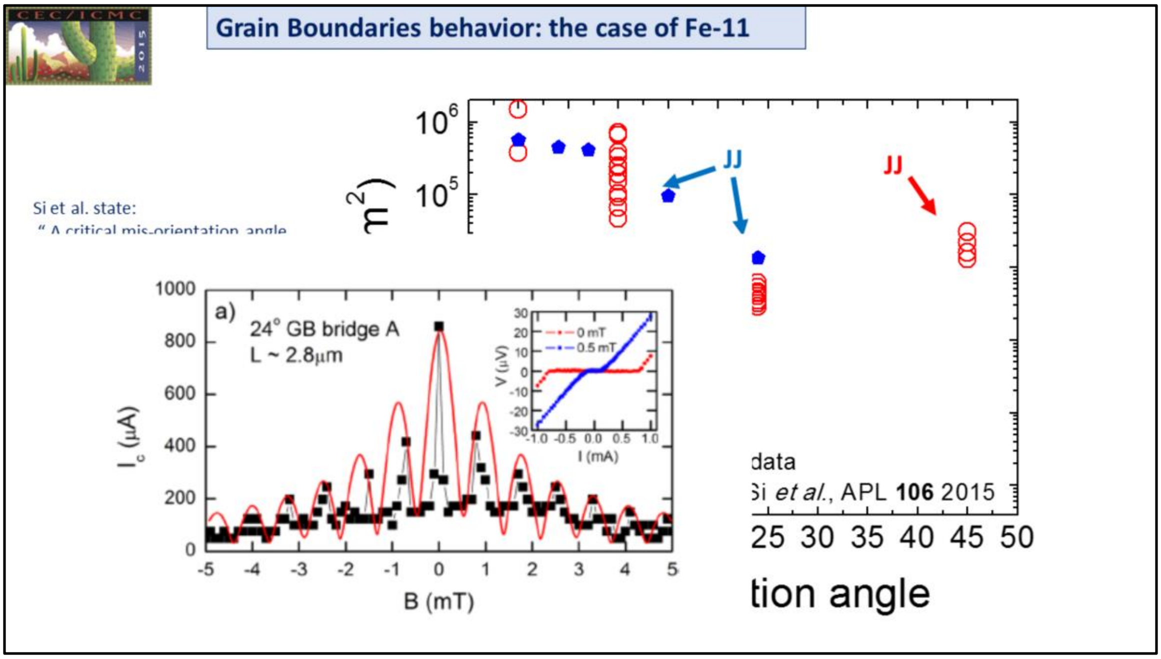
Here the critical fields for thin films are reported. We can observe a strain induced enhancement of Hc2 (ref. Tarantini).



GBs behavior for YBCO and Fe-122. See the comments on the figures.



GBs behavior in the case of Fe-11. In blue the data of Si el al. They 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". In red, our data. The agreement seems only partial.



They 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. On the contrary, we observe modulated behavior only with 45 degrees: next slide.

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Properties of high-angle Fe(Se,Te) bicrystal grain boundary junctions

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We report on the characterization of Fe(Se,Te) grain boundary junctions fabricated on a 45° tilt symmetric bicrystal substrate. The junctions show critical current densities of the order of 10^4 A/cm^2, showing a decay of about one order of magnitude with respect to the critical current density of the film, which is very promising for applications. The current-voltage characteristics show resistively shunted junction-like behaviors, with very low normal-state resistance values, typical of superconducting/normal-metal/superconducting junctions. The magnetic field dependences of the critical currents show the typical behaviors of large Josephson junctions. Nevertheless, the critical current is uniformly distributed across the barrier. We present a simplified theoretical model accounting for this effect. The small influence of faceting along the grain boundary on the electrical parameters may be consistent with an s-wave symmetry of the order parameter. © *2014 AIP Publishing LLC*. [http://dx.doi.org/10.1063/1.4871864]

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162601-3 Sarnelli *et al.*

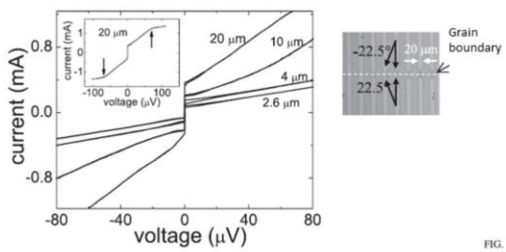


FIG. 2. Current-voltage characteristic of 45° [001] tilt bicrystal GBJs with different sizes measured at T = 4.2 K. In the inset, the IV characteristic of a junction 20 μm wide shows peaks due to flux motion.

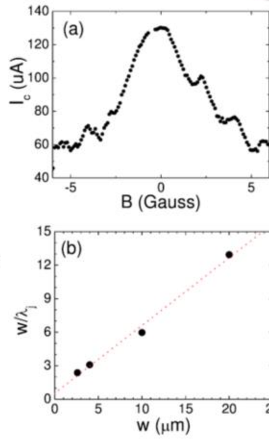
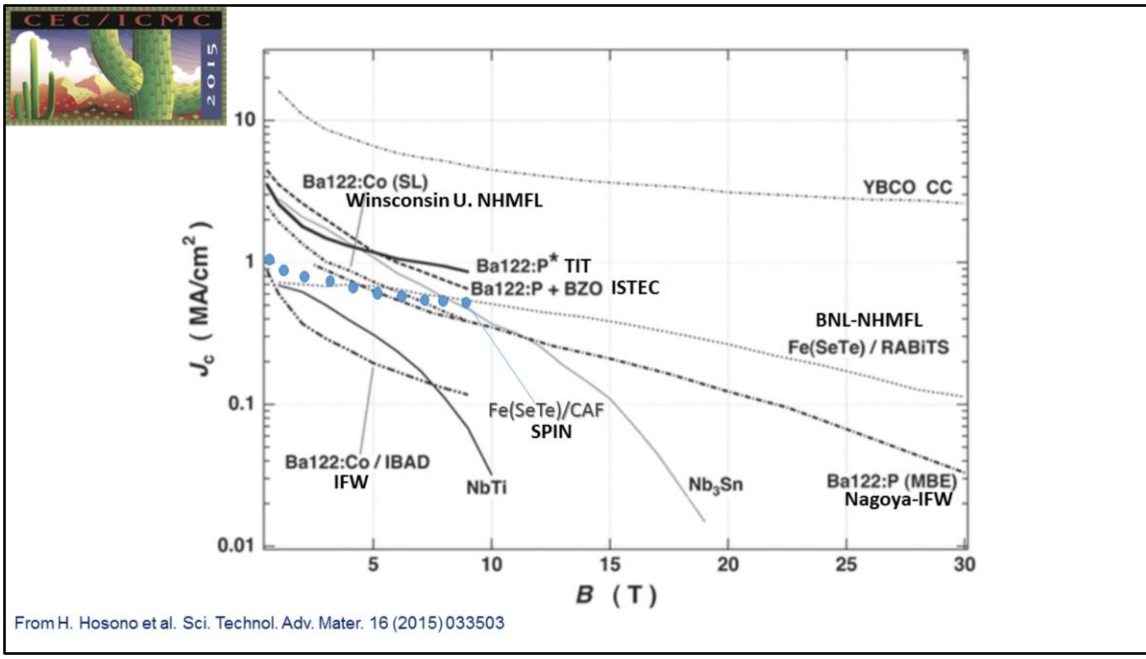


FIG. 3. (a) Magnetic field dependence of the critical current of 45° [001] Fe(Se,Te) symmetric bicrystal GBJ 2.6 μm wide at T = 4.2 K; (b) plot of the junction dimensions normalized to the Josephson penetration depth as a function of the junction width. The dashed curve is a linear fit of data.

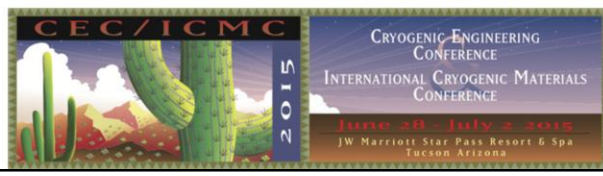
Therefore we conclude that more investigations are necessary to completly define the GBs behavior in the Fe-11 family.



Here a comparison (taken from Hosono et al. Review) of Jc of different Coated Conductors versus magnetic field, at liquid helium temperature. 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. In blue, our results.

CONCLUSION

- ✓Possibility of tuning the superconducting properties in Fe-chalcogenide by thin film technology.
- ✓T_c up to 23 K, much higher than the bulk value, appreciable critical current density values up to 30 T, enhanced H_{c2} (greater than 50T).
- ✓Promising prospects of Fe-chalcogenide for low temperature (≤ 4.2 K) and very high field (≥ 25 T) applications.
- ✓Search for low cost metallic oriented templates without oxide buffer layers



In conclusion: in Fe-11 it is possible to enhance, with strain, critical fields and critical temperatures. In addition, the pinning mechanisms also appears tunable.
It can be interesting to develop low cost metallic oriented template, without expensive oxide buffer layers: here the GBs mis-orientation appears less critical in respect to HTS.