



UNIVERSITÀ DEGLI STUDI DI SALERNO



Dipartimento di
Fisica E.R. Caianiello

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Scanning Probe Microscopy studies
of magnetically and electrically coupled
Superconductor/Ferromagnet systems

SPnM Laboratory

Scanning Probe Microscopy and nano-Matter



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We are here!



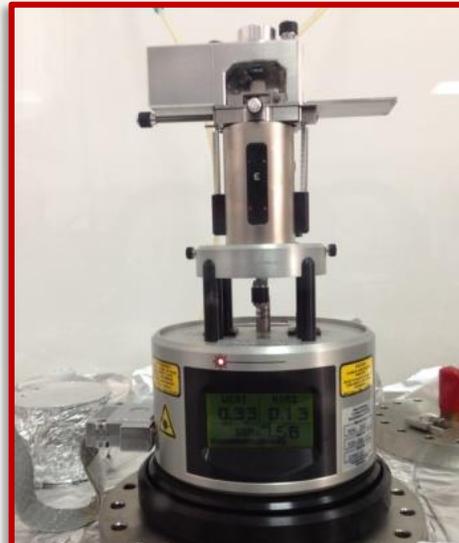
Top view of UniSa campus

Facilities at the SPnM Laboratory

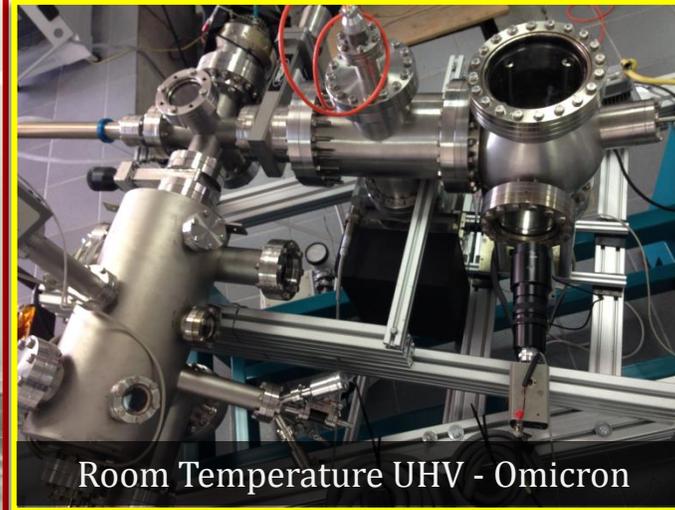


Sputtering DC

Sputtering DC equipped with 3 cathodes

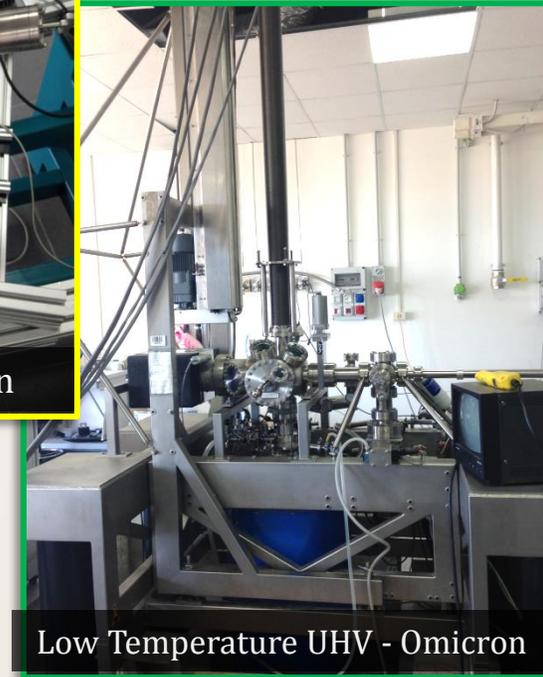


Mul



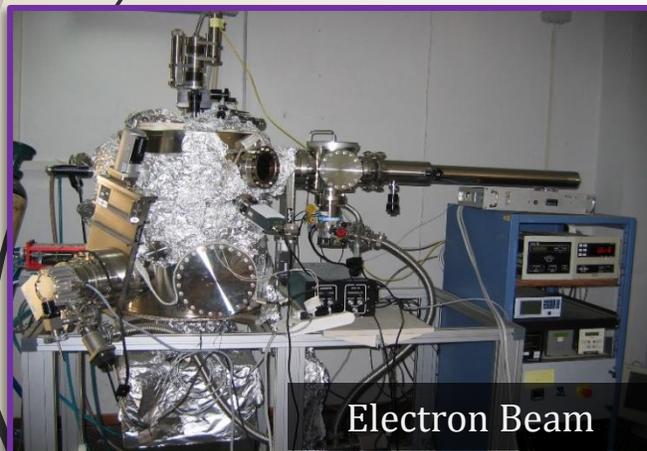
Room Temperature UHV - Omicron

Room Temperature-UHV Atomic Force Microscope/ Scanning Tunneling Microscope



Low Temperature UHV - Omicron

Cryogenic-UHV Atomic Force Microscope/ Scanning Tunneling Microscope, equipped with 7T out-of-plane superconducting magnet. Base temperature: 5K



Electron Beam

Electron-beam gun equipped with 5 crucibles



NanoWizard III - JPK

Room Temperature and Pressure Atomic Force Microscopes.

The team of the SPnM Laboratory



spnm.fisica.unisa.it



Fabrizio Bobba
Associate Professor



Anna Maria Cucolo
Full Professor



Matilde Sublimi
Post Doc



Cinzia Di Giorgio
Post Doc



Domenico D'Agostino
Ph.D. Student



Ofelia Durante
Ph.D. Student

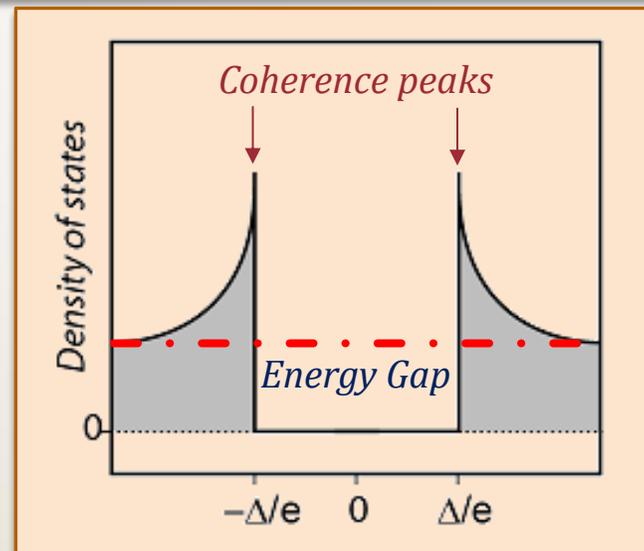
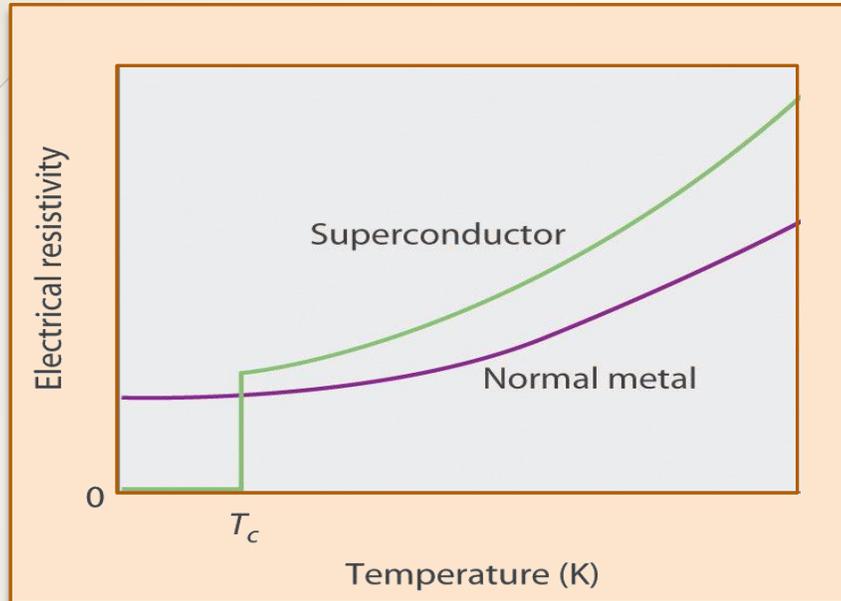
The activities of the SPnM Laboratory

- ▶ Study of **superconductivity** in S/F systems and novel superconductors by means of low-temperature UHV MFM and STM
- ▶ Study of the electric and electro-mechanical properties of **thin films** and **nano-structured semiconductors** by means of AFM, KPFM, PFM, C-AFM
- ▶ Study of **amyloids** and their aggregation on DOPC and DOPC/DHA bilayers by means of AFM and Quantitative-AFM
- ▶ Study of piezoelectric properties of ferroelectric **polymers** by PFM; Study of antimicrobial activity of polymers by AFM and Quantitative-AFM
- ▶ Study of electrical properties of **2D materials** and their dependence on stress/strain by means of AFM, Quantitative-AFM, C-AFM and KPFM
- ▶ Study of morphological and elastic properties of **mirror prototypes** for gravitational waves detectors by means of AFM, Quantitative-AFM, Force Modulation - AFM

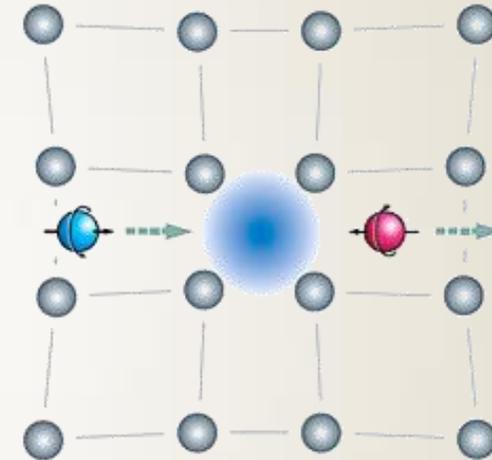
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Brief refresh on superconductivity



- ▶ Zero Electrical Resistance -> No dissipation due to the formation of the «Cooper pair»

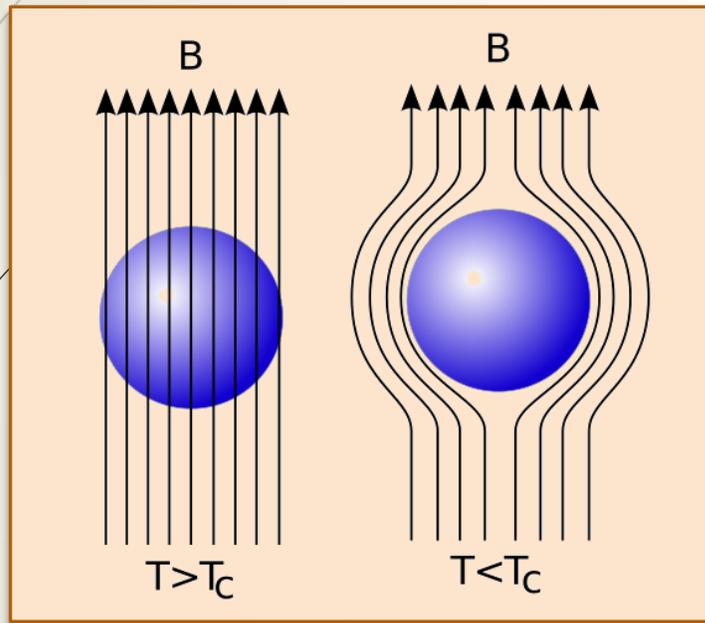


- ▶ Peculiar density of states, characterized by:
 - ▶ Energy gap (few meV)
 - ▶ Coherence peaks
- ▶ The coherence length ξ measures the size of the cooper pair

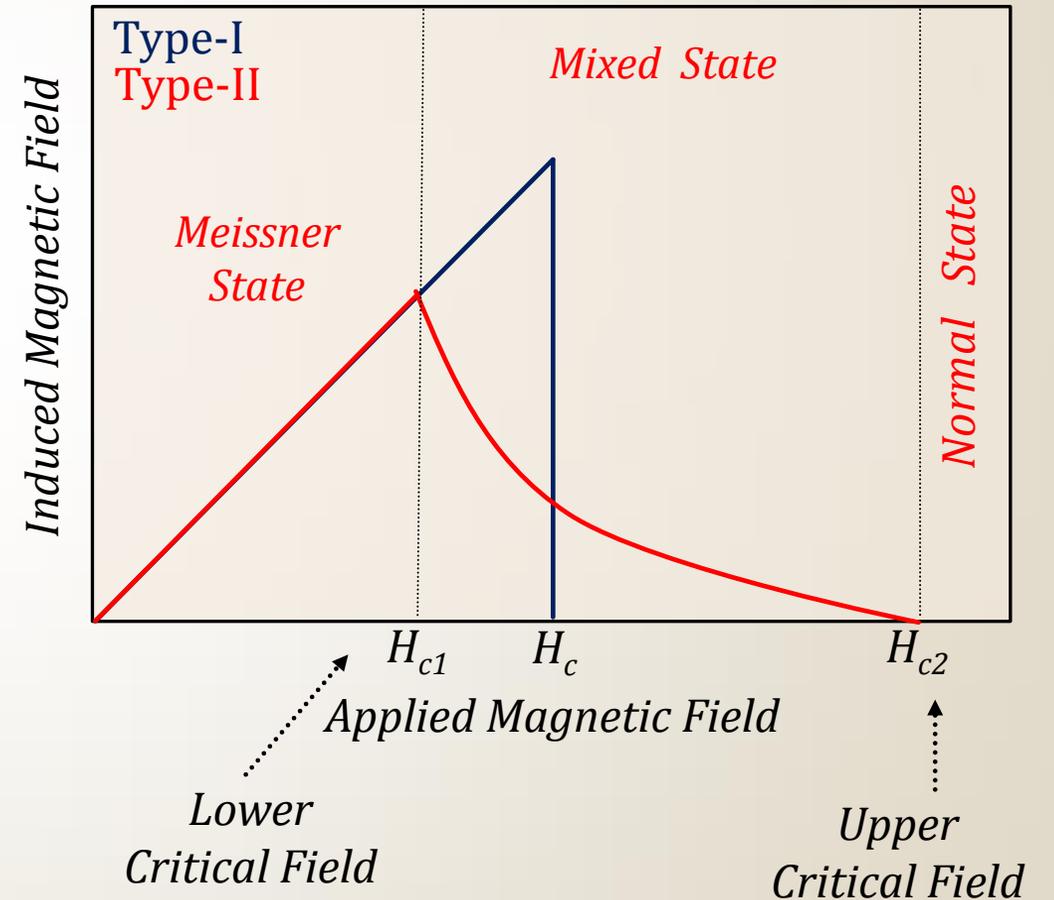
$$\xi_{BCS} = \frac{\hbar v_F}{\pi \Delta}$$

Brief refresh on superconductivity

Meissner Effect

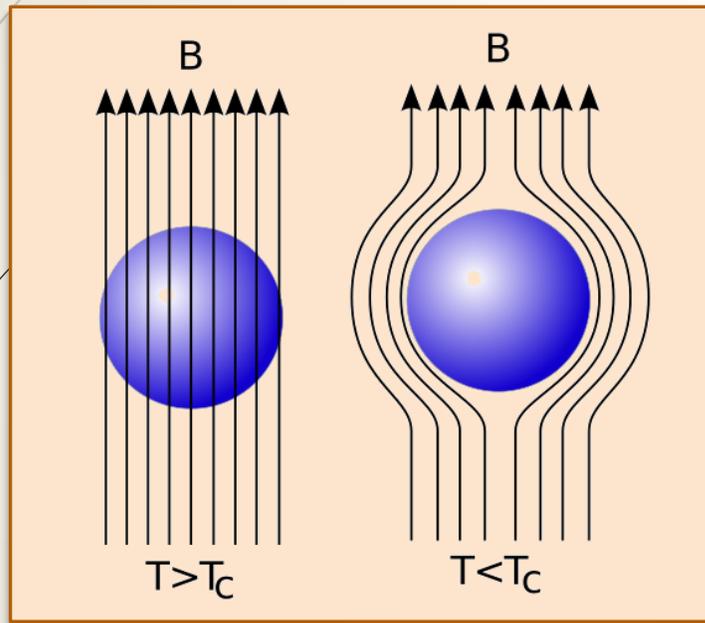


- Perfect Diamagnetism in Meissner state;
- 1st order transition in Type-I;
- 2nd order transition in Type II



Brief refresh on superconductivity

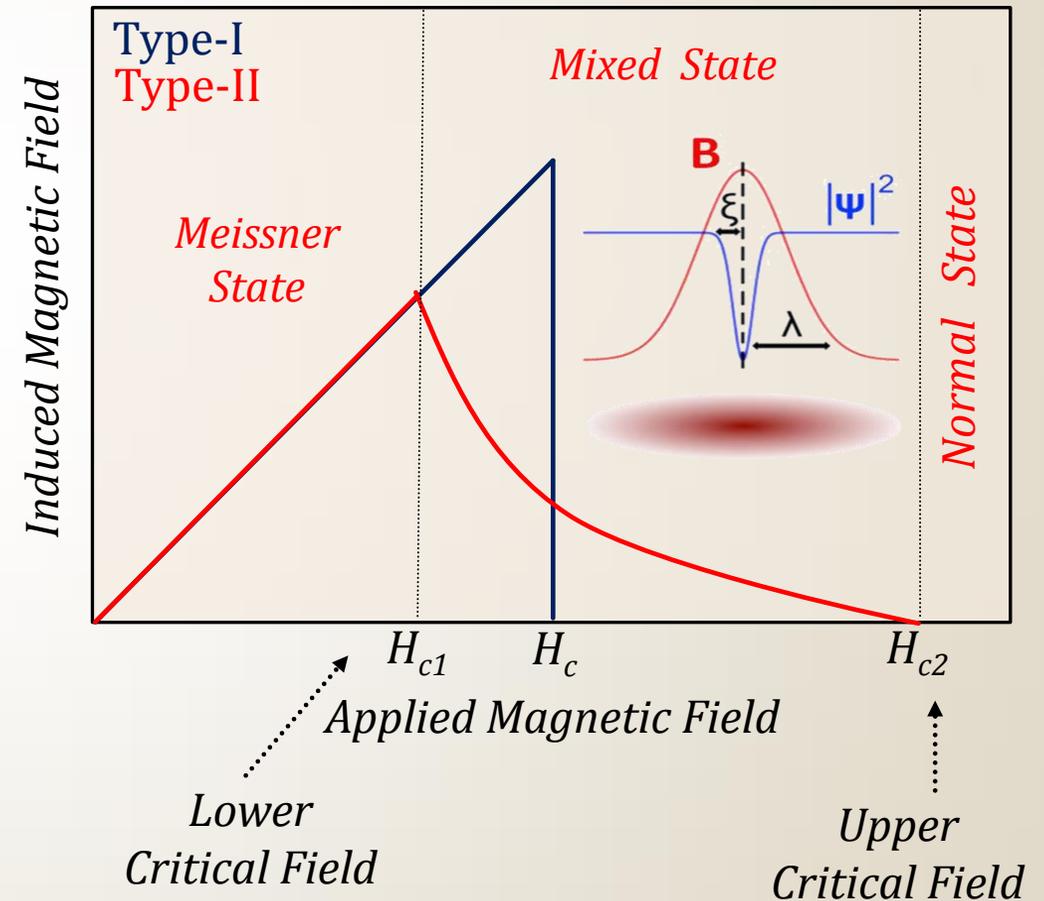
Meissner Effect



A superconducting vortex always supports a **magnetic quantum flux**:

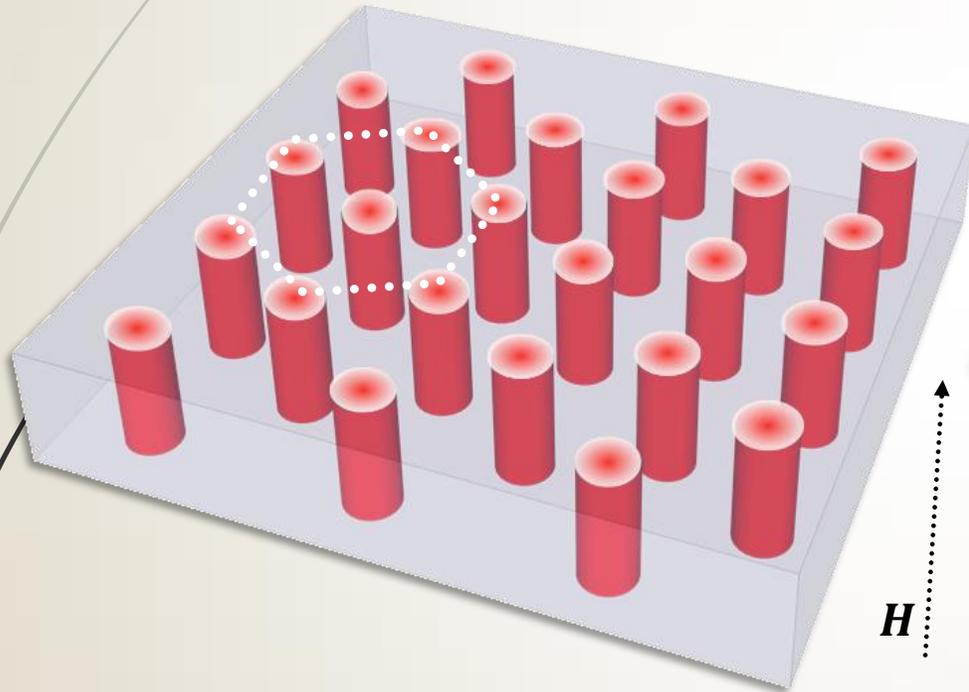
$$\Phi_0 = \frac{hc}{2e} = 2.07 \times 10^{-7} \text{ Gcm}^2$$

- Perfect Diamagnetism in Meissner state;
- 1st order transition in Type-I;
- 2nd order transition in Type II



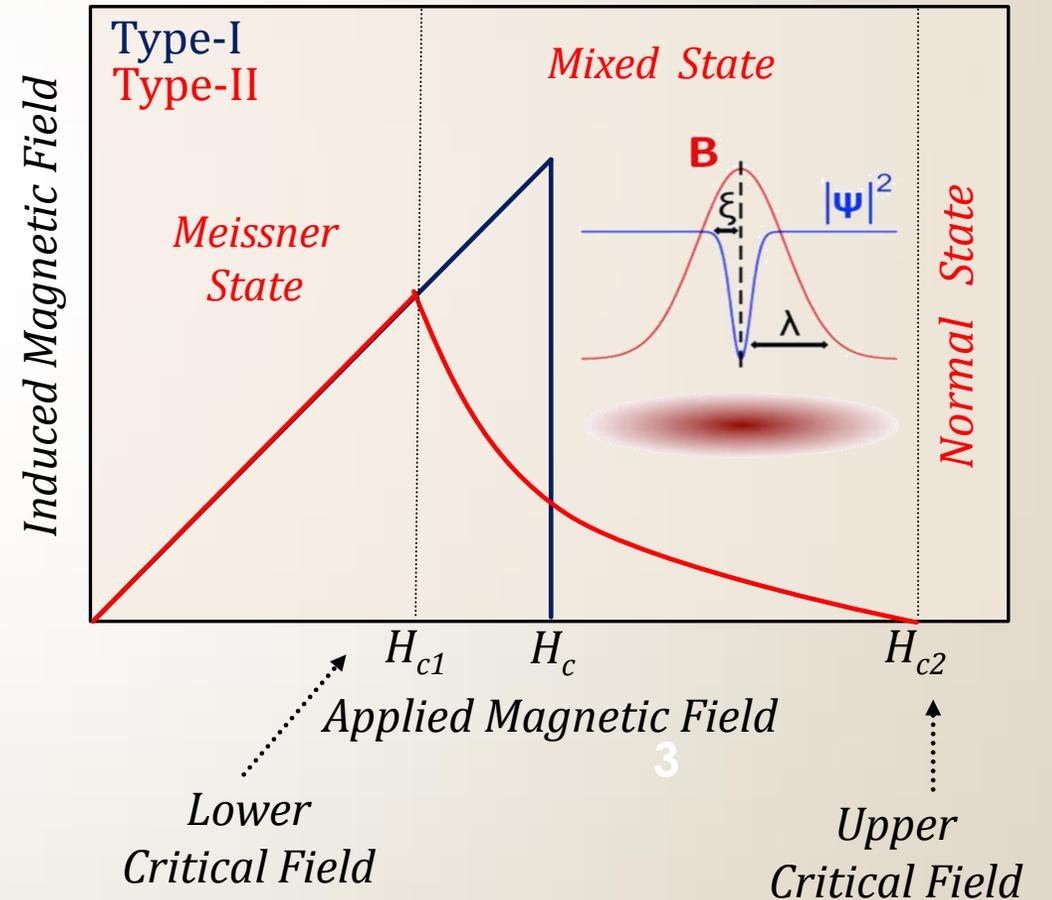
Brief refresh on superconductivity

Abrikosov hexagonal lattice



$$H = \frac{2 \Phi_0}{\sqrt{3} d^2}$$

- Perfect Diamagnetism in Meissner state;
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- 2nd order transition in Type II



Scanning Probe Microscopy techniques

- ▶ Scanning Tunneling Microscopy and Spectroscopy

Quantum Tunneling Current

$$I \sim e^{-kz}$$

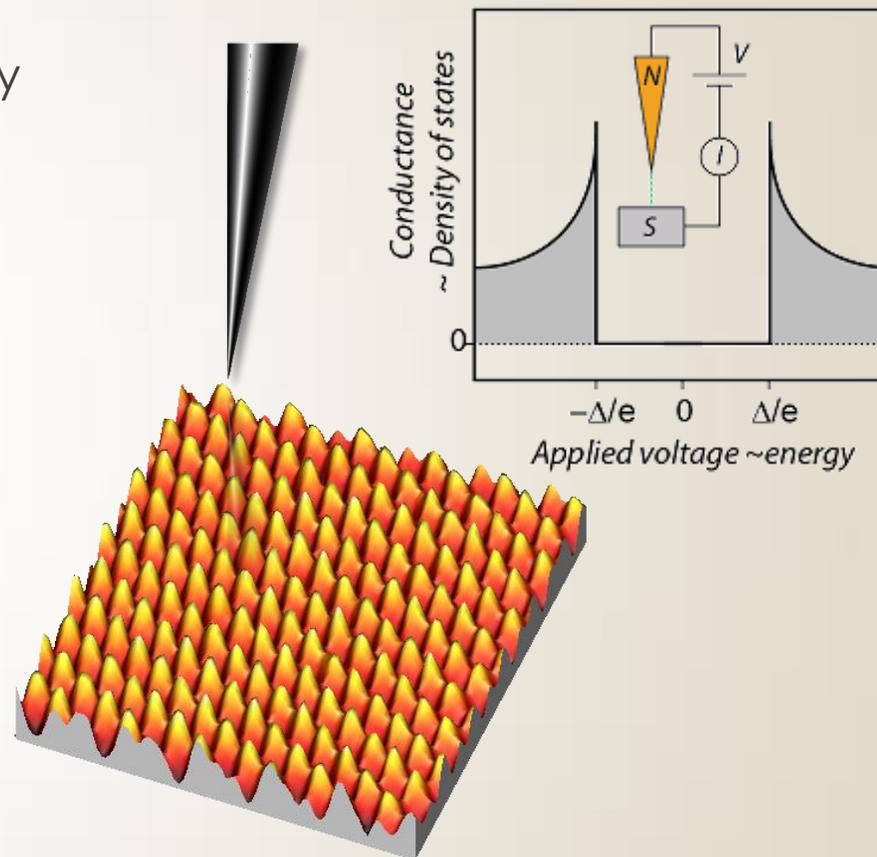
$$I \sim \frac{4\pi e}{\hbar} |M|^2 \rho_t(0) \int_{-eV}^0 \rho_s(\varepsilon) d\varepsilon \quad \longrightarrow \quad \frac{dI}{dV} \propto \rho_s(\varepsilon)$$

T=0K - Low Temperature approximation

- ▶ Magnetic Force Microscopy

Magnetostatic Interaction

$$df = \frac{f_0}{2k} \frac{dF_z}{dz}$$



Scanning Probe Microscopy techniques

- ▶ Scanning Tunneling Microscopy and Spectroscopy
Quantum Tunneling Current

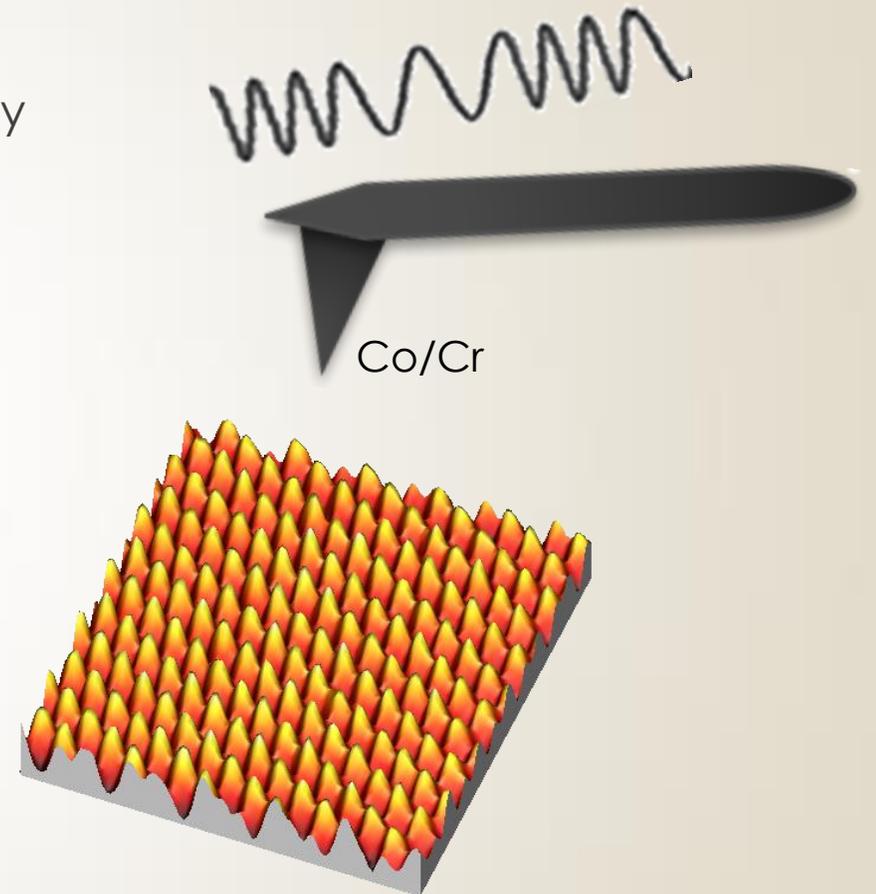
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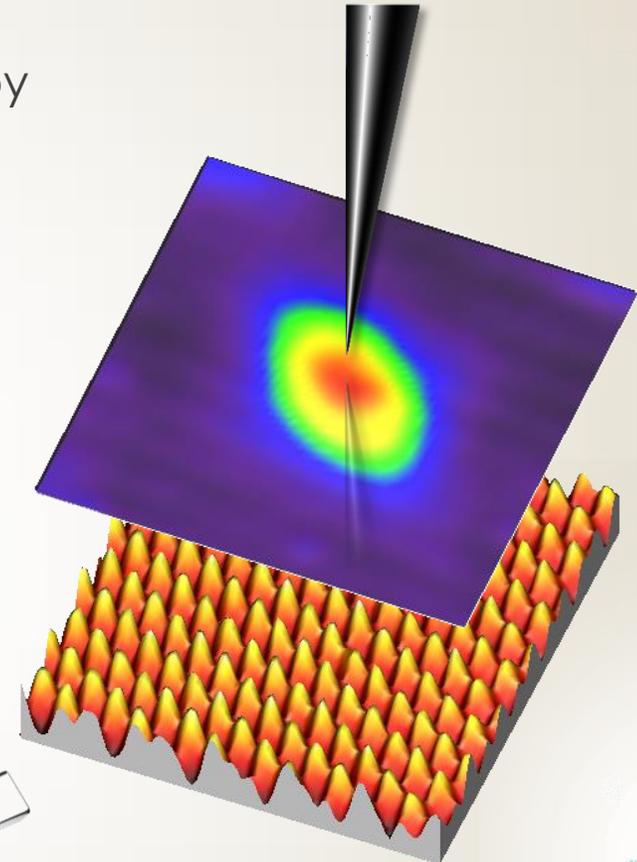
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T=0K - Low Temperature approximation

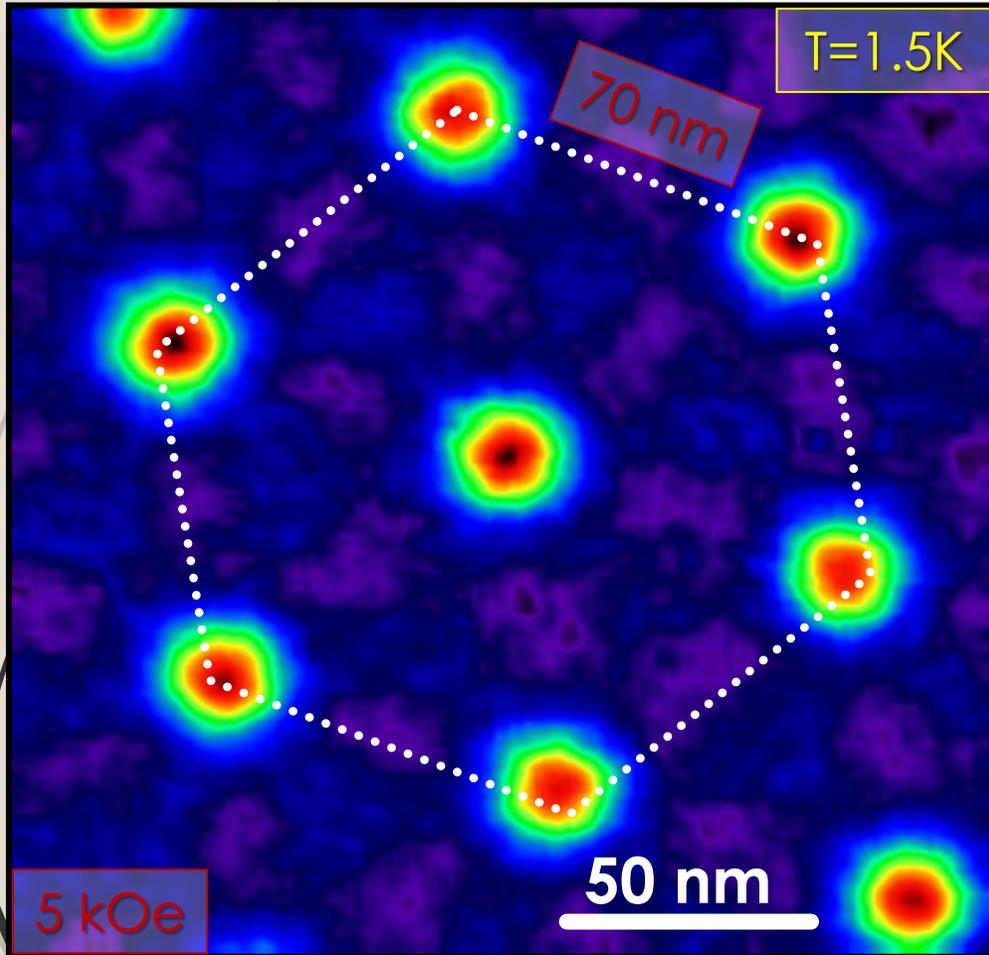
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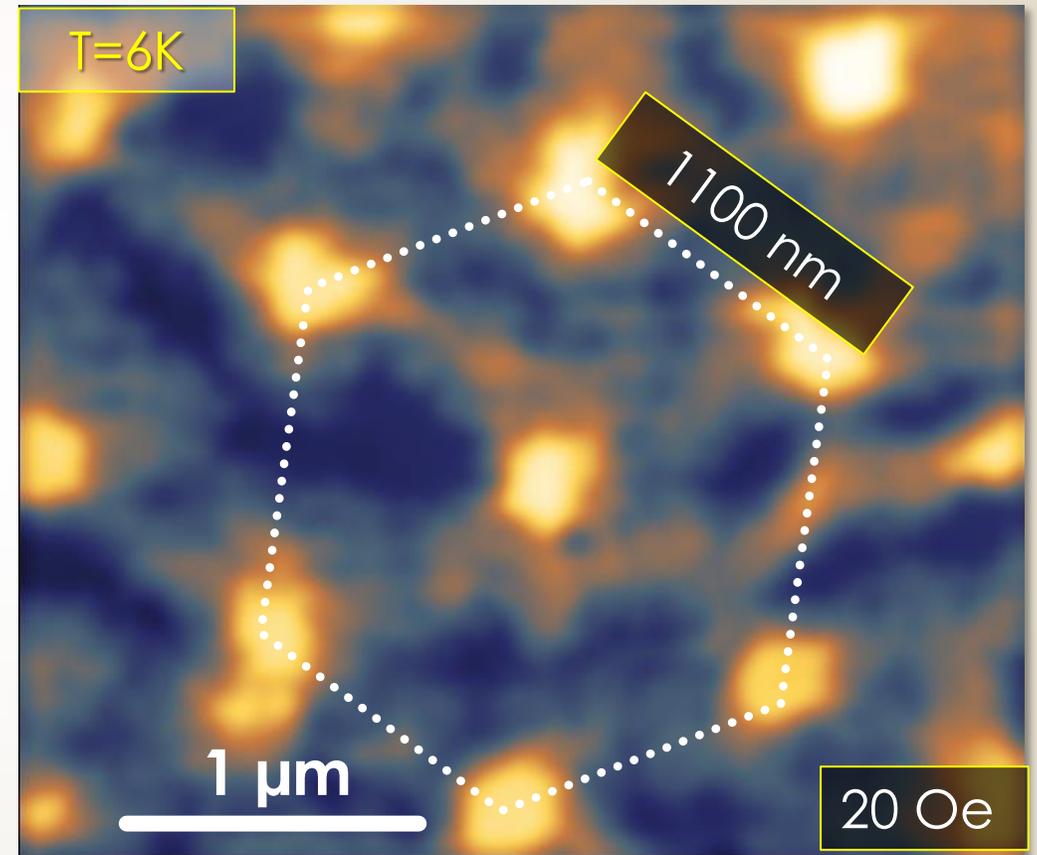


Scanning Probe Microscopy techniques

Low Temperature – STM on NbSe₂



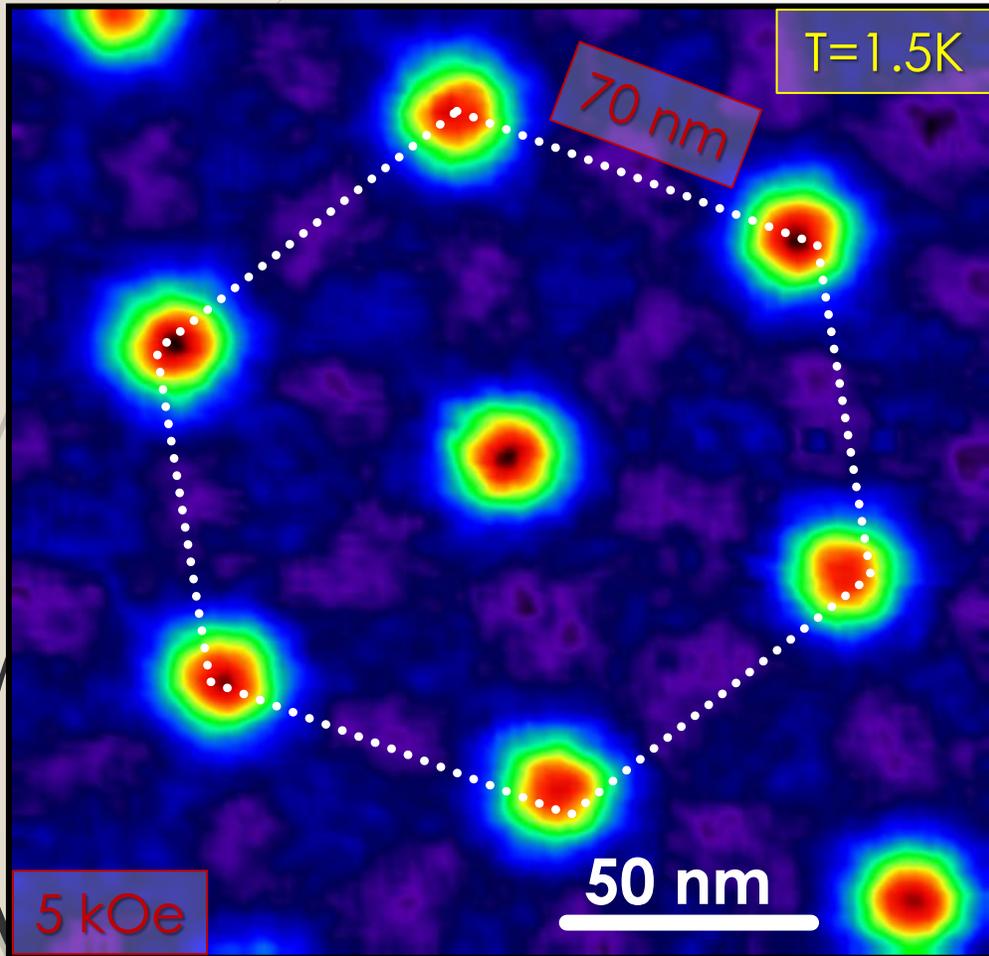
Low Temperature - MFM on NbSe₂



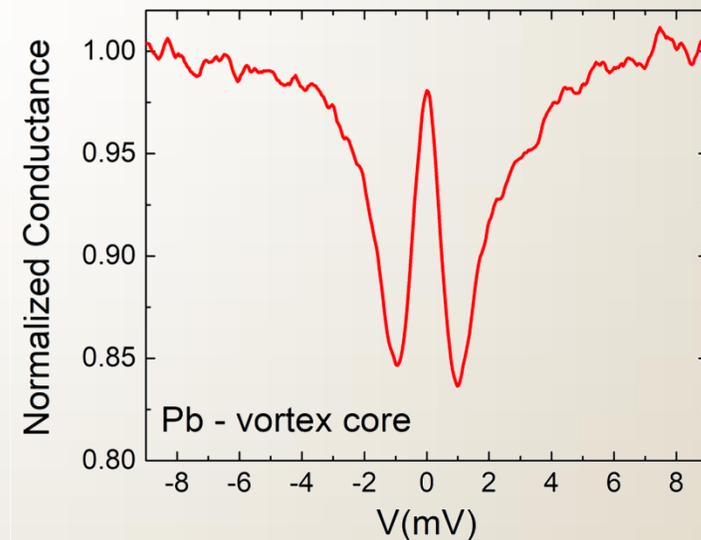
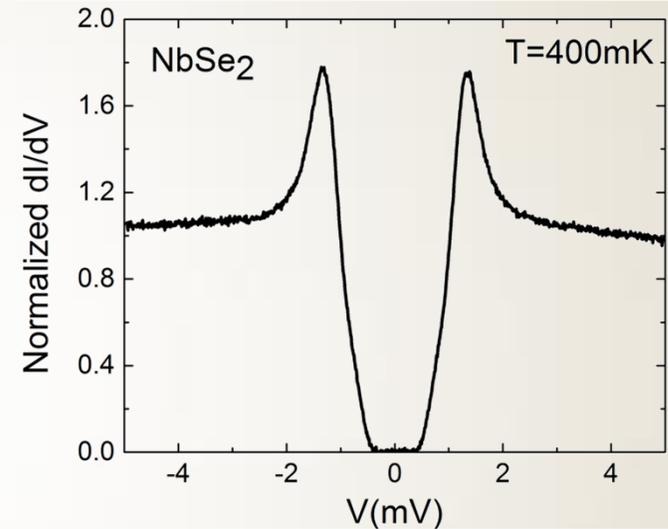
$$H = \frac{2 \Phi_0}{\sqrt{3} d^2} \quad \xi \approx 50 \text{ \AA} \quad \lambda \approx 200 \text{ nm}$$

Scanning Probe Microscopy techniques

Low Temperature – STM on NbSe₂



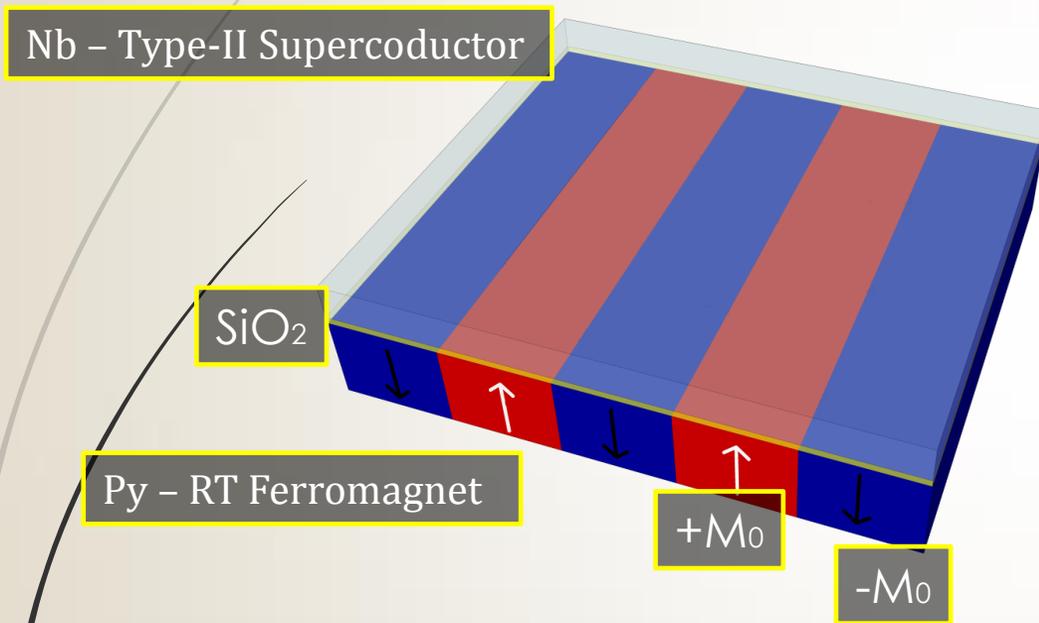
Low Temperature - STS



Superconductor/Ferromagnet systems

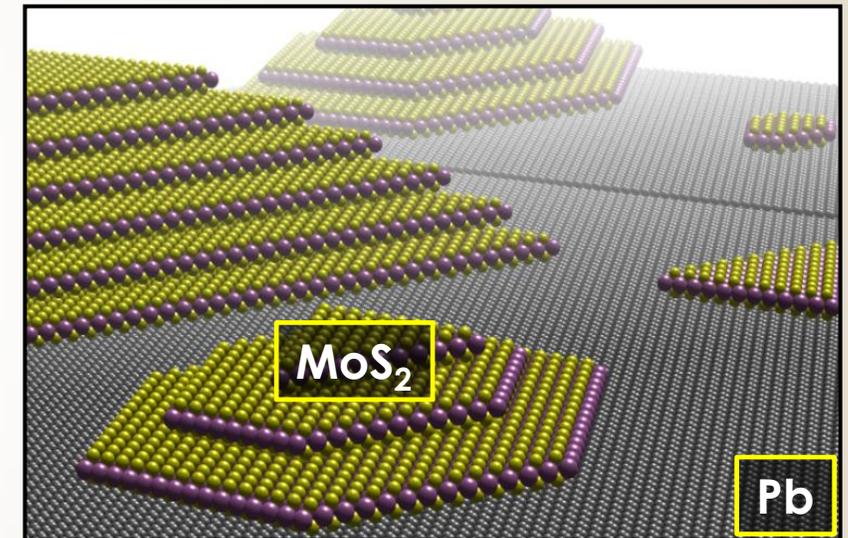
We used Low-Temperature MFM and STM/STS to investigate the following Superconductor/Ferromagnet systems:

Magnetically coupled Nb/Py ($\text{Ni}_{80}\text{Fe}_{20}$)



- Nb is a type-II superconductor with a T_c of 9.2K
- Py has stripe-configuration of magnetic domains
- A thin layer of SiO₂ electrically decouples Nb and Py

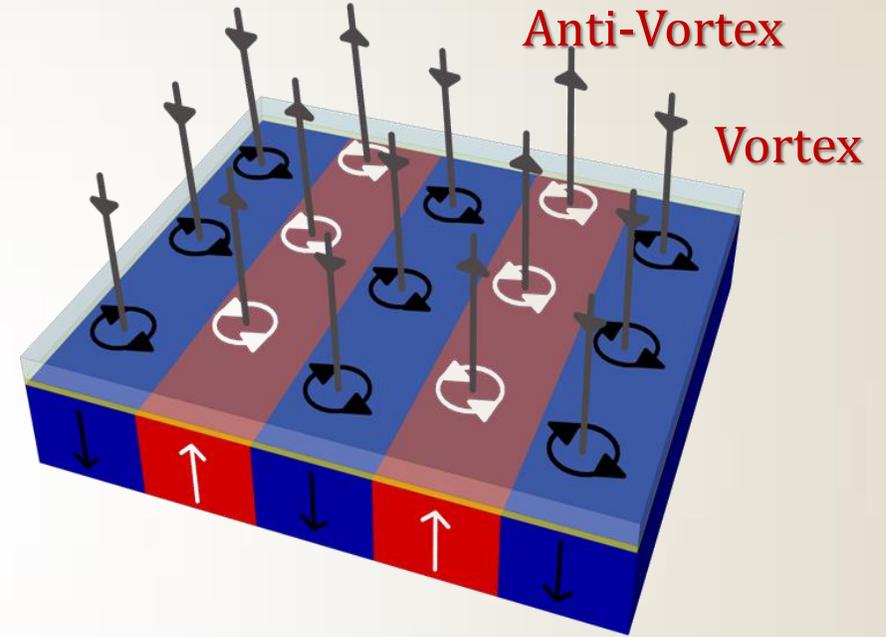
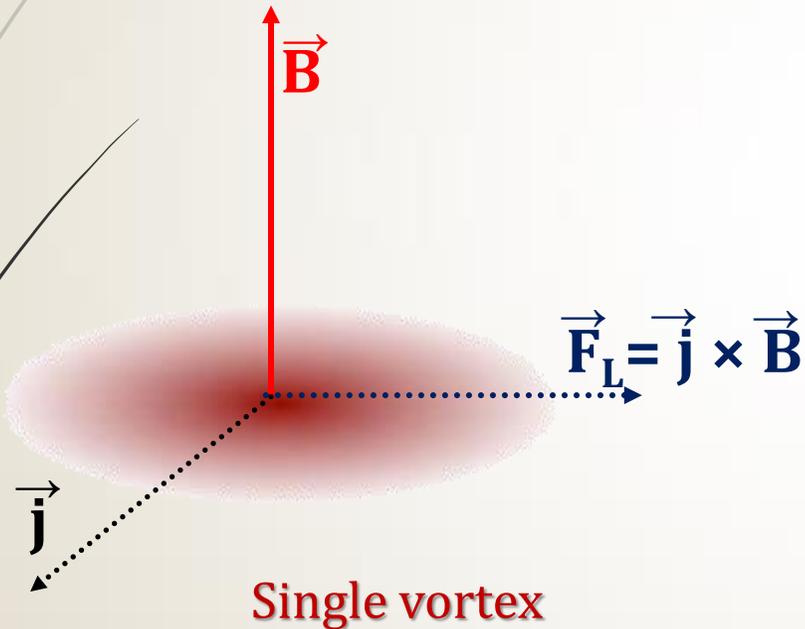
Electrically coupled MoS₂/Pb



- Pb is a type-II superconductor with a T_c of 7K
- MoS₂ is generally a semiconductor, but a non-zero magnetization is predicted to appear at the edge states of mono-layer islands

Magnetically coupled S/F systems - motivations

Vortices are forced into motion by the Lorentz Force causing dissipation.

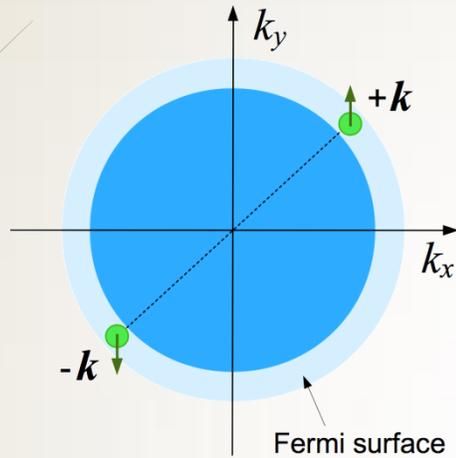


- Vortex motion across the stripes is prohibited
- Vortex motion along the stripe is strongly suppressed

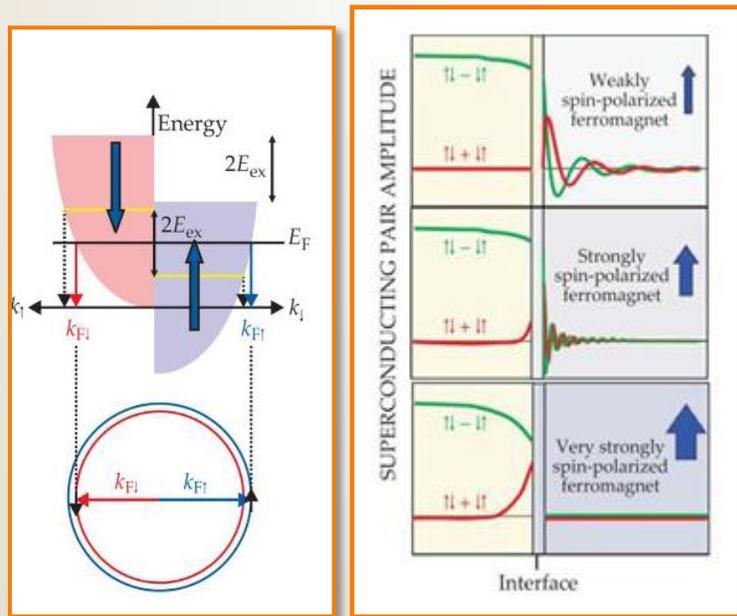


Vortex pinning is increased³
Dissipation free regime is kept at higher magnetic field

Electrically coupled S/F systems - motivations



- The BCS theory predicts the existence of a sc condensate of Cooper pairs, with zero-center of mass momentum, opposite spin (zero total spin) and charge $2e$



- The exchange field of F splits the energy bands for up and down spin, thus killing the Cooper pair (which has opposite spins)
- In order to survive the Cooper pair can acquire equal spins, giving rise to **supercurrents totally spin-polarized**

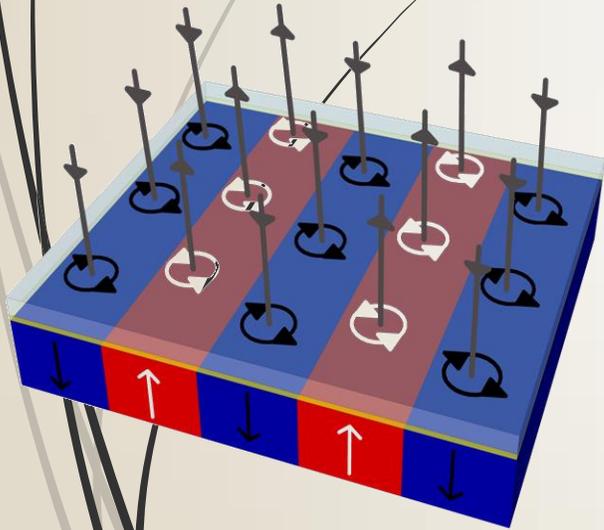


Magnetically coupled S/F systems - motivations

We studied **magnetically coupled S/F** systems in strong collaboration with Physics Department of Temple University (Philadelphia, USA)

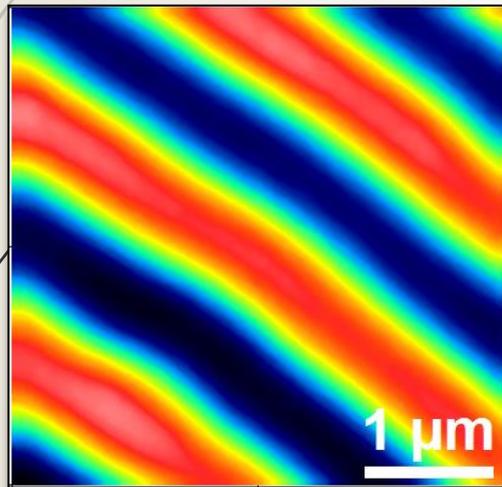
Our activity can be wrapped up in three main results:

- ▶ Nucleation of vortices in S/F;
- ▶ Study of enhanced pinning effect due to magnetic defects;
- ▶ Advances in Quantitative-MFM.



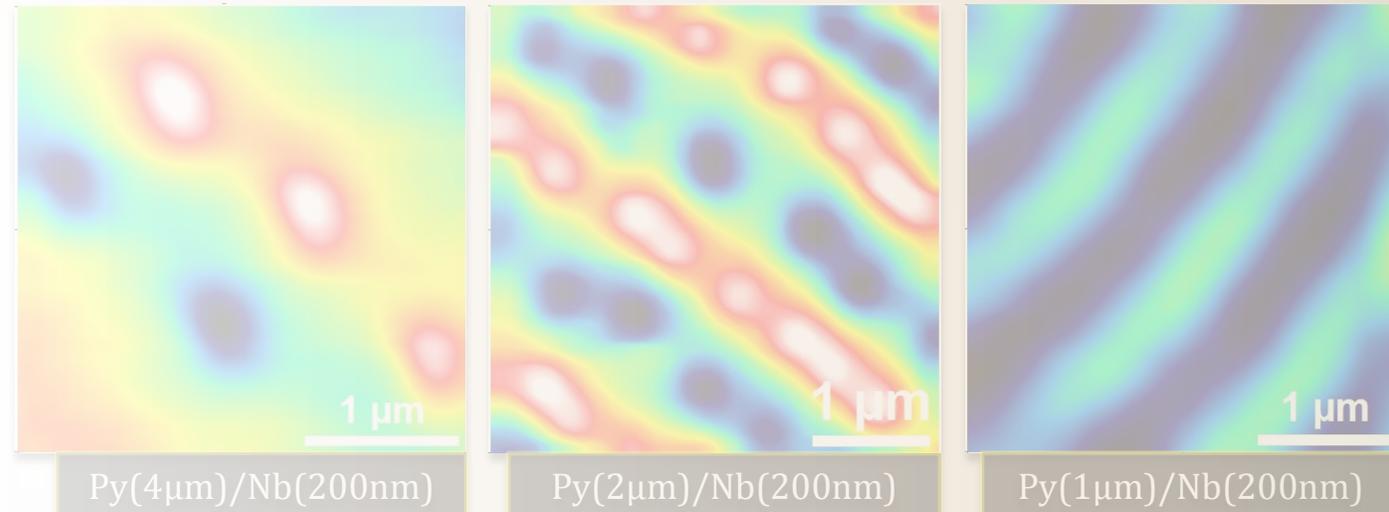
Vortex Nucleation

Typical MFM image of Py at $T=13\text{K}$ (above $\text{Nb } T_c$)



- Peculiar stripe-arrangement of magnetic domains
- Stripe width proportional to Py thickness

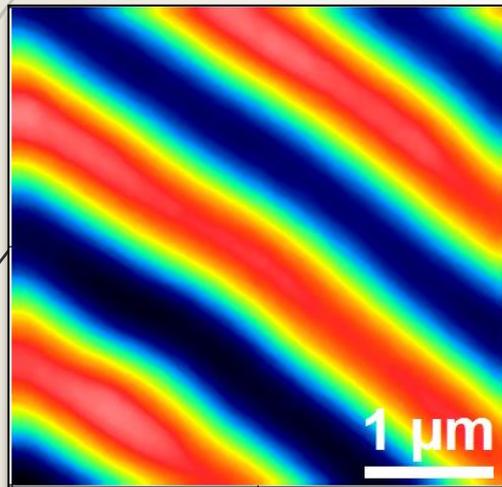
Zero field cooling – $T=6\text{K}$ (below $\text{Nb } T_c$)



- Spontaneous Vortex-Antivortex pairs are formed in Py(4μm)/Nb(200nm) and Py(2μm)/Nb(200nm) whereas no vortex occurrence is recorded in Py(1μm)/Nb(200nm).
- Opposite polarized vortices correctly lie on proper magnetized stripes.

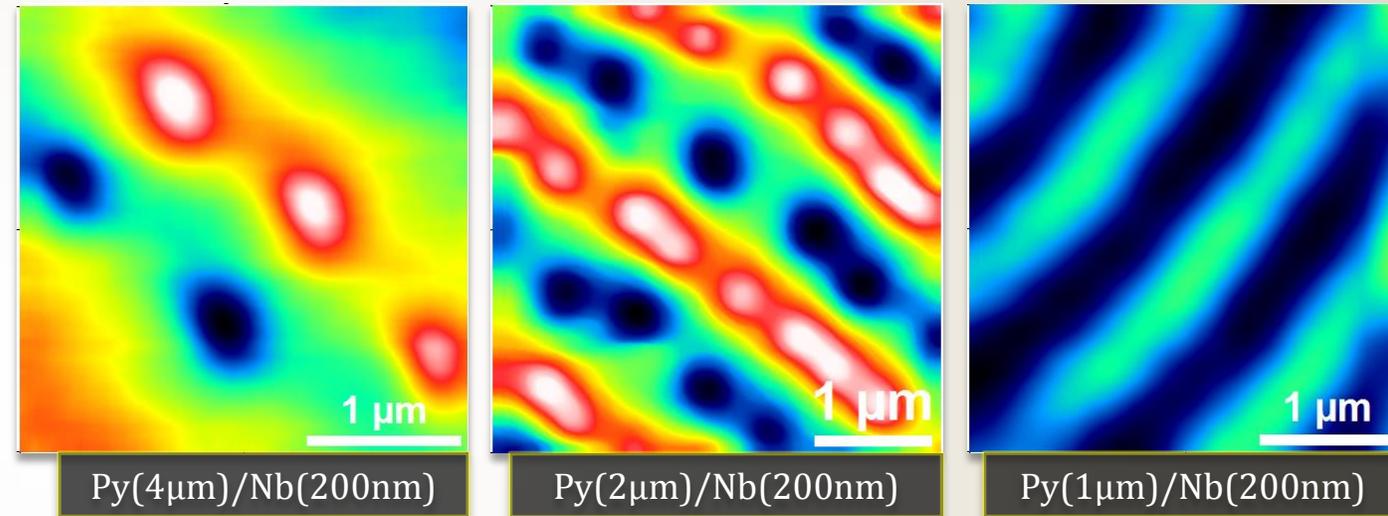
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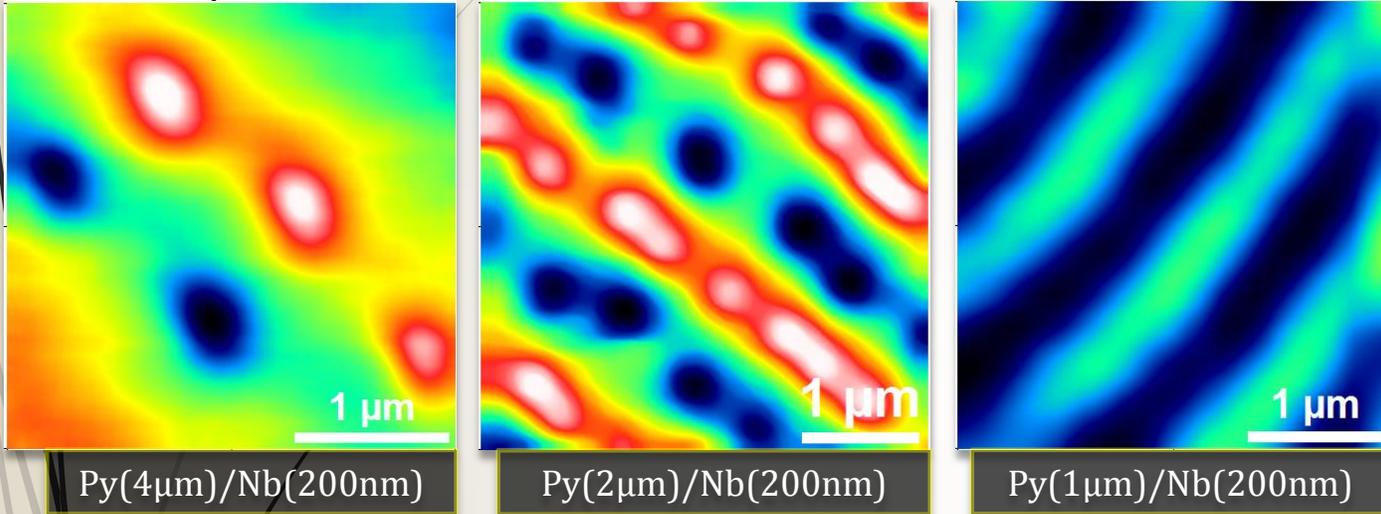
Zero field cooling – $T=6\text{K}$ (below $\text{Nb } T_c$)



- Spontaneous Vortex-Antivortex pairs are formed in $\text{Py}(4\mu\text{m})/\text{Nb}(200\text{nm})$ and $\text{Py}(2\mu\text{m})/\text{Nb}(200\text{nm})$ whereas no vortex occurrence is recorded in $\text{Py}(1\mu\text{m})/\text{Nb}(200\text{nm})$.
- Opposite polarized vortices correctly lie on proper magnetized stripes.

Vortex Nucleation

Zero field cooling – $T=6K$ (below Nb T_c)



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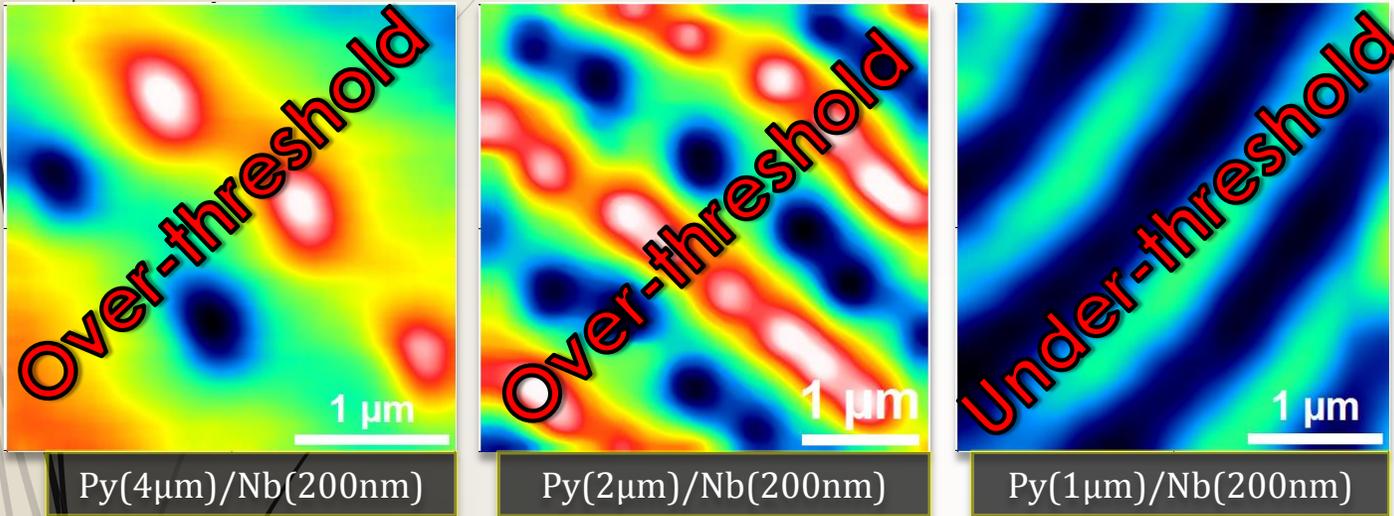
Theoretical Model

- ▶ Spontaneous V-AV can be formed in S/F only when the M_0 value of F is over-threshold

$$M_0 > M_c$$
$$M_c = 0.2 \frac{d_s}{w} H_{c1}$$

Vortex Nucleation

Zero field cooling – T=6K (below Nb T_c)



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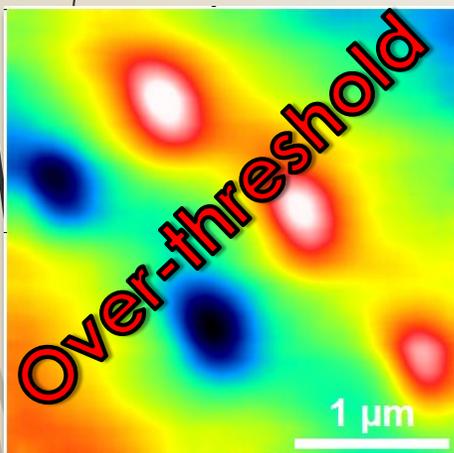
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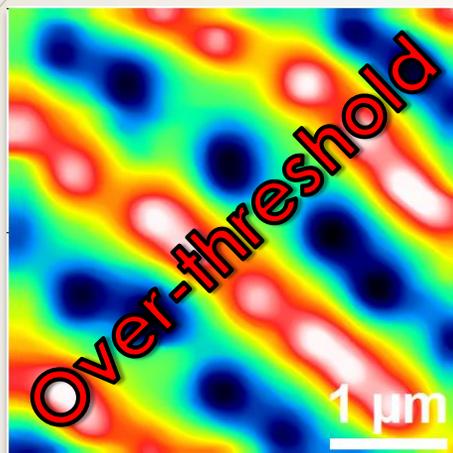
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Vortex Nucleation

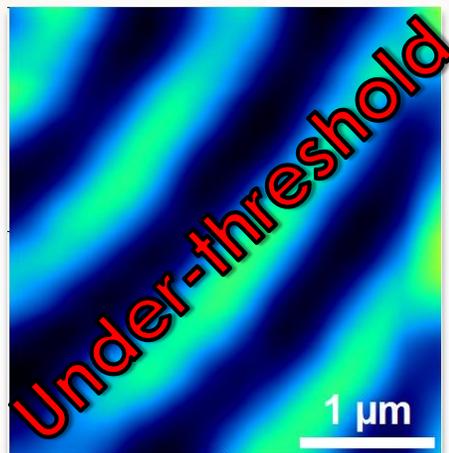
Zero field cooling – T=6K (below Nb T_c)



Py(4μm)/Nb(200nm)



Py(2μm)/Nb(200nm)



Py(1μm)/Nb(200nm)

Nb [nm]	Py [μm]	M _c (6K) [G]
200	4	16.6
200	2	11.9
200	1	33.9

Threshold value

Theoretical Model

- Spontaneous V-AV can be formed in S/F only when the M₀ value of F is over-threshold

$$M_0 > M_c$$

$$M_c = 0.2 \frac{d_s}{w} H_{c1}$$

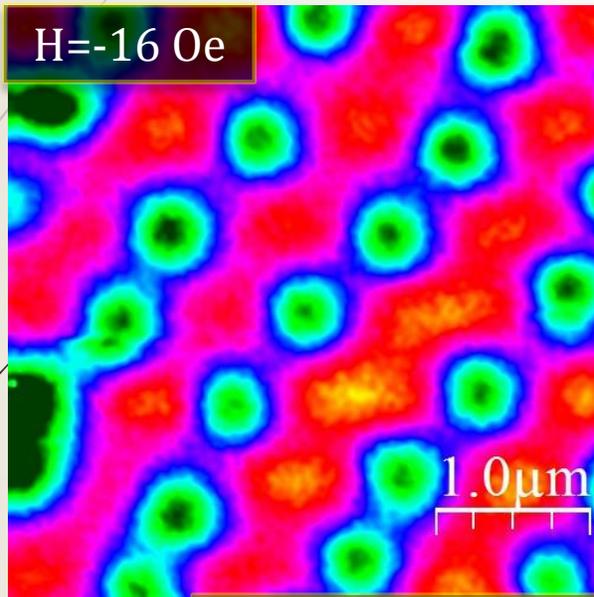
$$M_0(\text{Py} - 4\mu\text{m}) > 16.6\text{G}$$

$$M_0(\text{Py} - 2\mu\text{m}) > 11.9\text{G}$$

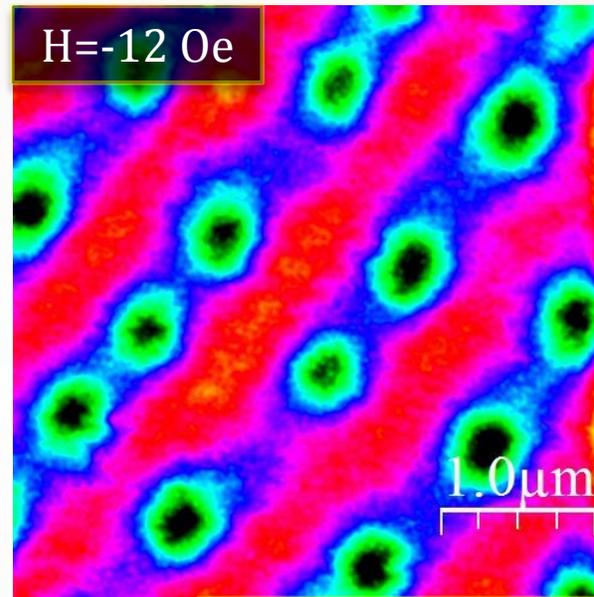
$$M_0(\text{Py} - 1\mu\text{m}) < 33.9\text{G}$$

Vortex Nucleation

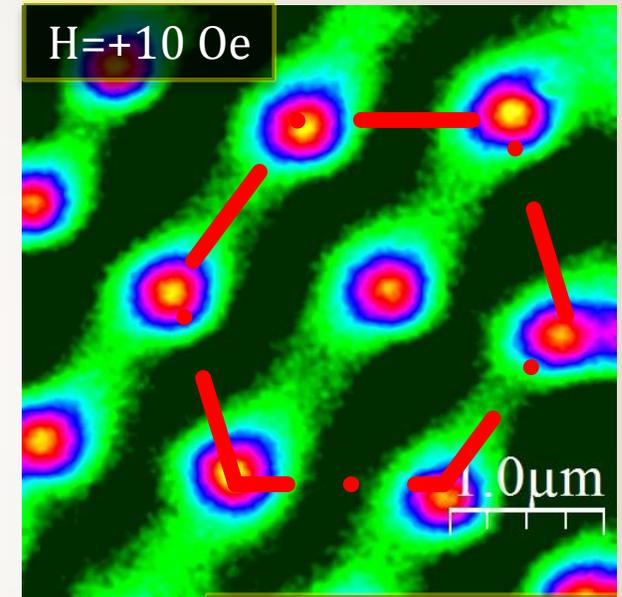
Field cooling – $T=6\text{K}$ (below Nb T_c)



Py(1 μm)/Nb(360nm)



Py(1 μm)/Nb(200nm)



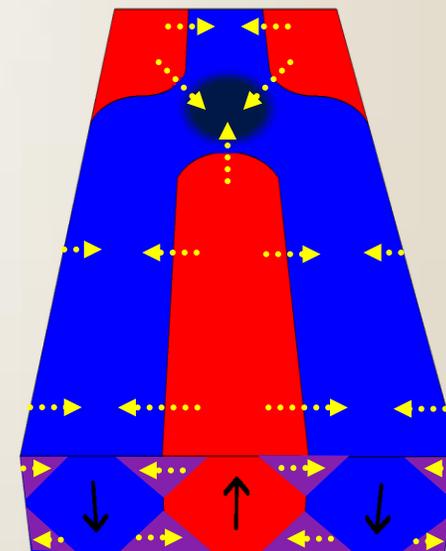
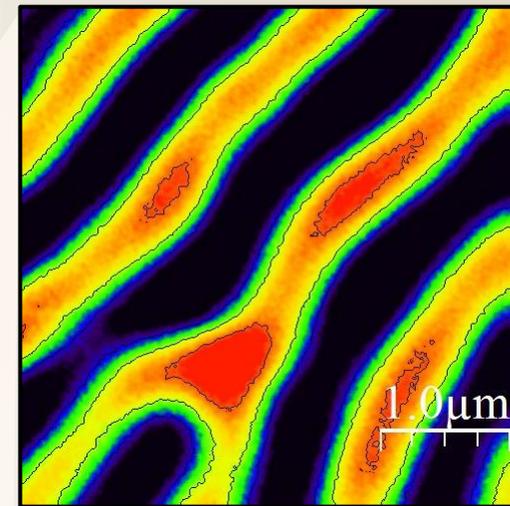
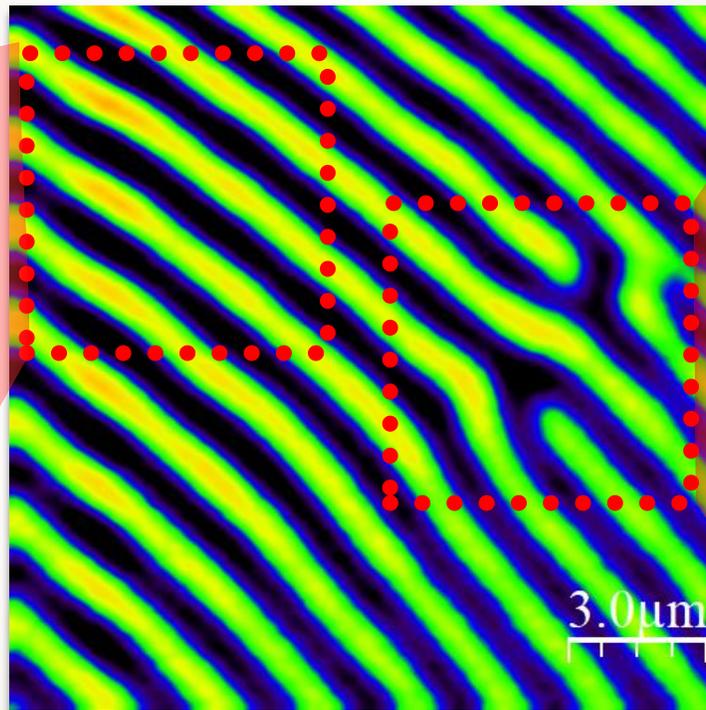
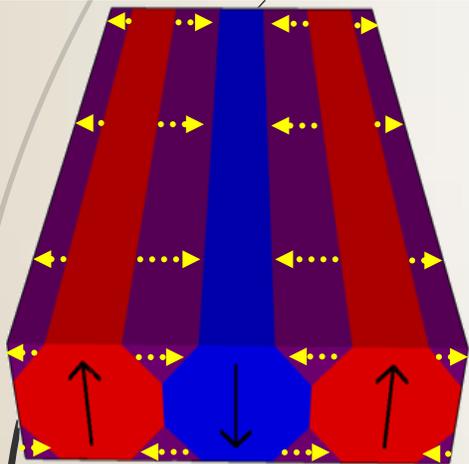
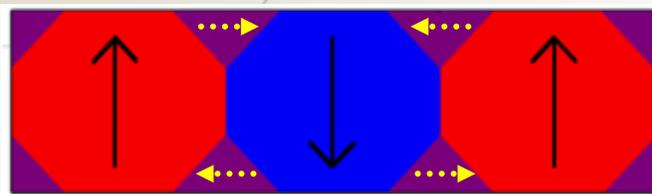
Py(1 μm)/Nb(150nm)

- Field included Vortices (or Antivortices) are formed with the same polarity as the external magnetic field on the proper polarized stripes;
- The tuning of the magnetic field intensity with respect to the stripe periodicity can lead to the formation of the hexagonal lattice (**Matching Field**)

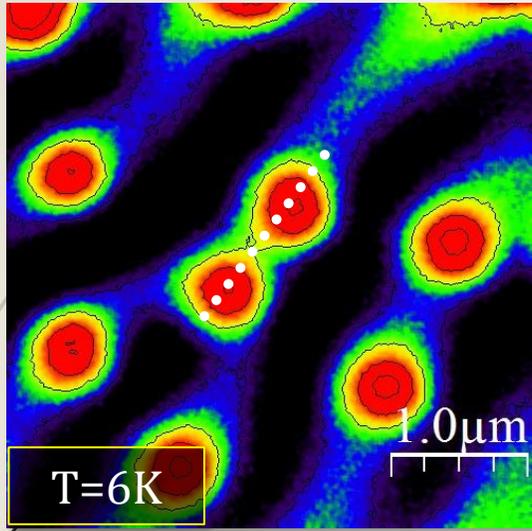
$$H = \frac{2 \Phi_0}{\sqrt{3} d^2}$$

Pinning Enhancement

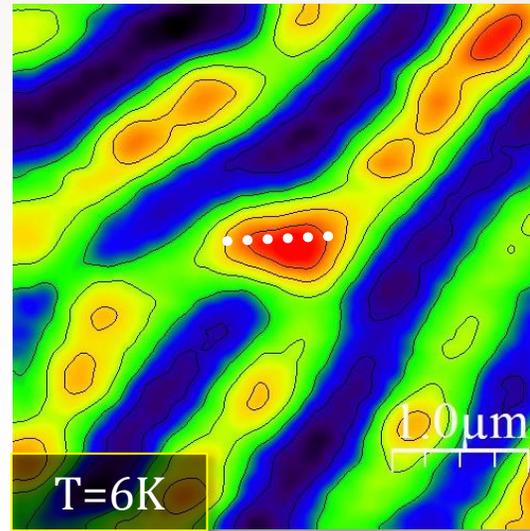
Typical MFM image of Py
at $T=13\text{K}$ (above $Nb T_c$)



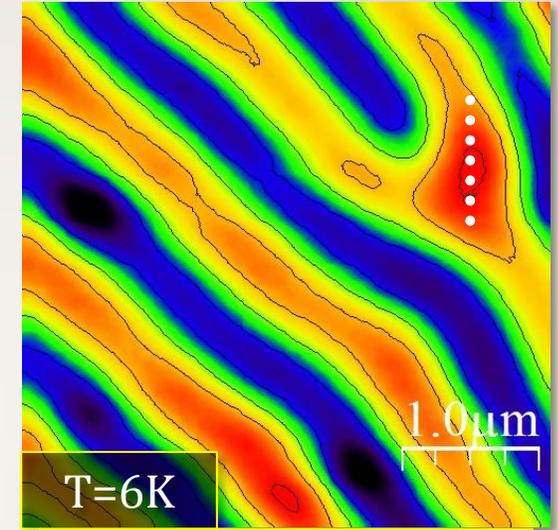
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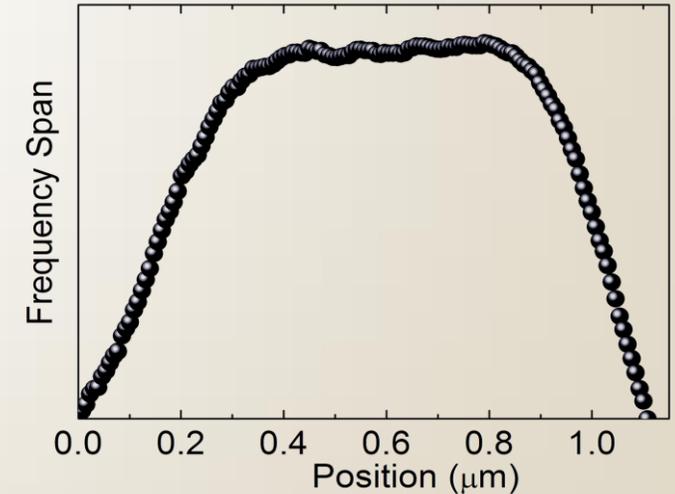
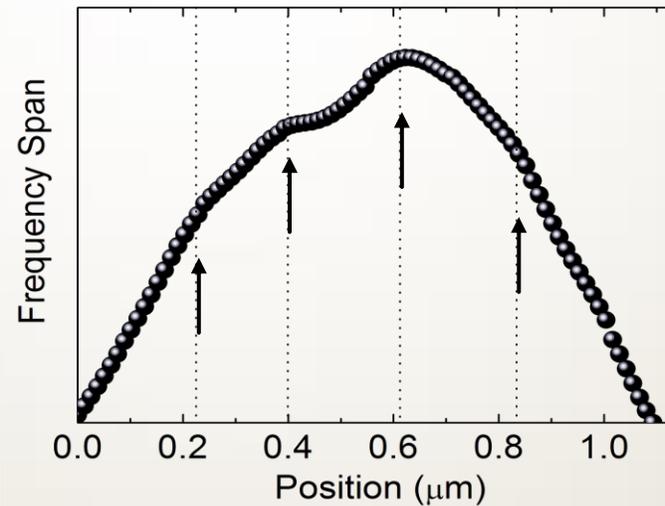
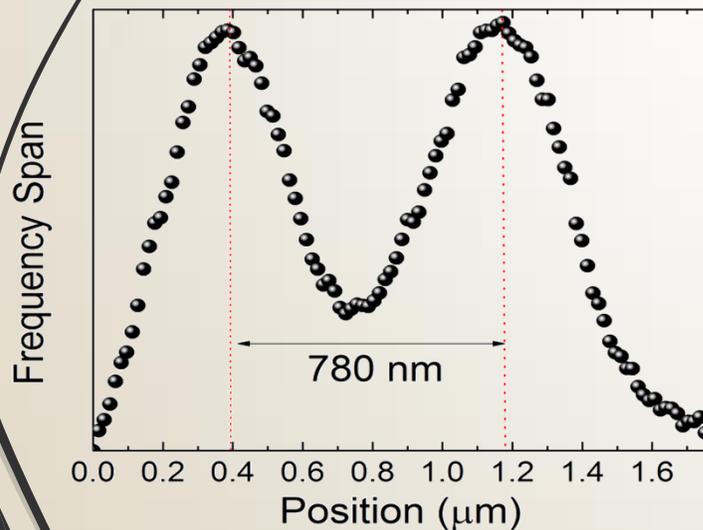
Field cooled in magnetic tip's field



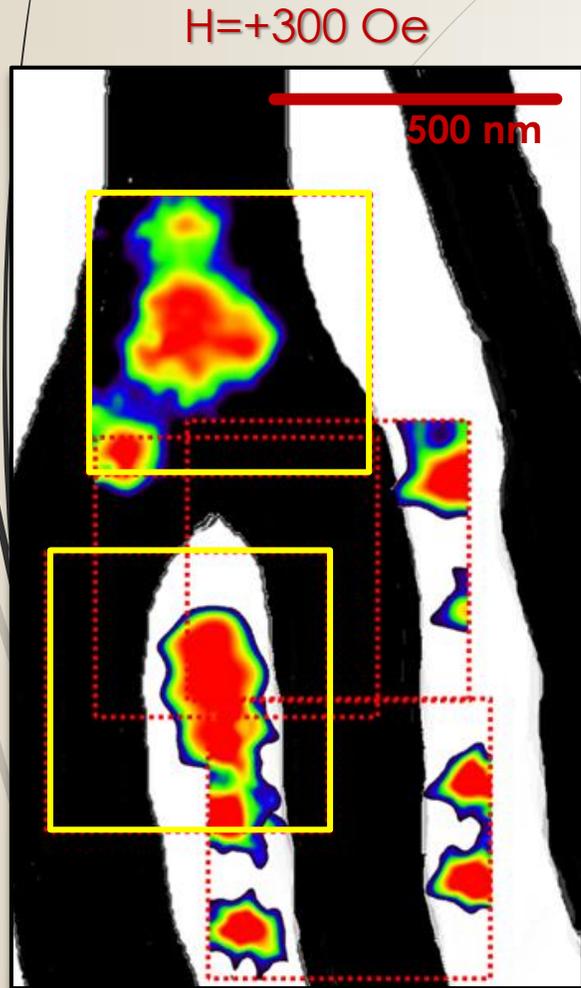
Field cooled in H=30 Oe



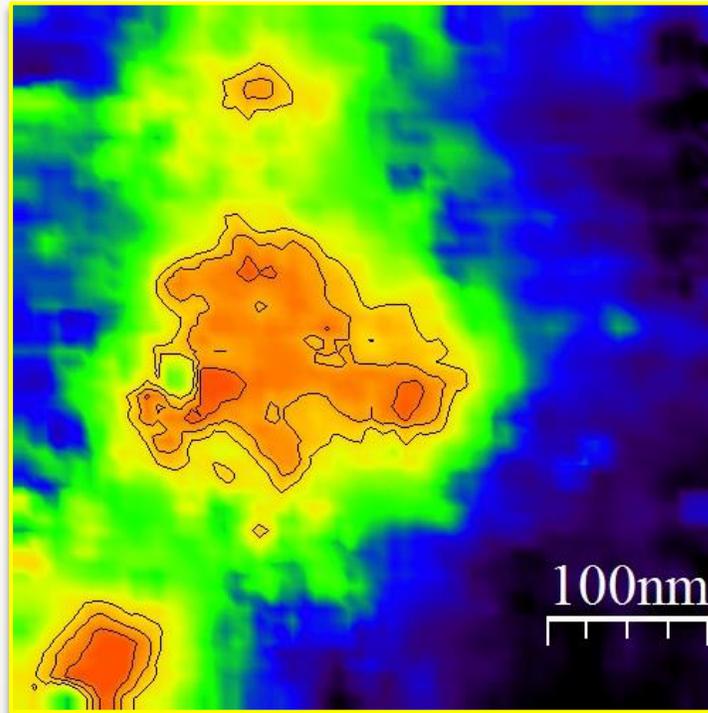
Zero Field cooled



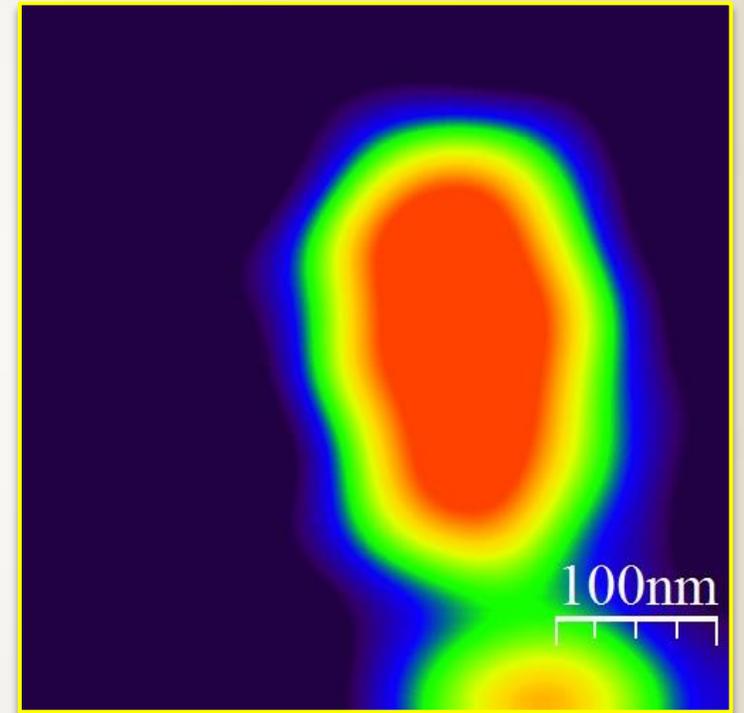
Pinning Enhancement



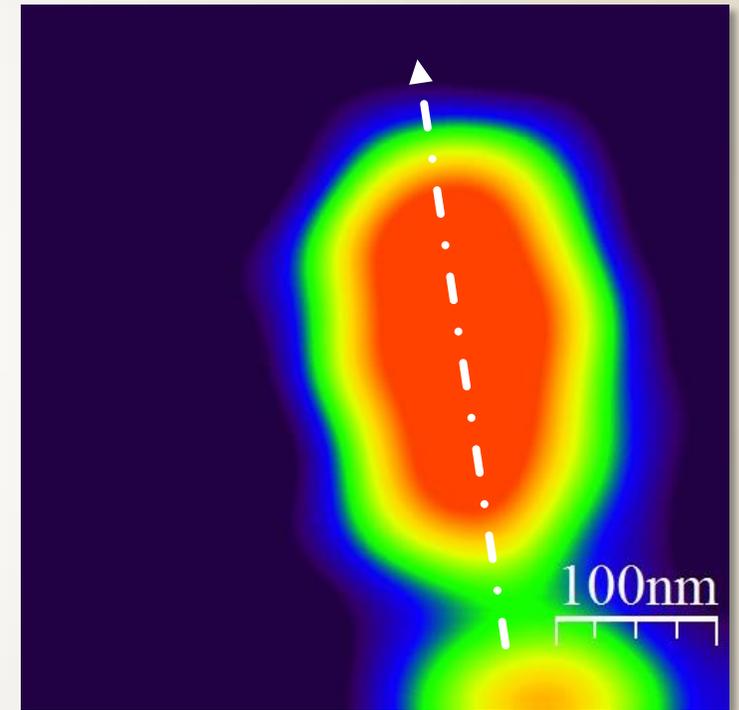
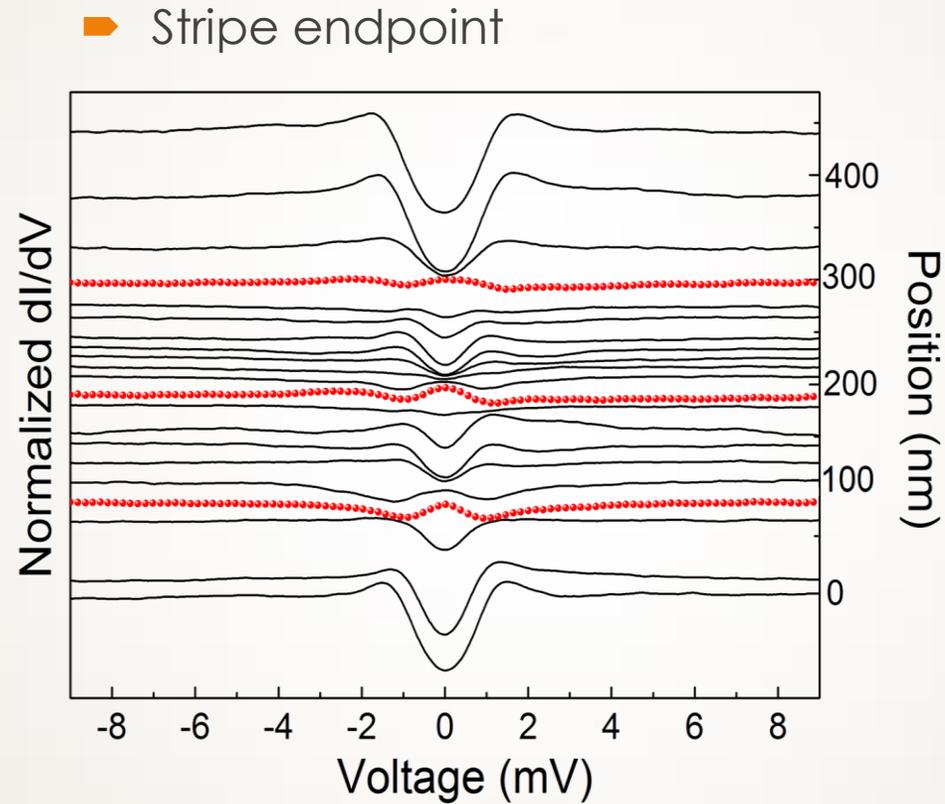
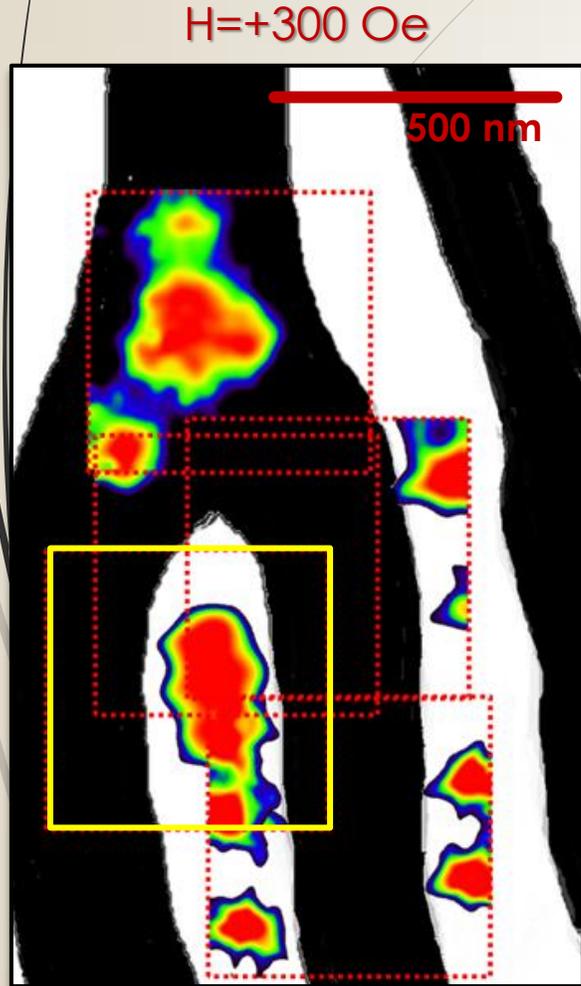
Top of the fork



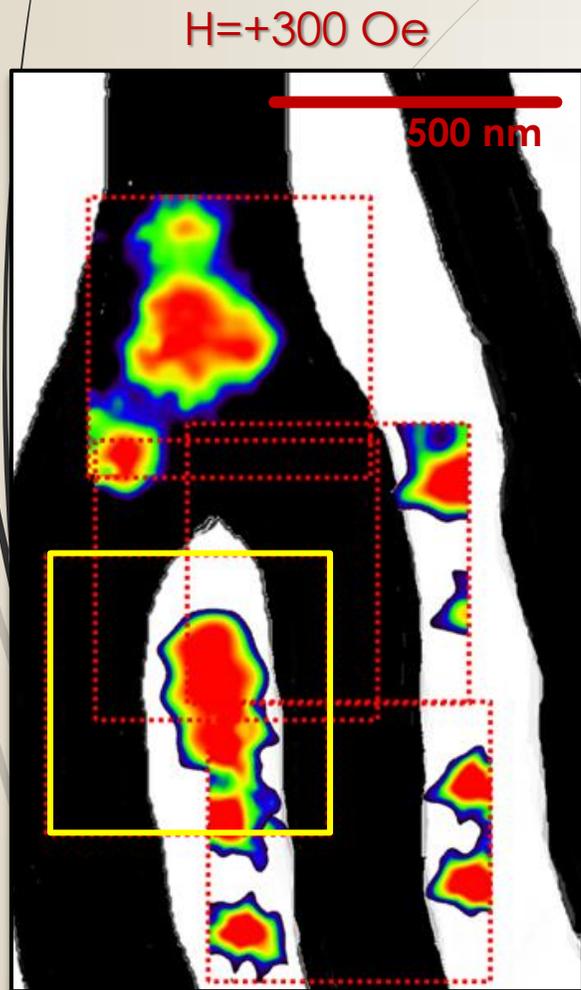
Stripe endpoint



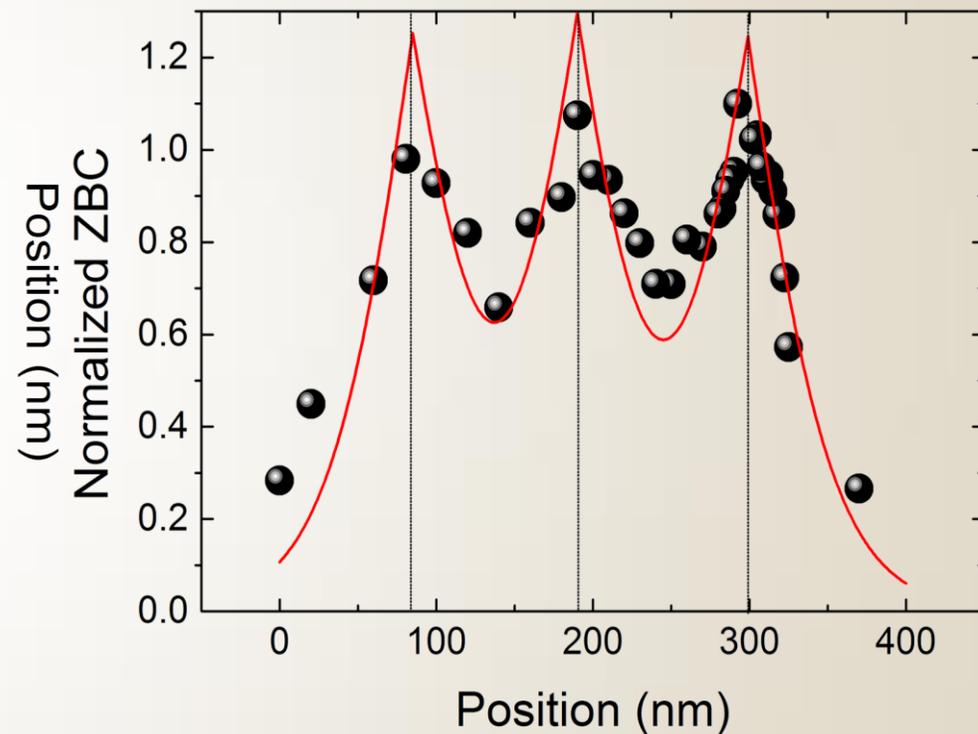
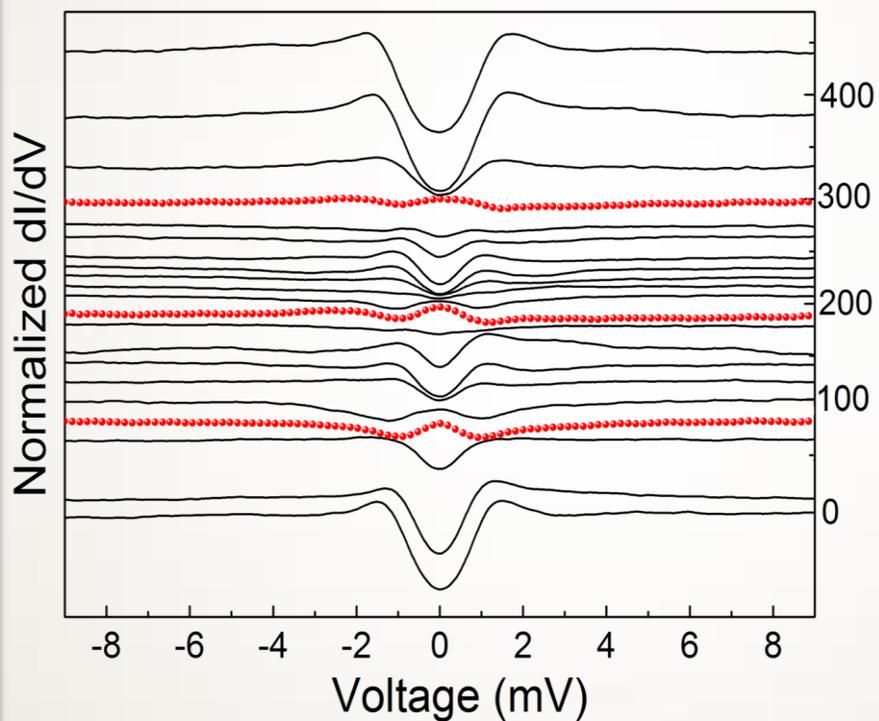
Pinning Enhancement



Pinning Enhancement

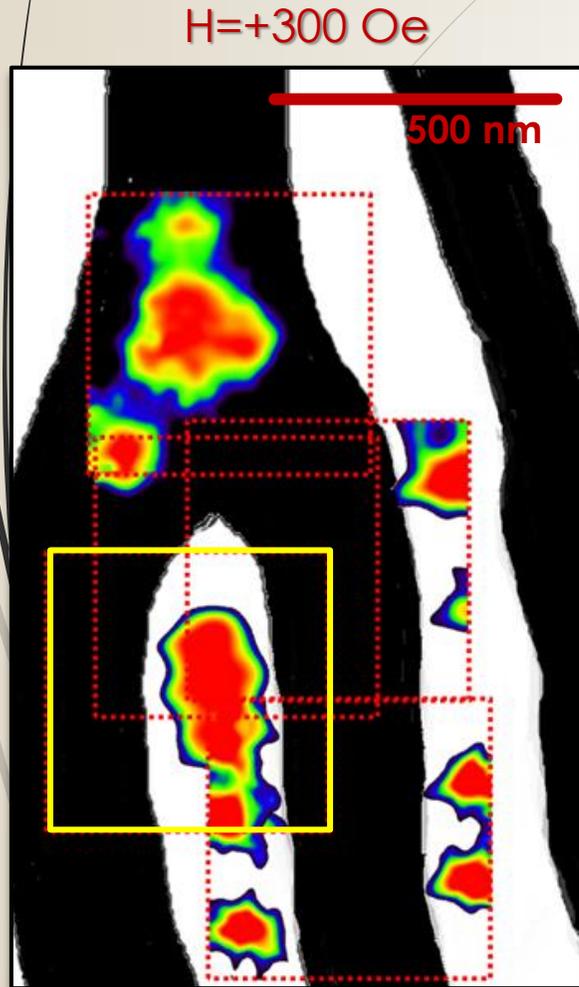


Stripe endpoint



V-V distance (nm)	ξ (nm) - 1.5K
105-109	55

Pinning Enhancement



V-V distance (nm)	ξ (nm) - 1.5K
105-109	55

- Condition for fully separated vortices at H_{c2} :

$$H = \frac{2 \Phi_0}{\sqrt{3} d^2} \quad d(H_{c2}) \approx 2.8 \times \xi$$

- Bifurcations can induce **vortex clusters** with **intervortex distance shorter than the minimum value achievable at H_{c2}** .

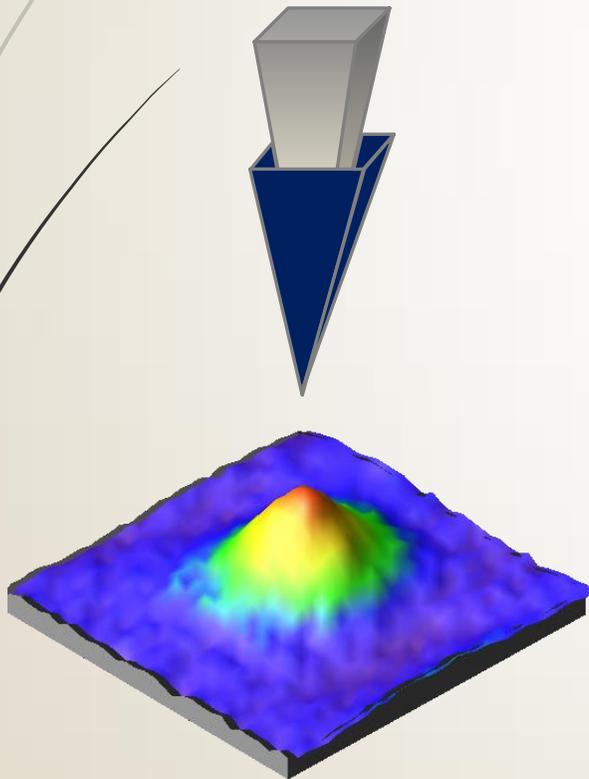
Advances in Quantitative-MFM



We always deal with samples carrying out a well know magnetic field: the superconducting vortex!

A superconducting vortex always supports a magnetic quantum flux:

$$\Phi_0 = \frac{hc}{2e} = 2.07 \times 10^{-7} Gcm^2$$



Advances in Quantitative-MFM



We always deal with samples carrying out a well know magnetic field: the superconducting vortex!

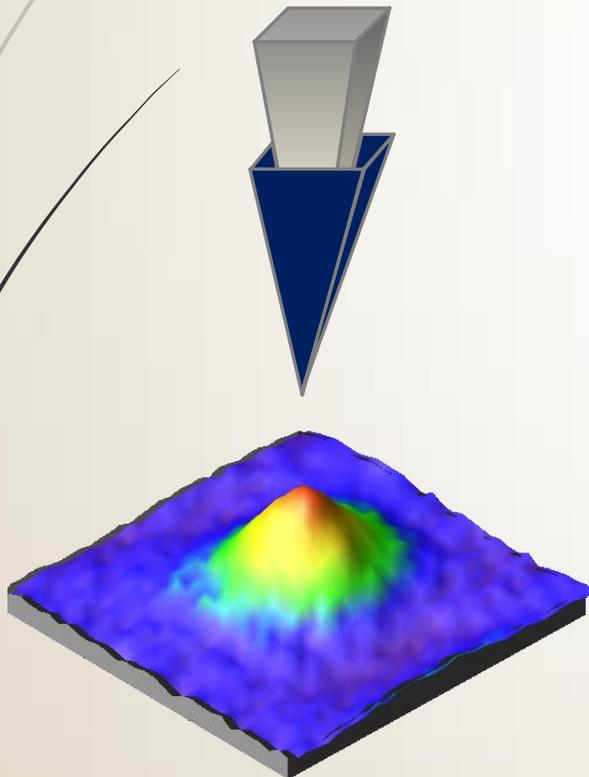
A superconducting vortex always supports a magnetic quantum flux:

$$\Phi_0 = \frac{hc}{2e} = 2.07 \times 10^{-7} Gcm^2$$

Many tip characterization procedures have been proposed in the approximation of:

- ❖ *Monopole* ; U. Hartmann, Adv. El. El. Phys. **47**, 49 (1994)
- ❖ *Dipole*; U. Hartmann, Phys. Lett. A **137**, 475 (1989)
D. Litvinov *et al.*, Appl. Phys. Lett. **81**, 1878 (2002)
- ❖ *Pseudopole*. T. Haberle, *at al.*, New J. Phys. **14**, 043044 (2012)

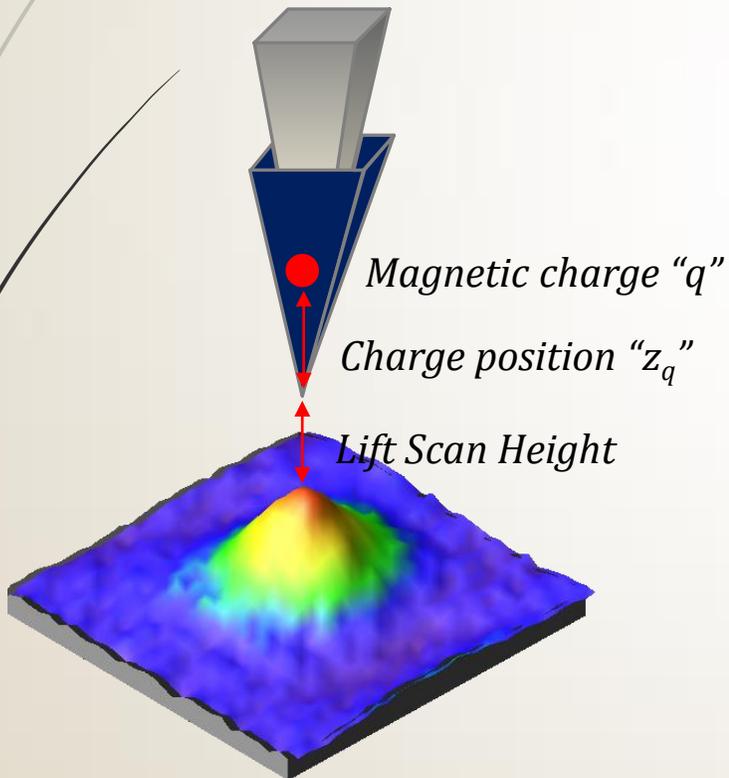
on different samples: wires, rings, dots,...



Advances in Quantitative-MFM



We always deal with samples carrying out a well know magnetic field: the superconducting vortex!



A superconducting vortex always supports a magnetic quantum flux:

$$\Phi_0 = \frac{hc}{2e} = 2.07 \times 10^{-7} Gcm^2$$

In the point probe approximation, the force exerted on the tip by the vortex :

$$F_z = qH_z = \frac{q}{\mu_0} (\vec{\nabla} \times \vec{A})_z$$

can be used to determine q and z_q .

Advances in Quantitative-MFM

Cantilever frequency shift due to tip-V and tip-AV interaction:

$$df = -\frac{f_0}{2k} \frac{dF_z}{dz} = -\frac{f_0}{2k} \left(\frac{dF_{z,tip-V}}{dz} + \frac{dF_{z,tip-AV}}{dz} \right)$$

Force exerted by V (AV) on the magnetic tip:

$$F_{z,tip-V(AV)} = qH_z = \frac{q}{\mu_0} (\vec{\nabla} \times \overrightarrow{A_{V(AV)}})_z$$



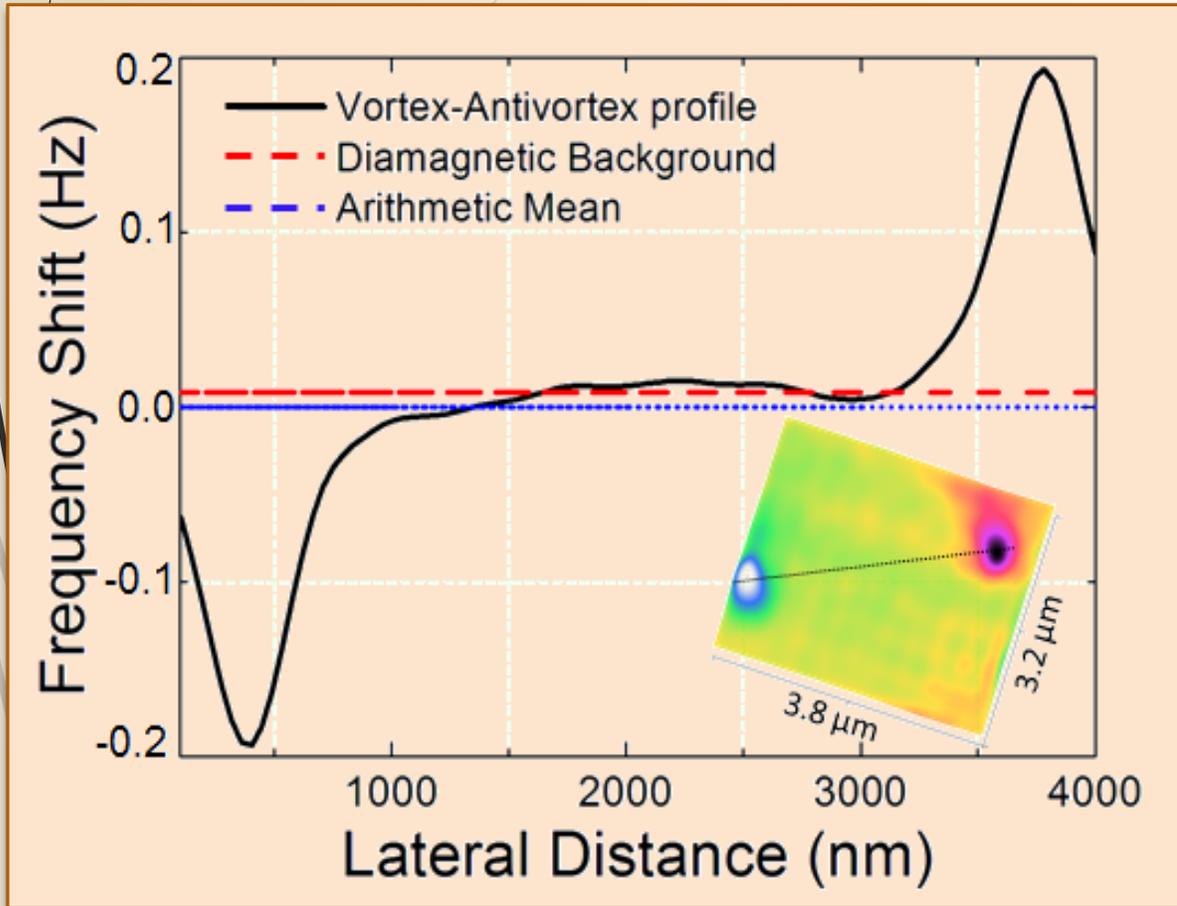
tip magnetic charge

$$F_{z,tip-V(AV)} = +(-) \frac{q}{\mu_0} \int_0^\infty G(x) \Phi(x) e^{-xz} J_0(xr) x dx$$

$Z_q + Z_{lift\ scan}$

diamagnetic contribution

Advances in Quantitative-MFM

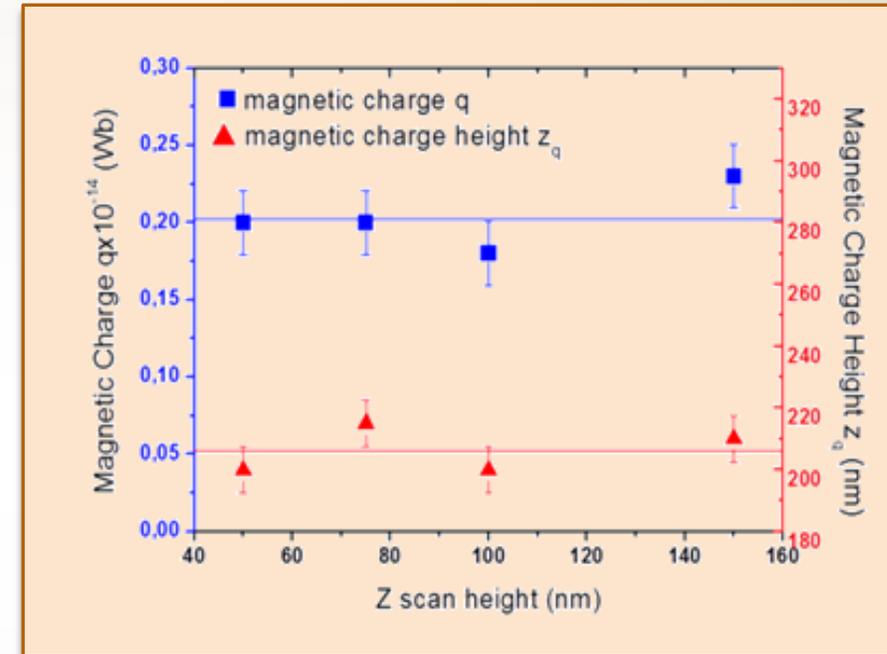
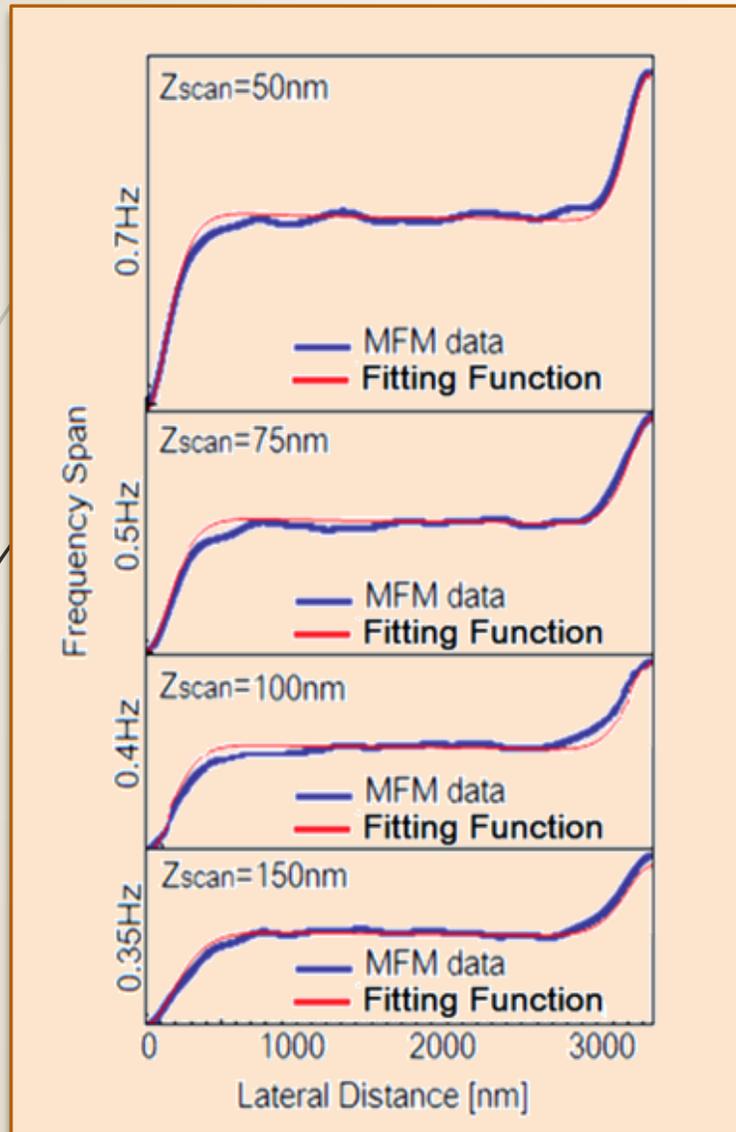


- ▶ We nucleated a V-Av pair in Nb-200nm single layer;
- ▶ We measured the MFM signal along the V-AV profile, which shows the unbalancing in V-AV height due to the diamagnetic background;
- ▶ We fitted the measured profile by considering the contribution of V, AV and diamagnetic background to the frequency shift df ;

$$F_{z,tip-V(AV)} = +(-) \frac{q}{\mu_0} \int_0^{\infty} \underbrace{G(x)}_{\text{diamagnetic contribution}} \Phi(x) e^{-x(z)} \underbrace{J_0(xr)}_{Z_q + Z_{\text{lift scan}}} x dx$$

- ▶ We extracted q and z_q as fitting parameters.

Advances in Quantitative-MFM

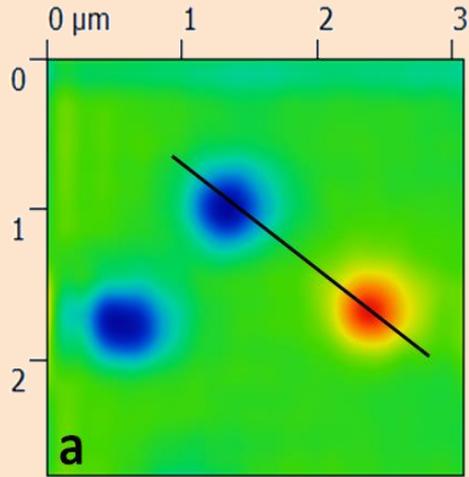


- q and z_q representative of the used MFM tip have been derived from the zero-slope linear fit, resulting in:

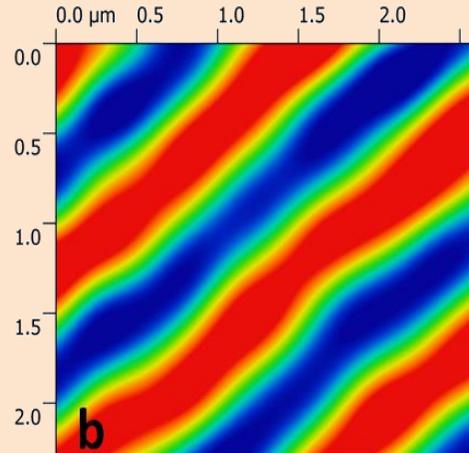
$$q = (0.20 \pm 0.01)10^{-14}Wb$$

$$z_q = (206 \pm 4)nm$$

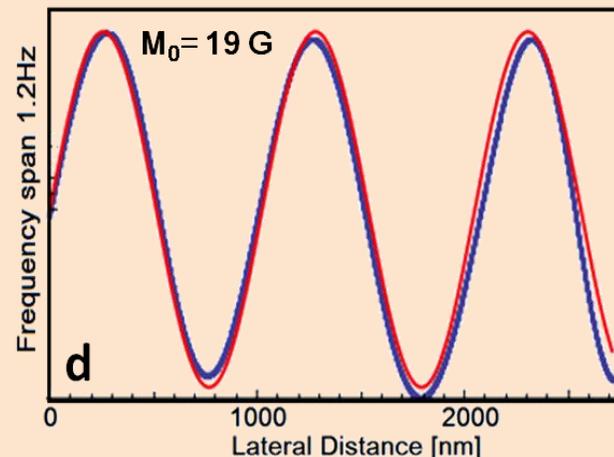
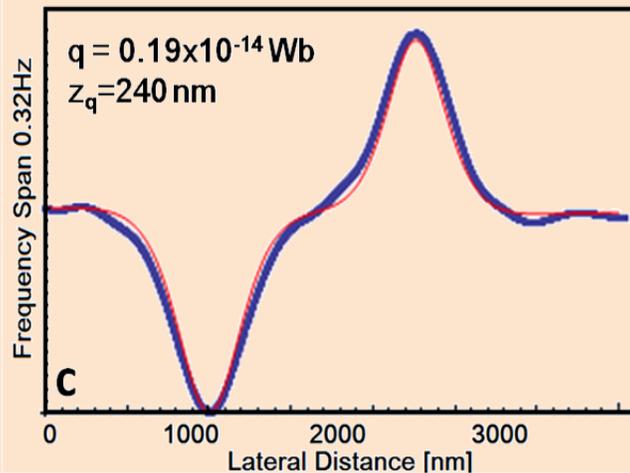
Advances in Quantitative-MFM



Frequency span 0.32Hz



Frequency span 1.2Hz



- ▶ We characterize the MFM tip on V-AV pair in Nb;
- ▶ We used the same tip to measure a sample of Py(1μm);
- ▶ We calculated analytically the frequency shift due to Py stray field;

$$df = -\frac{f_0}{2k} q \frac{dH_{Py}}{dz}$$

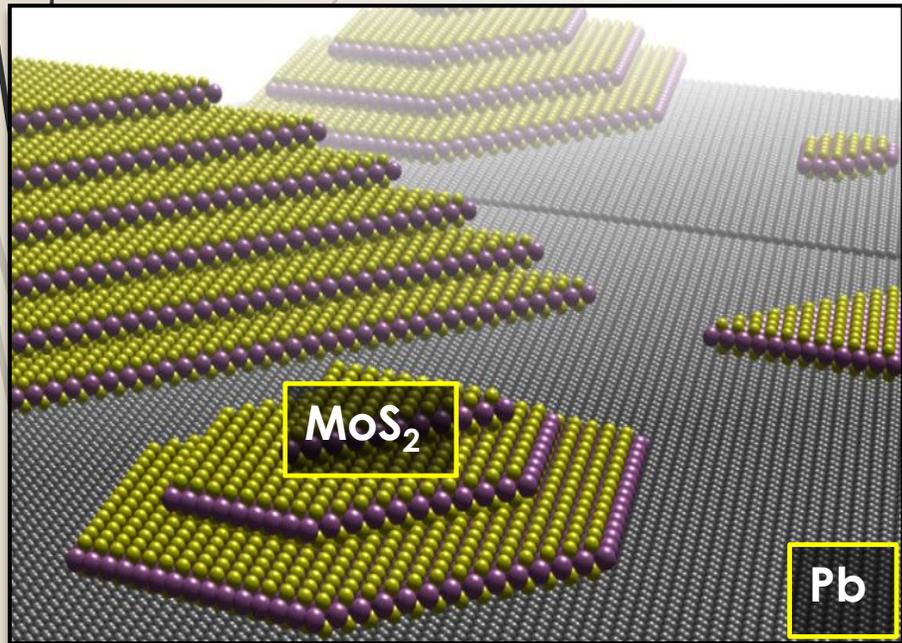
$$H_{Py} = 8M_0 \sum_k \frac{(2k+1) \left(\frac{\pi}{w}\right) \sin \left[\left((2k+1) \left(\frac{\pi}{w}\right) \right) x \right]}{(2k+1)^2 \left(\frac{\pi}{w}\right)} e^{-\frac{(2k+1)\pi}{w} z}$$

- ▶ We fit Py magnetic signal by keeping M_0 as fitting parameters and we found:

$$M_0(Py - 1\mu m) = 19G$$

Electrically coupled S/F systems

We studied **electrically coupled S/F** systems by performing LT-STM experiments @ Physics Department of Temple University (Philadelphia, USA)



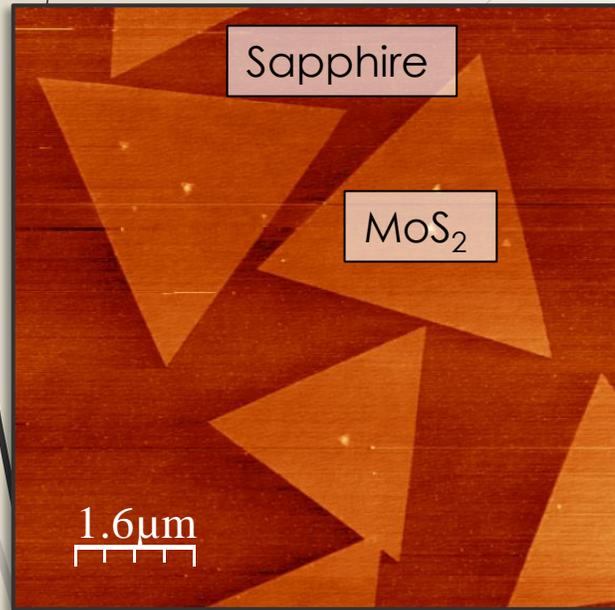
We aimed at investigating **inverse proximity effect** between a superconductor (Pb) and the ferromagnetic localized states at the edge of MoS₂ monolayer islands.

We found something different but....



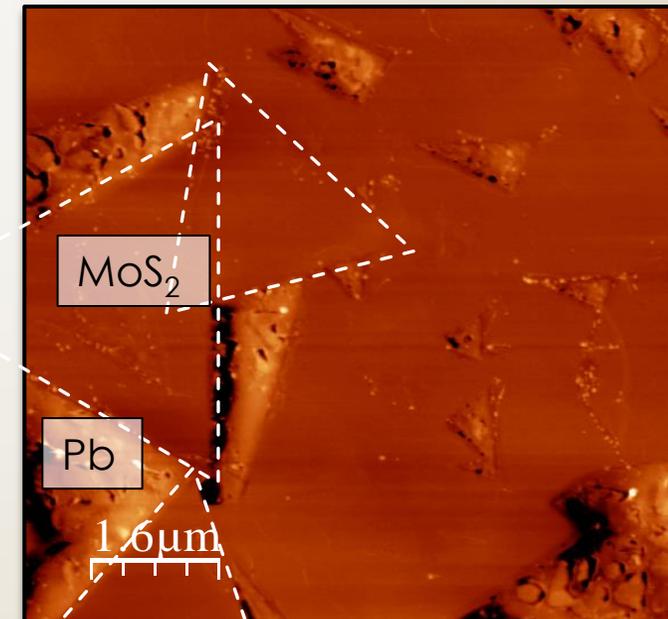
....we haven't got to the top yet!

Sample preparation

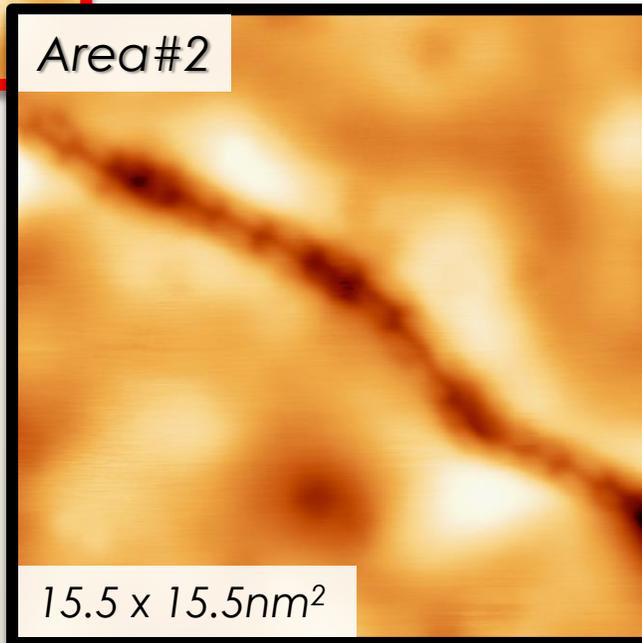
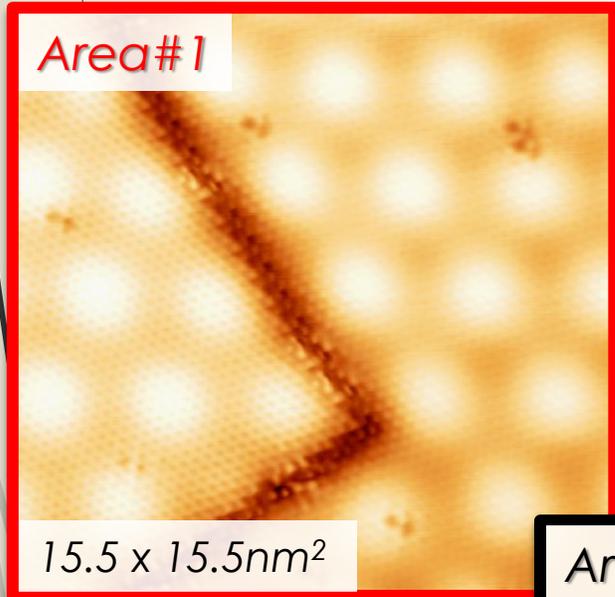


- ▶ We grew CVD-MoS₂ islands on sapphire substrate;
- ▶ We brought the samples in a UHV-chamber, annealed it and grew more than 300nm of Pb;
- ▶ We cleaved the bilayer from the bottom (in-situ) by peeling off the substrate (sapphire);

- ▶ We performed STM measurements on the bottom side of the so-grown bilayer.



Semiconducting vs Metallic MoS₂



We found two possible topographies:

- ▶ **Area#1:** very good atomic resolution of MoS₂ and Moiré pattern



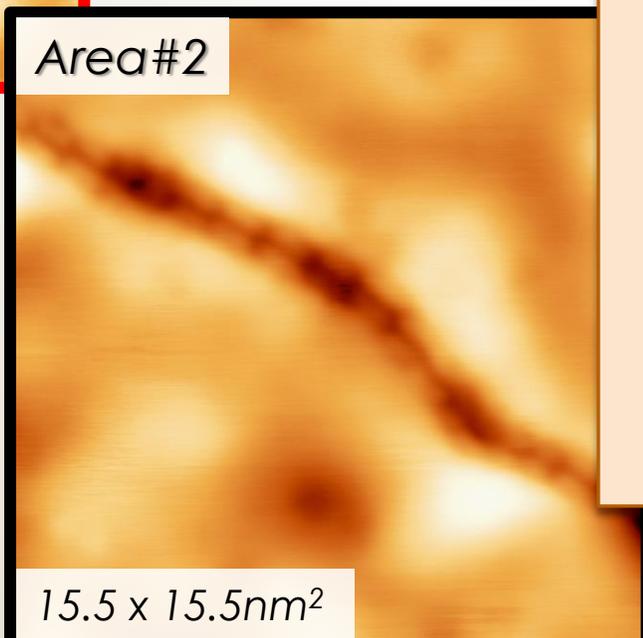
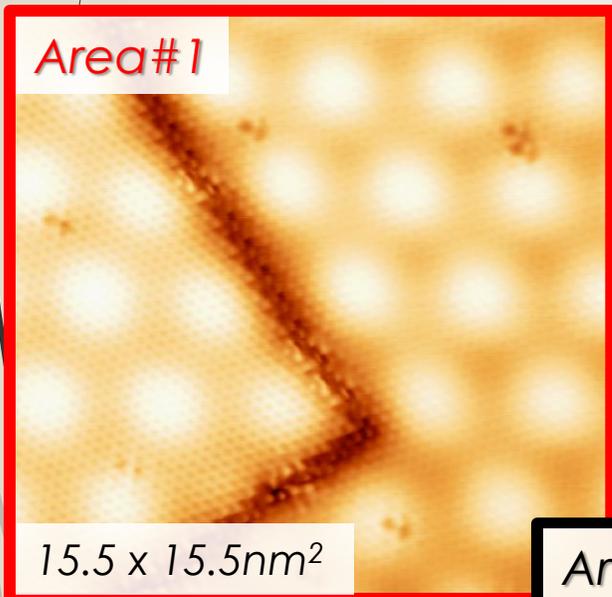
Good electric coupling with Pb underneath

- ▶ **Area#2:** no good resolution of MoS₂ and no Moire pattern

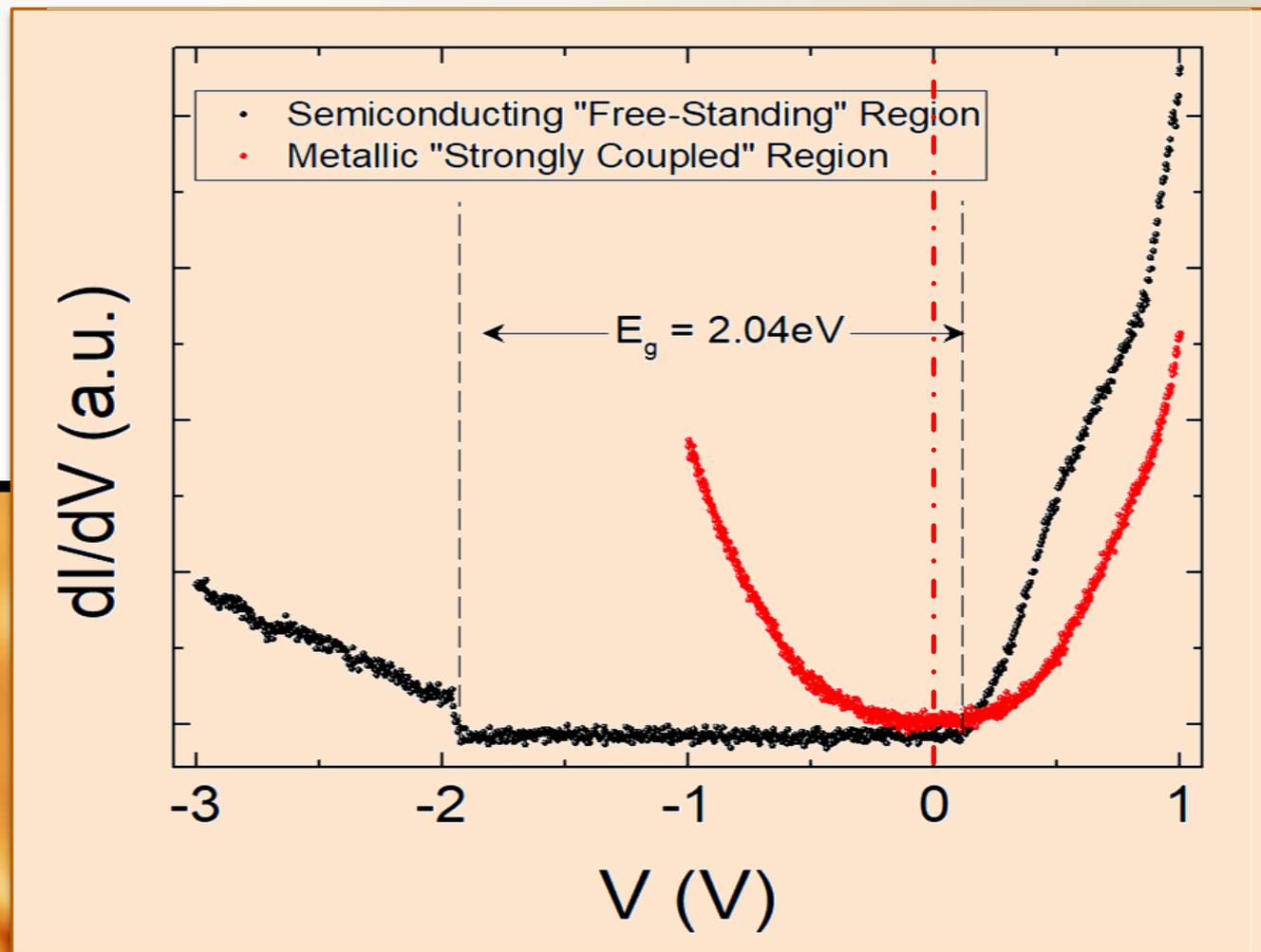


Bad electric coupling with Pb underneath
(almost free standing)

Semiconducting vs Metallic MoS₂

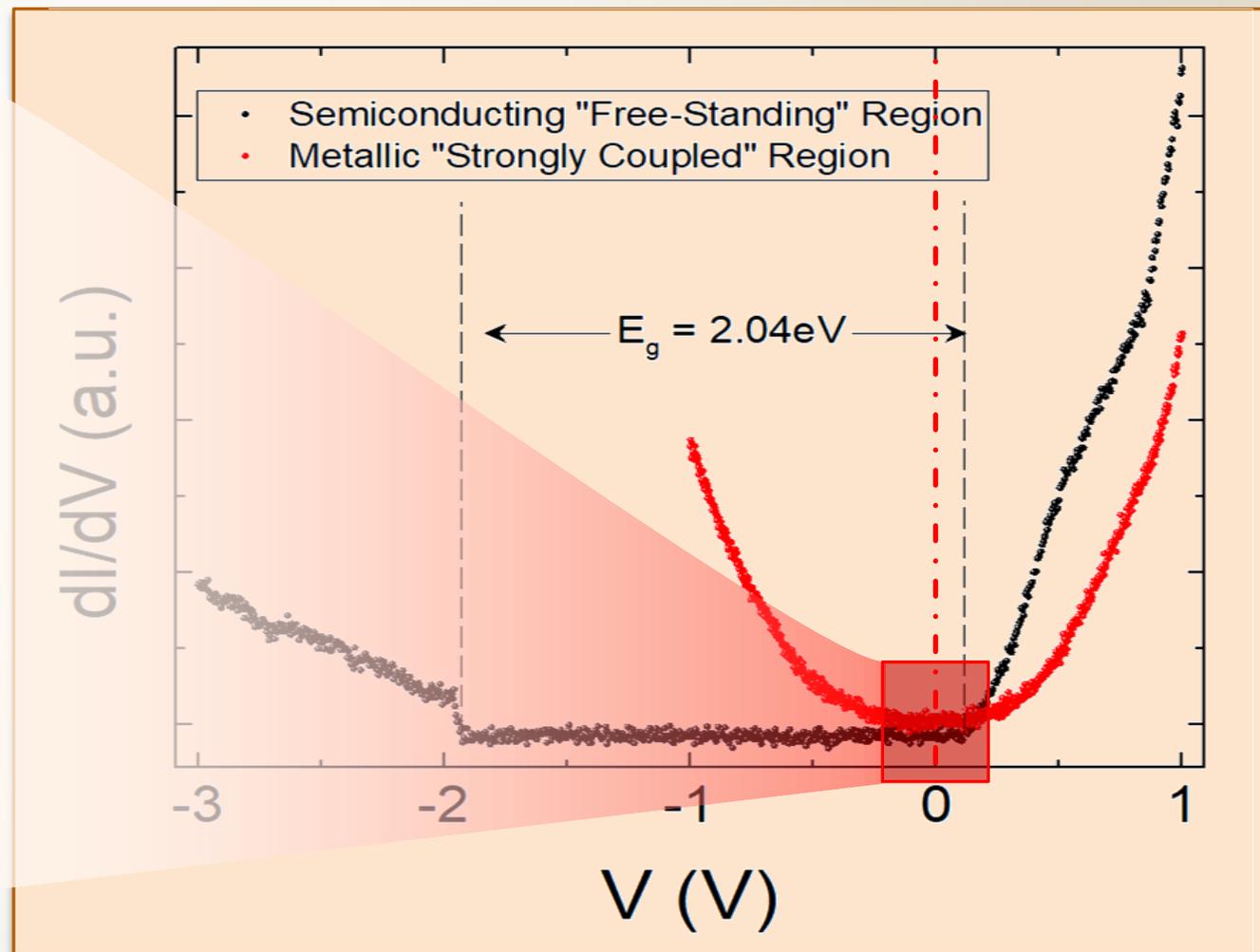
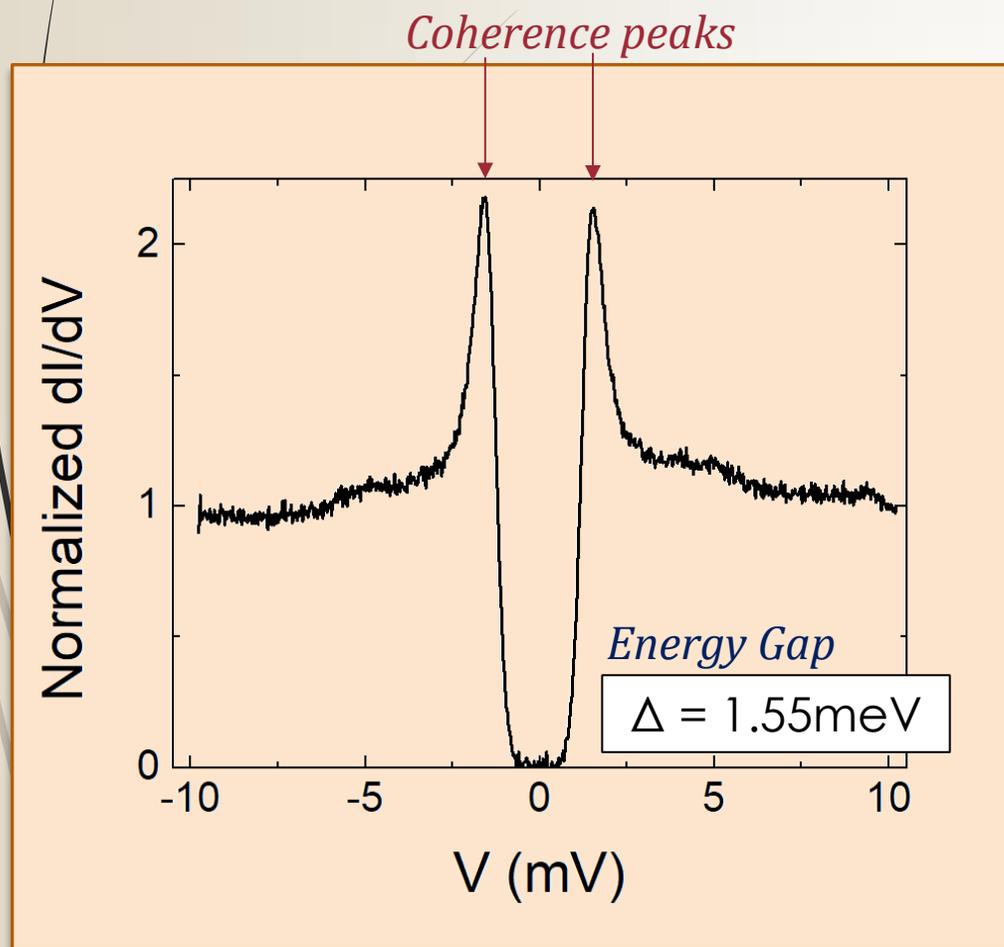


MoS₂ lattice parameter 3.22 Å



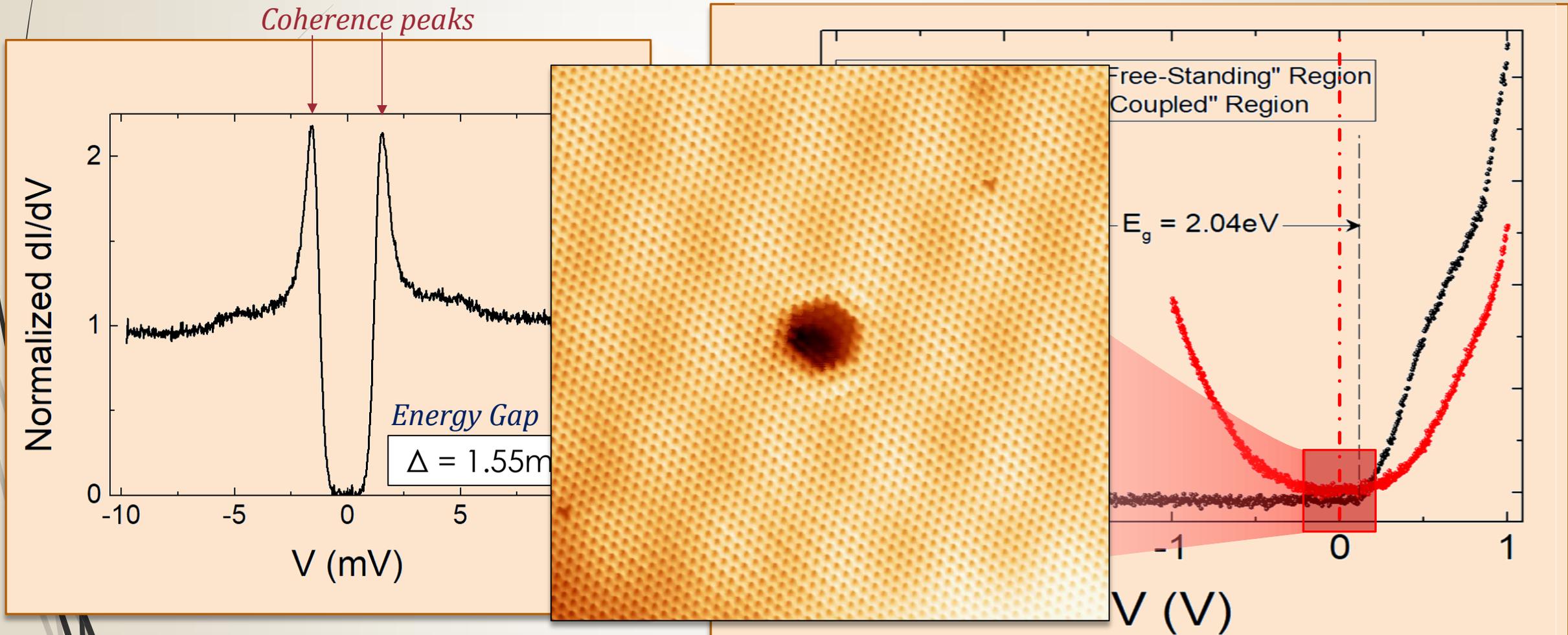
$$I \sim \frac{4\pi e}{\hbar} |M|^2 \rho_t(0) \int_{-eV}^0 \rho_s(\epsilon) d\epsilon$$

Semiconducting vs Metallic MoS₂



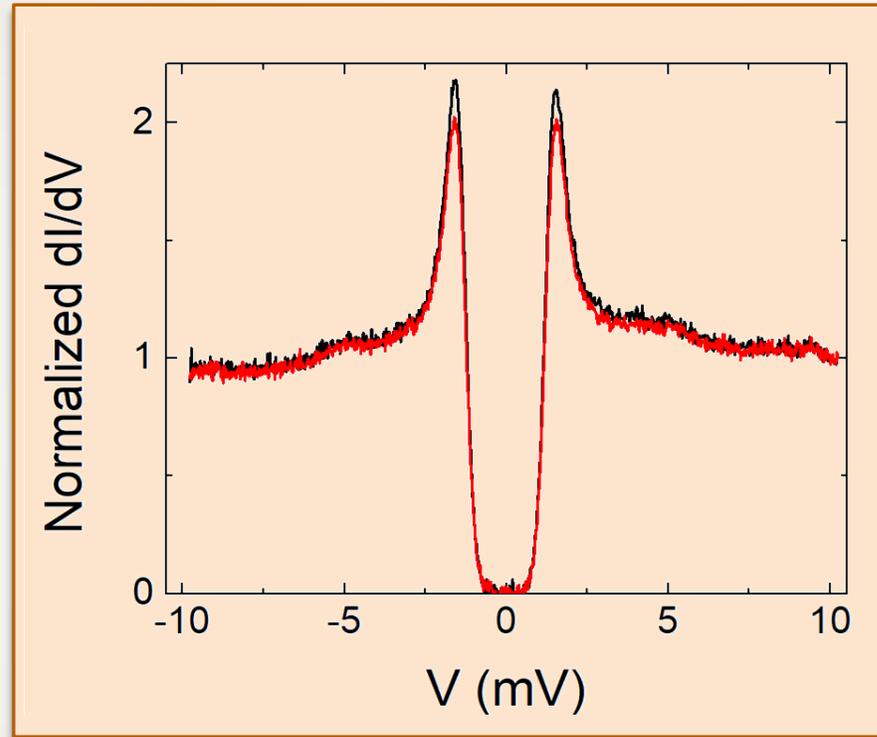
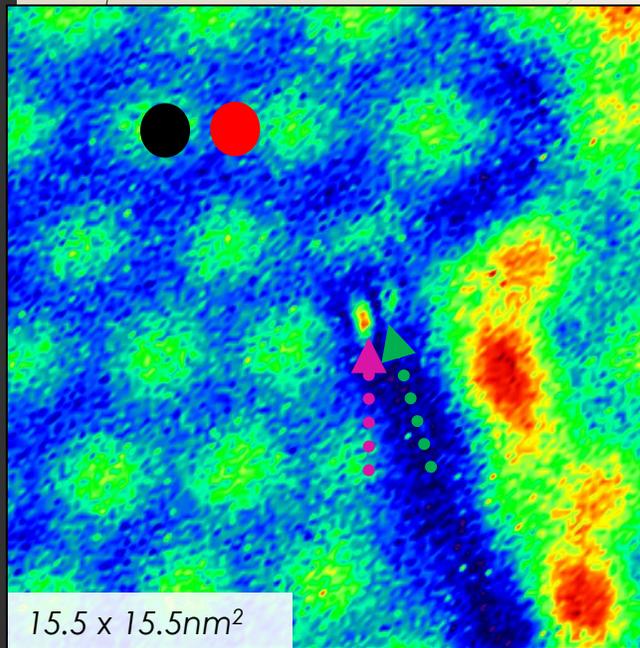
$$I \sim \frac{4\pi e}{\hbar} |M|^2 \rho_t(0) \int_{-eV}^0 \rho_s(\varepsilon) d\varepsilon$$

Semiconducting vs Metallic MoS₂

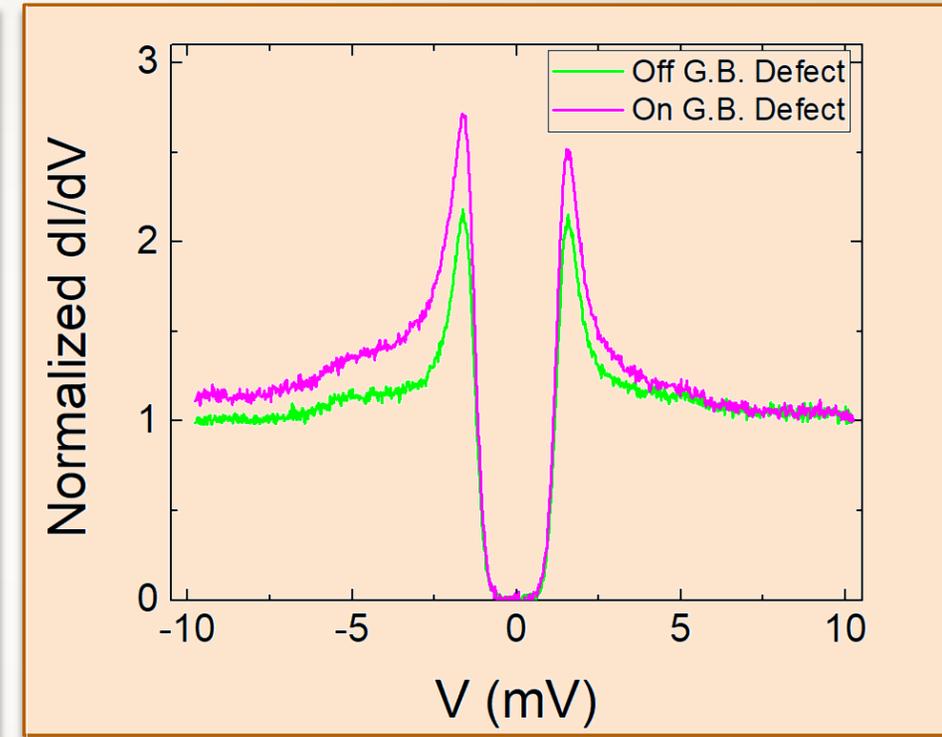


$$I \sim \frac{4\pi e}{\hbar} |M|^2 \rho_t(0) \int_{-eV}^0 \rho_s(\varepsilon) d\varepsilon$$

Spectroscopy on Moiré pattern and defects



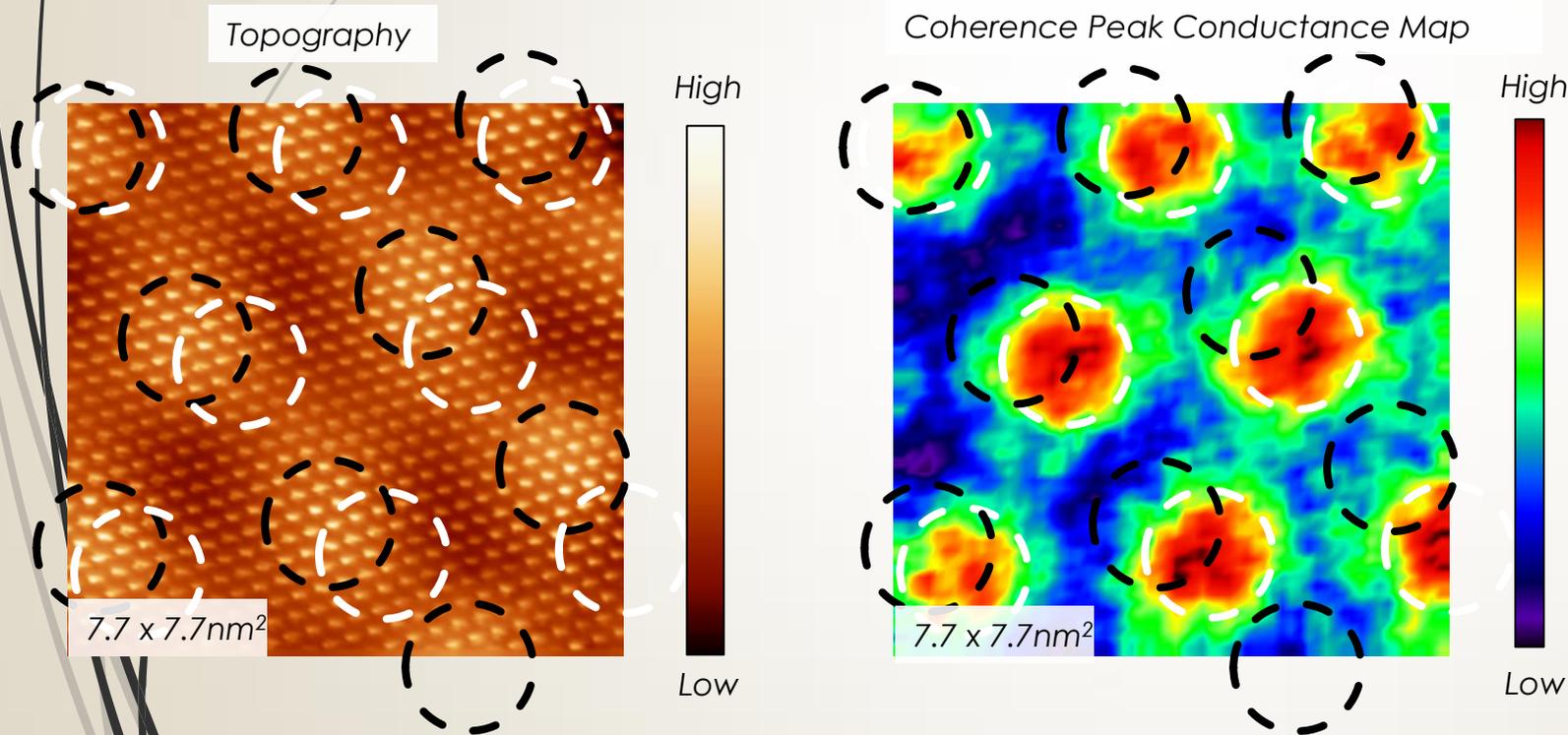
Change in coherence peak height across the green spots



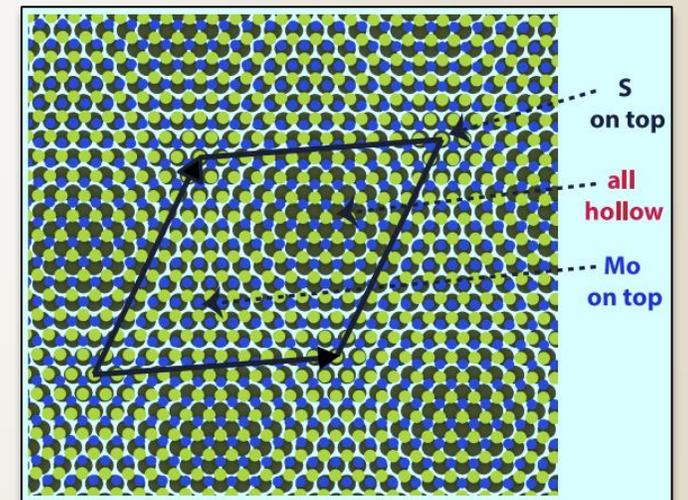
Change in coherence peak height across the defects

Moiré pattern and grain boundary

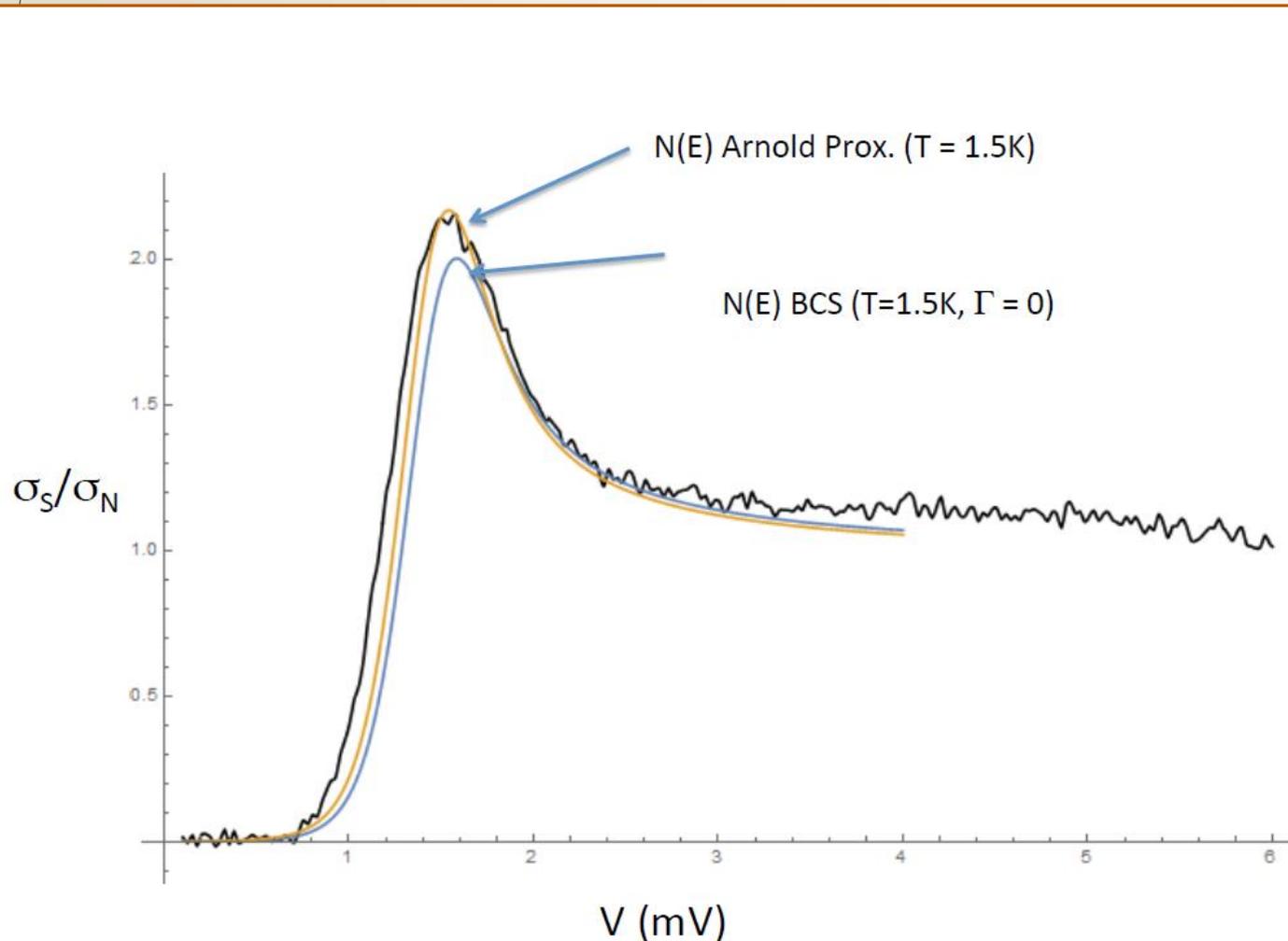
- ▶ If we acquire simultaneous topography and conductance maps at the coherence peak we see a mismatch of the topographic and conductance maxima



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Fit of Spectroscopy

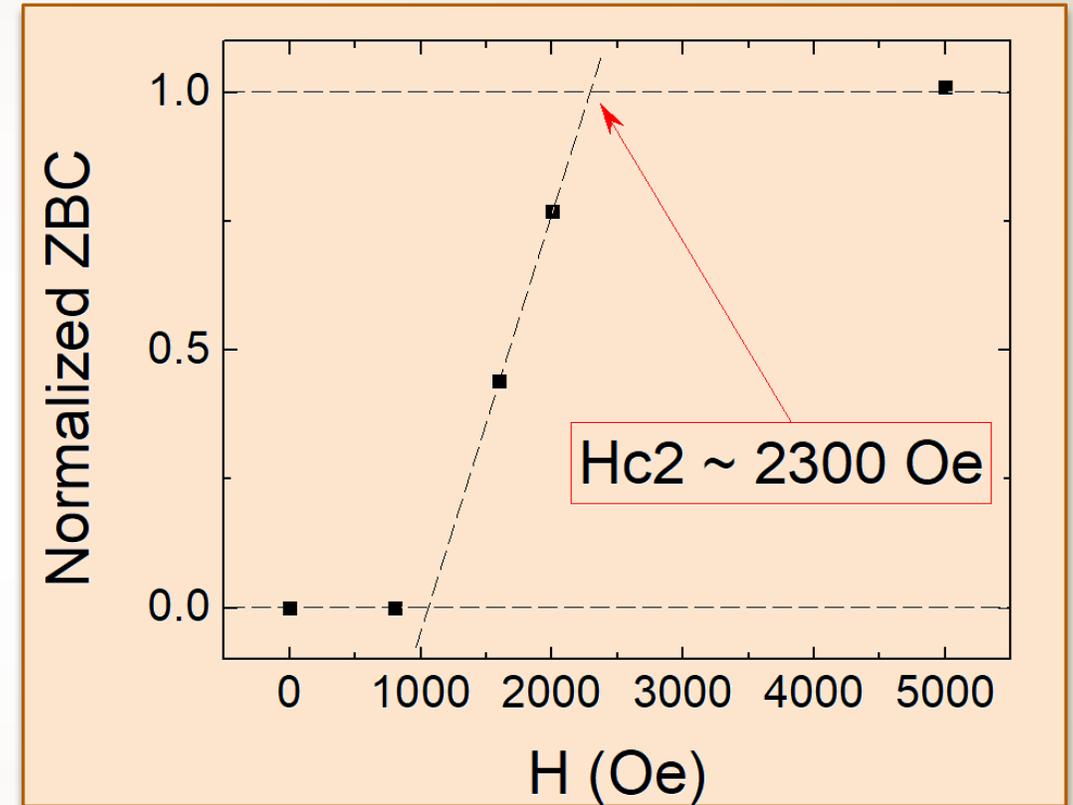
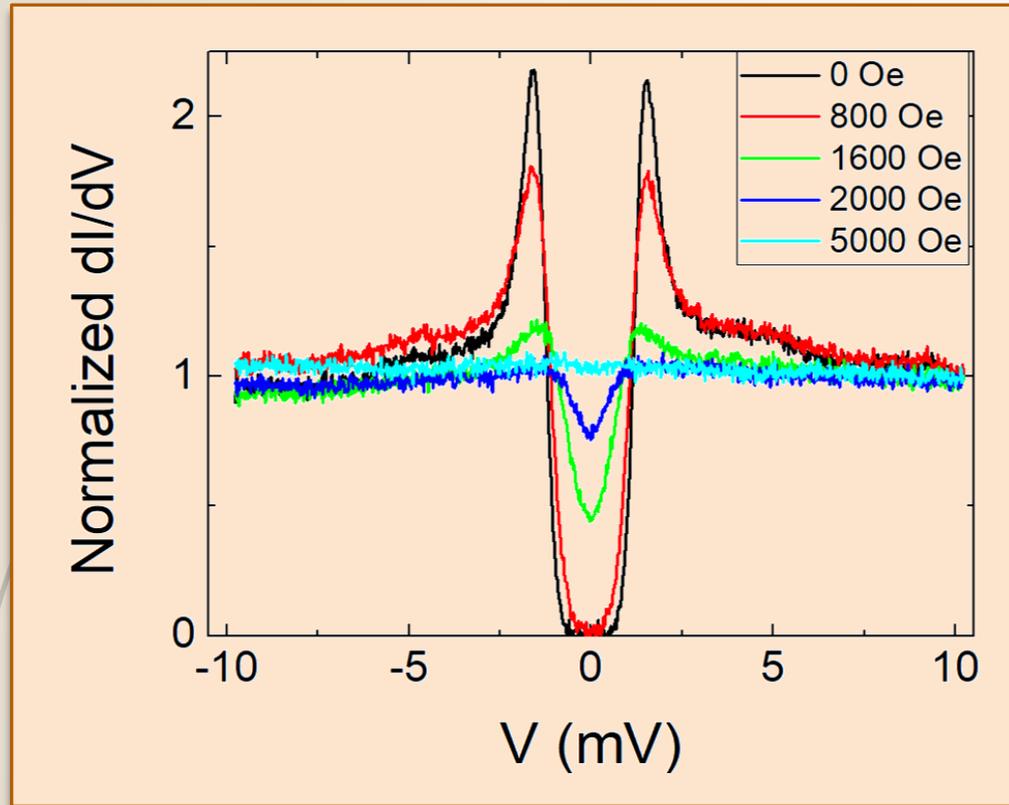


- ▶ If we fit the LDOS with standard BCS theory, we are not able to reproduce the coherence peak height;
- ▶ If we fit the LDOS with a model taking into account the **proximity** between a **SC** and a **metal**, it is almost perfect on the peak height.



Does MoS₂ monolayer become metallic and then superconductor when proximized to Pb?

Magnetic Field Dependence



- We observed the suppression of superconductivity around 2300 Oe;
- The critical field estimated from ZBC vs H is higher than the one in single-Pb.



Typical feature of proximized structures!

Conclusions

- ▶ **Magnetically coupled S/F** have been studied to get out insights into vortex nucleation and pinning at the nanoscale;
- ▶ **Electrically coupled S/F** are studied aiming at finding triplet superconductivity but, to date, performed STM experiments gave indications of possible metallicity of MoS₂ monolayer, when coupled to Pb, and its proximation to SC below Pb T_c.



Thank you for the attention!!!