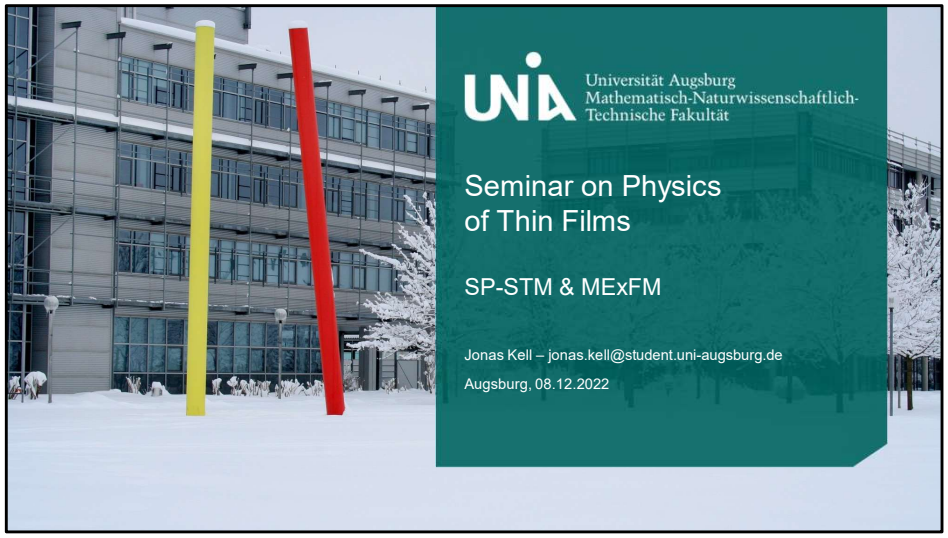

Universität Augsburg
Mathematisch-Naturwissenschaftlich-
Technische Fakultät
Seminar on Physics
of Thin Films
SP-STM & MExFM
Jonas Kell – jonas.kell@student.uni-augsburg.de
Augsburg, 08.12.2022

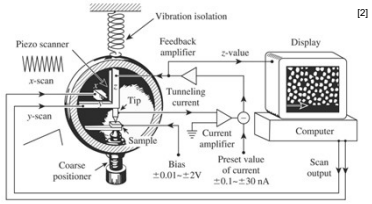
Outline: Spin-Polarized-Scanning-Tunneling-Microscope

1. General Functionality & Construction
2. Spin Resolved/Polarized STM (SP-STM)
3. Experiments & Applications
4. Outlook: Magnetic Exchange Force Microscopy (MExFM)
5. Summary and Conclusion

Construction & Current Feedback Mechanism

General Functionality & Construction

- Piezo electric crystals allow for very fine control
- Distance sample ↔ tip < 1nm
- Overlapping electron clouds allow for tunneling current to flow between tip/sample
  - Direction reversable, depending on bias voltage sign
- Feedback circuit keeps the perceived tip ↔ sample distance constant
- Combination with different control signals produces different scanning modes
  - A-Scan (dot, often time resolved)
  - B-Scan (line)
  - C-Scan (surface)



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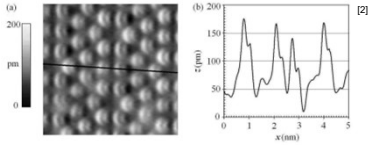
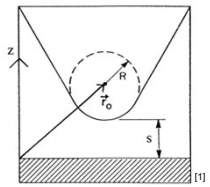


[2]

Mathematics of the Tunneling Process

General Functionality & Construction

- The tunneling current depends:
  - On the "local density of states" (LDOS, here $\mathbf{n_S}$)
  - The bias voltage **U**
  - The distance to the sample **S**
    - The exponential dependency is key to the high resolution capability



[1]

$$I = \frac{2\pi e}{\hbar} U \sum_{\mu,\nu} |M_{\mu\nu}|^2 \delta(E_\nu - E_F)\delta(E_\nu - E_F)$$

$$M_{\mu\nu} = \frac{-\hbar^2}{2m} \int dS(\Psi_\mu^* \nabla \Psi_\nu - \Psi_\nu \nabla \Psi_\mu^*) \qquad n_S(E_F, \vec{r}_0) = \sum_\nu |\Psi_\nu(\vec{r}_0)|^2 \delta(E_\nu - E_F)$$

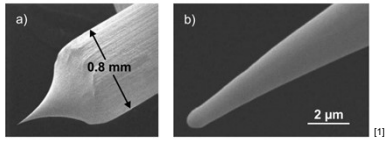
$$\Psi_\mu = \frac{1}{R} e^{-\kappa R} \qquad I \propto \exp(-2\kappa s)$$

- The equation for the tunneling current is derived from first-order time-dependent perturbation theory
- Psi_mu is the electronic state of the tip, Psi_nu the of the sample/surface
- S-type wavefunction, kappa is the inverse decay length !exponential decay!
- M is the tunneling matrix, where the wavefunctions are describing the unperturbed states
- n_S is the Local density of states at the fermi level

Environment & Generating Tips

General Functionality & Construction

- Temperature range generally in the region of mK to some K
  - Too high temperatures cause thermal fluctuations to overshadow measured effects
- Measuring at atom-scale resolution generally requires ultra-high-vacuum (UHV) setups
  - Kinetic energy of gas molecules causes noise through collisions
  - Tips get contaminated with oxides
- Tip preparation
  - Generally performed *in situ* to avoid contaminations
  - Pulling procedures
  - Electrochemical etching methods
  - Cleanup through electron/ion bombardment



[1]

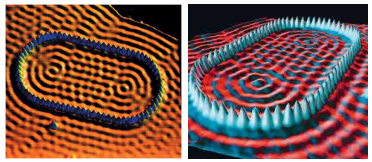
- In situ (in the UHV experiment) / as opposed to ex-situ (before pulling the vacuum)
- Pulling to generate nanowires generally a one-step process that is quite refined if performed in situ. Otherwise cleanup
- Electrochemical etching leaves often oxides and other residue. Cleanup through bombardment is generally required

Example: Manipulation of individual atoms

General Functionality & Construction

- Active manipulation of the sample on the per-atom basis
- Constructive process to “drag” atoms into desired locations
- Multiple applications
  - Data storage
  - Sample preparation (for other measurements)
  - Validation of theorized wavefunction behavior

- “Quantum Corrals”



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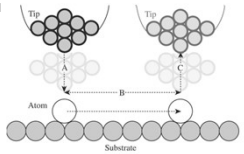

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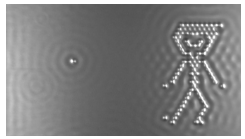
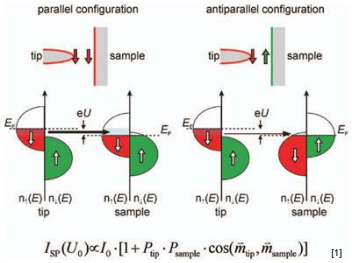


- Process to move individual atoms:
  - Locate atom
  - Increase tunneling current to bring tip closer
  - Interaction energy reaches the “diffusion activation energy”
  - Atom can be dragged over the potential wall between the stable atom lattice positions
  - Gradually decrease set tunneling energy to move tip away from the ad-atom
- Stray Atoms on surface are called “ad-atoms” (adsorbed atom) <-> is opposite of vacancy
- Quantum corrals show electronic density as wavefunction, not only shape of standing waves in a potential well, but standing wave effect that shows reflection of the wall (only on one location in the focus points of the ellipse is an atom. The other site is empty. Nevertheless they show the same electronic signature.)
- Images curtesy to the research at IBM (Don Eigler)

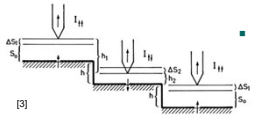
Theory of the Spin Resolved STM

Spin Resolved/Polarized STM (SP-STM)

- The model for a spin-dependent ferromagnet-ferromagnet tunnel junction is applied
- The local densities of states for tip and sample
  - $n_t = n_t^\uparrow + n_t^\downarrow, \quad n_s = n_s^\uparrow + n_s^\downarrow$
  - $m_t = n_t^\uparrow - n_t^\downarrow, \quad m_s = n_s^\uparrow - n_s^\downarrow$
  - $P_t = m_t/n_t, \quad P_s = m_s/n_s$
- First time experimentally confirmed for stepped antiferromagnetic coordinated, stepped chromium surface



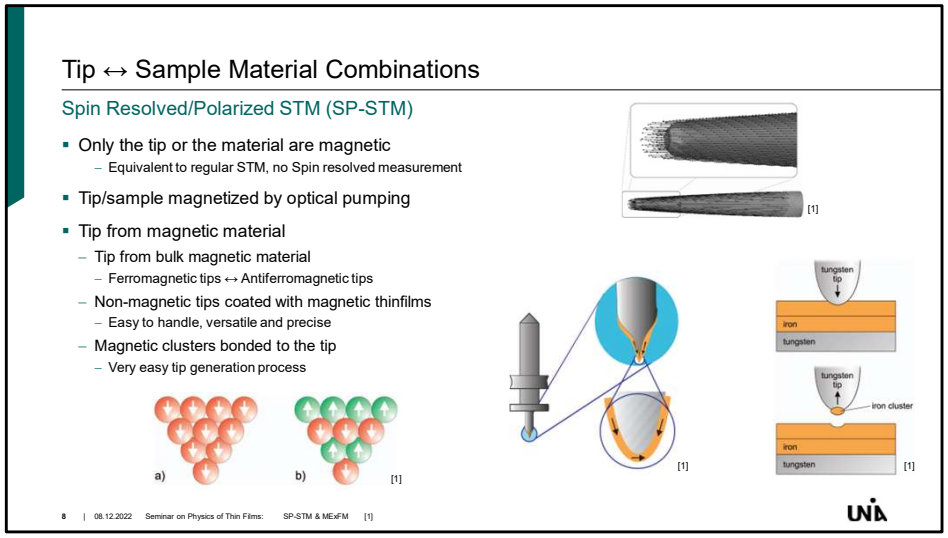
$$I_{SP}(U_0) \propto I_0 \cdot [1 + P_{tip} \cdot P_{sample} \cdot \cos(\vec{m}_{tip}, \vec{m}_{sample})] \quad [1]$$



[3]

- Sample composition
  - Cr(001)
  - Steps non-magnetic: 0.144nm
  - Steps magnetic: 0.12nm/0.16nm

- The tunnel junction exactly provides the 1 + cos(m*m) dependency
- n is the local density of states, m is the “magnetization”, P is the polarization
- First SP-STM experiment (1990)
  - Cr(001) (Chromium) surface was measured in STM with a tungsten tip. The step is monoatomic and has a height of 0.144nm
  - When switching to a CrO2 magnetic tip, the heights alternate between 0.12nm and 0.16nm. The contribution results from the antiferromagnetically polarized sample layers



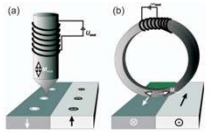
- Optical pumping was the first used method (initially GaAs, galliumasenid was proposed).
    - Either the tip or the sample could be optically pumped, however pumping the tip did not produce great results
    - Has the advantage, that strength and polarization of the magnetization can be controlled
- Tip preparation must be performed “in situ” to avoid oxidation (should already have been said. Just reminder)
- Bulk magnets may pose problems because of the magnetic effects of the stray fields.
    - Antiferromagnets avoid this
    - Switching of the apex spin orientation is only possible with high external magnetic fields -> sample must be hard against magnetic changes
- Thinfilms are quite nice, because they can be applied with established deposition techniques over the surface of tips that are generated with established preparation methods
    - Clean with electron bombardment, remove oxides with high temperature flash, grow known combination of material and layer thickness to control the magnetization direction
    - Thinfilms allow for great control over the direction of the spin at the apex of the tip. (in-plane or out of plane)

- Voltage pulses on a non-magnetic tip over a magnetic surface may be used to obtain a cluster of magnetic material at the tip
    - Orientation of the magnet can be changed with additional voltage pulses
    - Simplest method for getting magnetic tip
    - Dipping into the material also a possibility

Modes of Operation

Spin Resolved/Polarized STM (SP-STM)

- Constant-current
  - Default imaging mode of STM
  - Feedback circuit makes sure, that the tunneling current is kept constant
  - The smallest magnetic superstructure will be imaged with SP-STM, not necessarily the atomic structure
- SR-spectroscopic
  - Aims to provide separation between topographic structure and electronic structure
  - Probing bias voltage is modulated
  - Extracts energy dependency of the local electron density of states (LDOS)
  - Need to be measured at fixed tip ↔ sample distance (either stationary or secondary ample-and-hold amplifier)
- Modulated tip
  - Aims to provide separation between electronic structure and magnetic structure
  - Magnetization of the tip is modulated periodically
  - Additionally requires bias voltage modulation and tip distance modulation



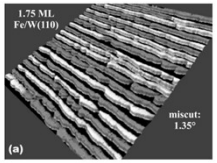
[1]

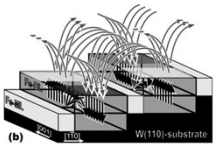
- Modulated tip is said in source [1] to be too complicated,
  - As it requires the use of the other two strategies alongside itself
  - As well as the fact that the magnetization of thin films can also be modulated with external magnetic fields
  - But can achieve close to atomic precision if needed
  - Stray fields from the ferromagnetic tip relatively easy affect measurements
- Basically, very tiny hysteresis loop

Nanostripes on stepped Surfaces

Experiments & Applications

- Nanostripes get formed when material is deposited on stepped substrate
- Magnetic effects may emerge that depend on the topology
  - Influence of the width?
  - Influence of the height?
  - Influence or the distance between stripes?





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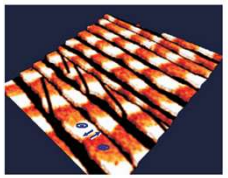
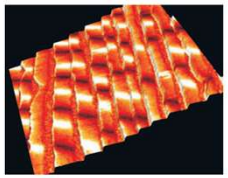
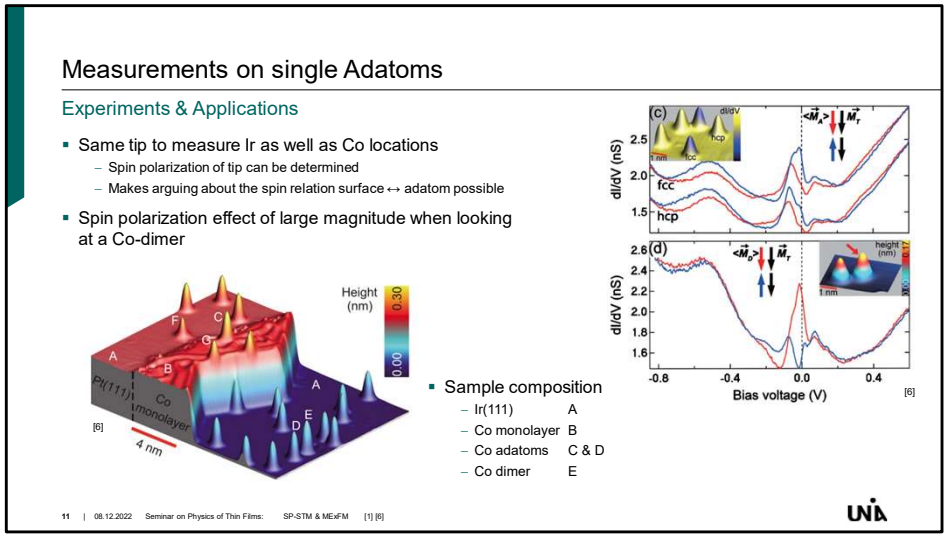




[1]

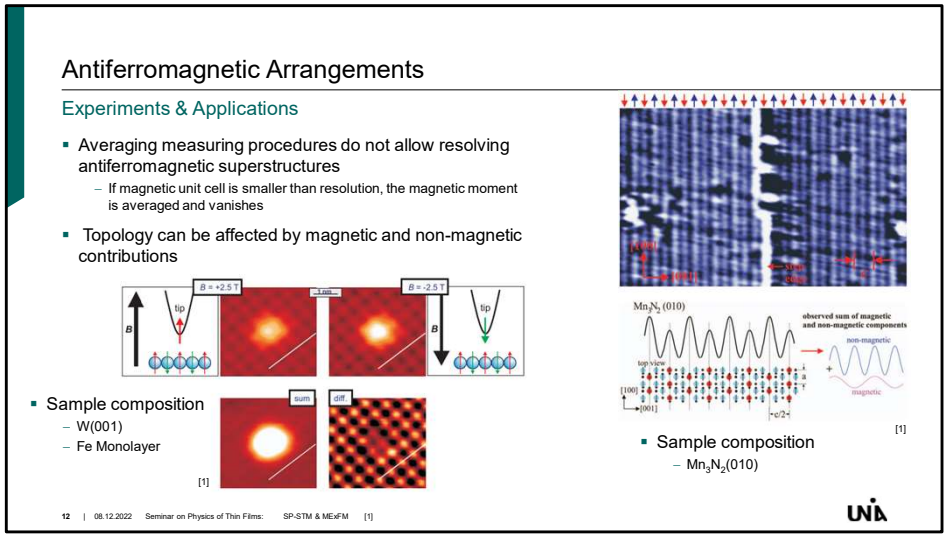
- Sample composition
  - W(110)
  - Fe monolayer
  - Fe doublelayer
  - Left, stripe-width > 10nm, right < 10nm

- W (tungsten, German: wolfram)
- Right image: grown in T~520K -> step-flow-growth
  - Monolayers magnetization is in plane, Double layer is out of plane
  - Has in particular strong effects on adatoms that are deposited close to nanowires/steps
  - Magnetic moment is neutral to the outside, as the magnetization of the double layer flips every other step
- Left image: for nanostripes with bigger width, the stray field is reduced by a periodic reversal of the direction of the magnetic moment inside the nanostripe



- Surface is stepped Pt(111) (platinum)
- On the surface, Co (copper) is deposited. It forms a nanostripe and deposits multiple adatoms on the base layer
- 300 mK STM, up to 12T B field, constant current mode, tip from polycrystalline tungsten wire, electrochemically etched and coated with ~45 atomic layers of antiferromagnetic Cr (chromium)
- Comparison to the flat spots at different magnetic field strengths (high coercivity of Cr!) allows for the alignment of tip <-> sample parallel and antiparallel on demand. This is used to calibrate the tip and get the spin direction of the tip
- Scans are performed mostly in location over one singular adatom or one dimer. They are stationary, but the bias voltage is modulated, to get an energy-dependent value for the LDOS (local density of states) -> different electron modes
- Differences can be seen, for the single adatoms, that can either adhere in fcc (face-center-cubic) or hcp (hexagonal-close-packed) configuration. These are mostly independent of the tip magnetization direction
- A large effect is noted for the scanning of a Co-dimer (two Co atoms, here two Co atoms, located as a close-packed dimer on two neighboring fcc sites)

- The effects basically resemble 1:1 the structure of the single adatom. This is expected, as the second atom should only broaden the line
- BUT at -0.2 V (! This resembles the fermi energy in that case !) a huge sign dependency of the magnetic moment can be noticed.
    - This is probably due to the following effect:
    - at the fermi energy, s, p and dz^2 electrons contribute the most because of their high symmetry
    - they might have a spin state opposite to the rest of the d-orbitals
    - For single adatoms a high degree of rotational symmetry is present -> large contribution
    - For dimers, the rotational symmetry is largely lifted and the contribution from the high-symmetry orbitals is reduced -> sign flip



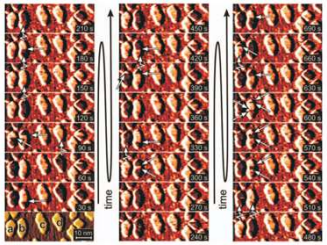
- W (tungsten, German: wolfram), Fe (iron)
- Left image: adsorbate atoms used as marker, to align the two scan passes properly and allow to take the difference
- External magnetic field of 2T is enough to re-align the tips magnetization direction
    - Allows to measure in magnetic and non-magnetic mode
    - Allows toggling between in-plane, out of plane and inversed magnetization direction
- Sum of passes gives information about the surface topography
- Difference of passes gives information about the magnetic superstructure (! It is even visible at the location of the adsorbate, indicating the superstructure is unaffected by the adsorbate)

- Mn_3N_2(010) (Manganese nitride)
- Right image: magnetic structure shows at room temperature
- Important: multiple strictures and periodicities stacked inside each other -> structure may be more complex, even if a quite high resolution is already chosen

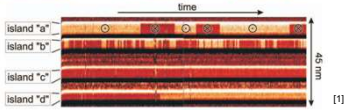
Time Resolved Experiments

Experiments & Applications

- SP-STM does not only image in real-space, but consequently also in the real-time domain
- Time resolution depends on imaging mode and spacial resolution
- Switching events most of the time thermally induced
  - May also be a result of the probe's influence or an external magnetic field

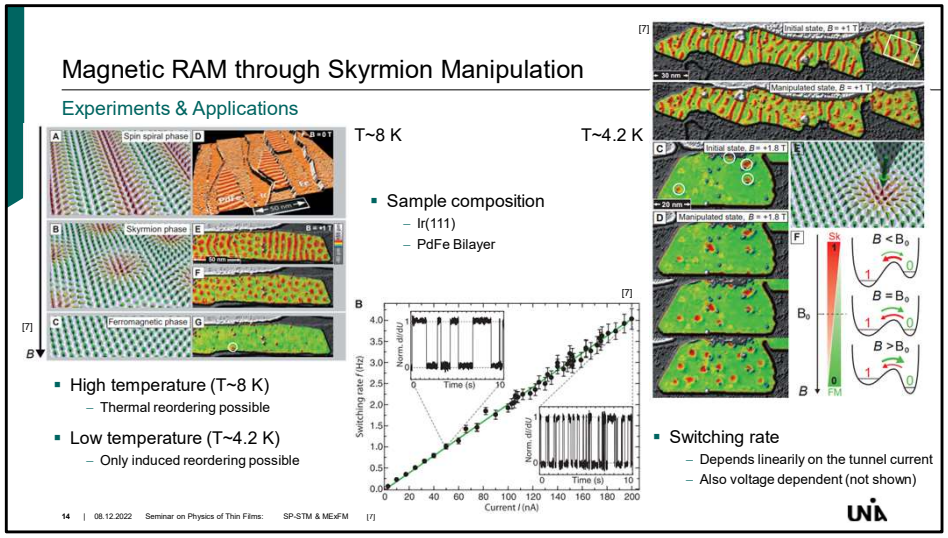


- Sample composition
  - Mo(110)
  - Fe islands (area < 40nm²)
- Antiferromagnetic tip required
  - Magnetic stray field of a ferromagnetic tip would influence switching behavior



[1]

- Upper image is relatively low time resolution, but the whole islands get imaged in 2D (C-Scan)
- The lower image is only a B-Scan. The time resolution may therefore be much higher.
- Switching to the A-Scan (Point-scan), will be performed on the next slide, even higher time resolution may be achieved
- Mo (molybdenum), Fe (iron)
- The switching of the spin polarization was observed for nanoislands smaller than 40nm^2
- The switching frequency was observed to depend on the size, as well as on the shape (more elongates switches more rapidly)
- Neighboring nanoislands have correlated switching (example islands a and b)



- Manipulation of Skyrmions is tested to be applicable as MRAM (magnetic random access memory) function.
- The branch that tests this (electronics using spin) is called “Spintronics”
- Ir(111) (Iridium) is chosen as a base, a Pd(Palladium)Fe(Iron) Bilayer is added (pd monolayer as a very defined and easily manipulatable magnetic structure)
- Tip is sometimes magnetically out-of-plane sensitive and sometime in plane (depending on figure, not really important)
- Left side: High temperature. The system undergoes two phase transitions if the magnetic field is raised from top (0T) to bottom. The system returns to the base state if the magnetic field is switched of, as thermal fluctuation’s energies are enough to traverse the phase transition energy gap
- Right side: Low temperature. The system undergoes the same transition but not on its own. Only when scanning with a comparably high tunnel bias voltage, the phase transition is undergone.
- In that metastable state, the individual Skyrmions can be created or deleted when the local area is scanned with tunnel electrons.
- The Skyrmions fluctuate between existing and not existing when scanned. For very small voltages the flipping is

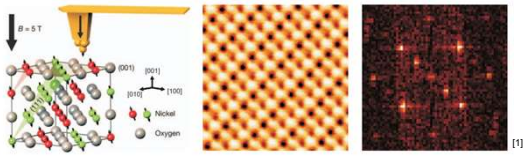
basically impossible. This allows for scanning without modifying the sample.

- For a high enough current, toggling the Skyrmions follows a frequency linear dependent on the current. This allows for precise control over the Skyrmion’s state

Spin Resolved Microscopy for Insulators?

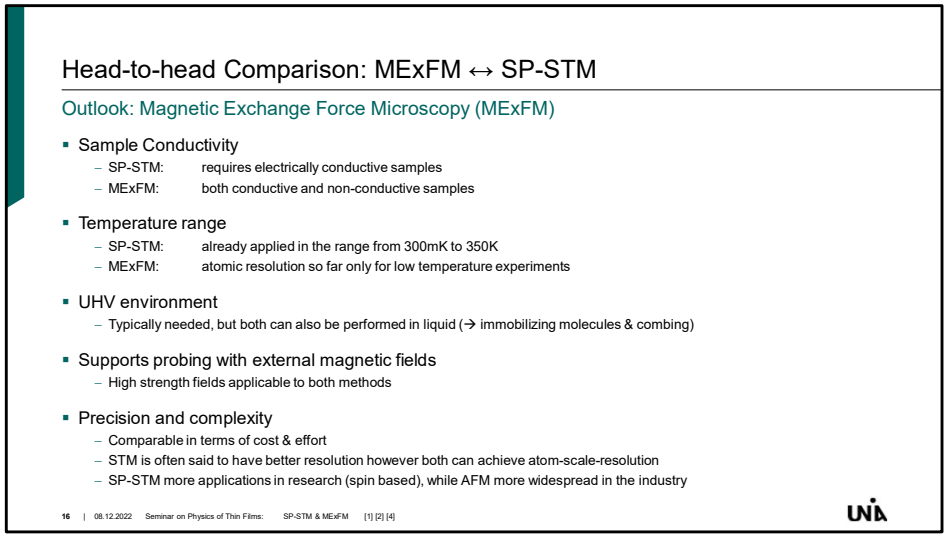
Outlook: Magnetic Exchange Force Microscopy (MExFM)

- No conductive samples needed
  - Electromagnetic exchange forces are used to deflect a cantilever that carries the probe tip
- Modes of operation
  - "Contact-mode": static mode of operation, tip is dragged along the surface
  - "Tapping-mode": dynamic mode of operation, cantilever is exited to oscillate and bring the tip very close to the sample
  - "Non-contact-mode": dynamic mode of operation, oscillating of the tip near resonant frequency, interaction forces change the resonant frequency
- Magnetized tips may be used to resolve spin dependent interaction forces



[1]

- Image:
  - 1) Schematic view of the NiO(001) (nickel, oxygen) being probed with an atomic force microscope
  - 2) Spin resolved image of the insulator
  - 3) Fourier transformation of the raw data. Shows two spots that correspond to the magnetic superstructure period of the antiferromagnetic spin ordering



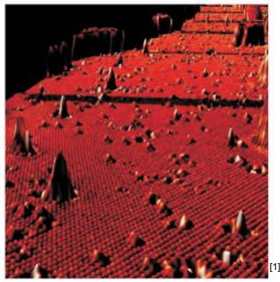
- SP-STM needs electrons to be conducted. MExFM does not require them
- Source [1] states the temperature rage. However if no atomic resolution is required, of course it is possible to use the methods in higher temperature environments
- Source [1] states the UHV is required. Other sources state that the solid-liquid interface is available to the STM AFM mechanic, while this was impossible for previous methods that required UHV
    - The ability to use liquids, opens the possibility to probe organic samples, that are only active in liquid solution
    - They are frequently moved or pinned with the probe tip. Flowing liquid across a pinned molecule can be used to orient it
- Mostly the spin-resolved research requires external magnetic fields for measuring and calibrating
- Most effort and time is required to prepare the tips. Source [1] states this is equally complicated.
- STM bit more advanced and better developed. Because tunnel effect has exponentially decaying reach, it is said to better guarantee only interaction with the tip atom, not the rest of the tip. AFM can have non-confined interactions -> less resolution
- Depending on the application, one or the other method may be better applicable

Final Overview

Summary and Conclusion

- Methods allow for probing on atomic scale
  - Measurements are performed in real-space, contrary to most atom-scale measurement methods
- Magnetic moment can be resolved locally
  - Avoids missing substructures with on average neutral magnetic moments
- Possible for metals, as well as insulators
  - SP-STM for metals
  - MExFM for insulators
- Comparably high time-resolution
  - Reducing resolution allows to study the changes of magnetic moments close to real time
- Active manipulation of sample possible (on measurement-scale)
  - Construct custom test environments for wavefunctions
  - Manipulate/create individual molecules for chemical research

Thank you for your kind attention


[1]

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