

ASPIC-reloaded

Upgrade of the UHV-system ASPIC for the investigation of surfaces and two-dimensional materials by ultra-low energy implantation and deposition of radioactive probe atoms

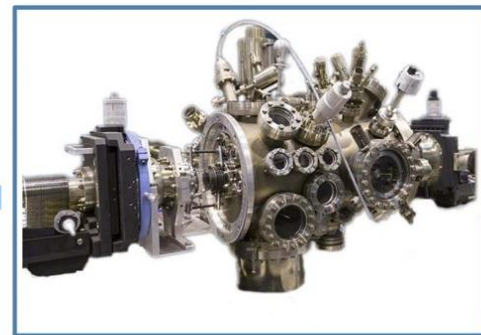
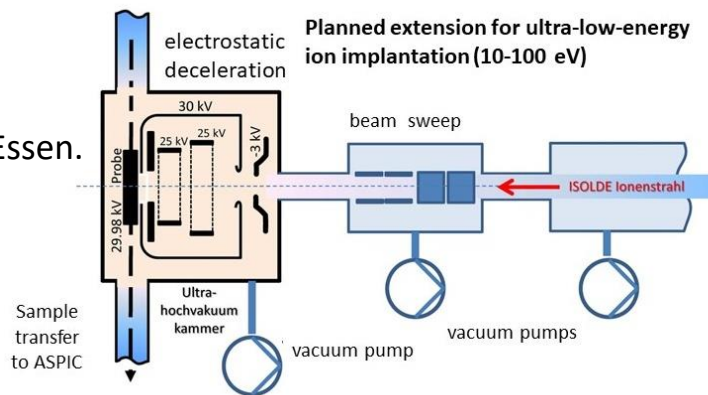
Hans Hofsäss and Koen van Stiphout, Faculty of Physics, University Göttingen

Consortial BMBF Project “Ecomarl”

- Hans Hofsäss from University Göttingen,
- Peter Schaaf from TU Ilmenau
- Doru Lupascu from University Duisburg-Essen.

BMBF 05K19 MG1:

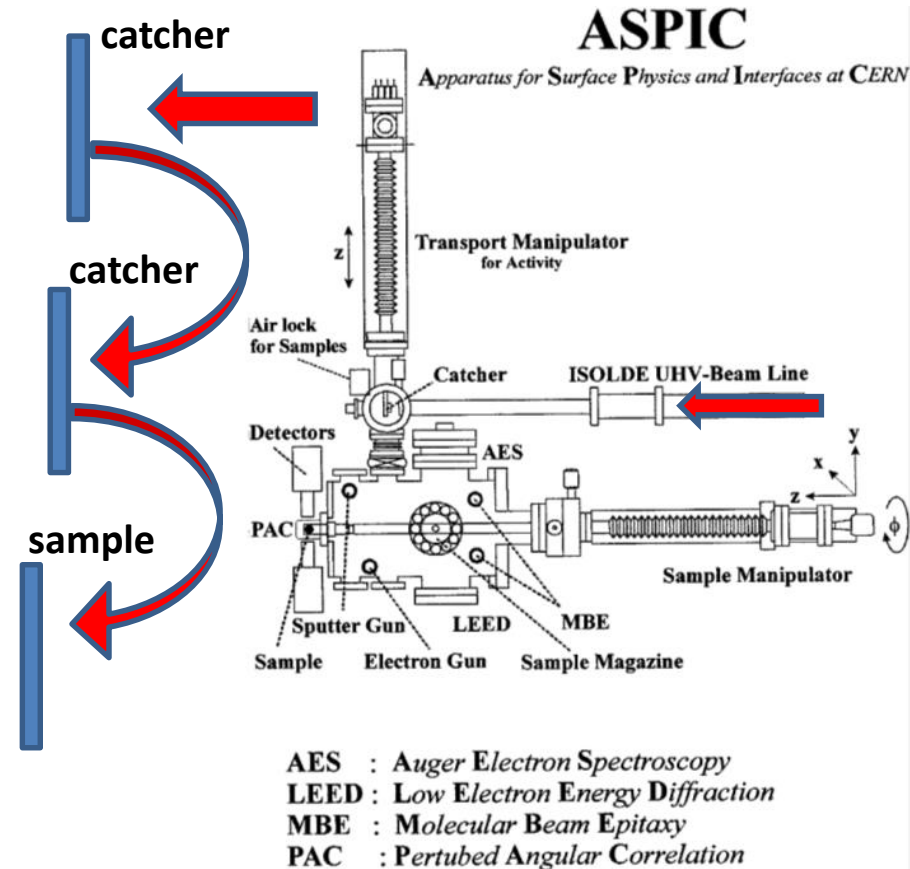
Total budget 2019 - 2022: 483.000 €



Existing apparatus for surface and interface physics (ASPIC)

History of ASPIC

- 1994 deposition of radioactive atoms on surfaces (H. Haas et al.)
 - Implantation into catcher, 2 stage evaporation onto sample surface
- 1995 Installation of Apparatus for Surface Physics at ISOLDE CERN (“ASPIC”)
 - Set up by HMI Berlin, now Helmholtz Center Berlin
 - Used until 2005
- 2014 proposed as UHV system at the beam-line VITO (versatile ion-polarized techniques online)
 - Only a UHV beamline was realized
- 2018 proposal to BMBF for ASPIC reloaded



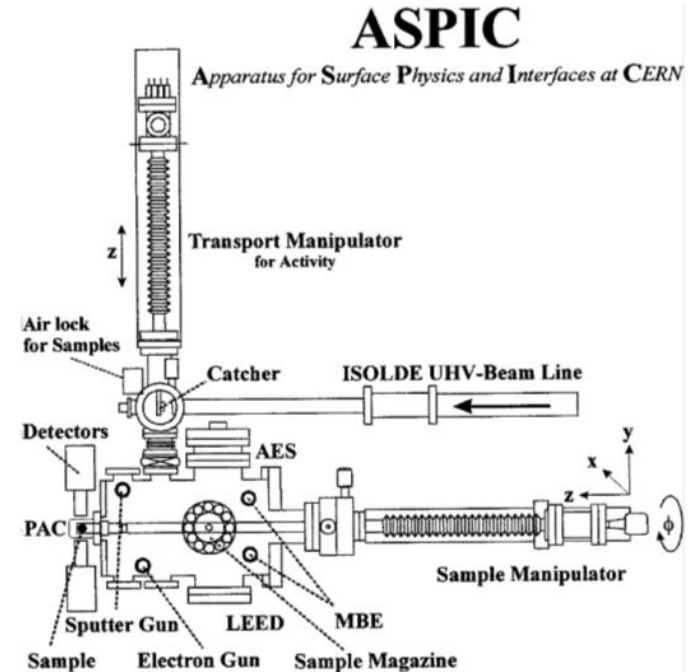
Features of the ASPIC UHV chamber

Versatile UHV chamber for surface physics

- **Sample surface analysis**
 - Auger electron spectroscopy (AES)
 - Low energy electron diffraction (LEED)

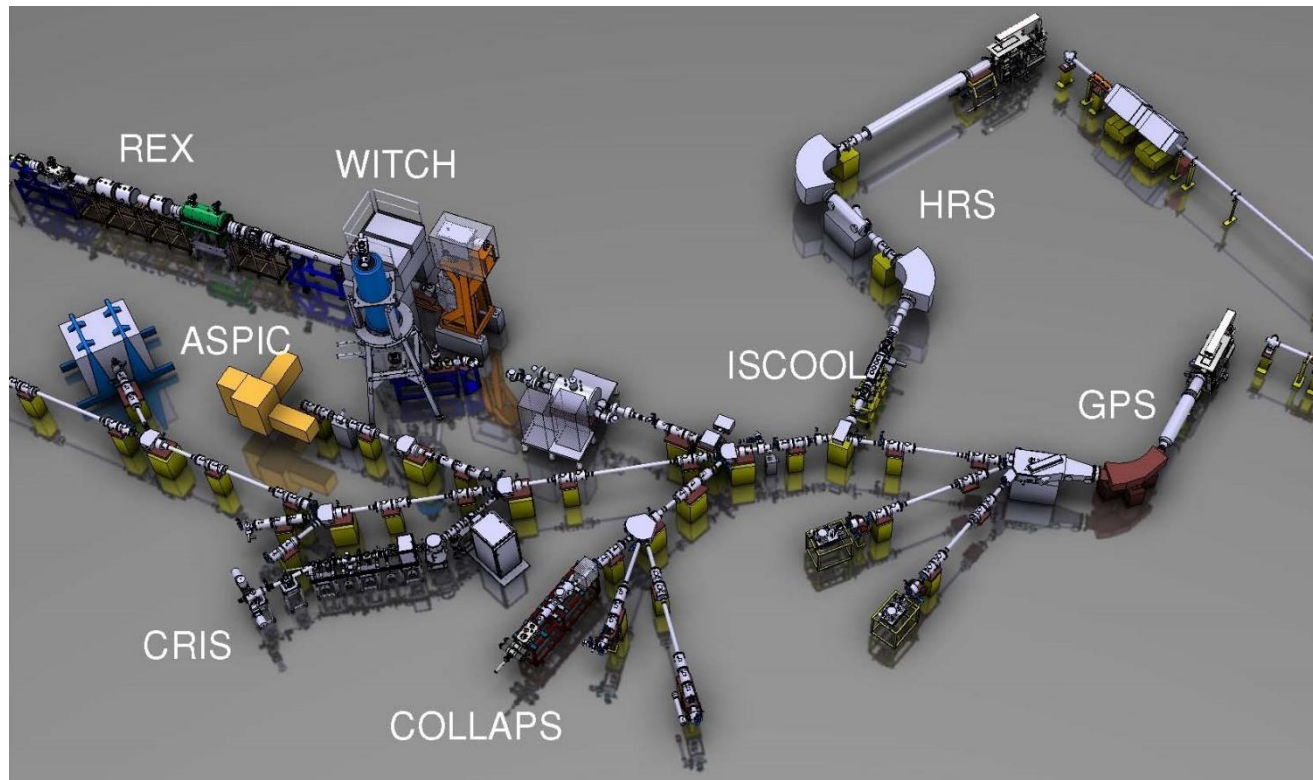
- **Sample preparation and manipulation**
 - Thin film deposition by molecular beam epitaxy (MBE) or vapor phase evaporation
 - Sample cleaning by sputter erosion
 - Sample annealing or cooling (to LN₂ temperature)
 - Electron beam heating

- **Perturbed $\gamma\gamma$ angular correlation**

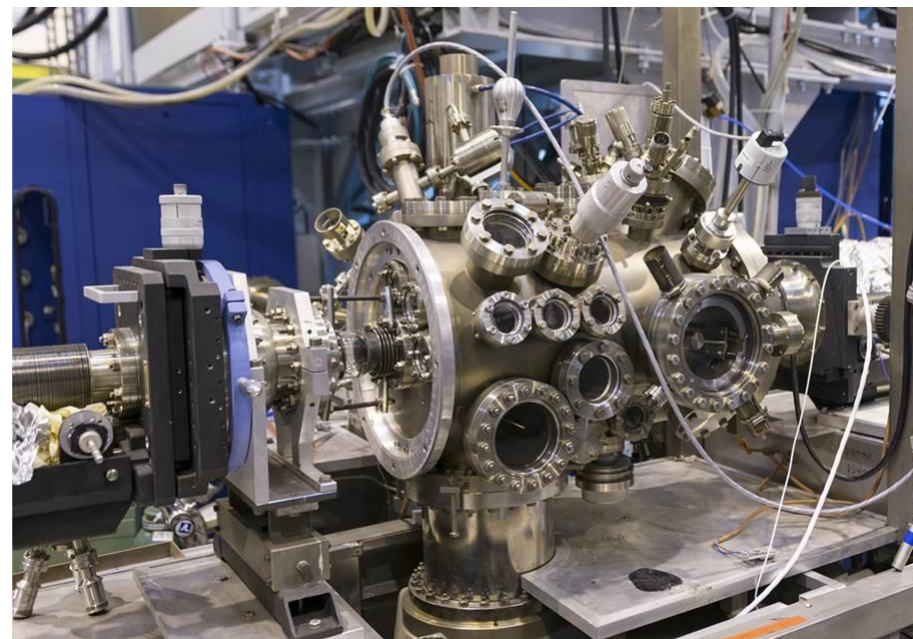
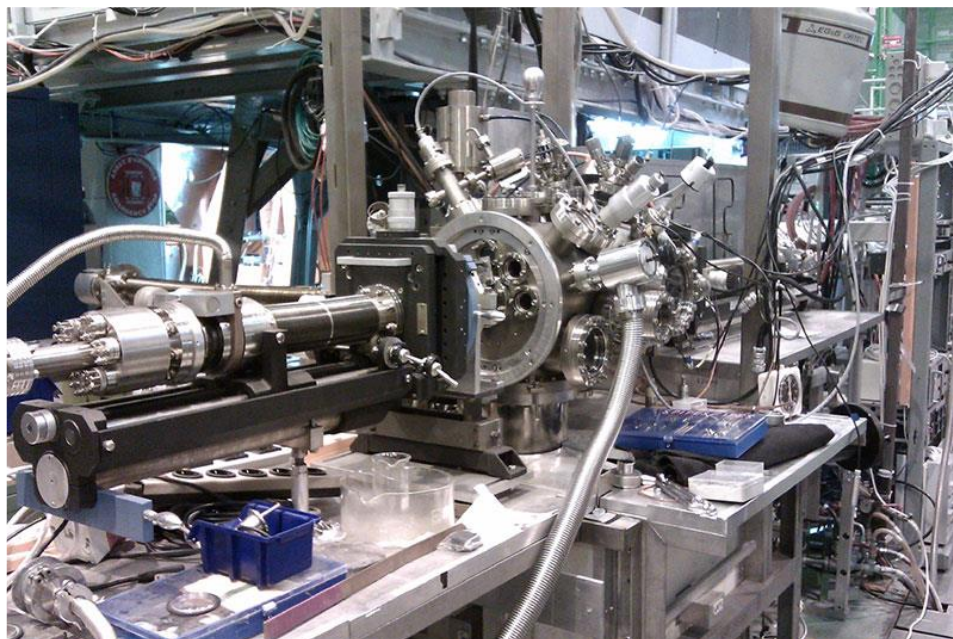


AES : Auger Electron Spectroscopy
 LEED : Low Electron Energy Diffraction
 MBE : Molecular Beam Epitaxy
 PAC : Perturbed Angular Correlation

ASPIC at the VITO beamline



ASPIC at the VITO beamline



Example: Adsorbate sites of In on a Ag(100) surface

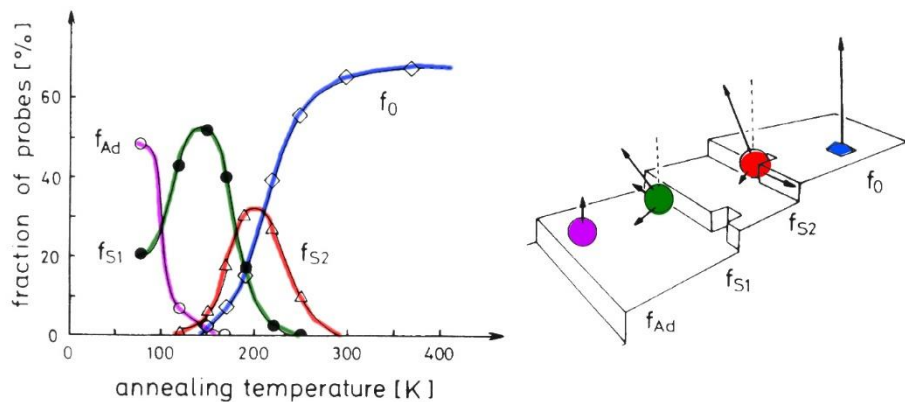


Fig. 5.23 Left side: Fraction of indium probes with different electric field gradients on stepped Ag(100) as a function of annealing temperature. Right side: Surface site model for indium probes suggested by the experimental parameters.

- Analysis of site fraction and electric field gradient orientation

R. Fink et al, Surface Science 225 (1990) 331

G. Schatz & A. Weidinger : Nuclear solid state physics

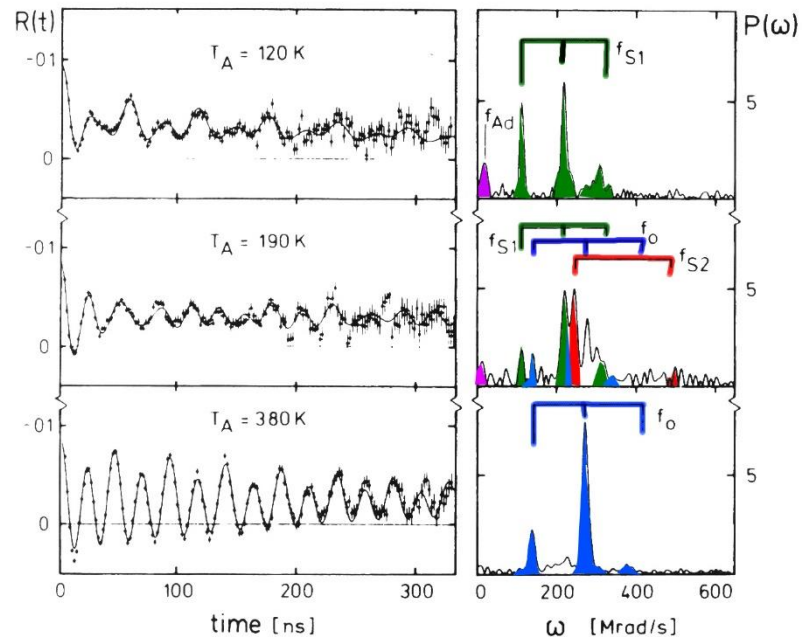


Fig. 5.22 PAC spectra along with their Fourier transforms for ^{111}In on stepped Ag(100) for three different annealing temperatures (annealing time 15 minutes, measuring temperature 77 K). The solid lines were fitted to the experimental points by Eq. (5.41).

Example: Magnetic hyperfine field for Cd on Ni surfaces

Substitutional terrace site

$$B = 6.6 \text{ T}, V_{zz} = 1.2 \cdot 10^{22} \text{ V/m}^2, \eta = 0$$

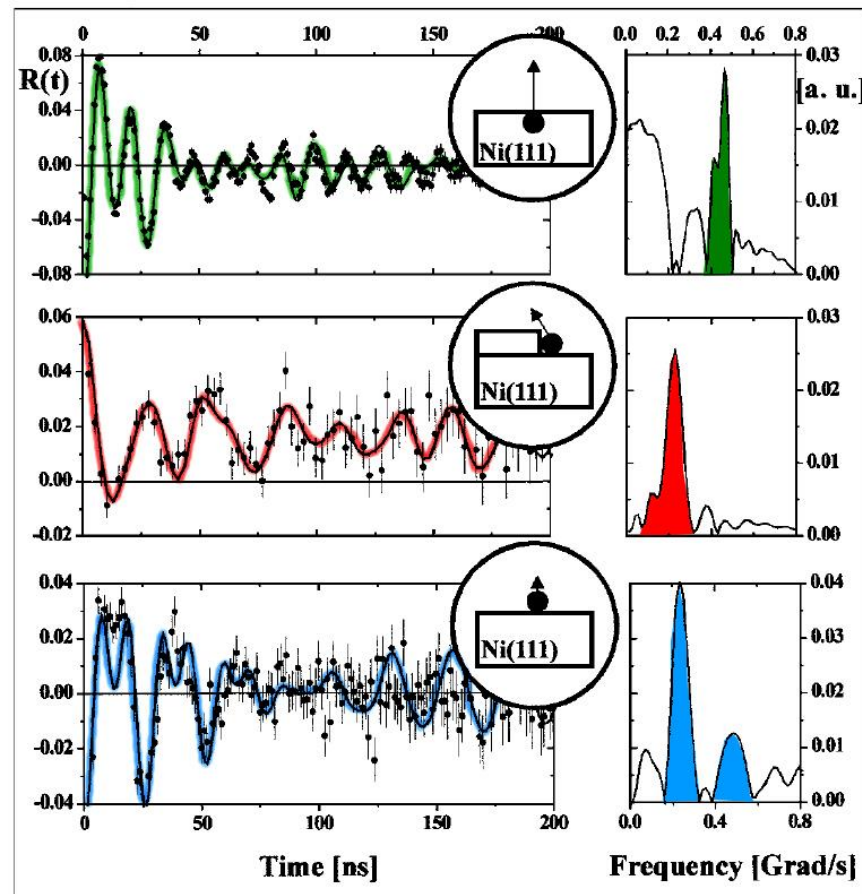
Edges and step sites

$$B = 1 \text{ T}, V_{zz} = 6 \cdot 10^{21} \text{ V/m}^2, \eta > 0$$

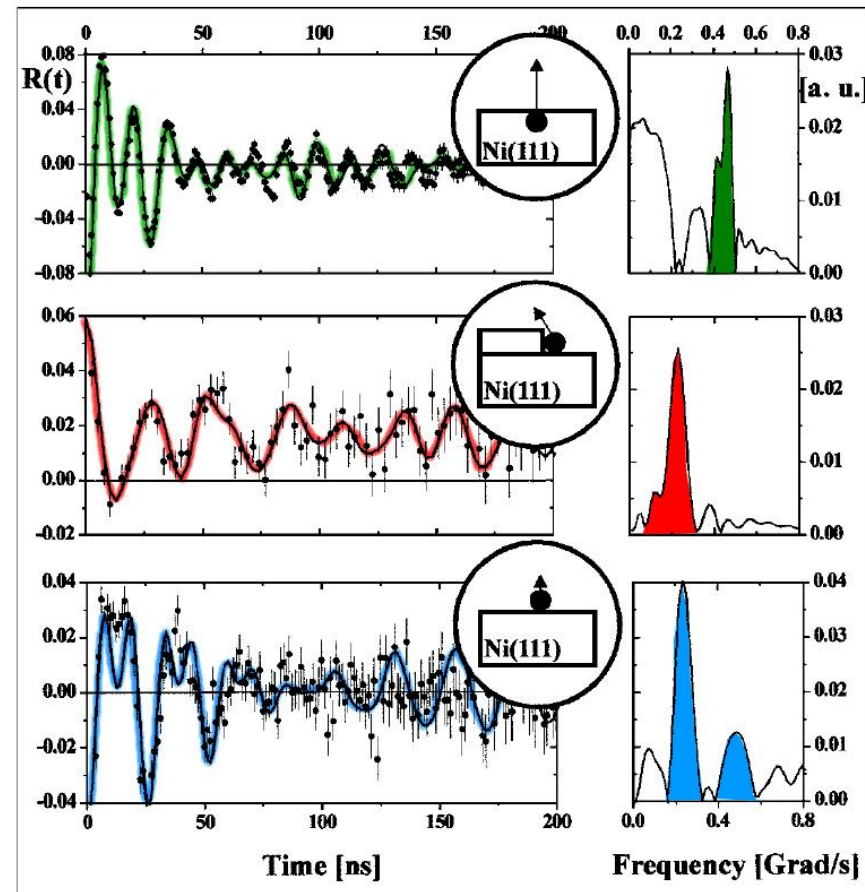
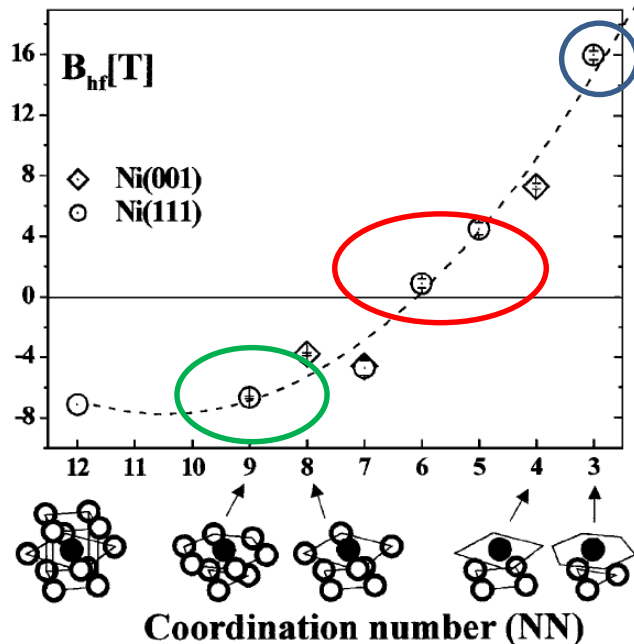
$$B = 4.11 \text{ T}, V_{zz} = 7.3 \cdot 10^{21} \text{ V/m}^2$$

Adatom site

$$B = 16 \text{ T}, V_{zz} = 1 \cdot 10^{21} \text{ V/m}^2, \eta = 0$$



Example: Magnetic hyperfine field for Cd on Ni surfaces



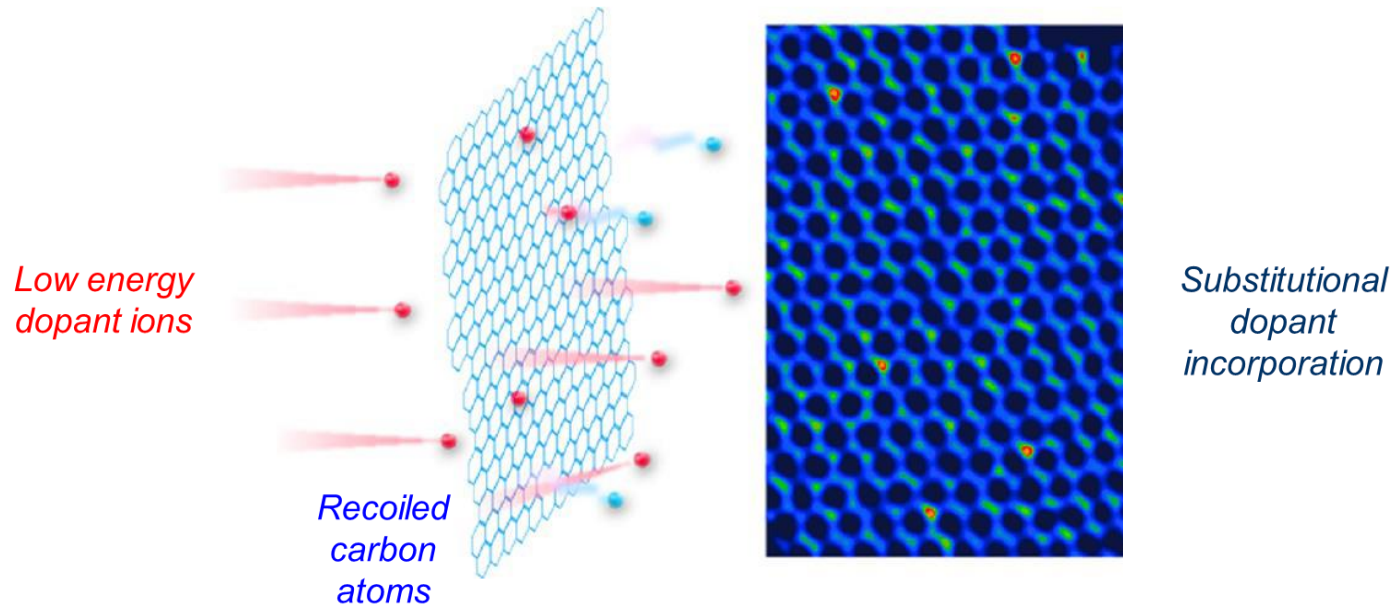
Potzger et al. , Phys. Rev. Lett 88 (2002) 247201-1

Ultra low energy ion implantation of graphene

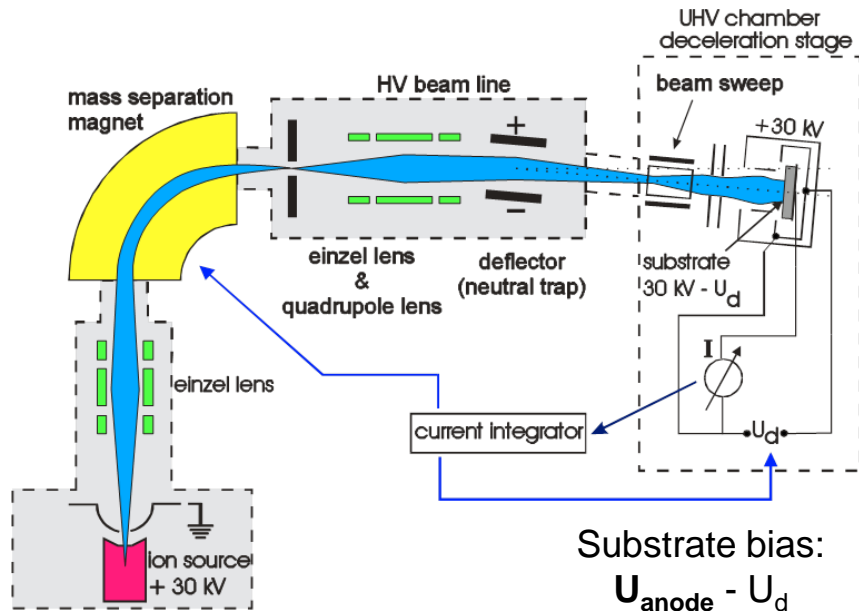
Why not a single binary replacement collision ?



Ultra-low energy, singly-charged mass-selected ion beams

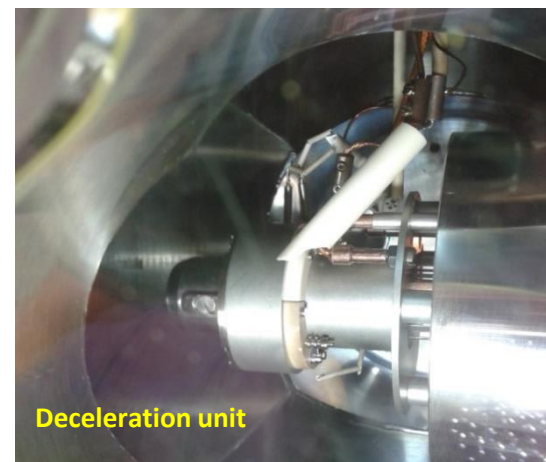


Mass selected ion beam deposition



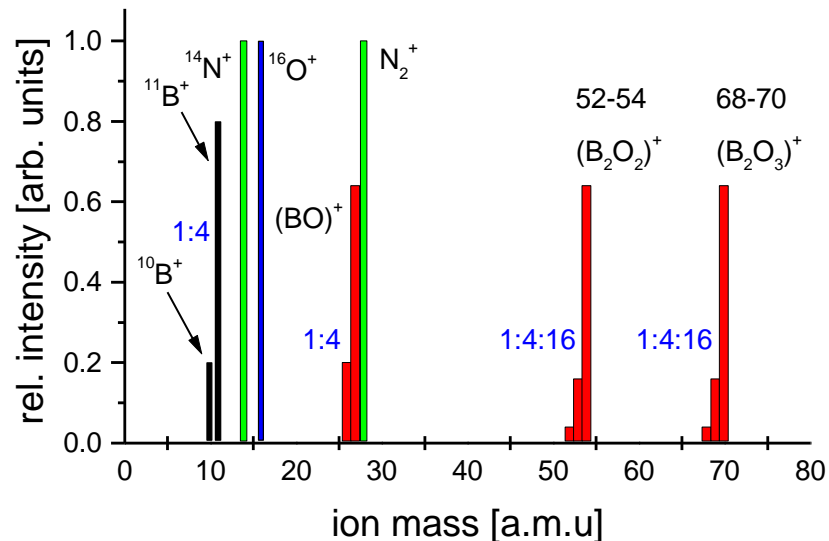
Substrate bias:
 $U_{\text{anode}} - U_d$

max. ion energy at the substrate
 $E_{\text{max}} = q \cdot U_d$

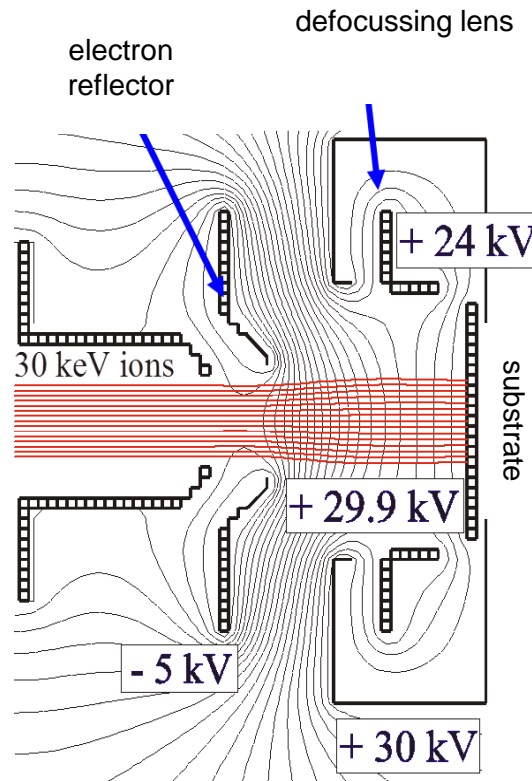


Features of the MSIBD system

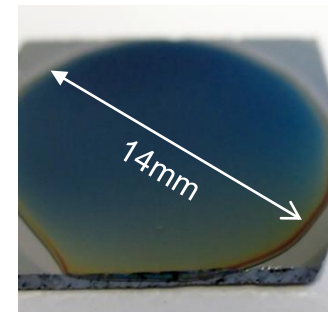
mass spectrum of ionized B_2O_3 vapor + N_2 -gas



- *Isotopically pure beams*
- *energy E_{max} down to about 10 eV*
- *Uniform deposition area 12 mm \varnothing*
- *Precise current measurement*
- *$P \approx 5 \cdot 10^{-10}$ mbar (UHV)*



a-C film from 100 eV C^+ ions



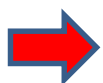
a-C film from 25 eV C^+ ions



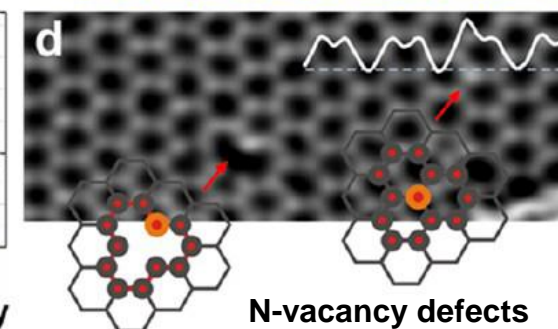
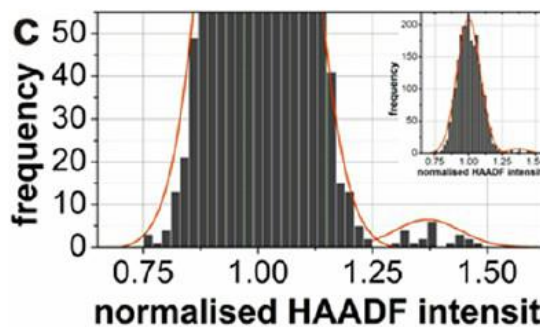
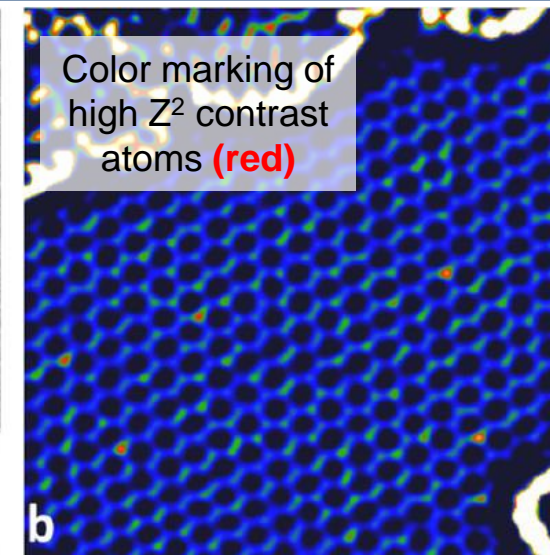
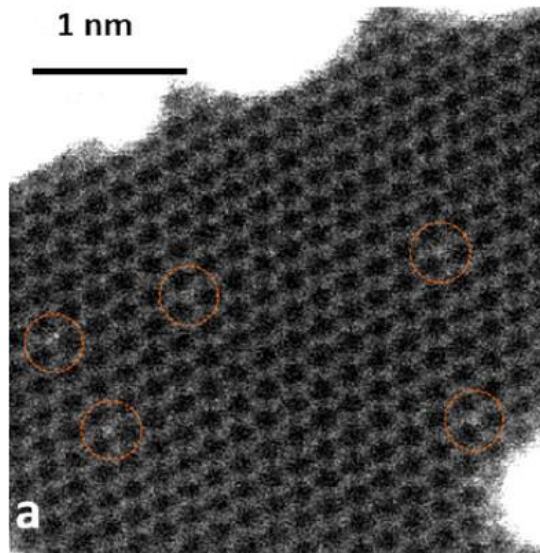
Ion implantation doping of free standing graphene

HAADF image of N-doped graphene

(High angle annular dark field)



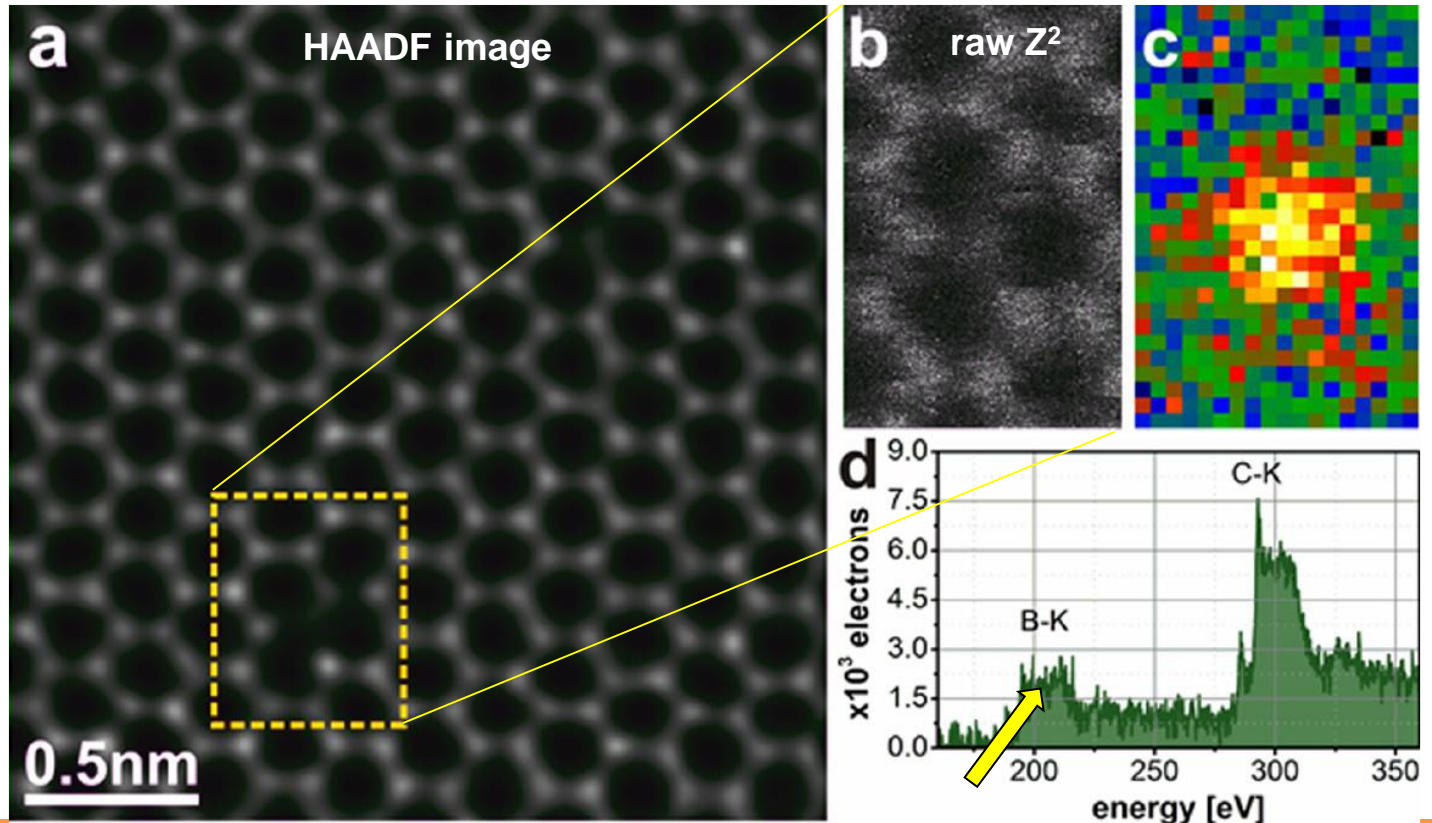
**substitutional
N-atoms**



U. Bangert, W. Pierce, D. M. Kepaptsoglou, Q. Ramasse, R. Zan, M. H. Gass, J. Van den Berg, C. Boothroyd, J. Amani, H. Hofsäss, *Nano Letters* **13** (2013) 4902

Ion implantation doping of free standing graphene

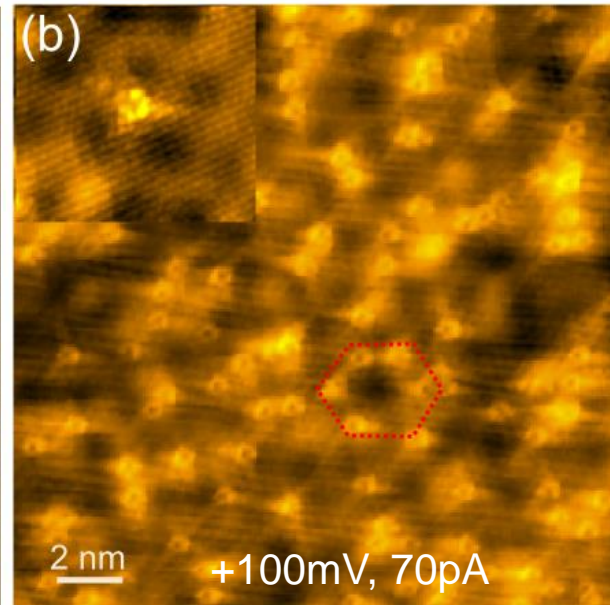
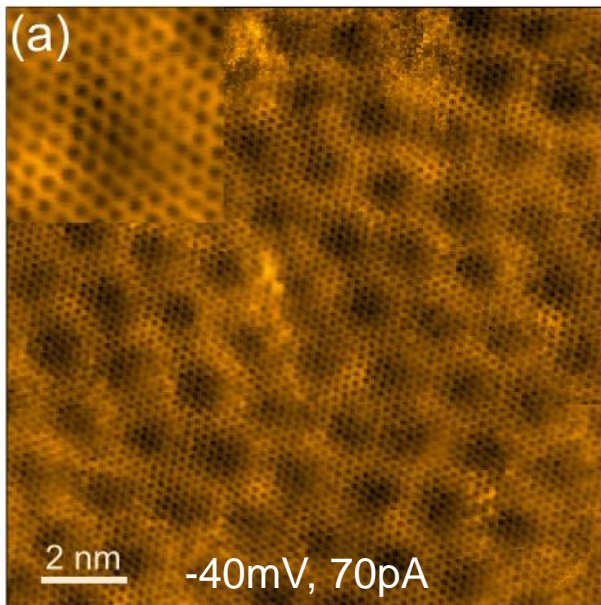
EEL @ 190-220eV



Ion implantation doping of graphene on SiC

Scanning tunneling microscopy

- a) pristine graphene on SiC:
graphene lattice &
6x6 corrugation
- b) N-doped graphene on SiC:
N scattering patterns with
3-fold symmetry

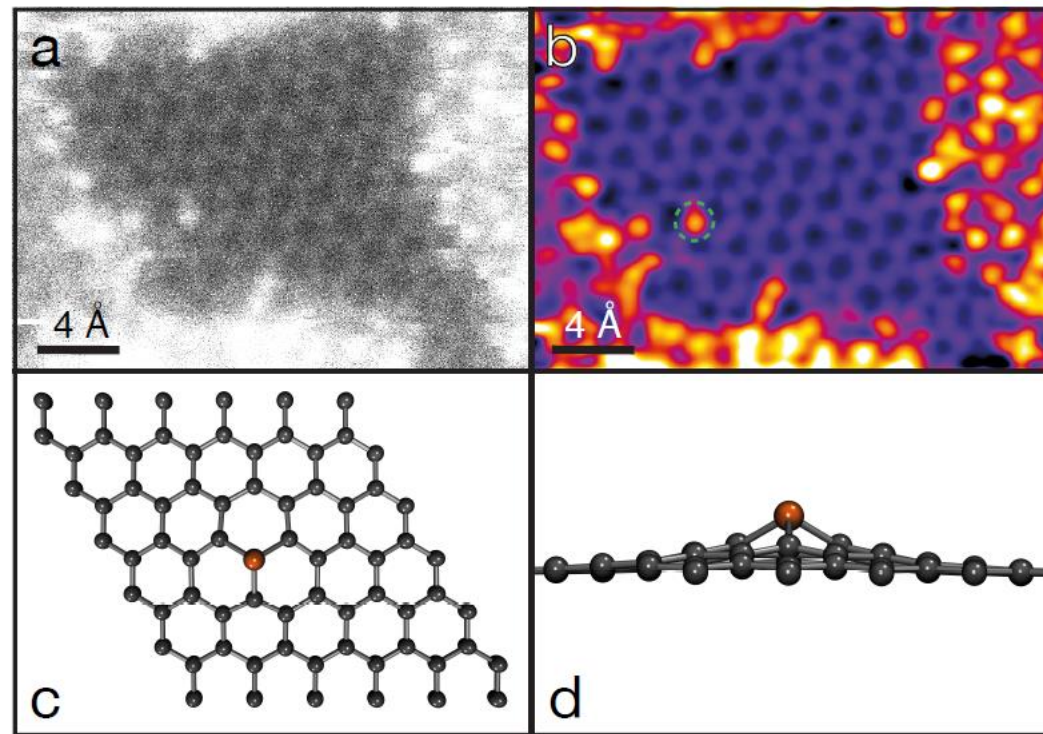


P. Willke, J. A. Amani, S. Thakur, S. Weikert, T. Druga, K. Maiti, H. Hofsäss,
and M. Wenderoth, **Appl. Phys. Lett.** **105** (2014) 111605.

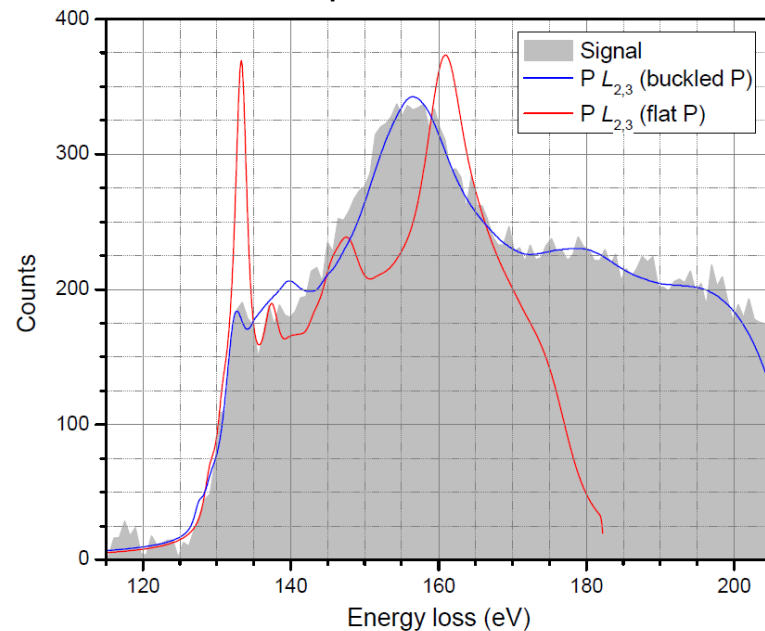
A substitutional P atom in graphene

unprocessed STEM/MAADF image

colored and filtered image



EELS Spectrum & simulations



T. Susi, T. P. Hardcastle, H. Hofsäss,
 A. Mittelberger, T. J. Pennycook,
 C. Mangler, R. Drummond-Brydson,
 A. J. Scott, J. C. Meyer, J. Kotakoski,
2D Mater. 4 (2017) 021013

A substitutional P atom in graphene

EEL spectrum averaged over the substitutional P dopant

energy windows from

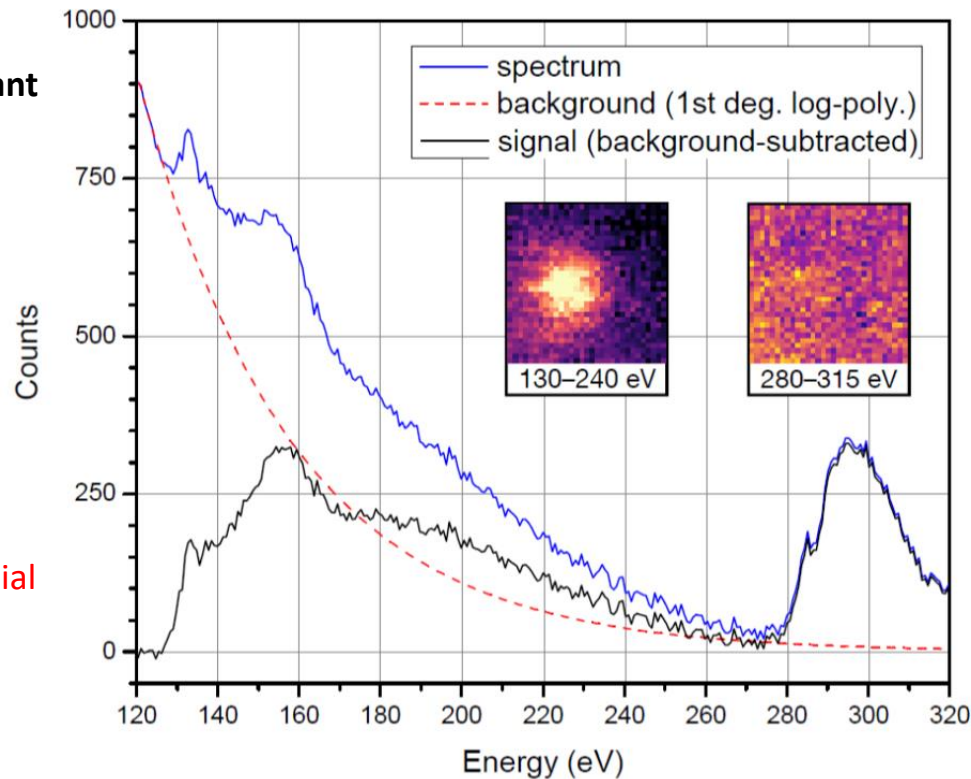
130–240 eV for the P L edge

280–315 eV K-edge.

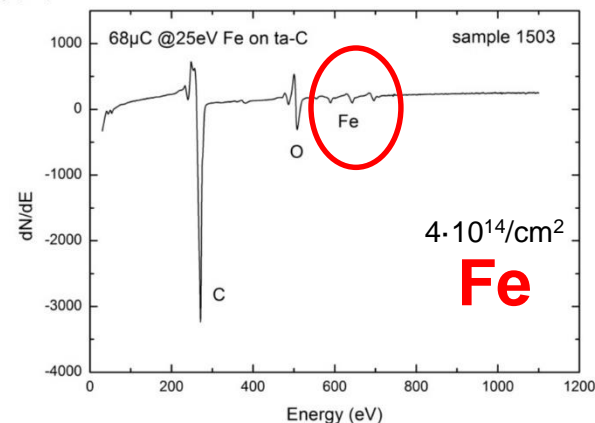
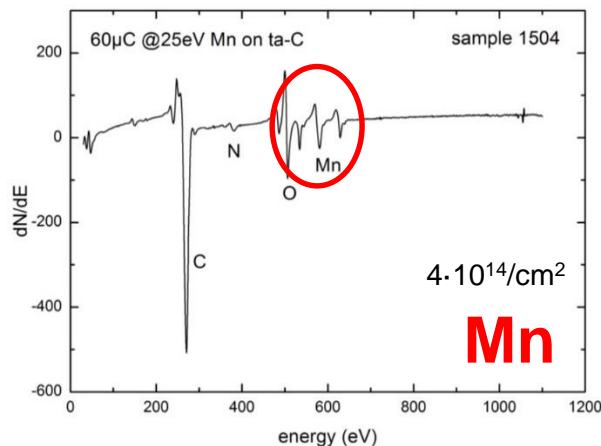
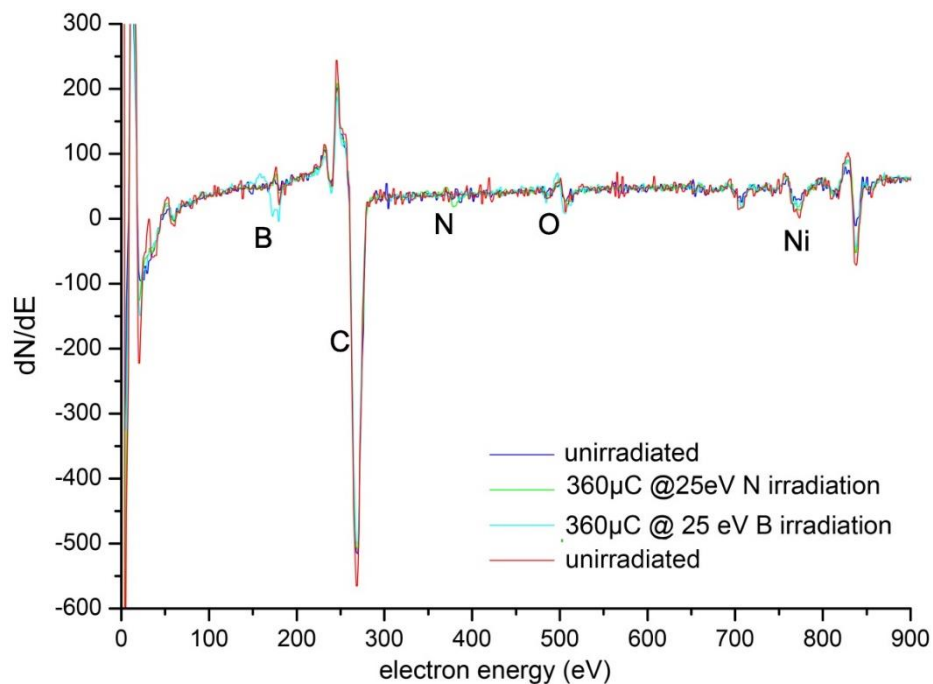
Blue line: original spectrum

red line: background fit using a 1st degree log-polynomial

Black line: the resulting signal

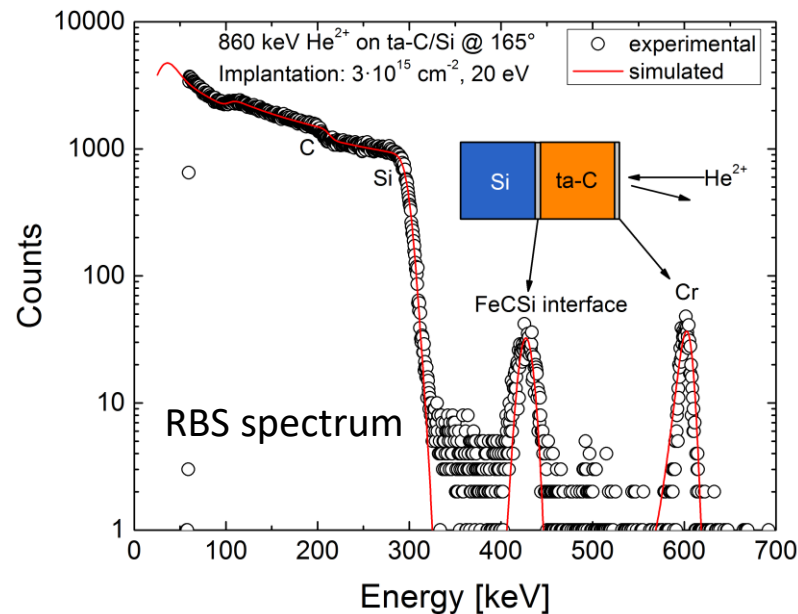
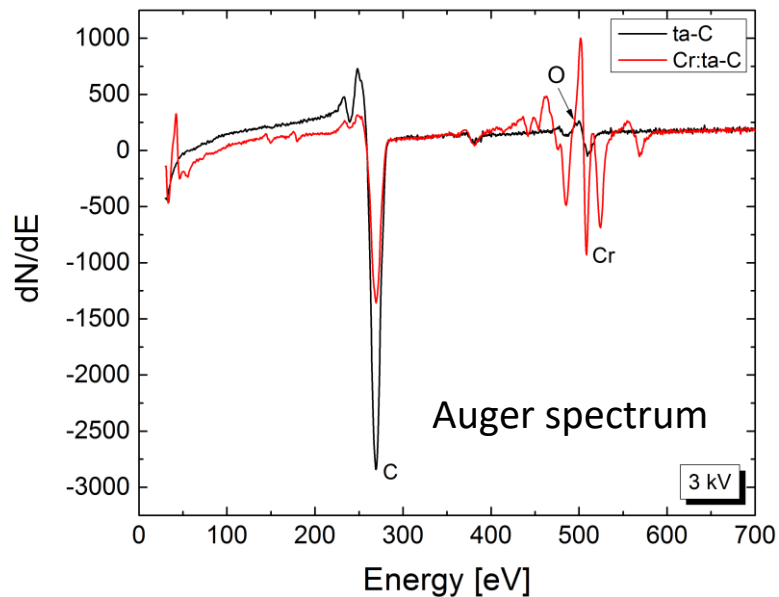


Auger spectra of implanted graphene and a-C



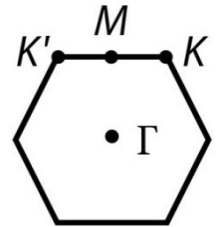
ULE Ion implantation of Cr into graphene and a-C

- Possible Kondo effect (spin scattering) impurity in graphene

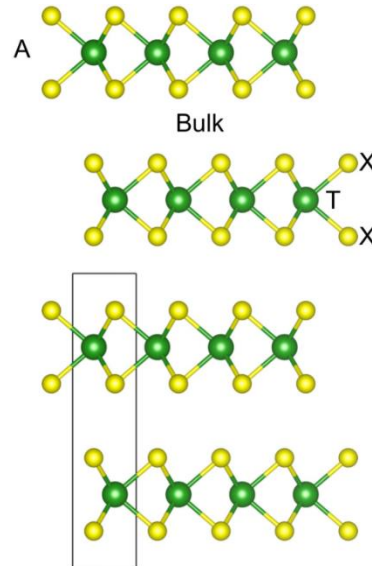


Transition metal dichalcogenides TMDs

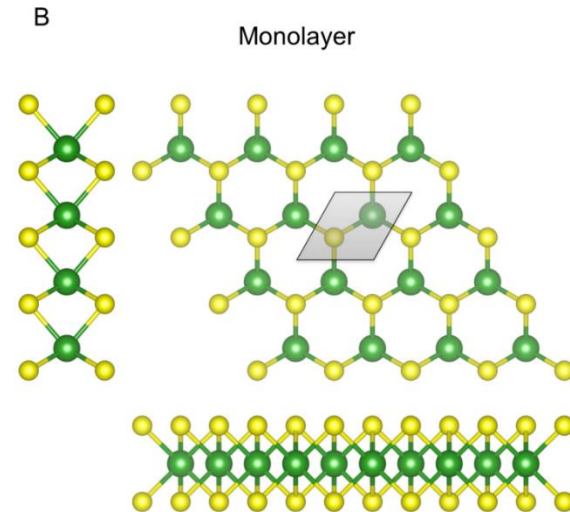
- Van-der-Waals bonds between individual layers
- Existence of a Bandgap in TMDs
 - Parabolic ($E_g = 1.8$ eV)



Bulk



Monolayer



X = Chalkogen

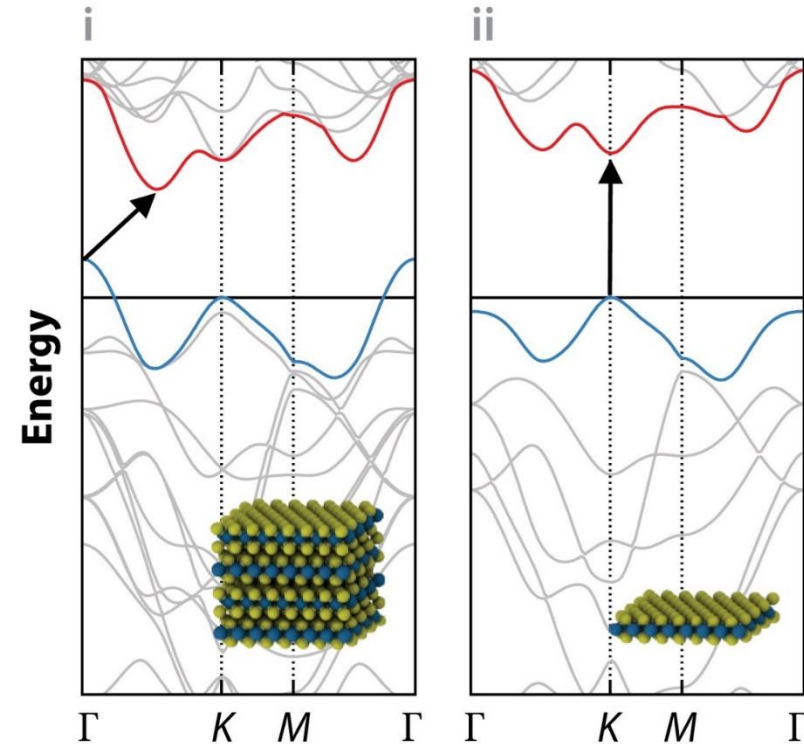
M = Übergangsmetall
M = transition metal

H																	He	
Li	Be											B	C	N	O	F	Ne	
Na	Mg	3									11	12	Al	Si	P	S	Cl	Ar
K	Ca	Sc	Ti	V	Cr	Mn	Fe	Co	Ni	Cu	Zn	Ga	Ge	As	Se	Br	Kr	
Rb	Sr	Y	Zr	Nb	Mo	Tc	Ru	Rh	Pd	Ag	Cd	In	Sn	Sb	Te	I	Xe	
Cs	Ba	La-Lu	Hf	Ta	W	Re	Os	Ir	Pt	Au	Hg	Tl	Pb	Bi	Po	At	Rn	
Fr	Ra	Ac-Lr	Rf	Db	Sg	Bh	Hs	Mt	Ds	Rg	Cn	Uut	Fl	Uup	Lv	Uus	Uuo	

Transition metal dichalcogenides TMDs

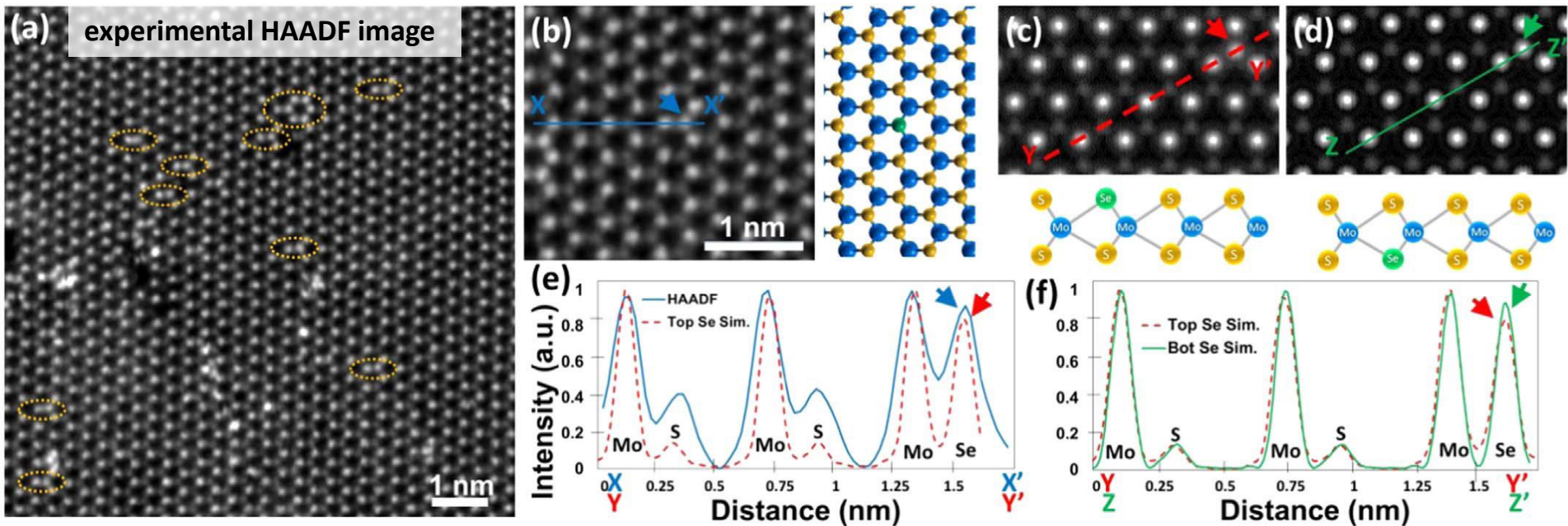
- Transition from bulk (i) to ML (ii)
- transition from indirect to direct band gap
- E_g^{dir} at K-Point of the BZ
- E_g^{ind} at Γ and $\Gamma - K$
- 2D TMDs:
 - Photon antibunching
 - Quantum confinement

→ **single photon source**



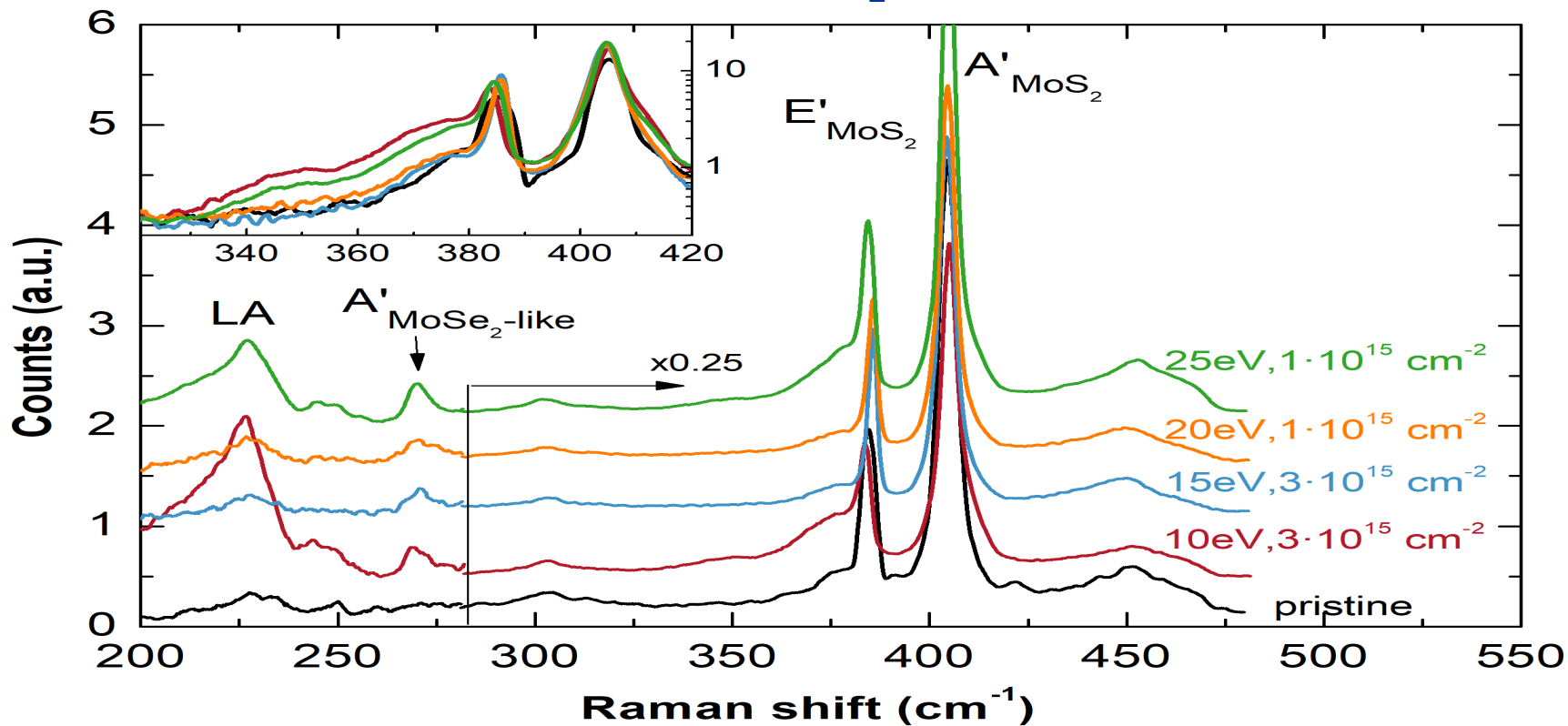
T. C. Berkelbach and D. R. Reichman, *Annu. Rev. Condens. Matter Phys.* **9** (2018), S. 379

Single layer MoS₂ implanted with Se

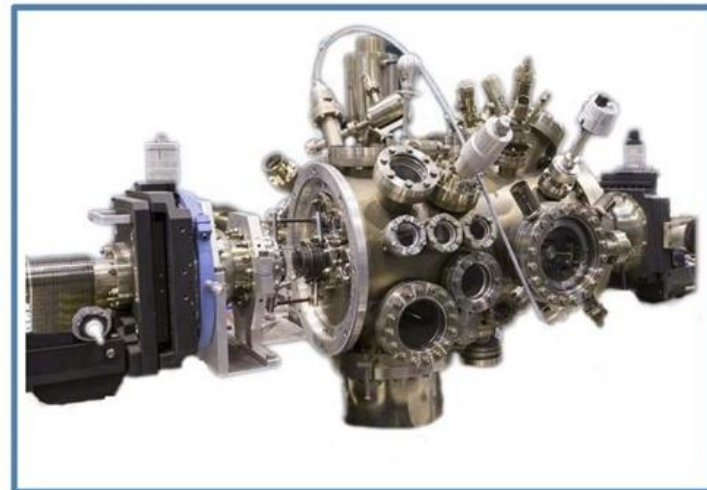
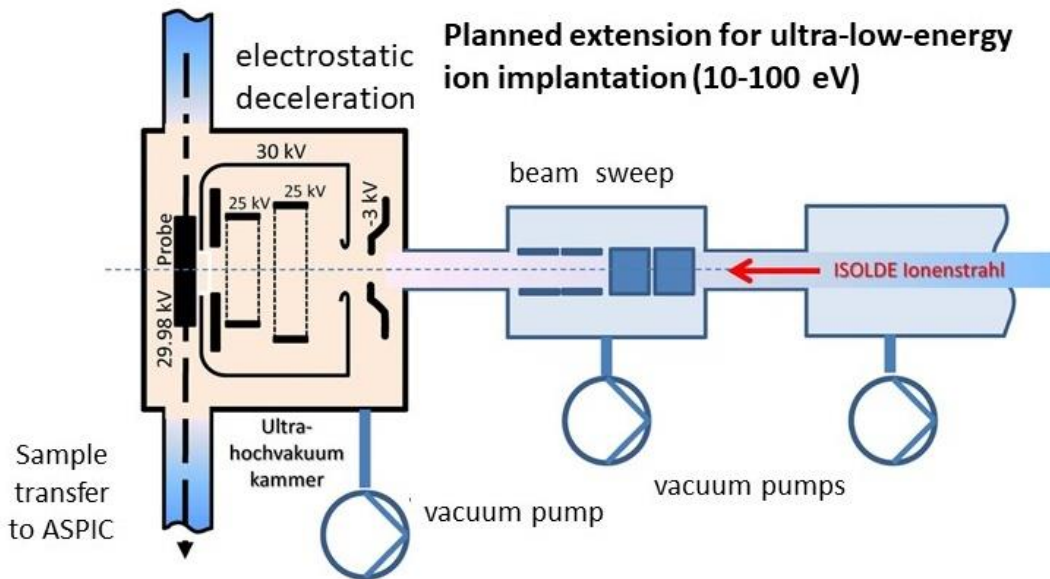


U. Bangert, A. Stewart, E. O'Connell, E. Courtney, Q. Ramasse, D. Kepatsoglou, H. Hofsäss, J. Amani, S. S. Tu, B. Kardynal, **Ultramicroscopy** **175** (2017), 021013

Raman spectra of Se implanted MoS₂

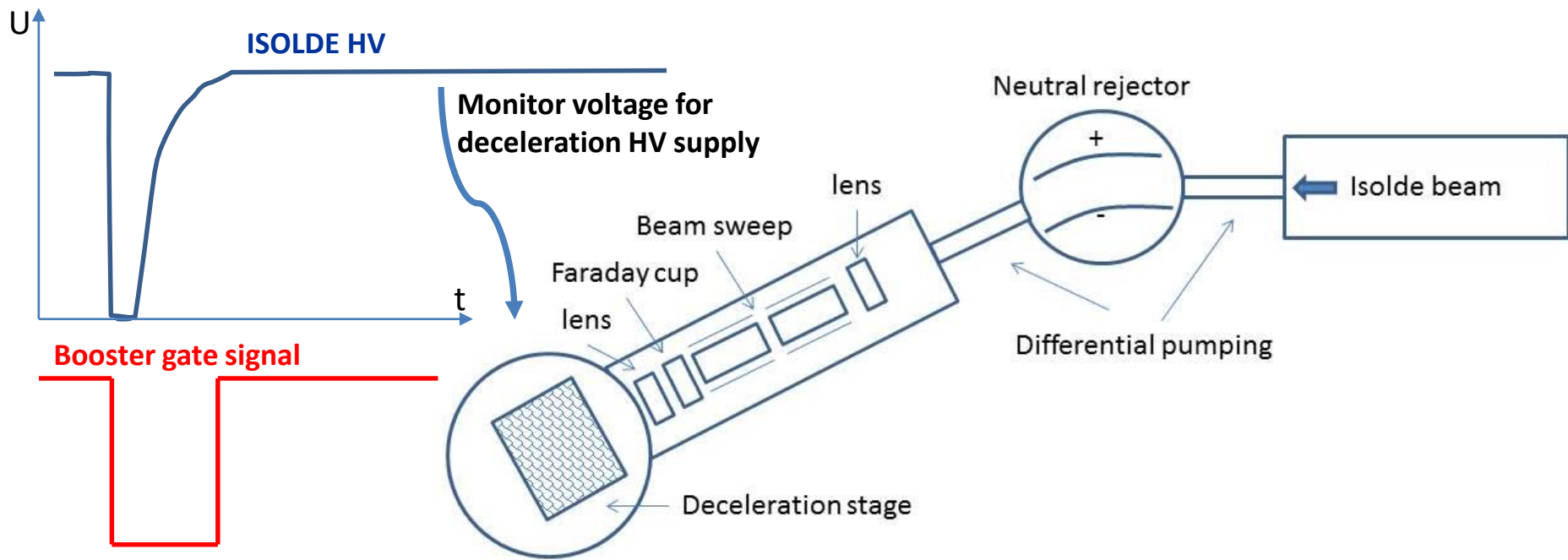


Upgrade of ASPIC for ultra-low energy ion implantation



Existing apparatus for surface and interface physics (ASPIC)

Schematics for deceleration and ion implantation



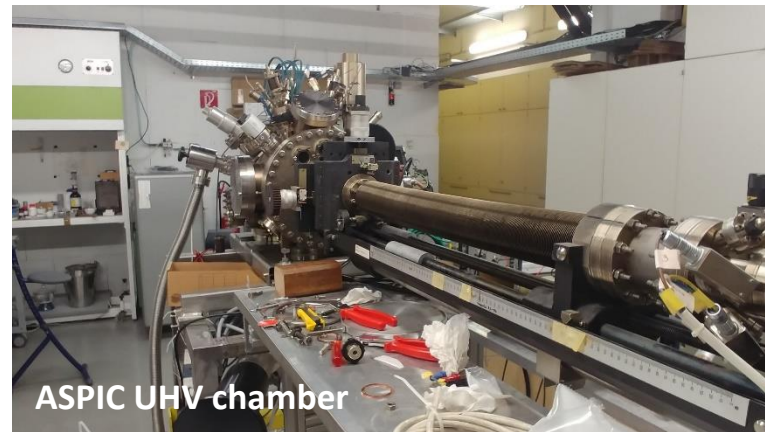
DC deceleration Voltage linked to ISOLDE HV power supply



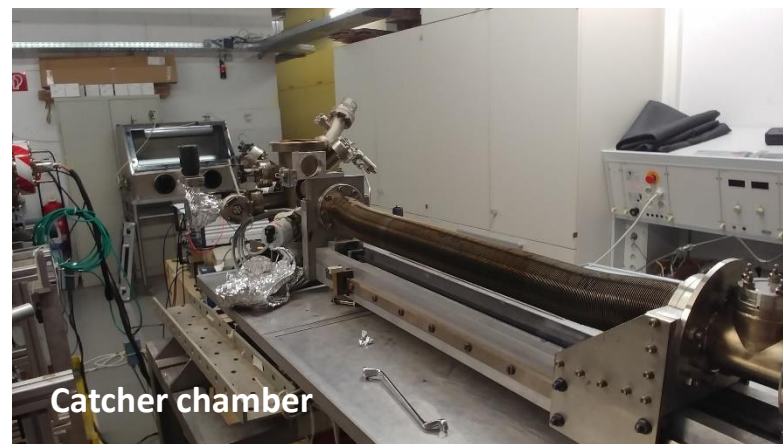
ASPIC UHV chamber



Catcher chamber



ASPIC UHV chamber



Catcher chamber

Catcher chamber

ASPIC UHV chamber



Power supplies and electronics

Planned upgrade of ASPIC

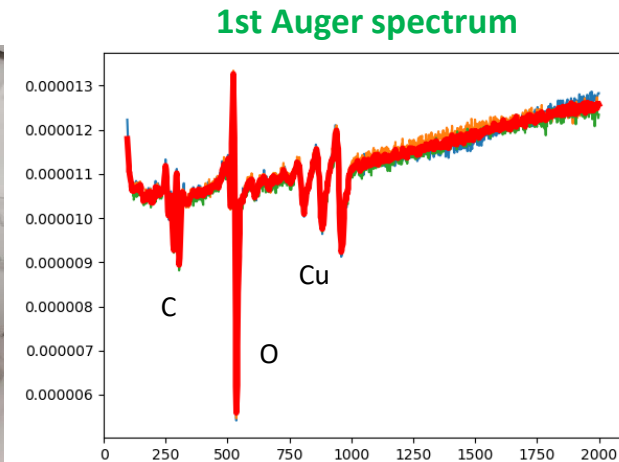
Proposal:

- Upgrade with state-of-the-art ion optics for deceleration of ions down to 10-20 eV.
 - Simulation using COMSOL and SIMION
- Renewal of the LEED and Auger Systems
 - **Auger is already working**
 - LEED is getting a new screen
- New and user-friendly sample transfer system
- New sample heating and cooling
- Renewal of the preparation and cleaning systems
- New process control (Lab View)

- Extension for surface Mössbauer spectrometry (SMS)
- Extension for emission channeling?



Disassembled LEED system



Proposed experiments using ASPIC

- **PAC and MS studies of 2D materials with very diluted impurities**
 - Cd in graphene and TMDs ^{111}Ag , $^{111\text{m}}\text{Cd}$, ^{111}In , ^{117}Cd
 - Mn/Fe magnetic impurities in graphene ^{57}Mn
 - Se in TMDs $^{77}\text{Br}/^{77}\text{Se}$; $^{73}\text{Se}/^{73}\text{As}$
 - Rare earth impurities in TMDs $^{140}\text{Ce}/^{140}\text{La}$; $^{156}\text{Gd}/^{156}\text{Eu}$
 $^{172}\text{Yb}/^{172}\text{Lu}$, ^{158}Gd , ^{158}Tb
- Possible on-line isotopes: ^{28}Mg , ^{48}Cr , ^{62}Zn , $^{111\text{m}}\text{Cd}$, ^{117}Cd , ^{116}Sb , ^{118}Sb ,
- **PAC and MS studies of (crystalline) surfaces**
- **PAC and MS studies of interfaces and very thin films**

See PhD thesis Matthias Nagl, 2014

