New magnetism of 3d monolayers grown with oxygen surfactant: Experiment vs. *ab initio* calculations

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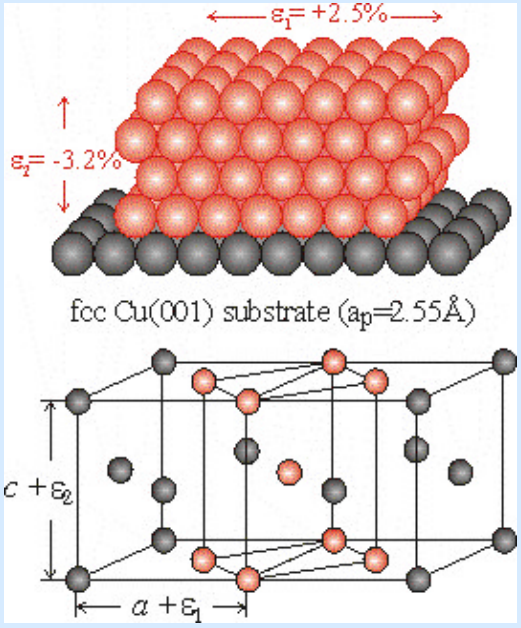


1. **Growth and structure**
2. **Magnetism and MAE**
3. **Induced magnetism at oxygen**

Goal: Theory should calculate:
XAS-, XMCD spectra,
magnetic moments at Ni and O

Þ **http://www.physik.fu-berlin.de/~ag-baberschke**

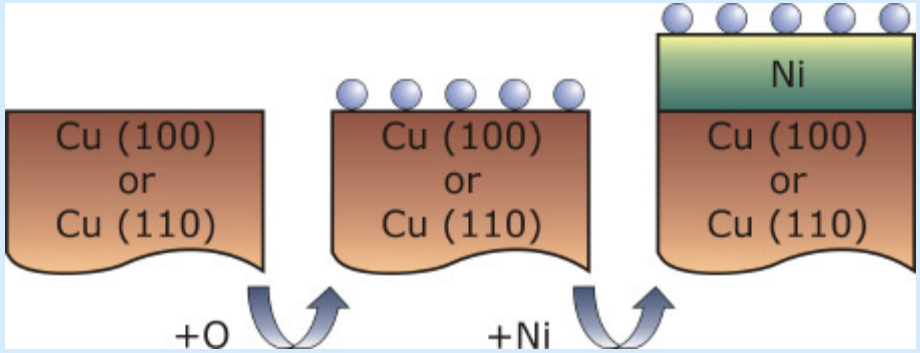
Acknowledgement

BESSY-crew: H. Wende, C. Sorg, N. Ponpandian, M. Bernien, (A. Scherz)

Lab. experiments: K. Lenz, T.Tolinski, (J. Lindner, E. Kosubek, C. Rüdt, R. Nünthel)

Support:
BMBF (BESSY), DFG (lab.)

1. Growth and structure: Surfactant Activity



Known:

- Improved growth

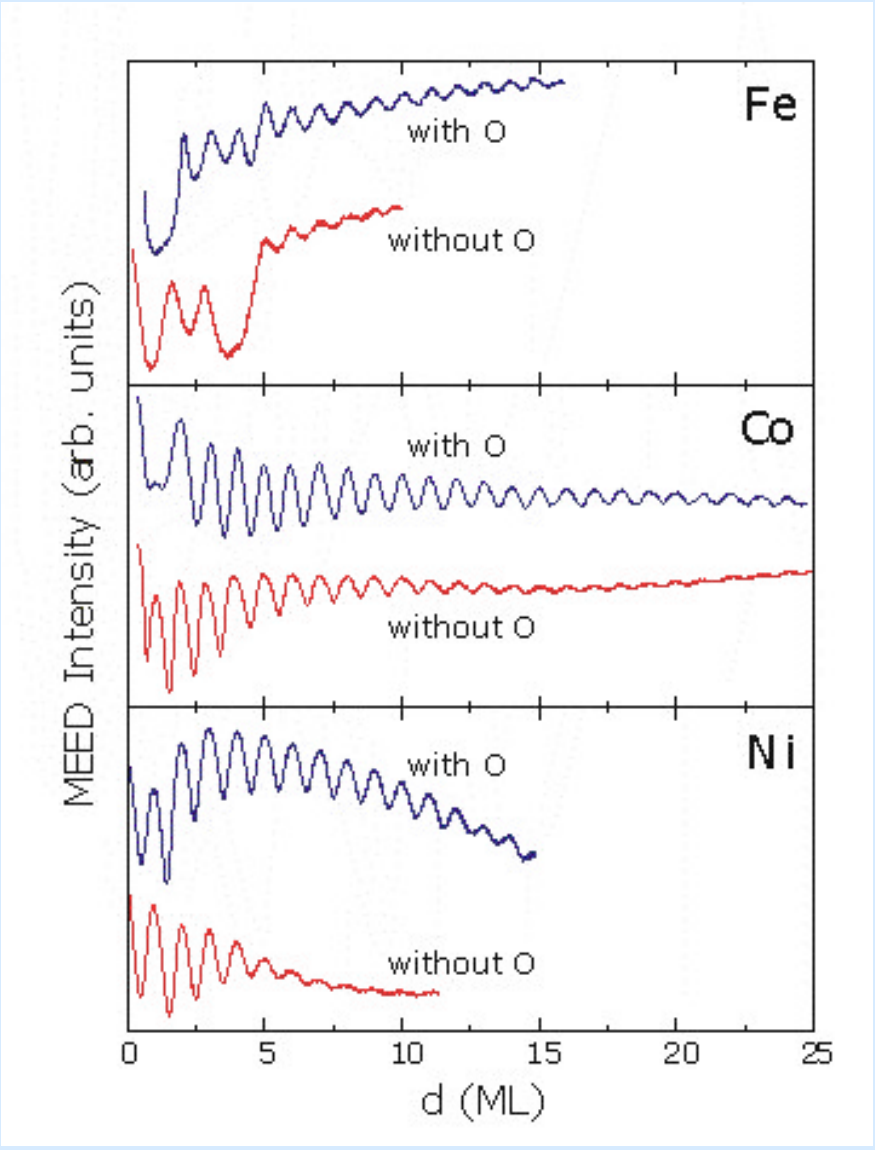
  R. Nünthel *et al.*,
  Surf. Sci. **531**, 53 (2003),
  Surf. Sci. **566-568**, 100 (2004).

- Shift of SRT to lower thickness

  J. Lindner *et al.*,
  Surf. Sci. **523**, L65 (2003).

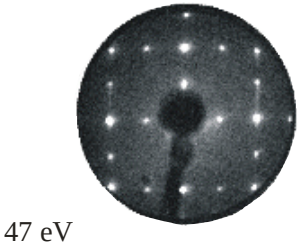
Open:

- Final evidence: O on top?
- NiO?
- Influence of O on spin and orbital moments of Ni?
- Induced magnetism of O?

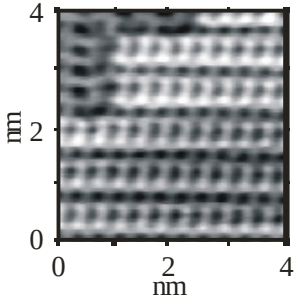

Fe
with O
without O
Co
with O
without O
Ni
with O
without O
MEED Intensity (arb. units)
0
5
10
15
20
25
d (ML)

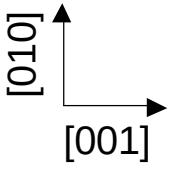
O($\sqrt{2} \times 2\sqrt{2}$)R45°/Cu(100)
missing row reconstruction

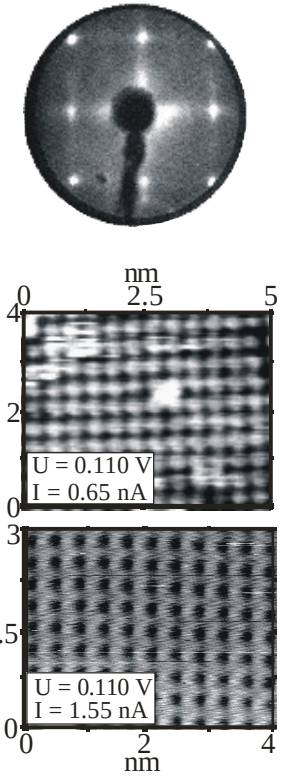
c(2 × 2)O/Ni/Cu(100)

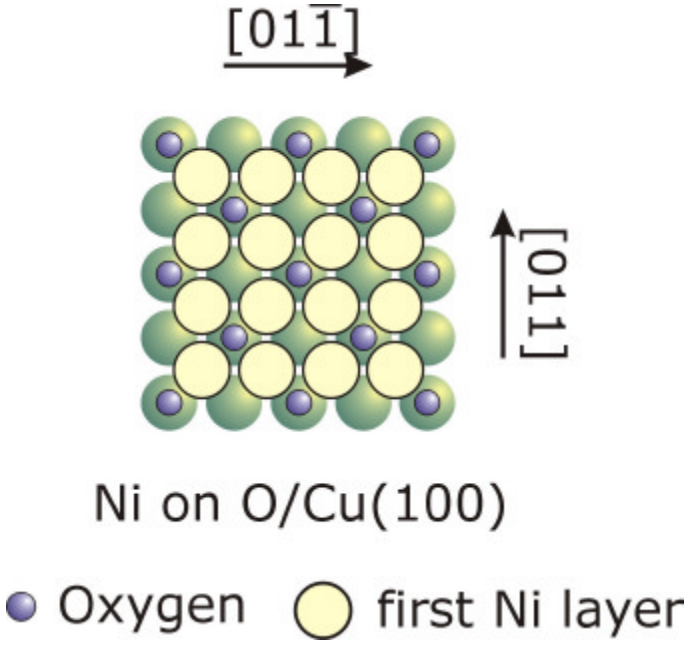


evaporate
5.5 ML Ni









from AES ⇒ oxygen floats on top of Ni film

R. Nünthel et al., *Surf. Sci.* **531**, 53-67 (2003)

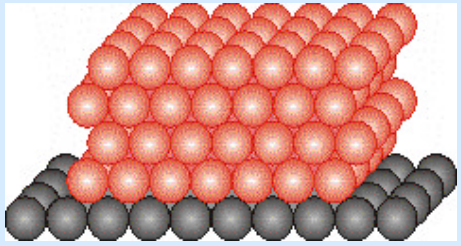
2. Magnetic Anisotropy Energy (MAE) in ultra thin films

There are only 2 origins for MAE: 1) dipol-dipol interaction ~ $(\vec{\mu}_1 \cdot \vec{r})(\vec{\mu}_2 \cdot \vec{r})$ and
2) spin-orbit coupling ? $\vec{L}\,\vec{S}$ (intrinsic K or ΔE_{band})

Growth of artificial nanostructures
bcc, fcc → tetragonal, trigonal

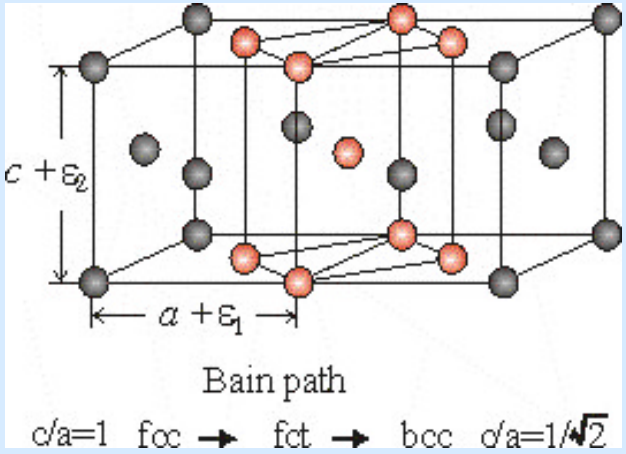
e_1= +2.5%

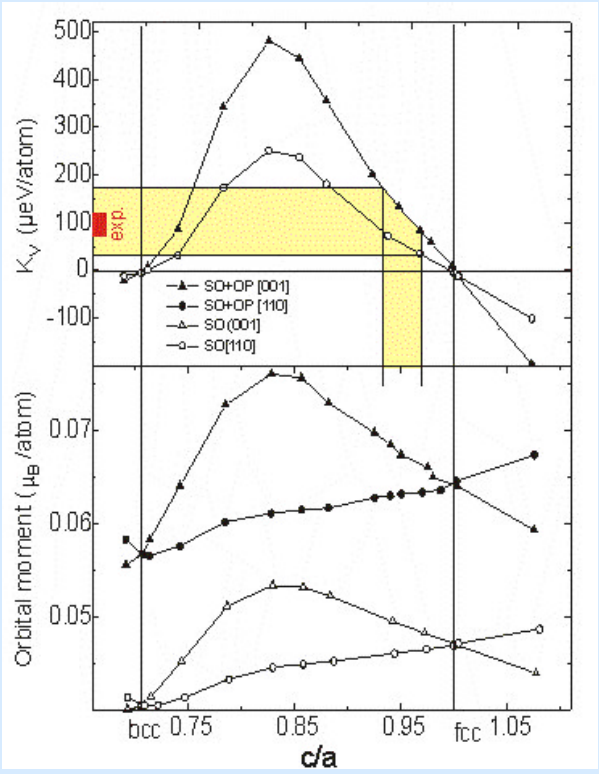
e_2= -3.2%



tetragonal
Ni(001)
($a_p^{Ni\ bulk}$=2.49Å)

fcc Cu(001) substrate (a_p=2.55Å)

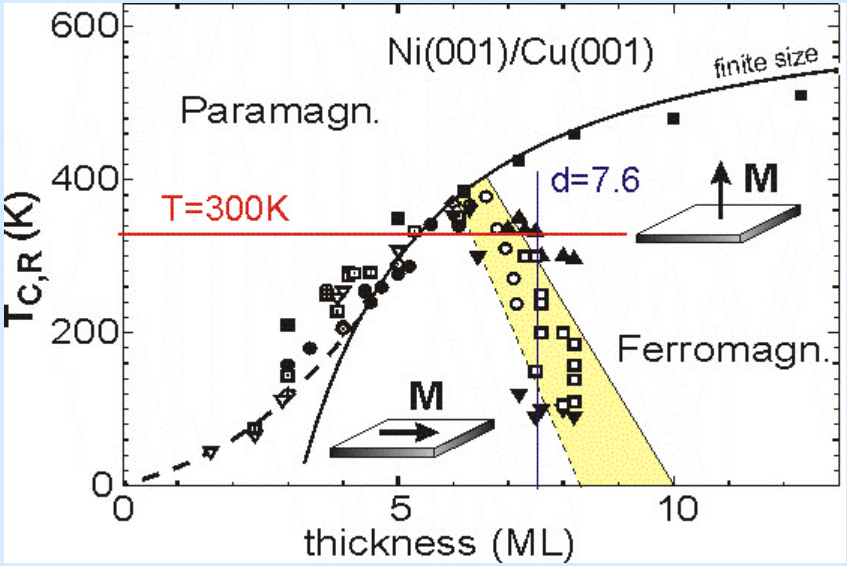


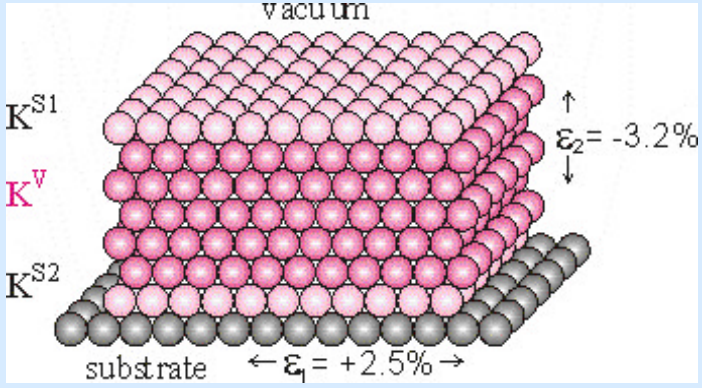


Structural changes by ≈ 0.05 Å increase MAE by 2-3 orders of magnitude (~0.2→100μeV/atom)

O. Hjortstam, K. B. et al. PRB **55**, 15026 ('97)
R. Wu et al. JMMM **170**, 103 ('97)

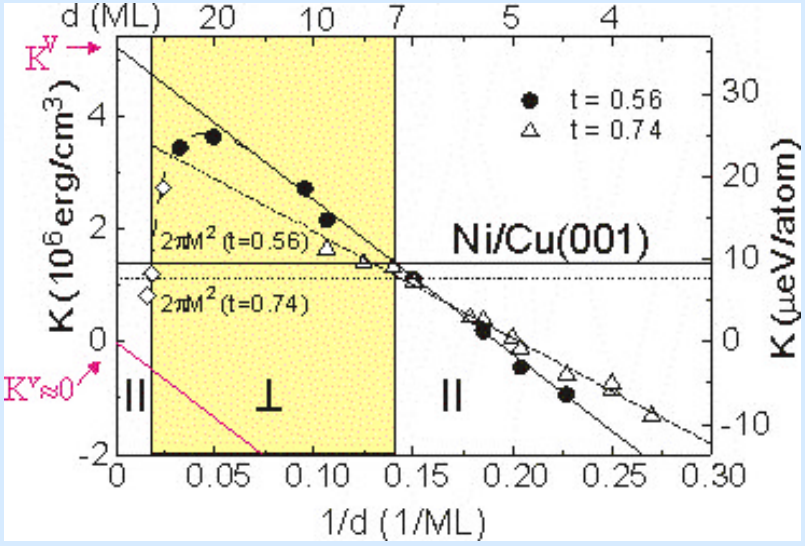
For thin films the Curie temperature can be manipulated





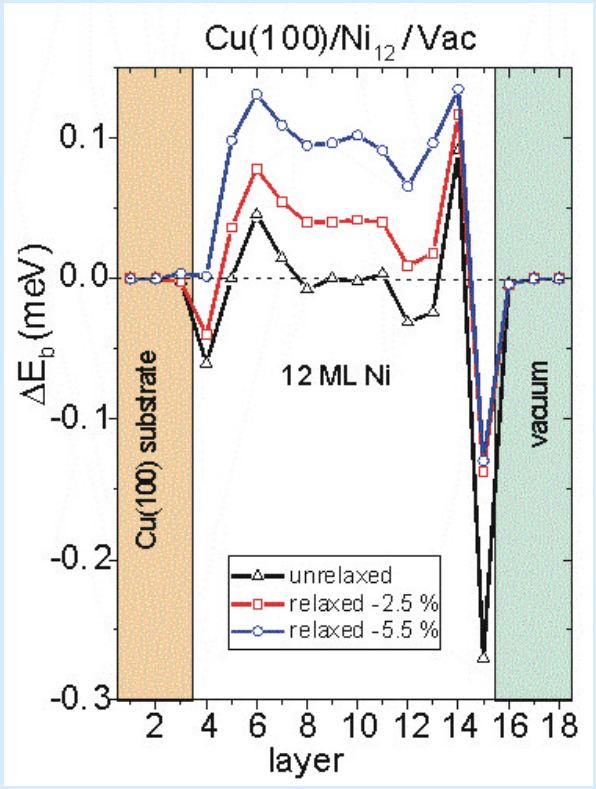
$$K_i = K_i^V + 2\frac{K_i^S}{d} \qquad t=T/T_C(d)$$

P. Poulopoulos and K. B.
J. Phys.: Condens. Matter **11**, 9495 (1999)



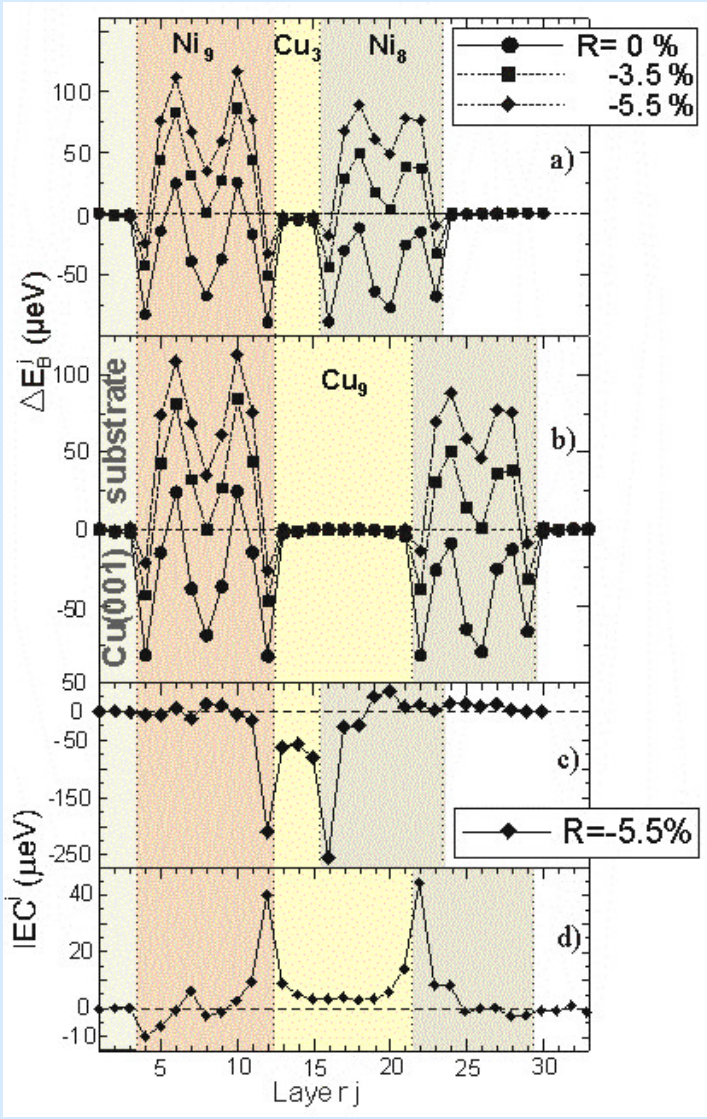
SP-KKR calculation for rigit fcc and relaxed fct structures

layer resolved $\Delta E_b = \Sigma K_i$ at T=0



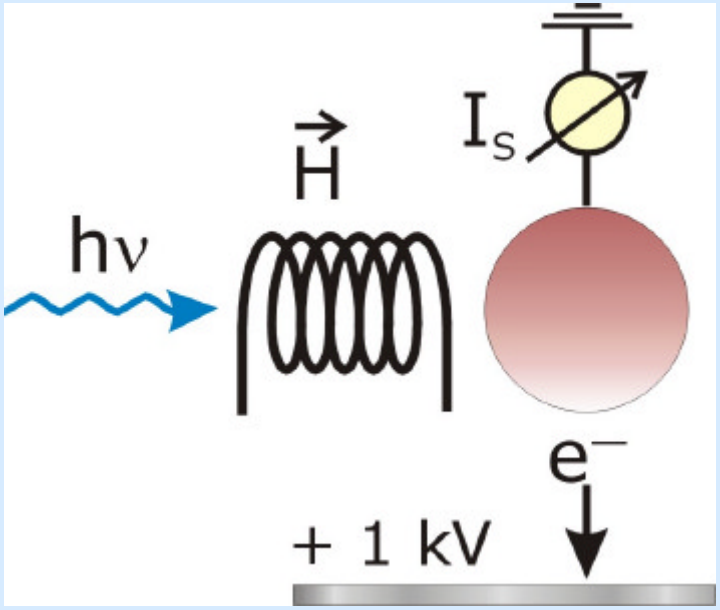
C. Uiberacker et al.,
PRL **82**, 1289 (1999)

R. Hammerling et al.,
PRB **68**, 092406 (2003)



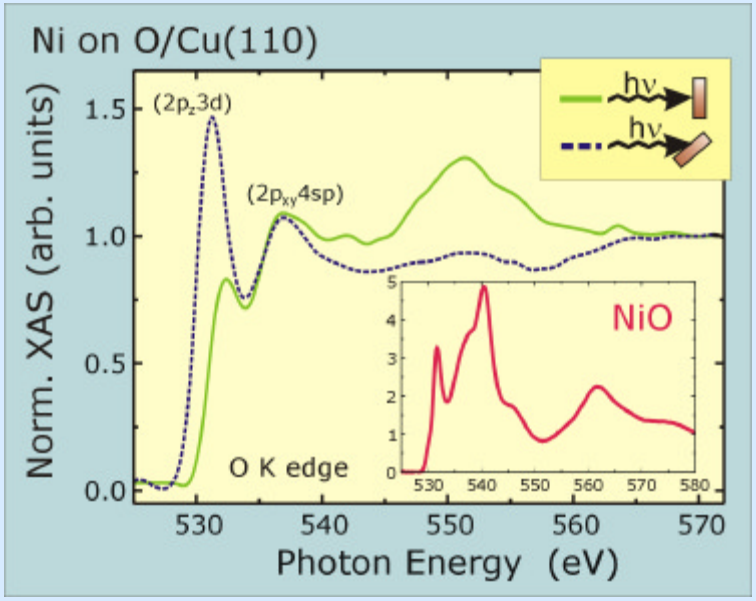
The surface and interface MAE are certainly large (L. Néel, 1954) but count only for one layer each. The inner part (volume) of a nano-structure will overcome this, because they count for in n-2 layers.

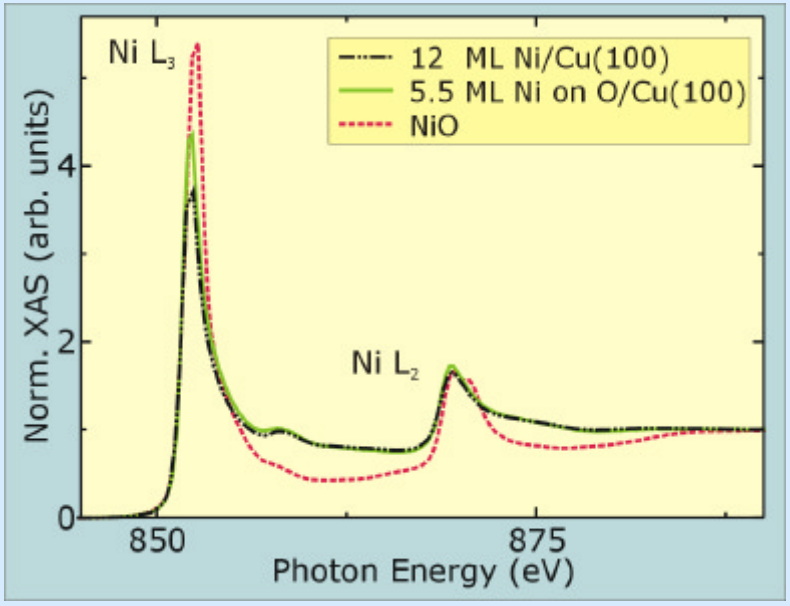
Experiment



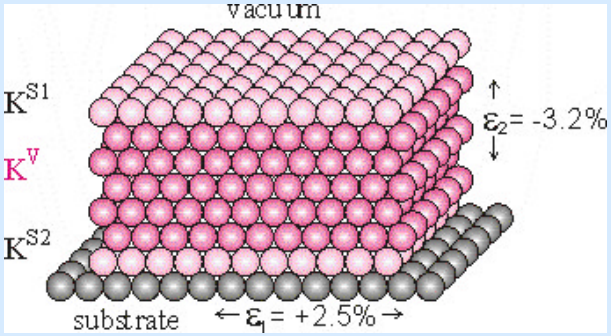
- X-ray absorption via total electron yield
- NEXAFS = μ(E)
  ⇨ fingerprint of the chemical bond
- XMCD = $\mu^+(E) - \mu^-(E)$
  ⇨ magnetic information
- element-specific method

Electronic Structure and Charge Transfer





- Strong angular dependence at O K edge
  - ⇨ oxygen on top
- NiO differs significantly from Ni on O/Cu
  - ⇨ no bulk-like NiO
- Enhanced XAS at Ni L_3 edge
  - ⇨ charge transfer from Ni to O

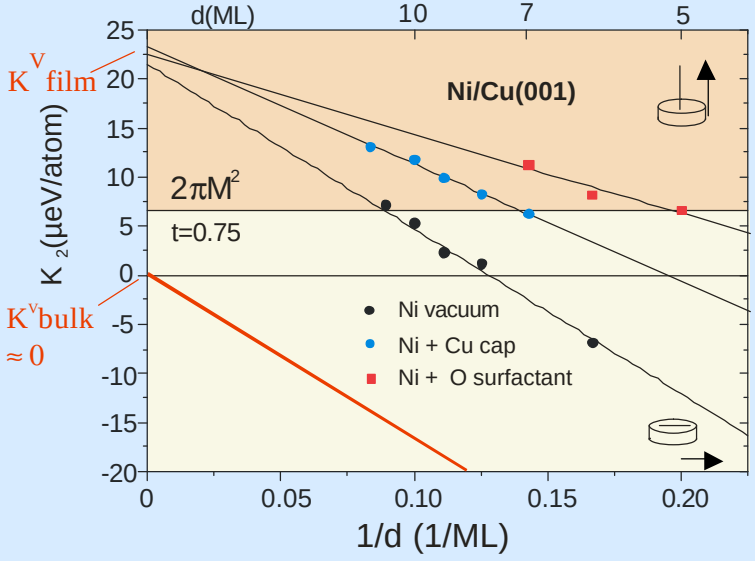


Interface	K_S (μeV/atom)	d_c (ML)
Ni/vacuum	-107	10.8
Ni/Cu	-59	7.6
Ni/CO (van Dijken et al.)	-81	7.3
Ni/H_2 (van Dijken et al.)	-70	6.8
Ni/O (surfactant)	-17	4.9

$$F \sim \left(2pM^2 - K_{2\perp}\right)\cos^2 q$$

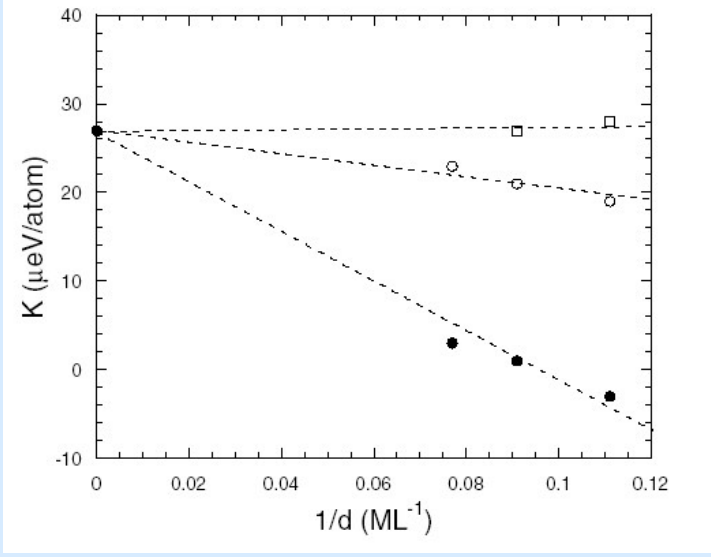
$$K = K^V + \frac{K^{S1} + K^{S2}}{d}$$

Experiment



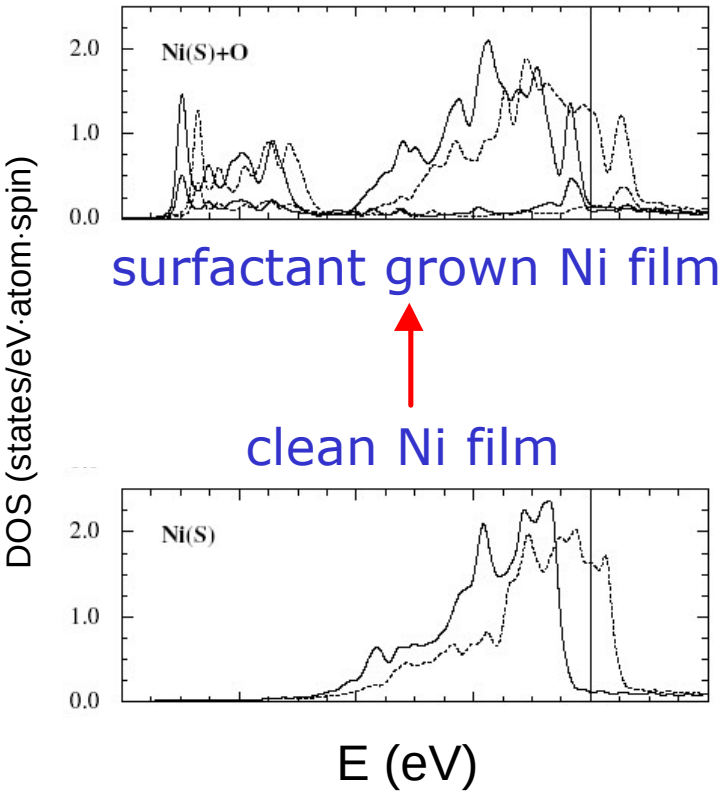
Theory (FLAPW)

Jisang Hong et al., *Phys. Rev. Lett.* **92**, 147202 (2004)

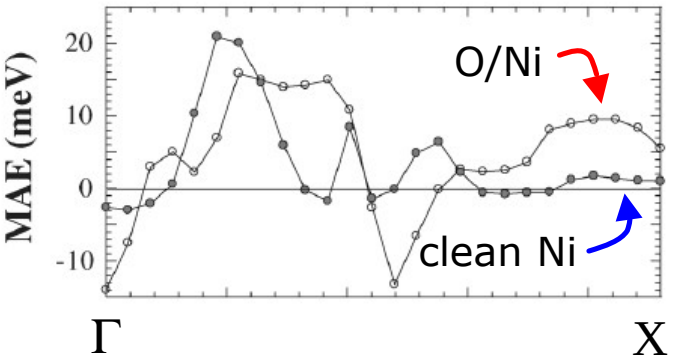


results of ab initio calculations

Density of states



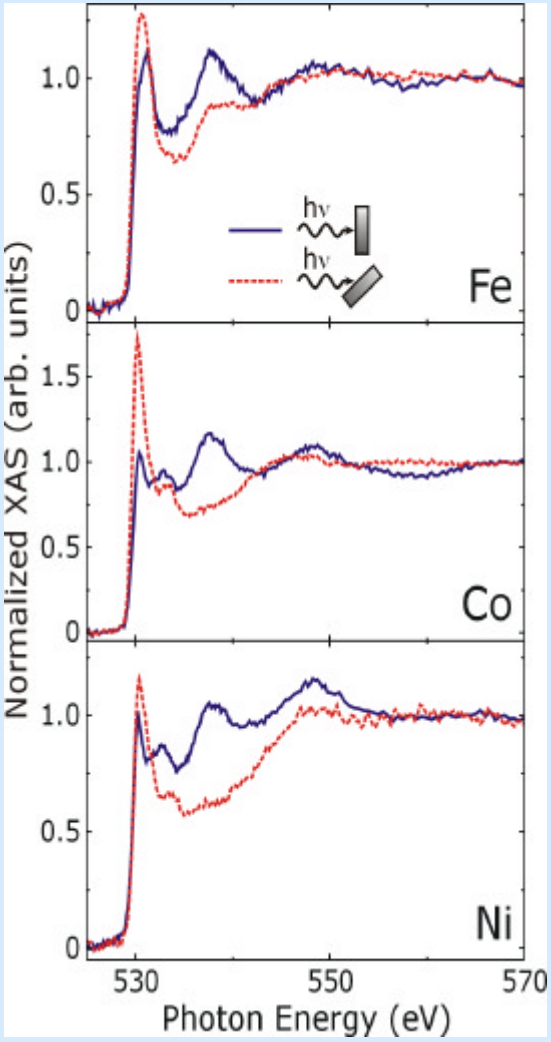
MAE along $\bar{\Gamma}\bar{X}$ axis

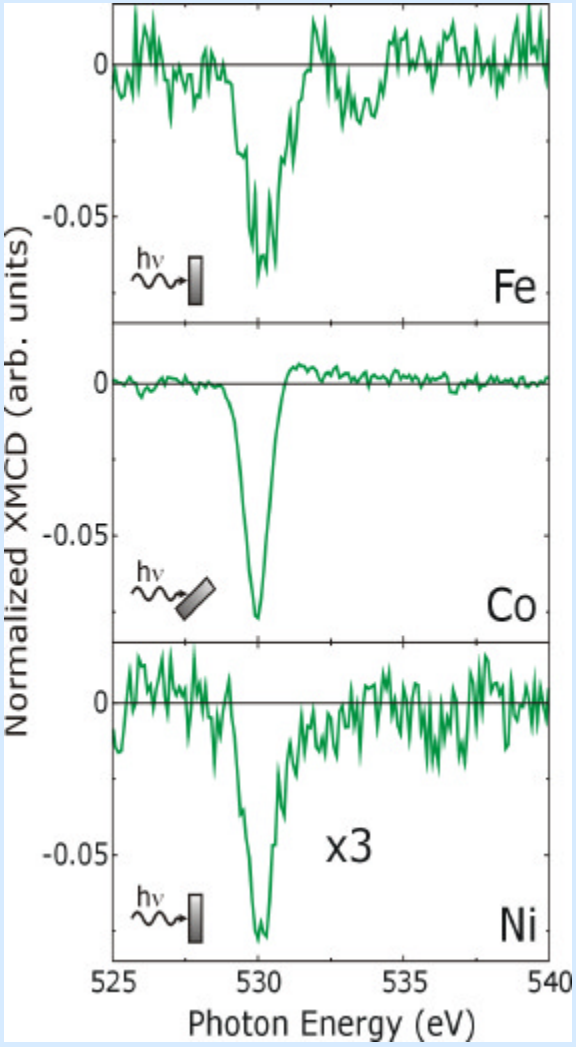


- DOS shows that topmost Ni moment is basically unchanged
- O-induced surface state seen in the vicinity of $\bar{X}$-point is responsible for change in MAE
- theory reveals induced moment in surfactant oxygen (~0.26μ_B)

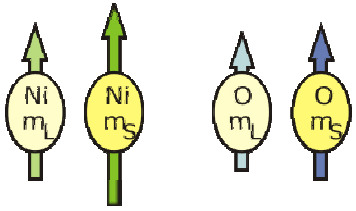
Jisang Hong et al., *Phys. Rev. Lett.* **92**, 147202-1 (2004)

3. Induced magnetism at oxygen NEXAFS and XMCD at O K edge

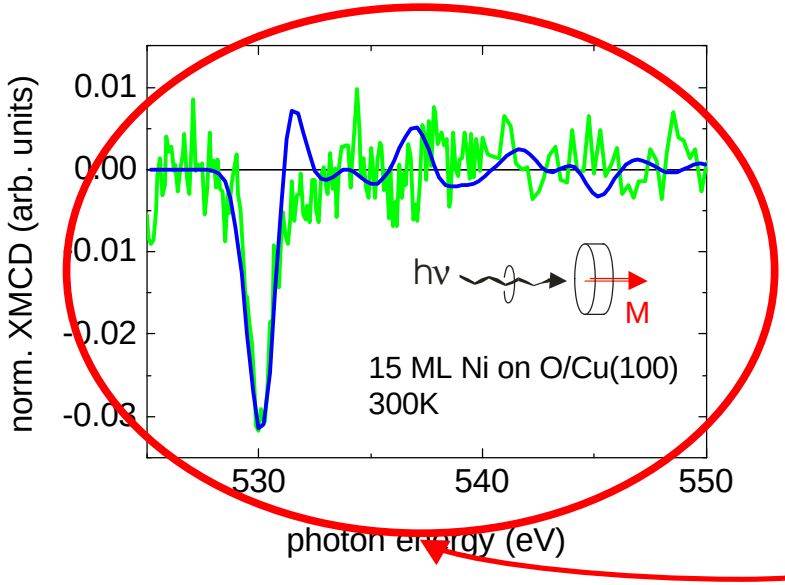




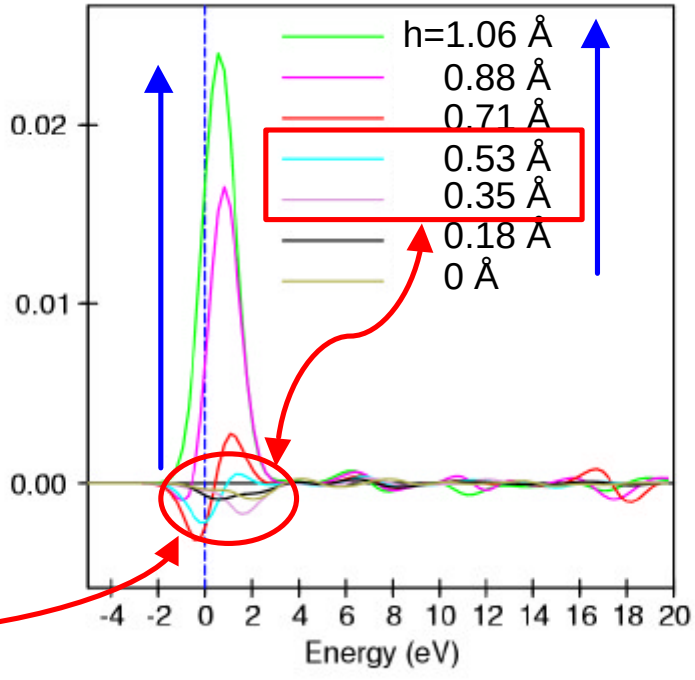
Induced magnetism in oxygen? Ni on O/Cu(100)



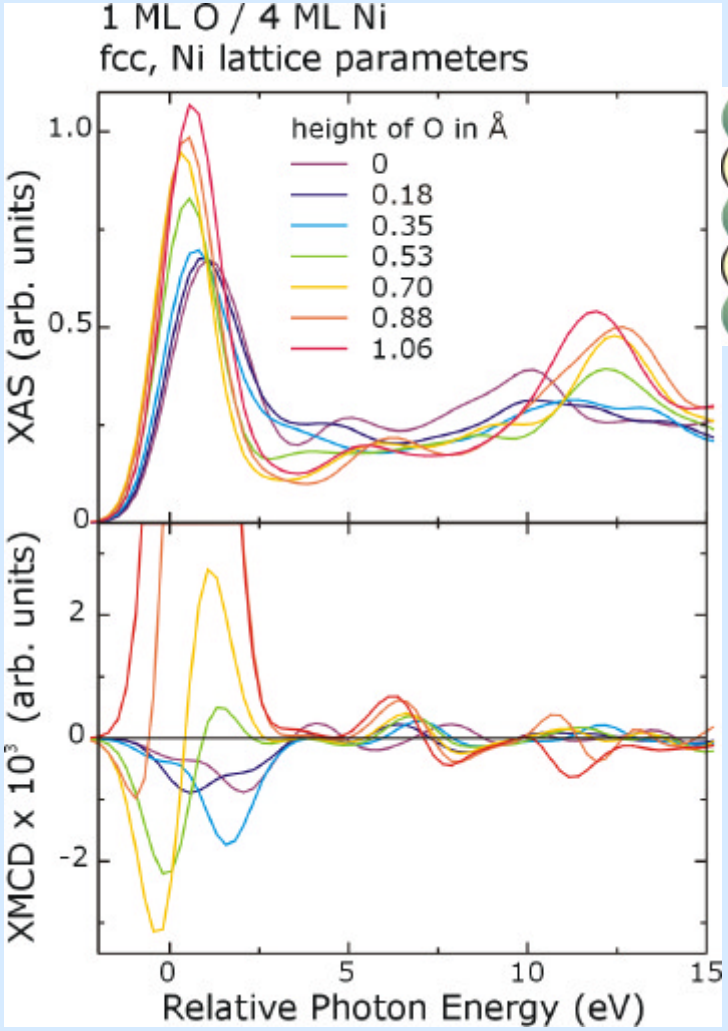
oxygen K-edge XMCD → orbital moment μ_L

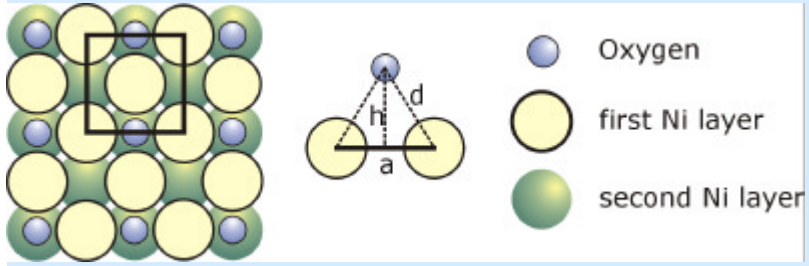


theory: group of H. Ebert (LMU)



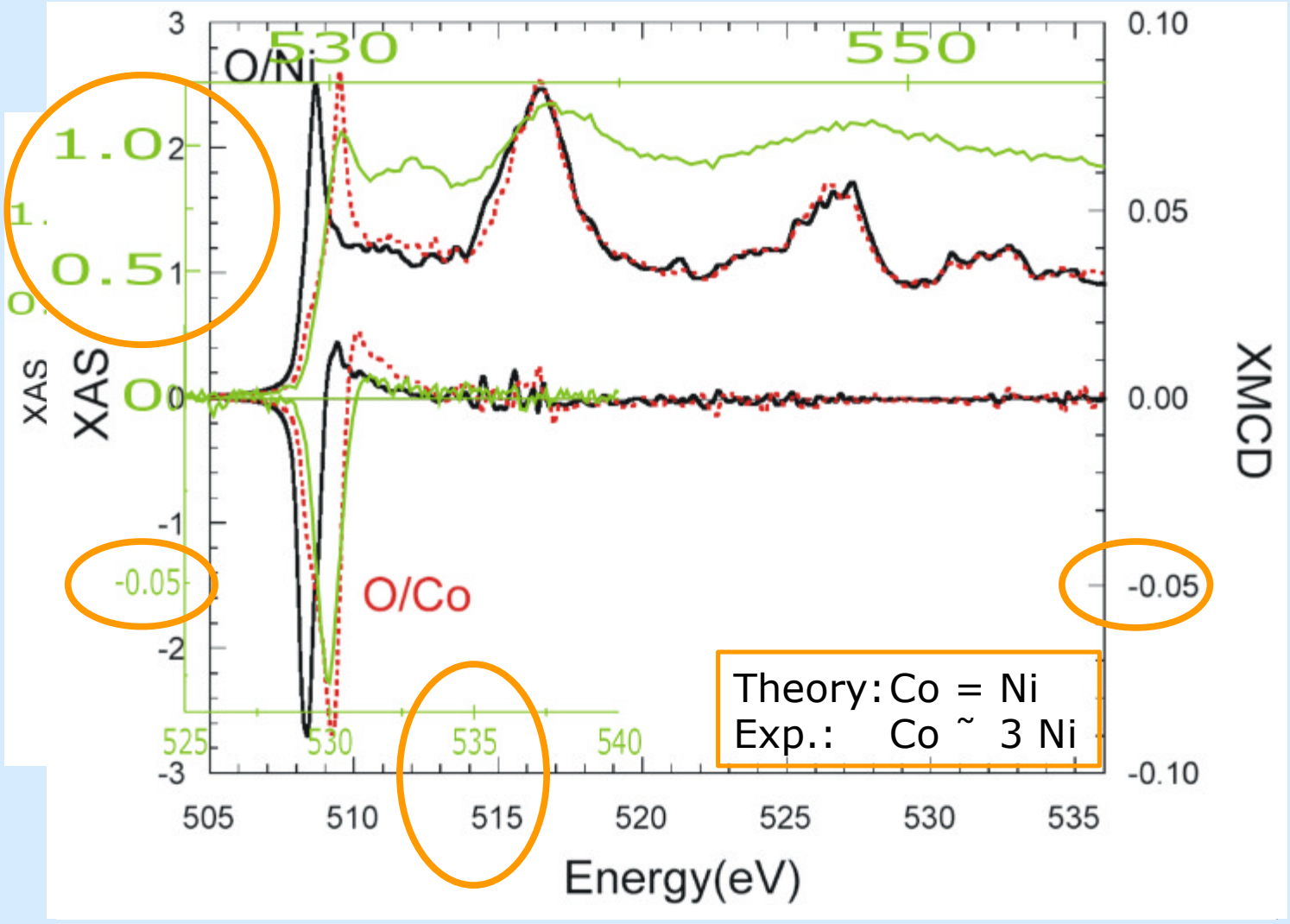
Experiment & Theory I (AG Ebert)

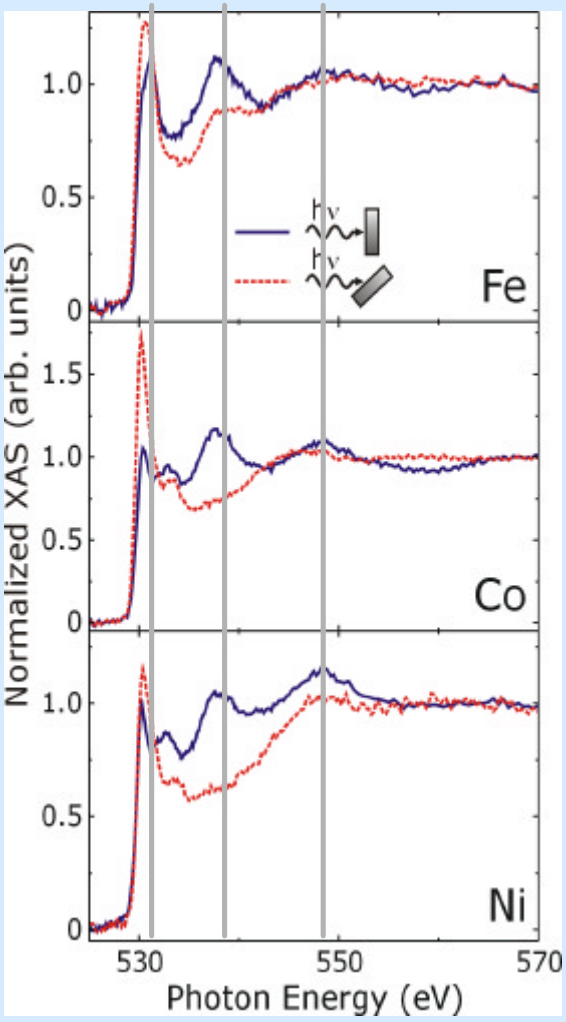




lattice constant	Ni-O distance	height	comment
3.52 (Ni) 3.61 (Cu)	**1.855** **1.855**	0.586 0.428	SEXAFS Aug. 2003, Ni on O/Cu(100)
3.52 (Ni) 3.61 (Cu)	**1.97** **1.97**	0.885 0.789	SEXAFS, O/Ni(100) bulk, Wenzel et al. PRB 36, 7689 (1987)
3.52 (Ni) 3.61 (Cu)	1.902 1.943	**0.72** **0.72**	LEED, O/Ni(100) bulk, K. Heinz et al., PRB 41, 10179 (1990)
3.640 3.634	**1.89** **1.87**	**0.51** **0.44**	Theory, T. S. Rahman in joint Surf. Sci. **531**, 53 (2003), 2 different structural scenarios
3.61	1.929	**0.68**	Theory, AG Wu in J. Hong et al. PRL **92**, 147202 (2004)

Experiment & Theory II (R. Wu)




Normalized XAS (arb. units)
1.0
0.5
0
1.5
1.0
0.5
0
1.0
0.5
0
hν
hν
Fe
Co
Ni
530
550
570
Photon Energy (eV)

Summary

The magnetism of nanostructures is a prototype case, which shows the close collaboration between theory and experiment

Theory: H. Ebert, LMU; J.J. Rehr, UW; O. Eriksson UU; P. Weinberger, TU Vienna; R. Wu, UCI; D.L. Mills, UCI; K.H. Bennemann, FUB; W. Nolting, HUB

- magnetism changes dramatically with surfactant: surface anisotropy is strongly reduced in magnitude
- Fe, Co and Ni induce magnetic moment in surfactant
- fair agreement with *ab initio* calculations