SYNCHROTRON-RELATED METHODS FOR INVESTIGATION OF NANOSTRUCTURES

SOME

SOME

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OUTLINE:

1. What can be done by x-ray scattering?
2. Standard applications – rather boring
3. Iso-strain method & anomalous diffraction
4. DAFS
5. In-situ scattering
6. Nanobeams & coherent scattering
7. Summary

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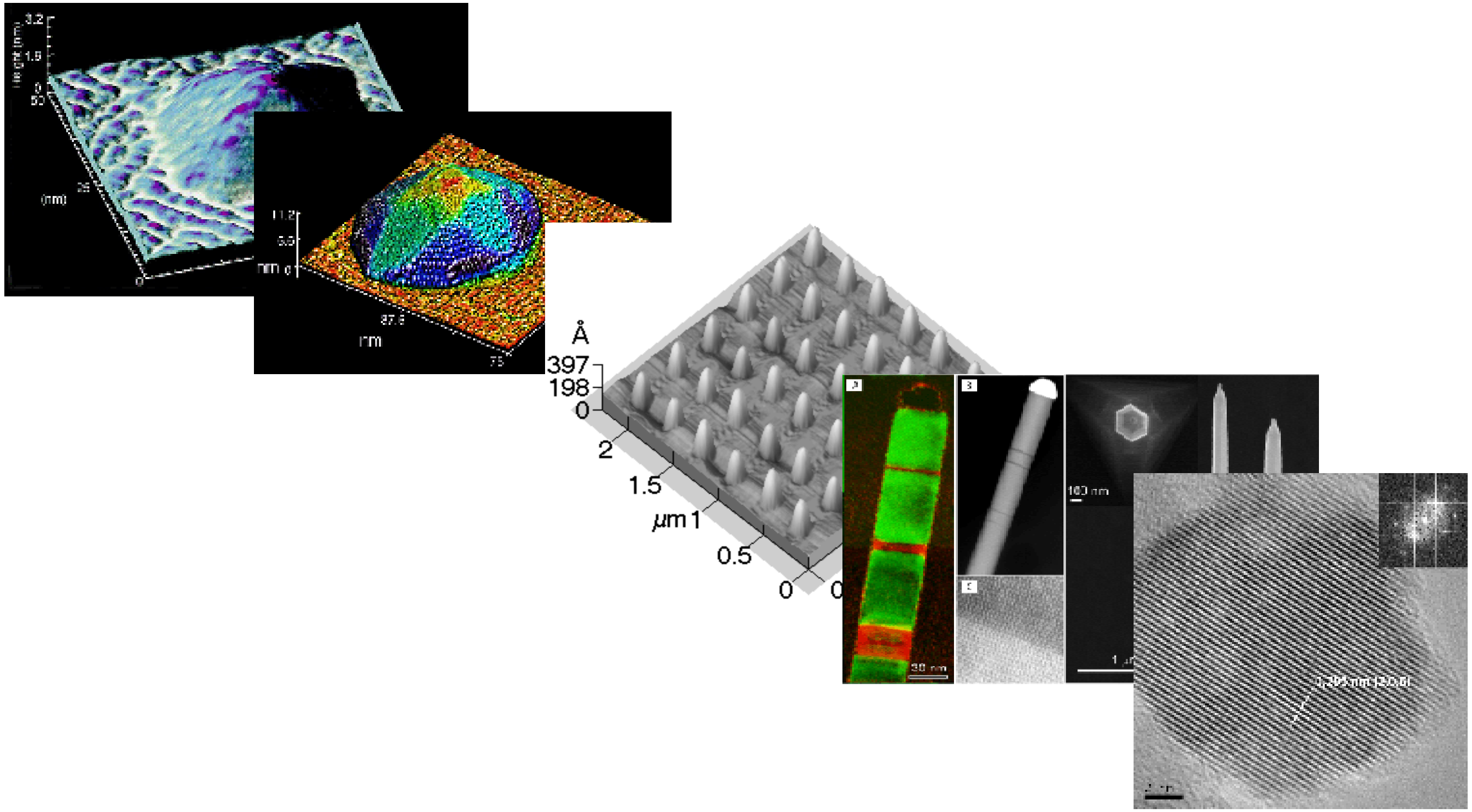
Various types of semiconductor nanostructures



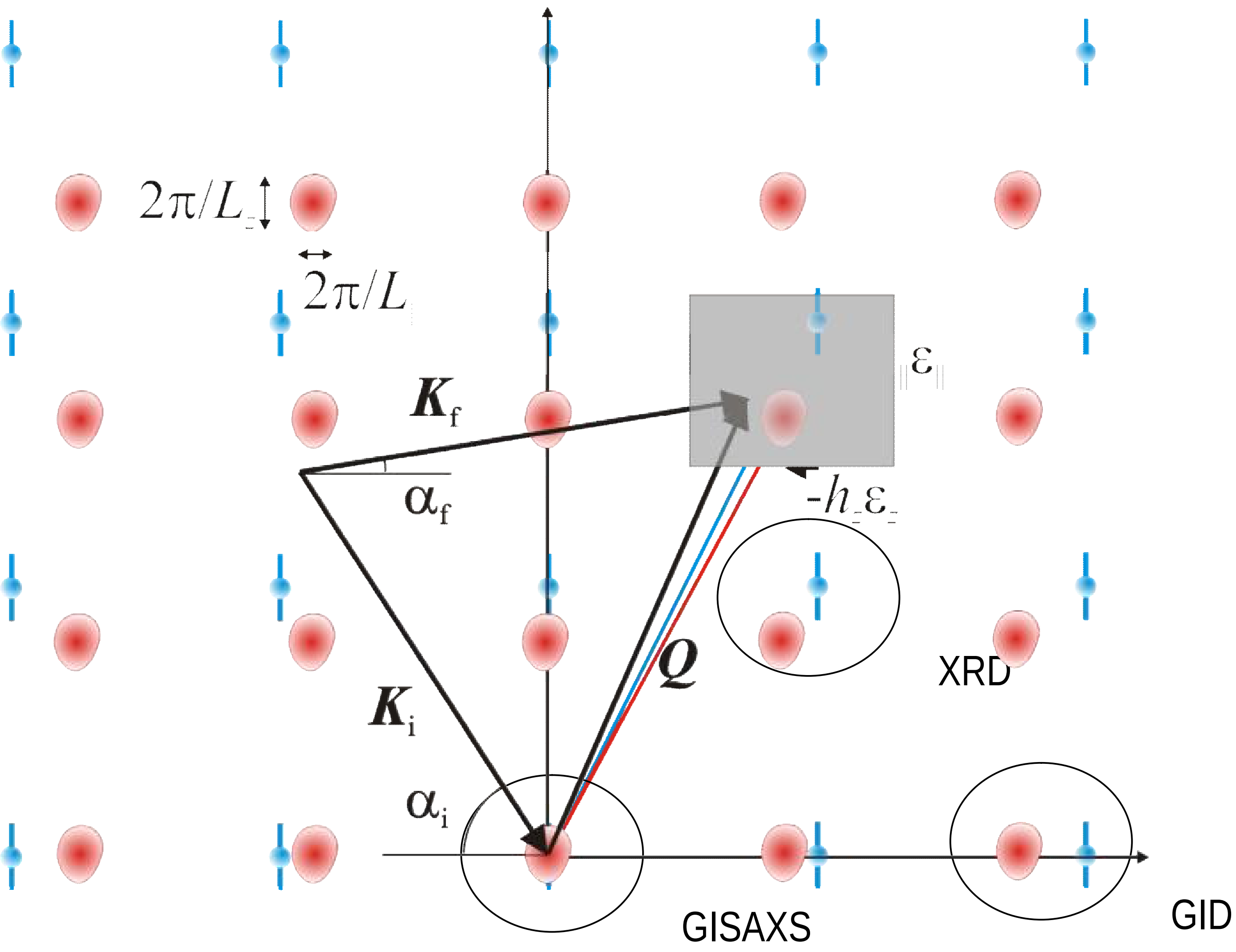
Structure of self-assembled nanostructures: position, sizes, chemical composition, atomic ordering

Methods local in real space: TEM, AFM, STM – investigation of few objects only

Methods local in reciprocal space: x-ray scattering assuming far-field limit – averaging over the irradiated volume (many objects). A direct interpretation of the data is impossible (the phase problem)

New methods: anomalous scattering, DAFS, coherent scattering – data can be interpreted almost directly without any data modeling

It is not possible to achieve both real-space and reciprocal-space resolution simultaneously (uncertainty principle) – resolution problem



$2\pi/L_z$
$2\pi/L_{||}$
K_f
α_f
$\varepsilon_{||}$
$-h_z\varepsilon_z$
Q
XRD
K_i
α_i
GISAXS
GID

Main problem:

very small volume of the nanoobjects ⇨ extremely weak intensity

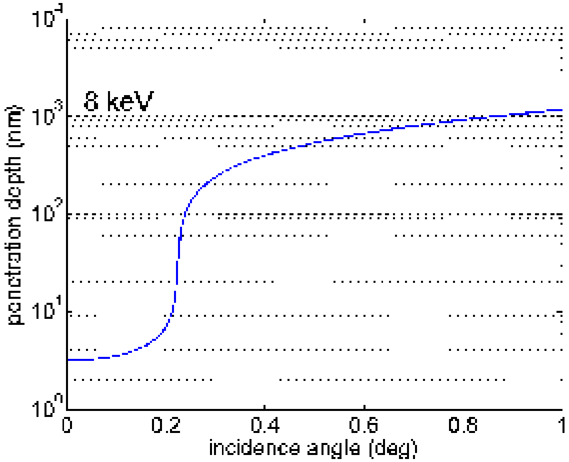
Possible solutions:

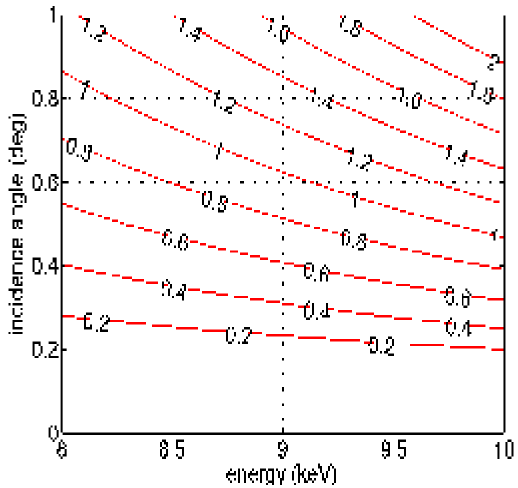
grazing-incidence geometry ⇨ reduction of the substrate signal

anomalous scattering ⇨ increase of the signal from the objects

Penetration depth of the primary radiation can be reduced down to few nm for incidence angles α_i close to the critical angle α_c of total external reflection. Analogously, the escape depth can also be tuned by changing the take-off angle α_f.

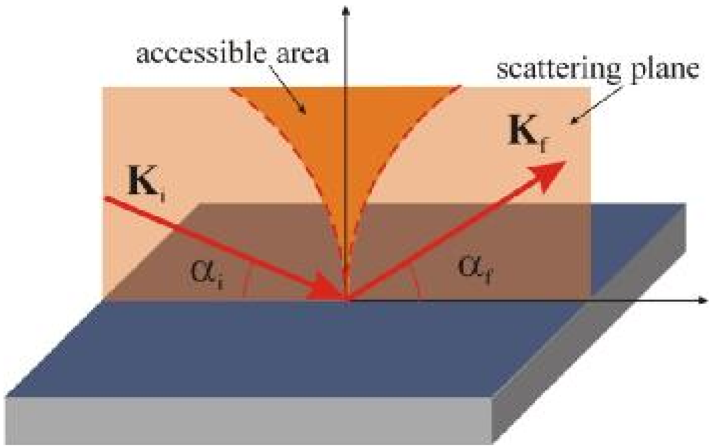
penetration depth of x-rays in Si



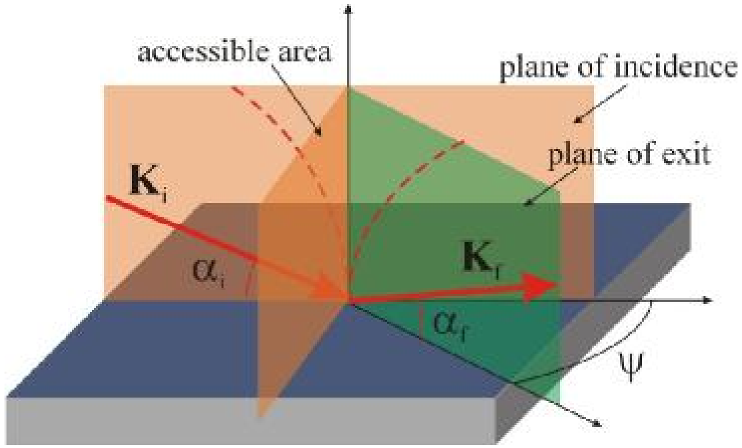


penetration depth in μm

coplanar arrangement – x-ray reflection (XRR) and x-ray diffraction (XRD)



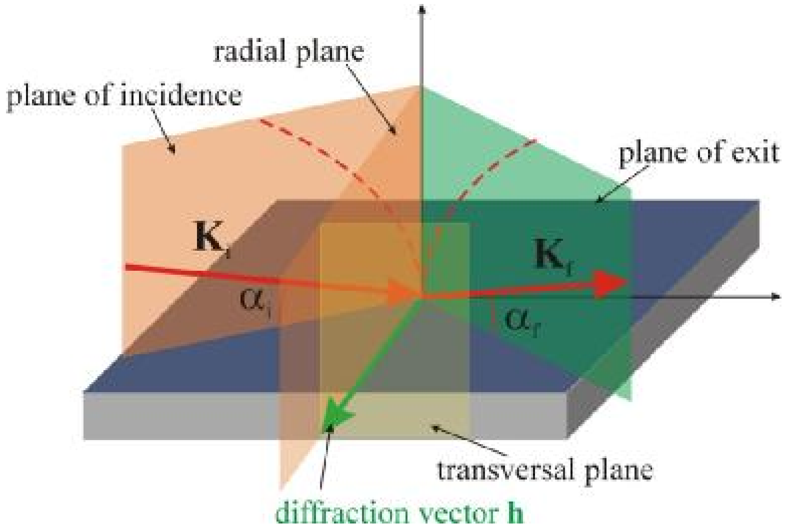
Non-coplanar grazing-incidence small-angle x-ray scattering (GISAXS)



scattering vector: $\boldsymbol{Q} = \boldsymbol{K}_f - \boldsymbol{K}_i$

reduced scattering vector: $\boldsymbol{q} = \boldsymbol{Q} - \boldsymbol{h}$

Grazing-incidence diffraction (GID)



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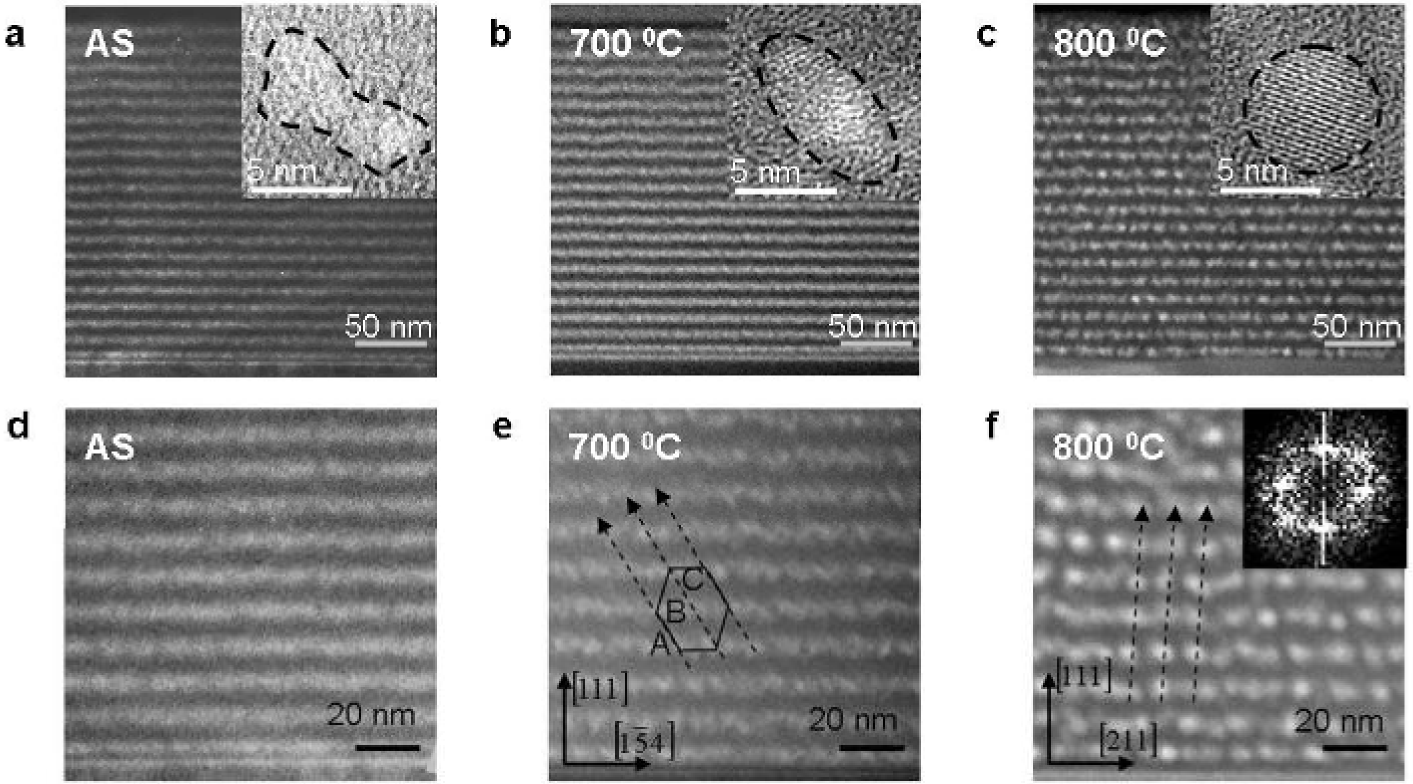
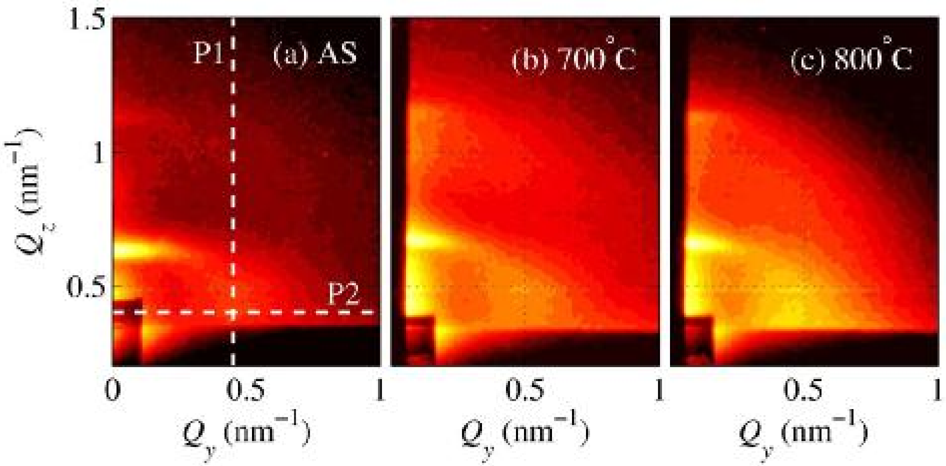
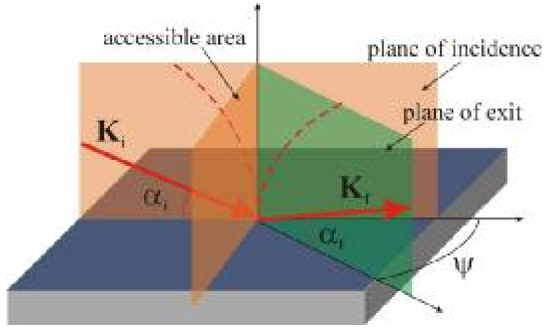
Ge nanocrystals in Ge+SiO_2/SiO_2 multilayer

magnetron sputtering deposition (IRB Zagreb)

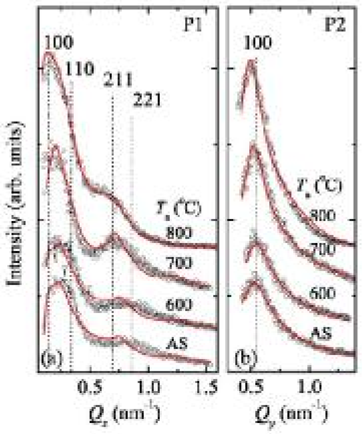


GISAXS measurement (SAXS beamline @ ELETTRA)

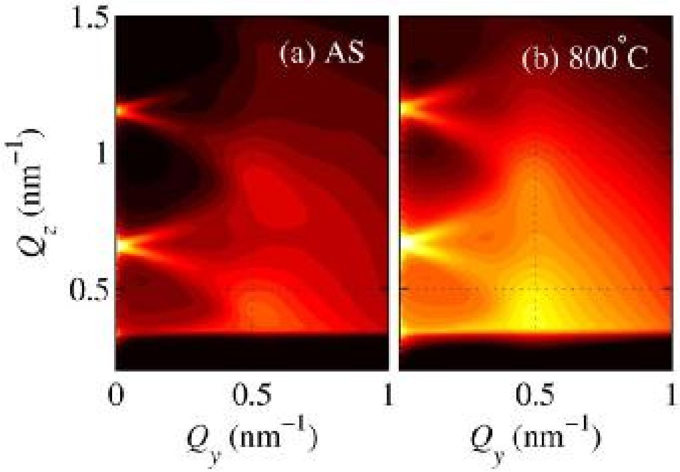




Fit to the theory – linescans:



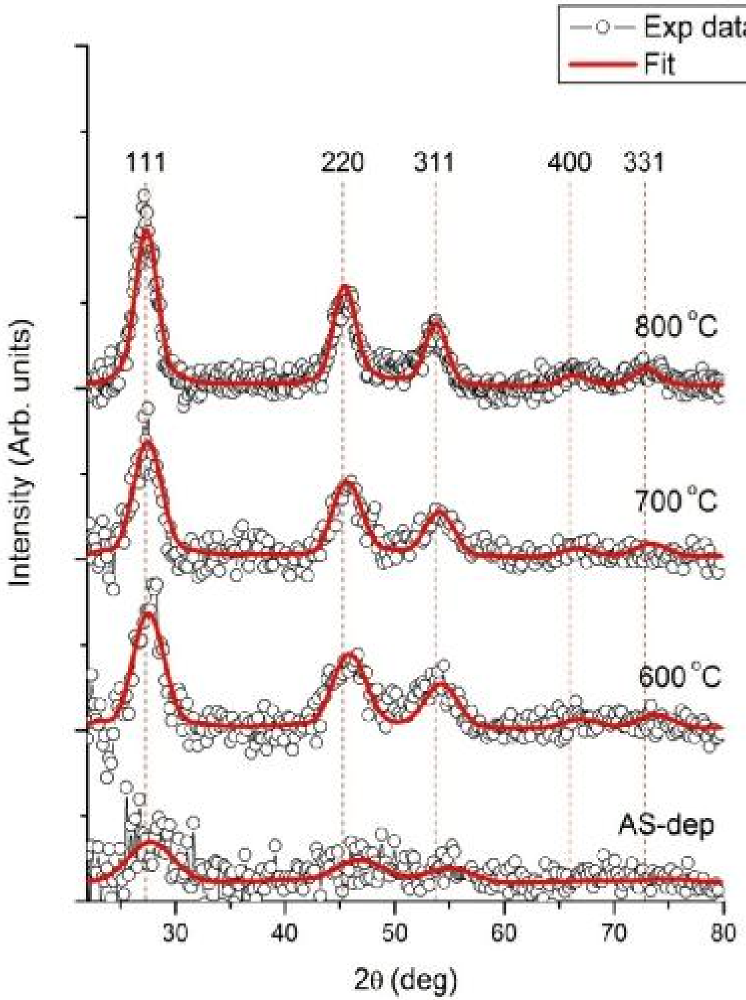
simulated intensity maps



⇒ size of individual nanocrystals, correlation parameters of their positions

See the talks by E. Majková and P. Šiffalovič

X-ray diffraction – standard laboratory diffractometer, parallel-beam geometry

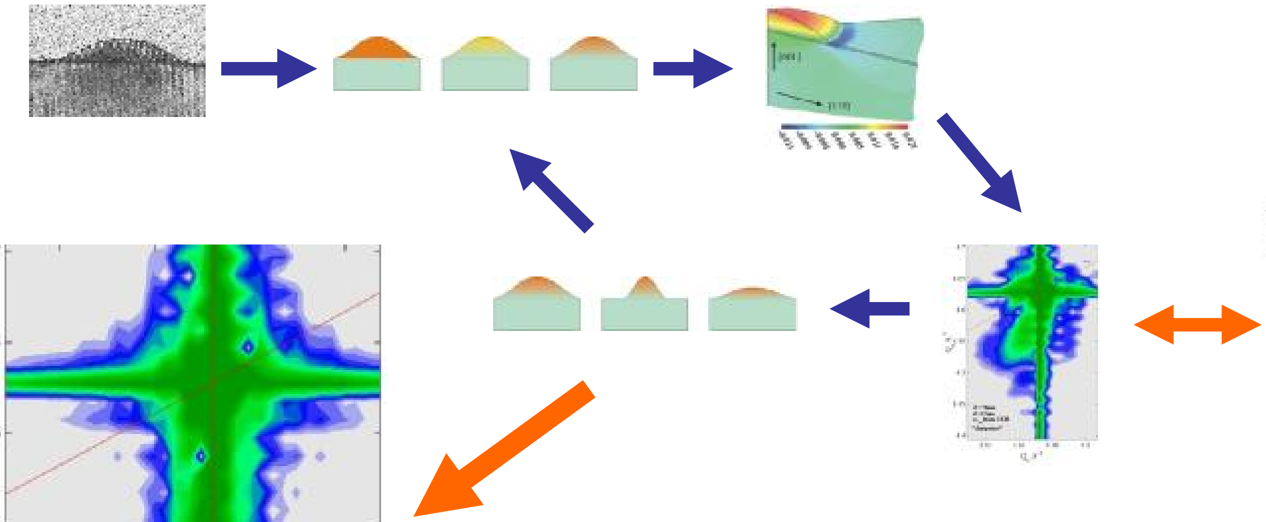


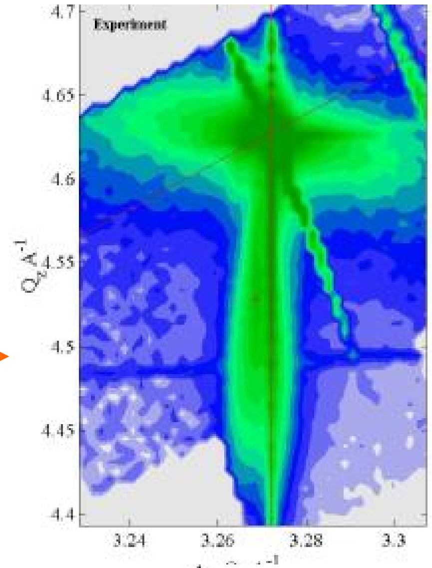
with increasing annealing temperature the density of defects (stacking faults, microtwins) increases

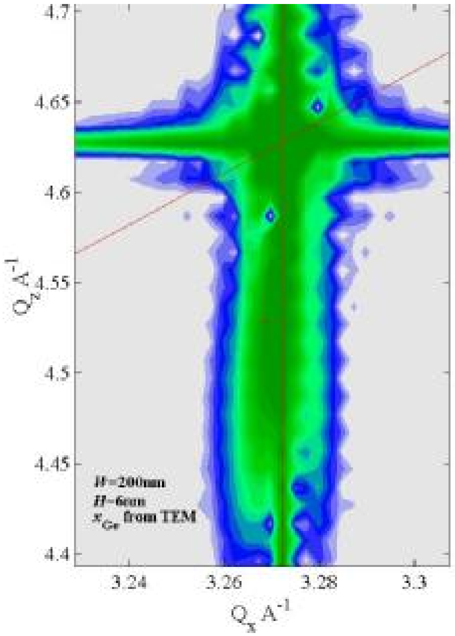
GeSi free standing quantum dots on Si sample grown by MBE, JKU Linz (Z. Zhong, G. Bauer, F. Schaeffler) ID10B@ESRF, J. Stangl, A. Hesse

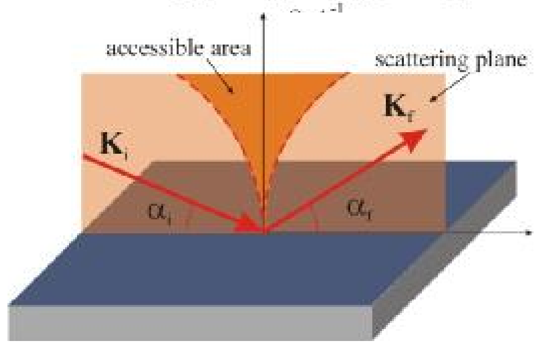
a “brute-force” approach

A. Hesse, J. Stangl, APL (2002)







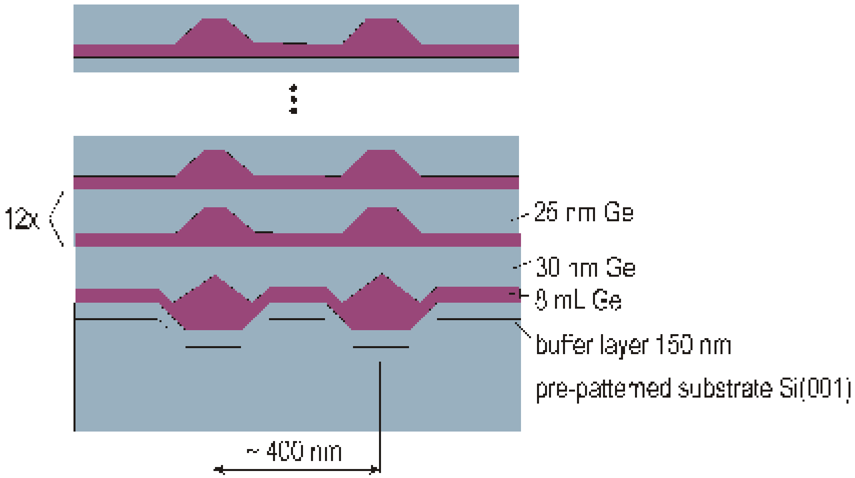


Very boring and extremely time-consuming!!!!!

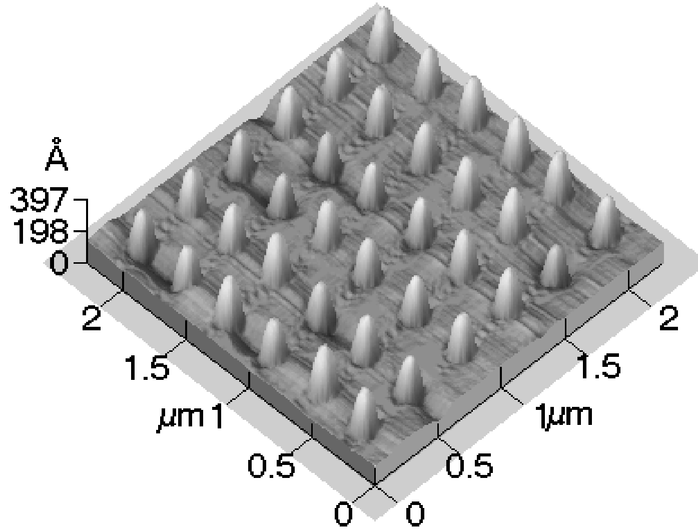
courtesy of J. Stangl

Ge quantum dots in a Ge/Si superlattice grown on a pre-patterned substrate

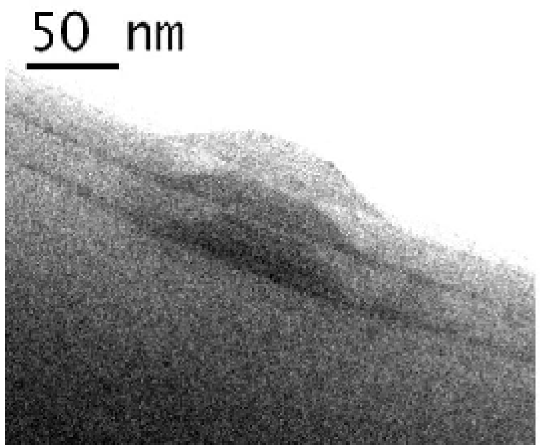
Samples grown by MBE at JKU Linz (prof. G. Bauer) and by D. Grützmacher (Jülich)



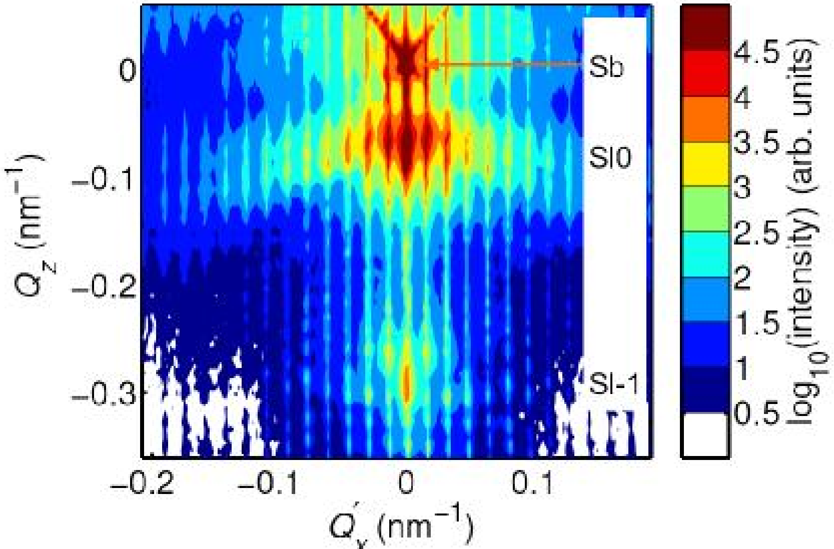
AFM: the first 2D layer of the dots

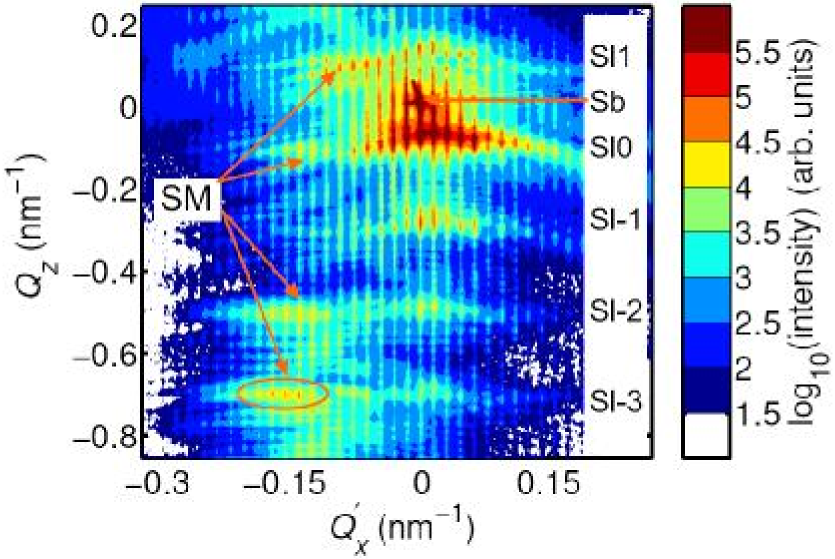


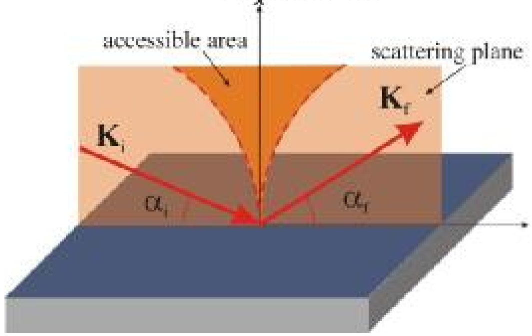
Cross-section TEM:



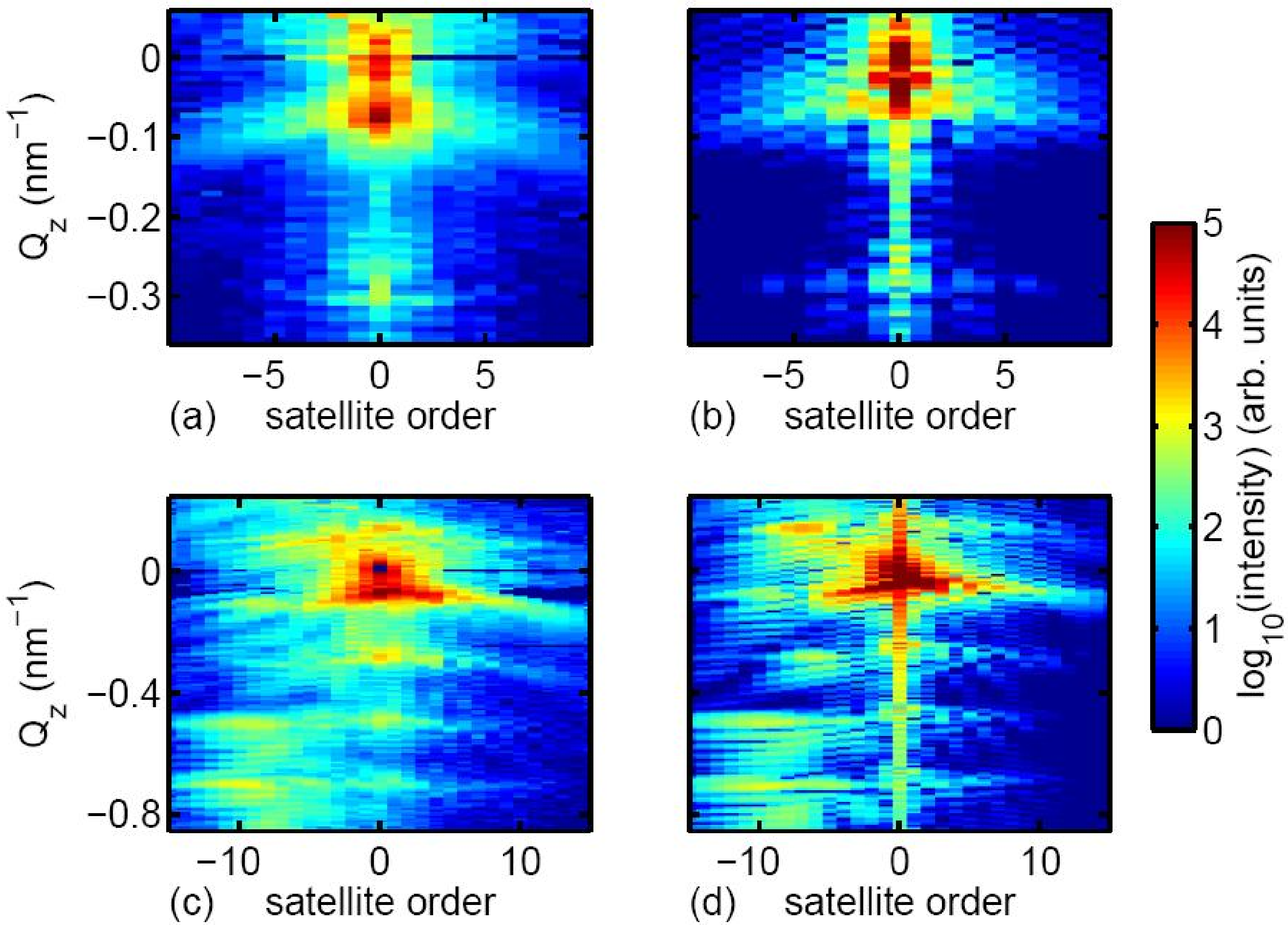
Coplanar 004 and 224 reciprocal space maps measured at the beamline ID10B, ESRF
λ = 1.547 Å

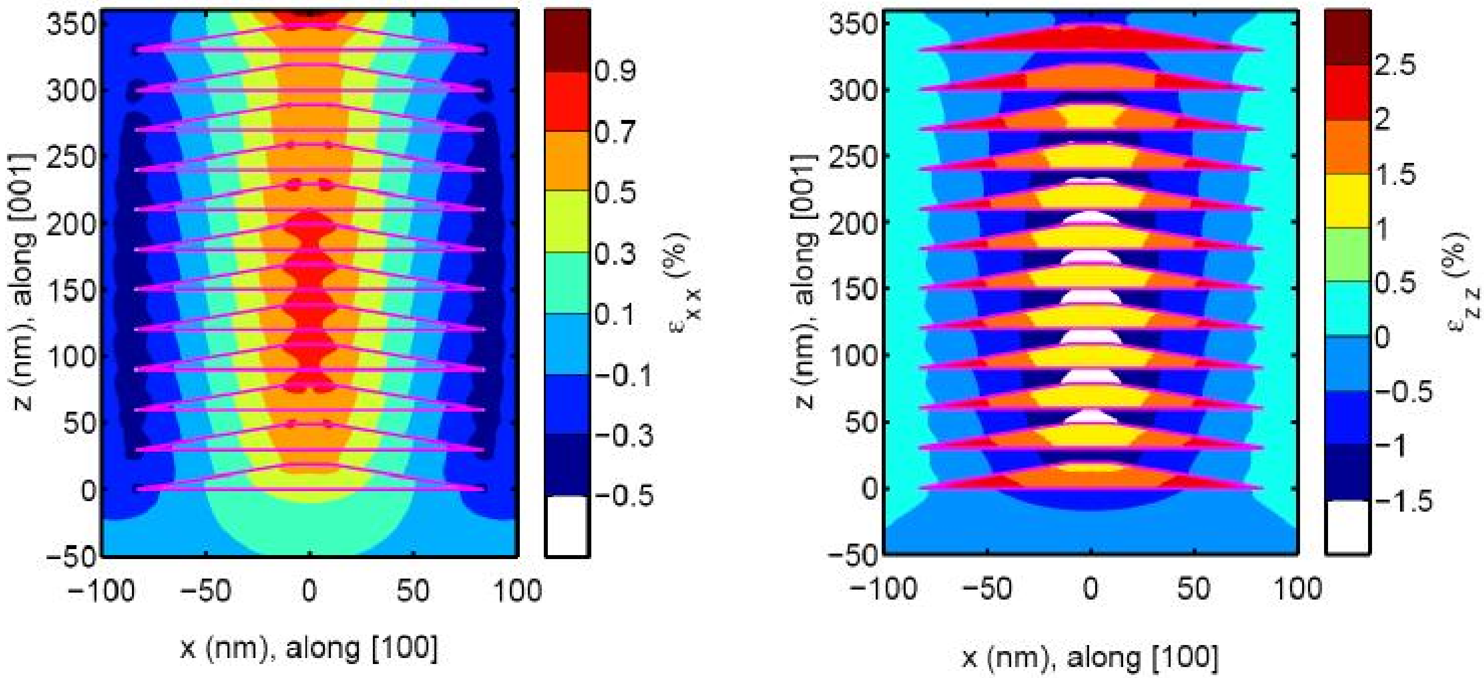






Experimental (left) and simulated (right) heights of the satellites in diffractions 004 (upper row) and 224 (lower row)





Resulting distribution of the strain tensor: the lateral (left) and the vertical components (the right panel). The Ge content increases from 30% (the bottom of the dot) to 45% at the dot apex.

J. Novák et al., J. Appl. Phys 98, 073517 (2005).
V. Holý et al. Phys. Rev. B 79, 035324 (2009).

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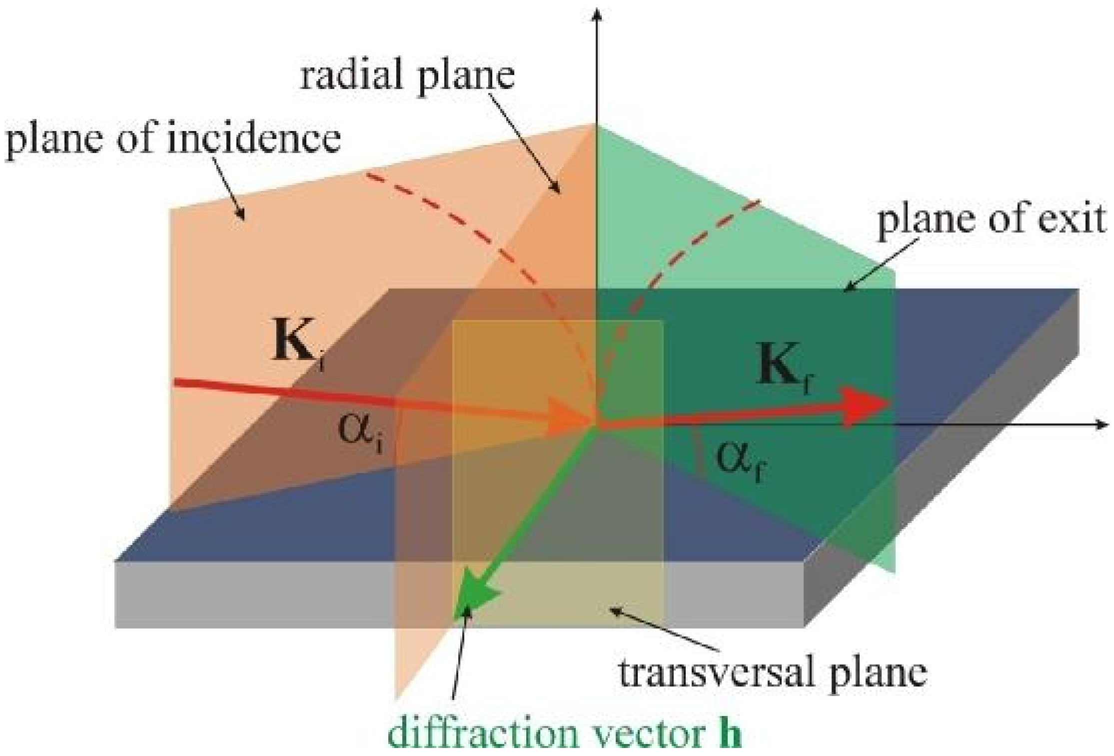
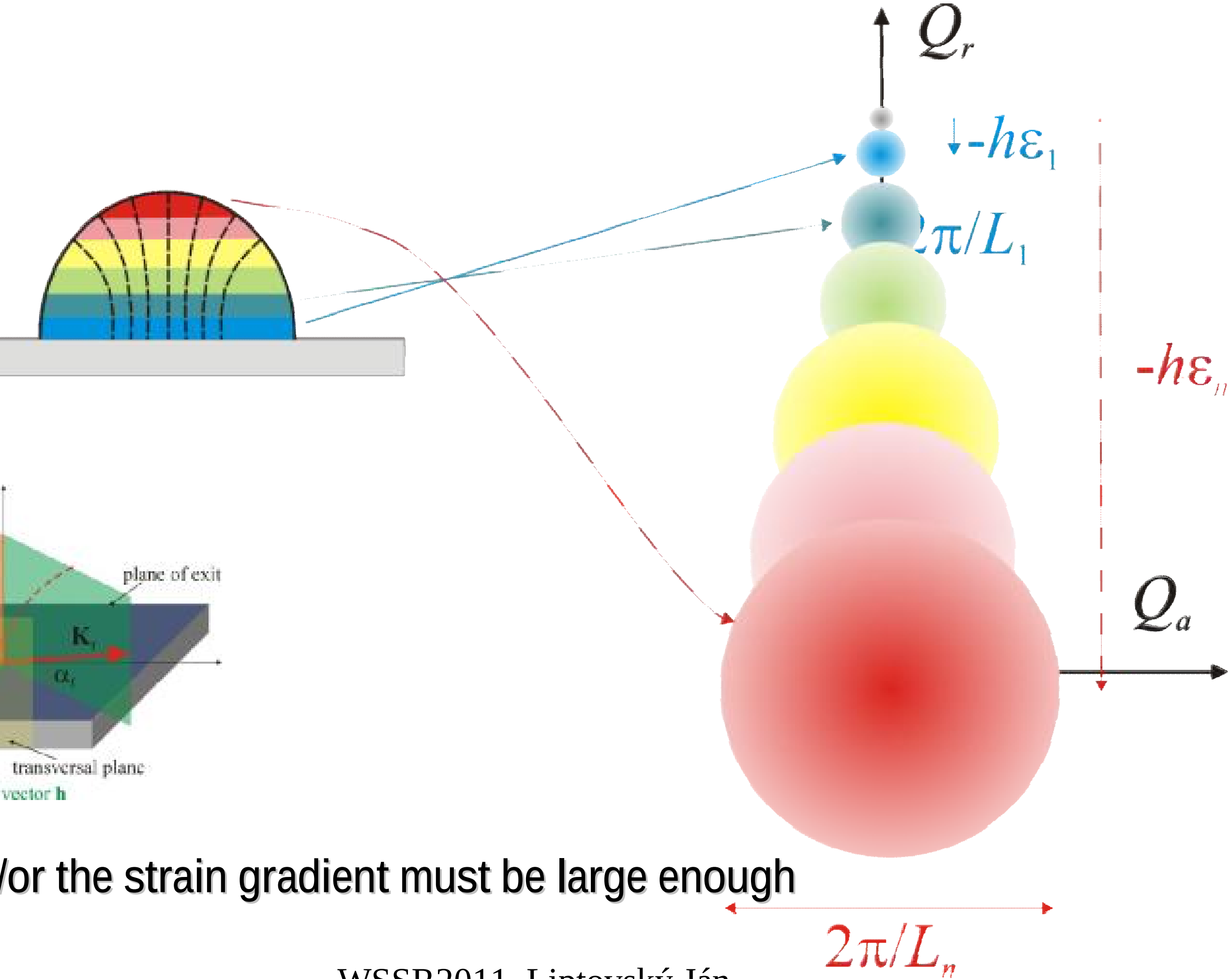
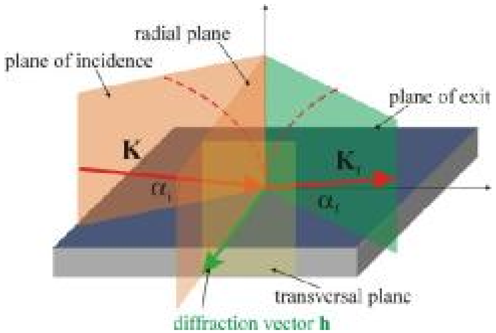
Iso-strain method:

decomposition of an island into disks of constant strain



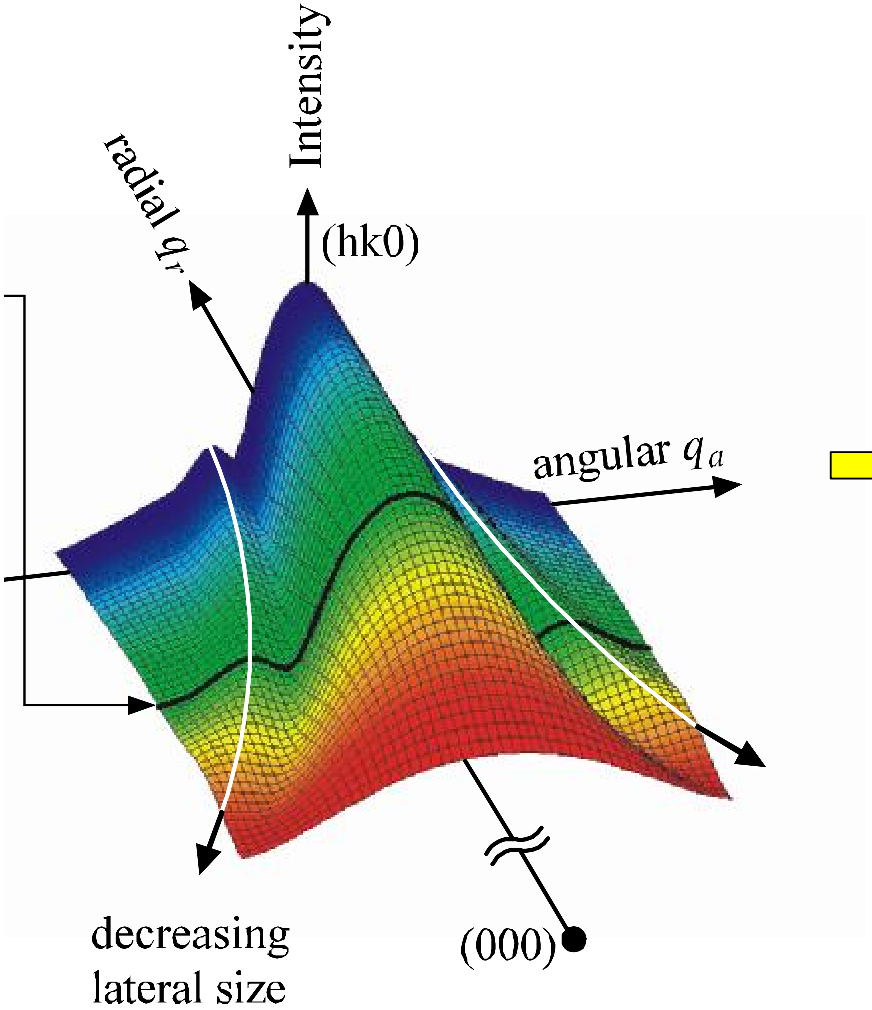
lateral size of iso-strain volumes

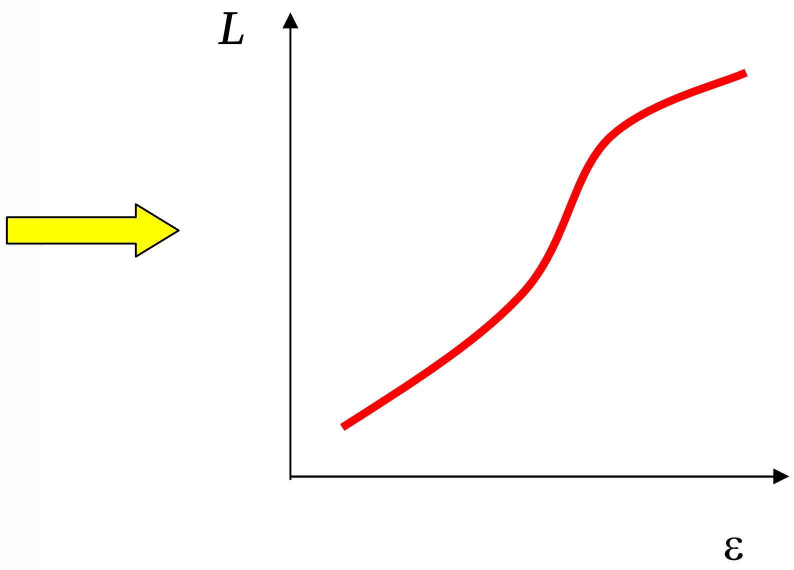




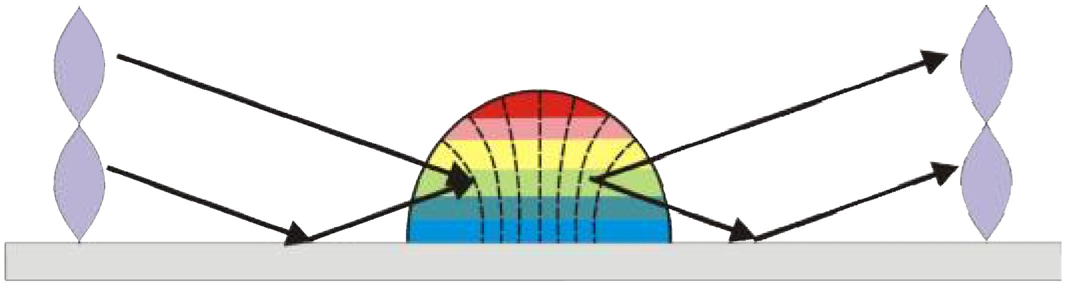
The dots and/or the strain gradient must be large enough

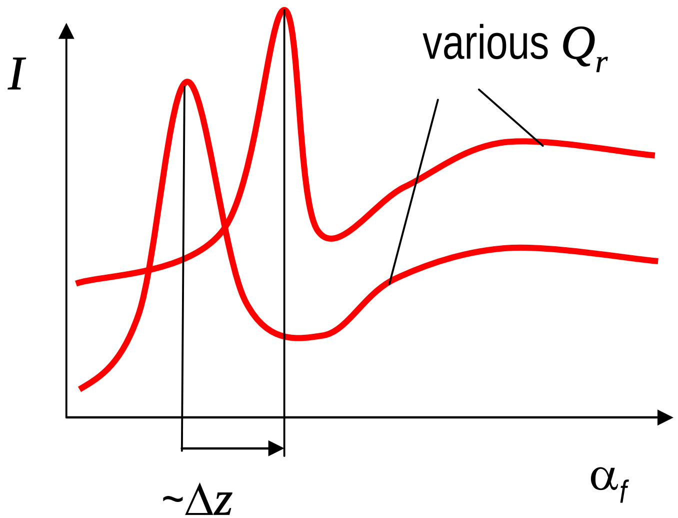
I. Kegel et al., Phys. Rev. Lett. 85, 1694 (2000); Phys. Rev. B 63, 035318 (2001).

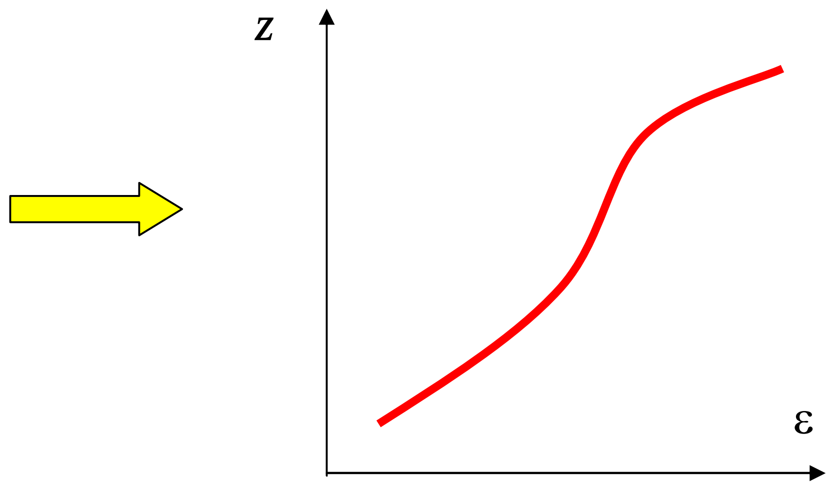




Vertical positions of iso-strain volumes

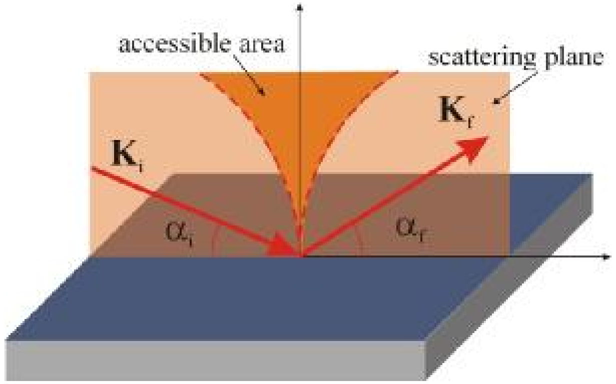


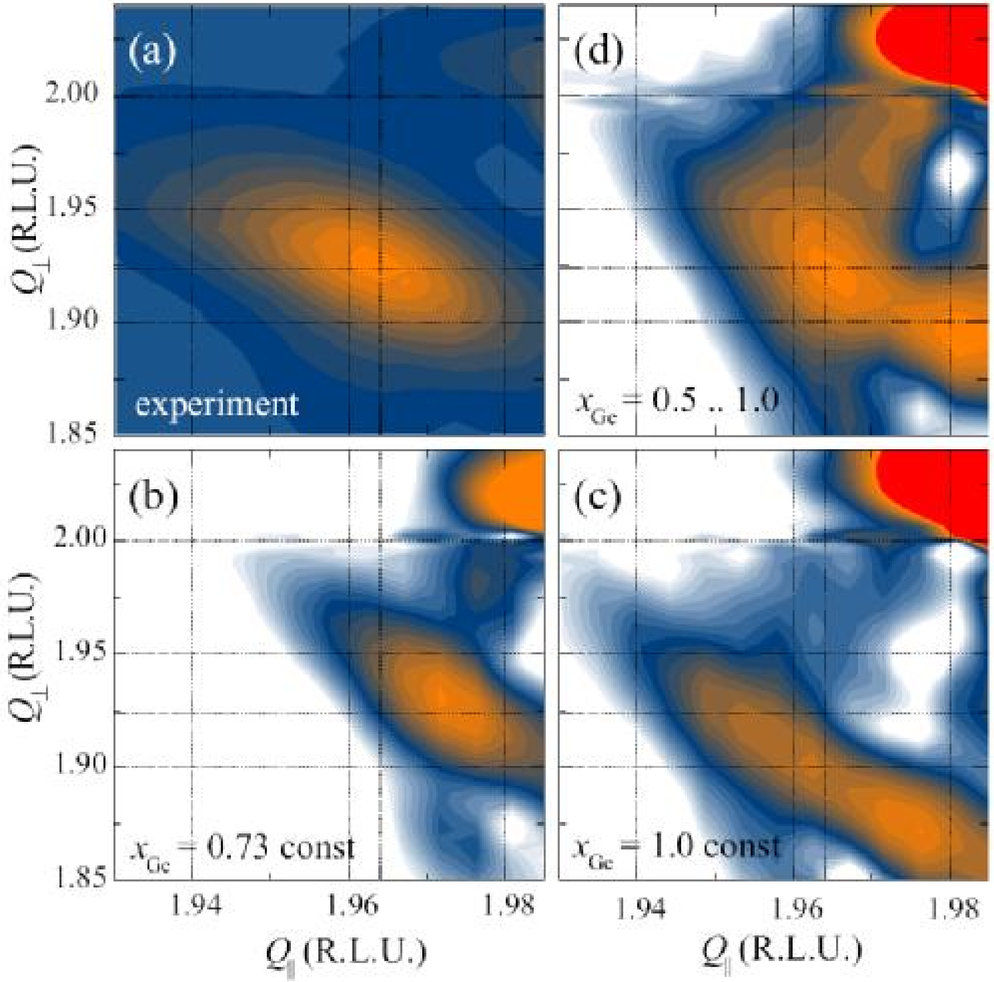




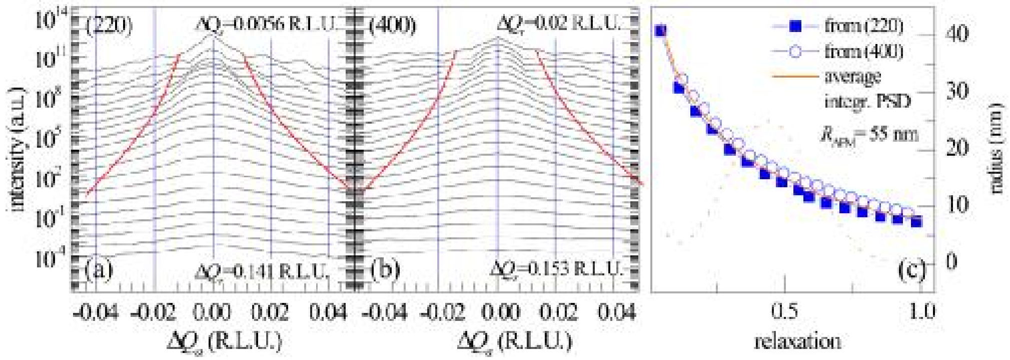
Ge islands on Si:

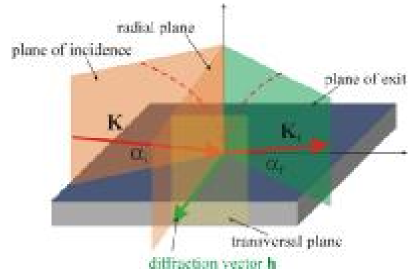
Conventional 202 reciprocal space map and its simulations



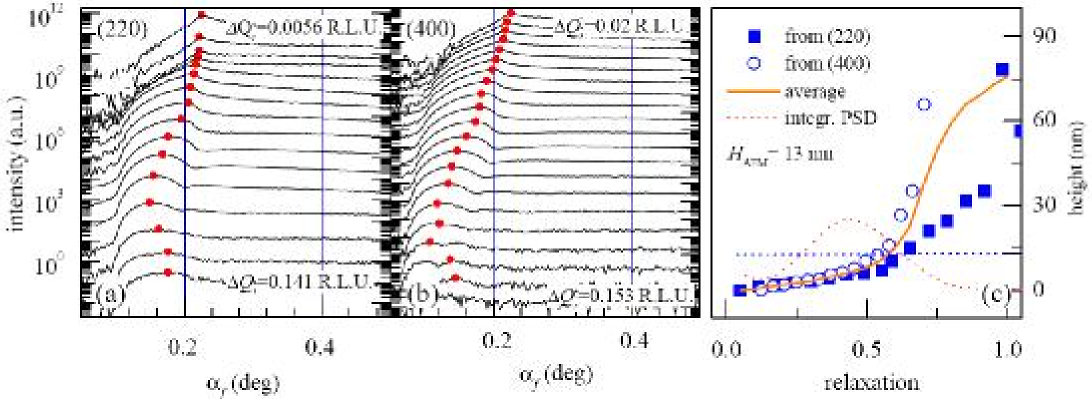


angular scans measured in 220 and 400 (a and b) and the resulting dependence of the radius of the iso-strain area on the relaxation degree (c)





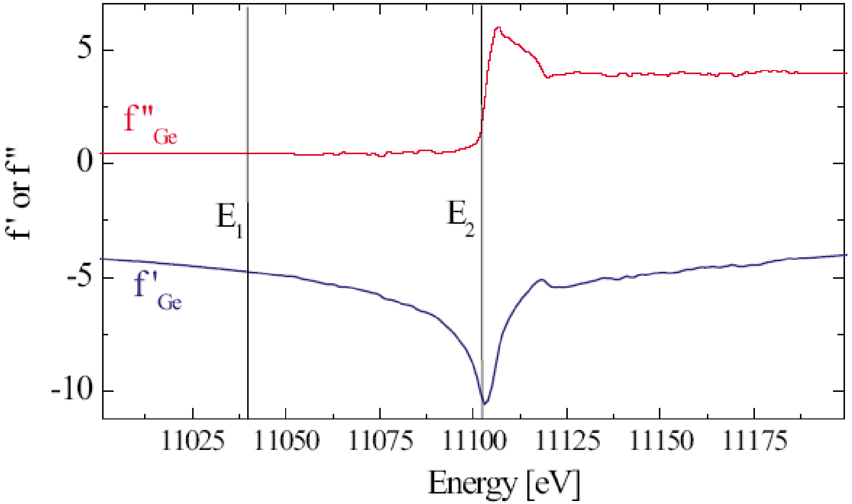
the α_f scans taken for various radial positions q_r in 220 and 400 diffractions (left) and the resulting dependence of the vertical position of the iso-strain area on the relaxation degree



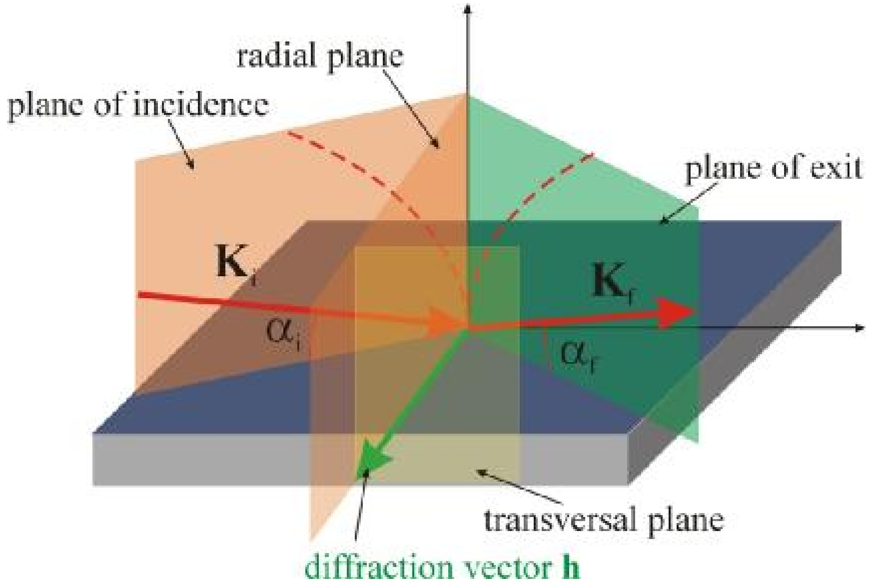
J. Stangl and V. Holy, *X-ray scattering methods for the study of epitaxial self-assembled quantum dots,* in: Quantum dots: Fundamentals, applications and frontiers, eds. B. A Joyce, P. C. Kelires, A. G. Naumovets and D. D. Vvedensky, NATO Sci. Series II, vol. 190, Springer, Dordrecht 2005

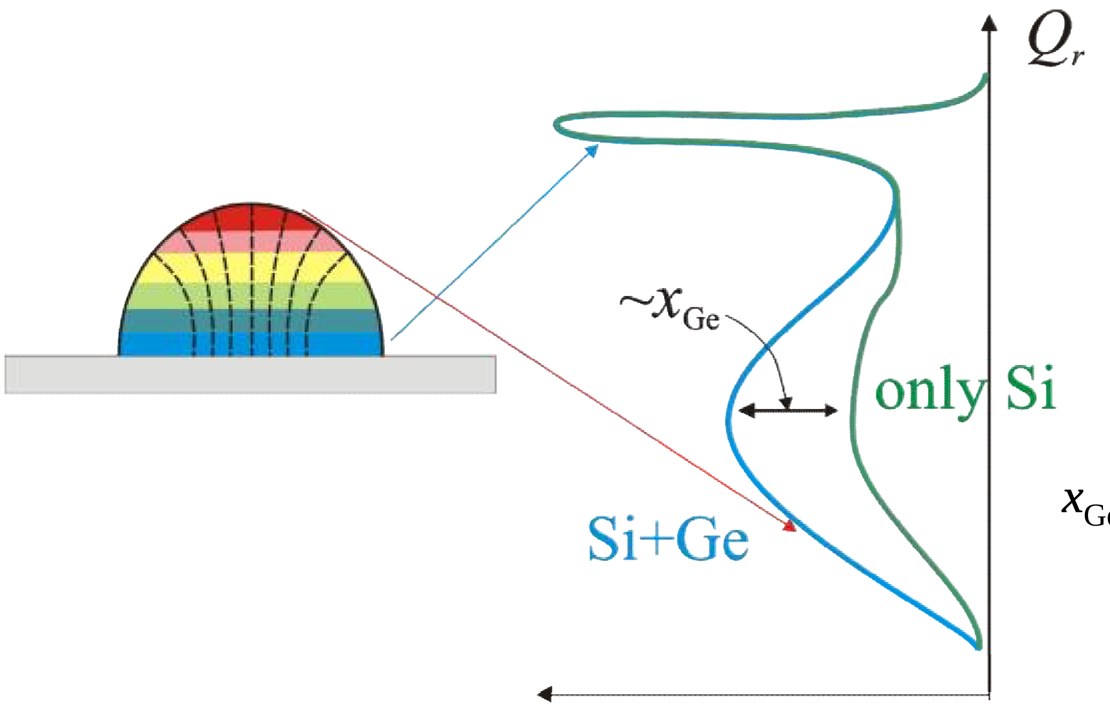
Local chemical composition: Anomalous diffraction

energy dependence of the dispersion corrections for Ge

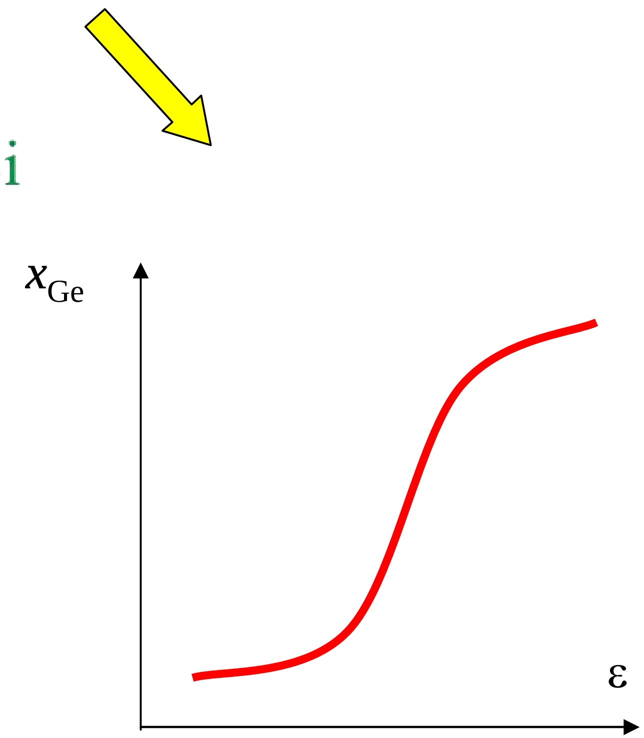


Grazing-incidence diffraction

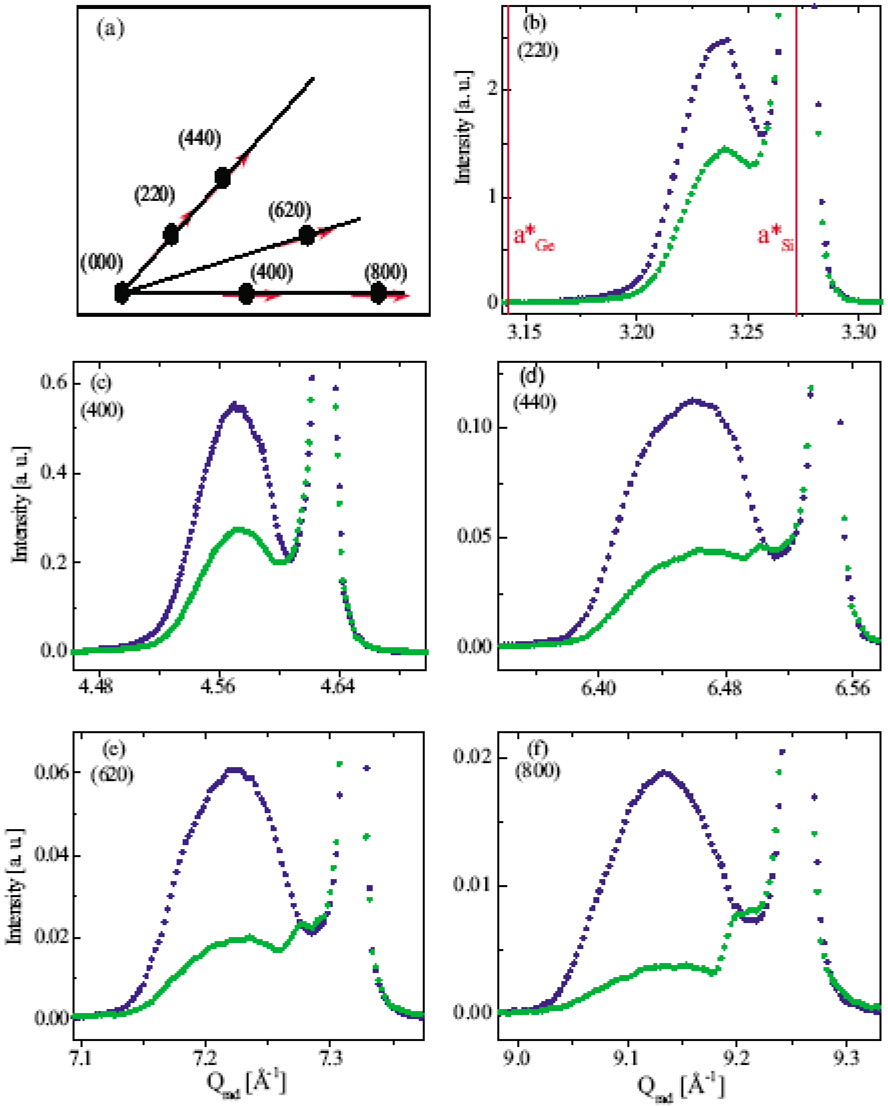




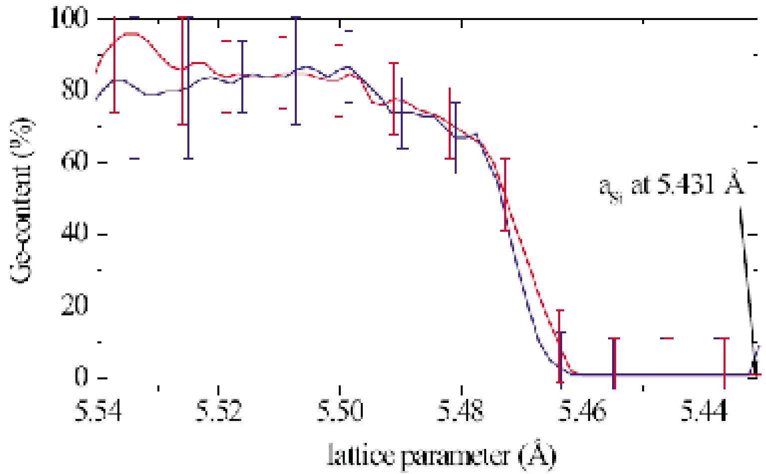
Q_r
$\sim x_{Ge}$
only Si
Si+Ge



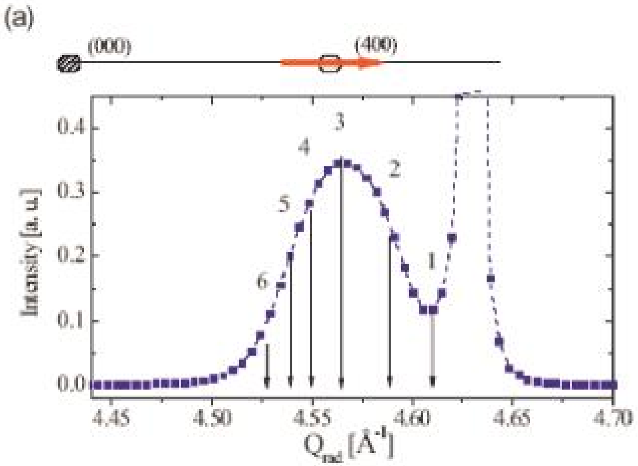
x_{Ge}
ε

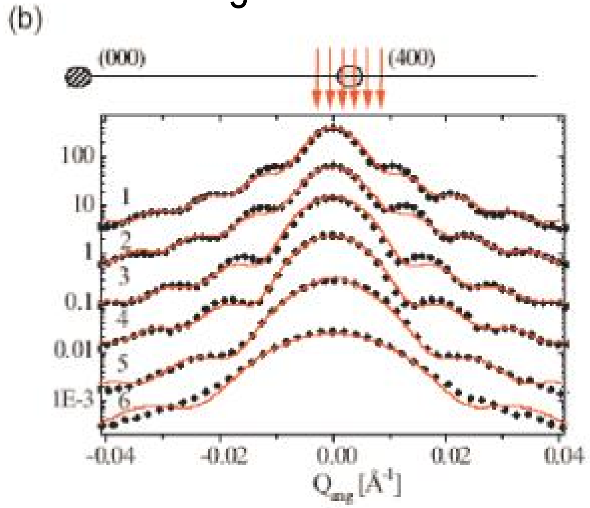


green dots – higher energy, only Si
blue dots – lower energy

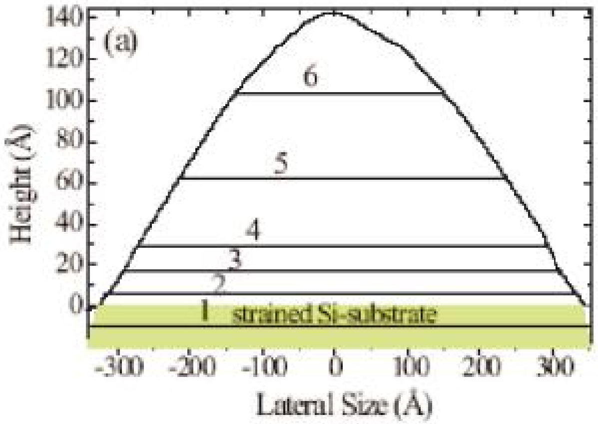


Courtesy of Tobias Schuelli, CEA Grenoble series of angular scans

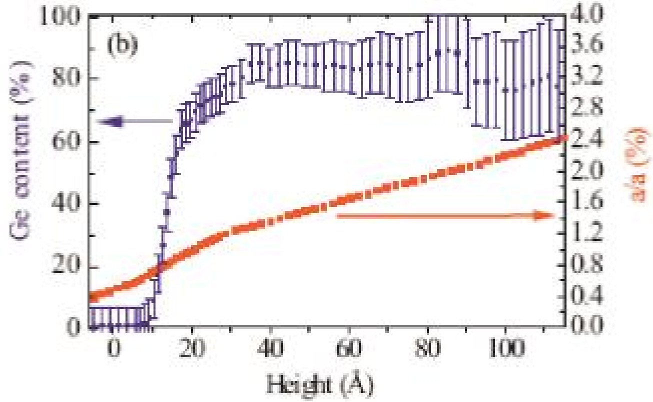




reconstructed dot shape



reconstructed profile of Ge content



T. U. Schuelli et al, PRL 90, 066105 (2003).

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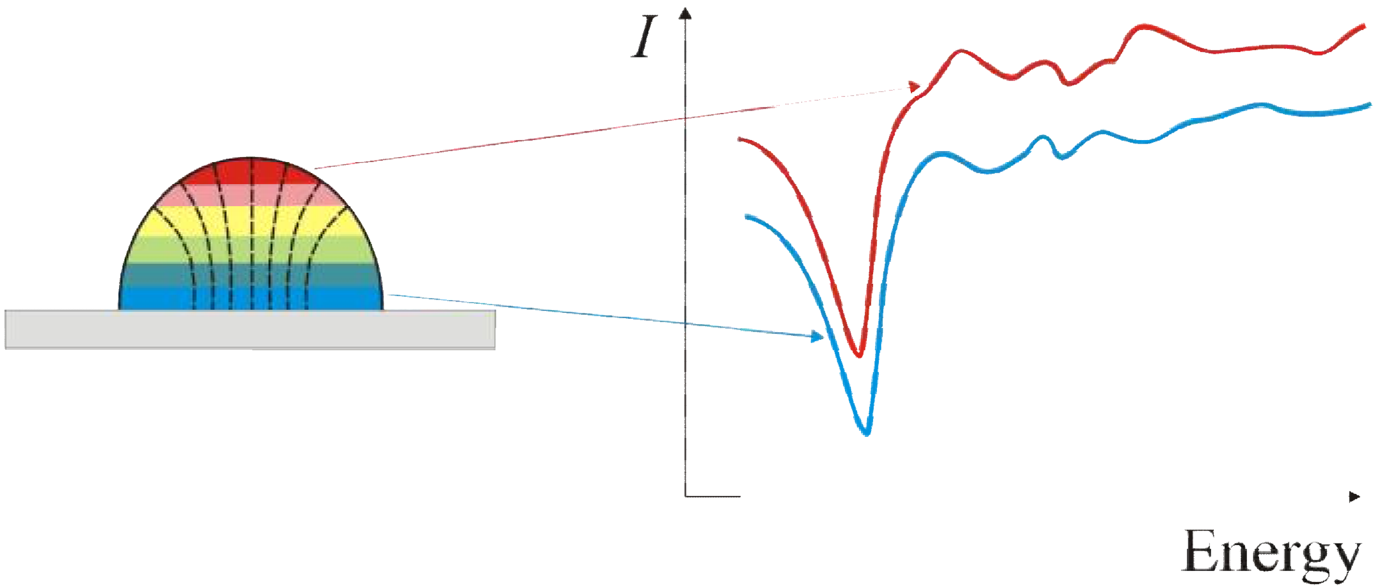
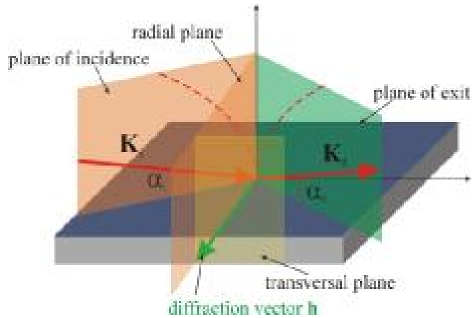
DAFS: Combination of x-ray absorption spectroscopy (EXAFS) and diffraction – measurement of the energy dependence of the diffracted intensity with constant scattering vector ***Q***

The scattered intensity stems from a certain part of the sample with given strain (iso-strain volume)

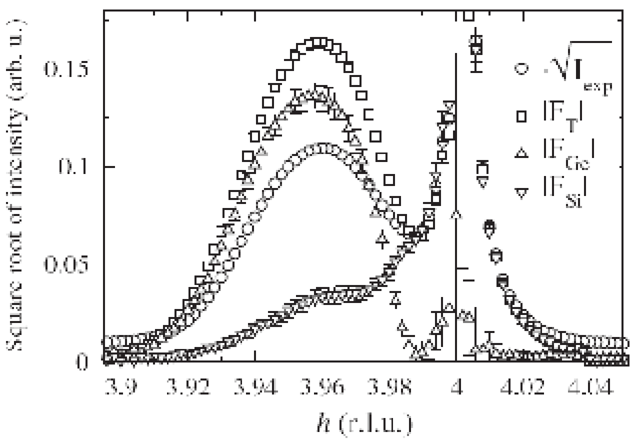


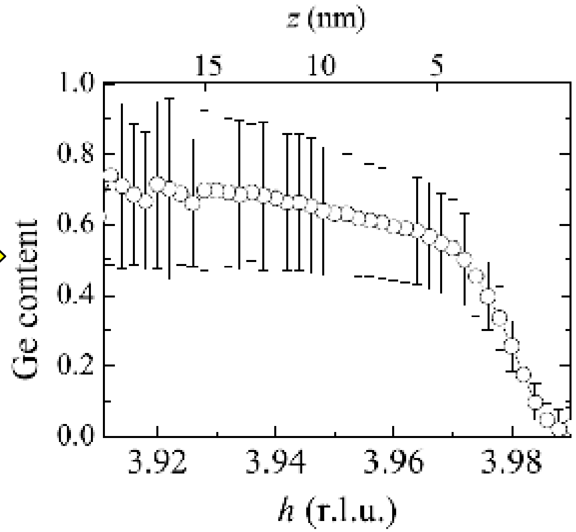
Ge quantum dots on flat and pre-patterned Si(001) substrates

T. U. Schuelli et al., Phys. Rev. Lett. 102, 025502 (2009)
M.-I. Richard et al., Eur. Phys. J. 167, 3 (2009)

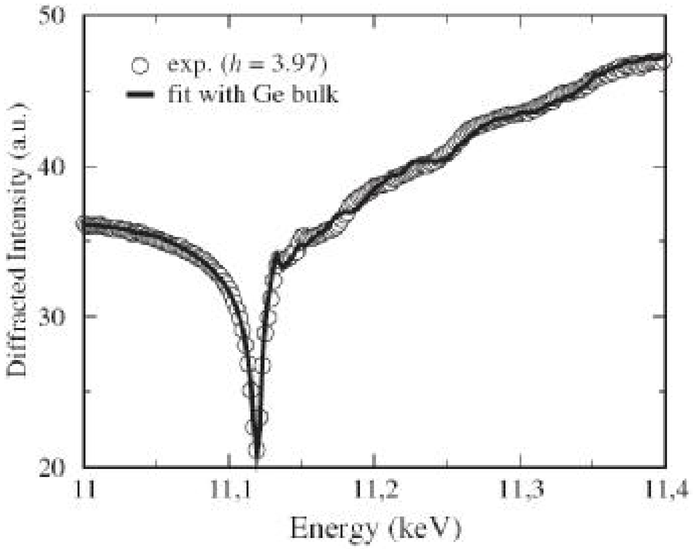


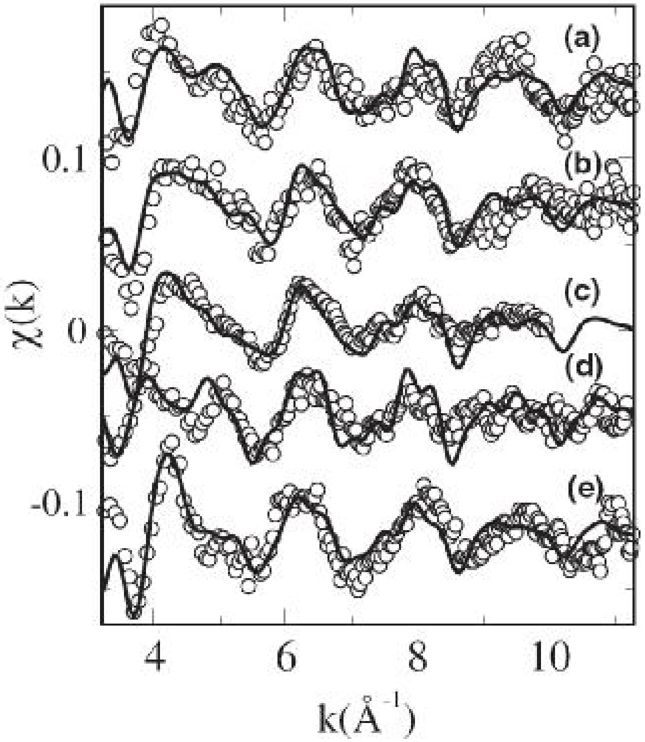
anomalous GID 400





example of a DAFS curve measured at (3.97 0 0)





measured (points) and VFF-simulated (lines) EXAFS oscillations measured in various reciprocal lattice points

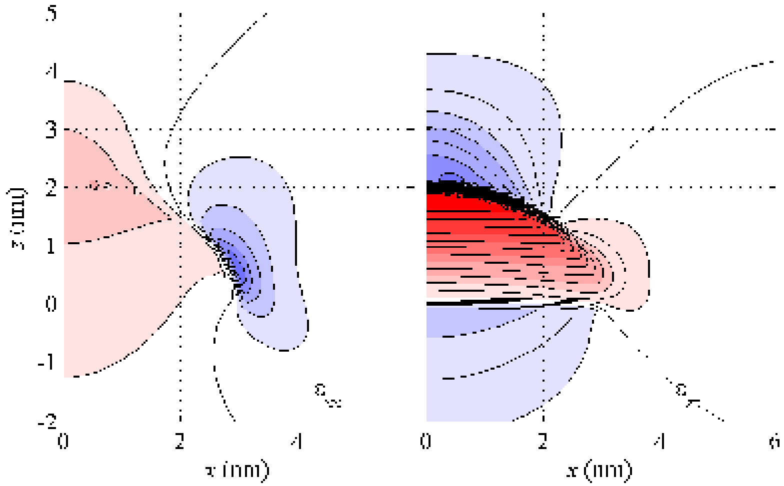
Bond leghts and local Ge content in various parts of the dot volume

CdSe/ZnSe quantum dots

Samples grown by MOVPE, D. Hommel, Univ. Bremen, measurement BM02@ESRF

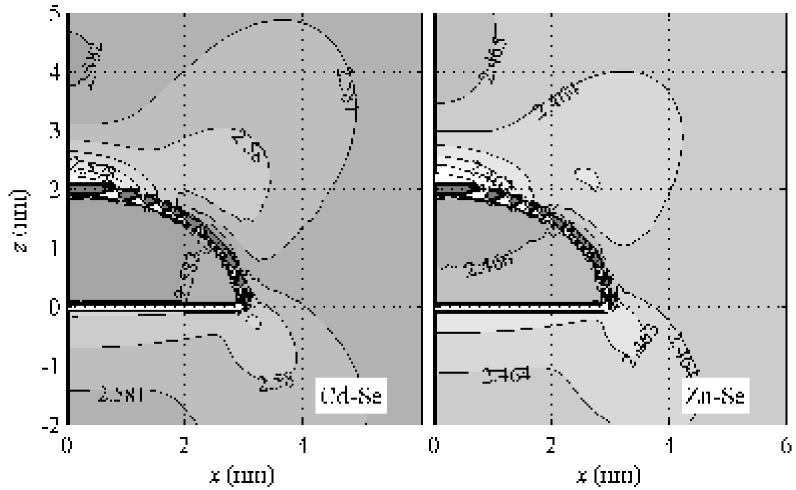
DAFS measurement around the SeK edge

Inhomogeneities in local elastic strains in and around the dots

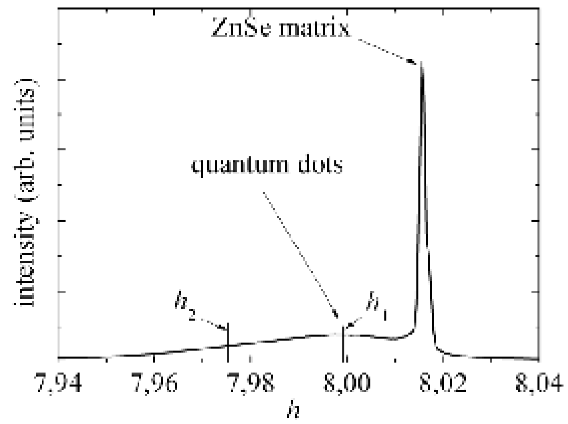


lead to

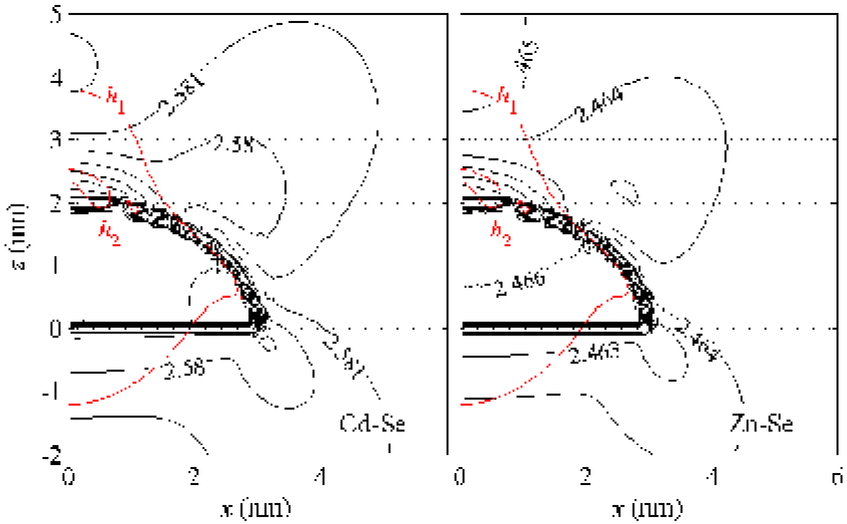
inhomogeneities in the Zn-Se and Cd-Se bondlengths
(calculated by the simplified valence-force field model – d'Acapito)



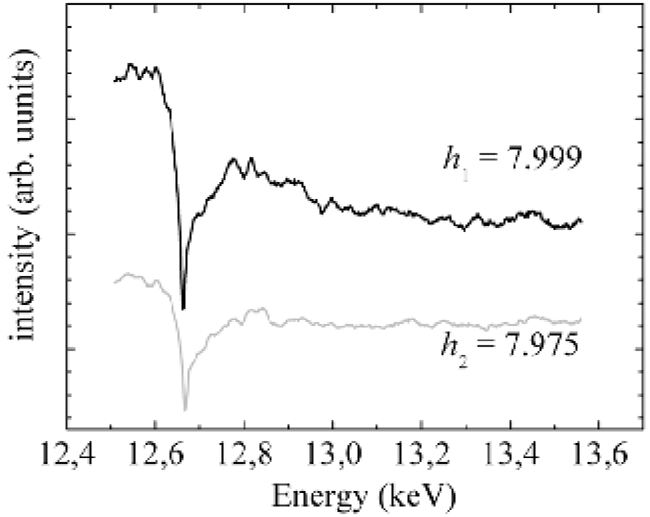
GID, diffraction 800, radial scans

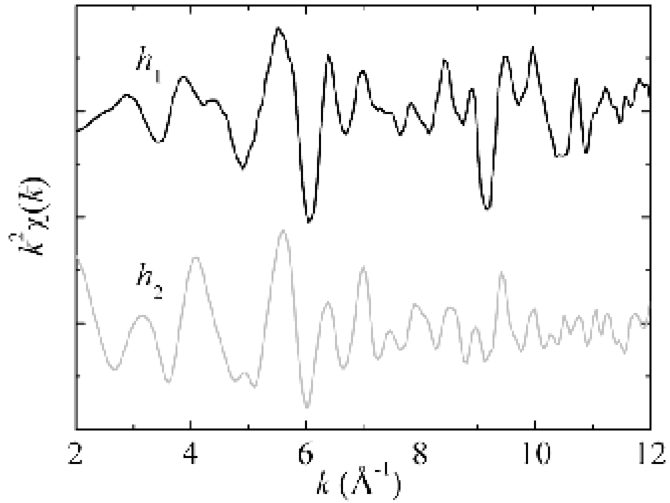


Choosing the working-point (h_1 or h_2) we change the iso-strain volume:



and the DAFS spectra are completely different



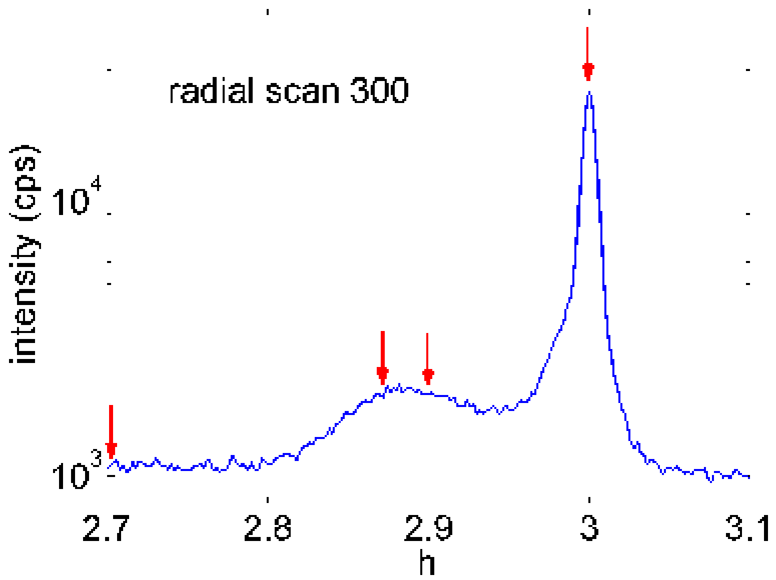


Not finished yet ☹

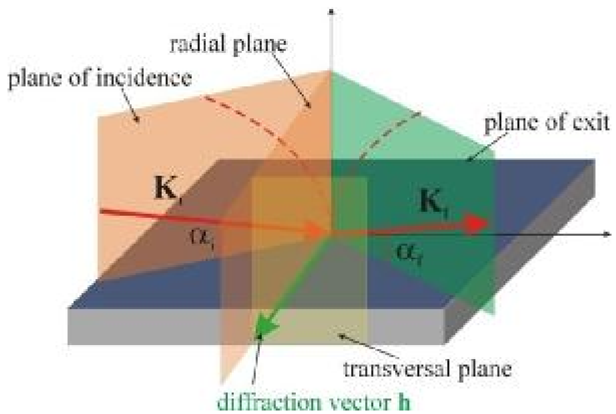
Free-standing InGaN quantum dots on GaN:

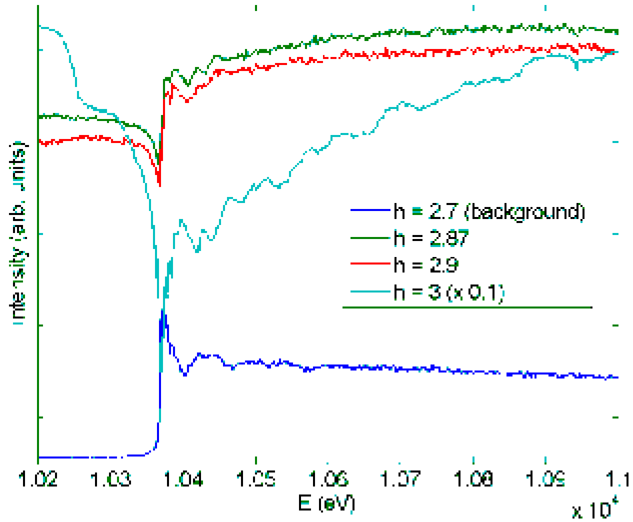
Samples grown by MOVPE, D. Hommel, Univ. Bremen, measurement BM02@ESRF

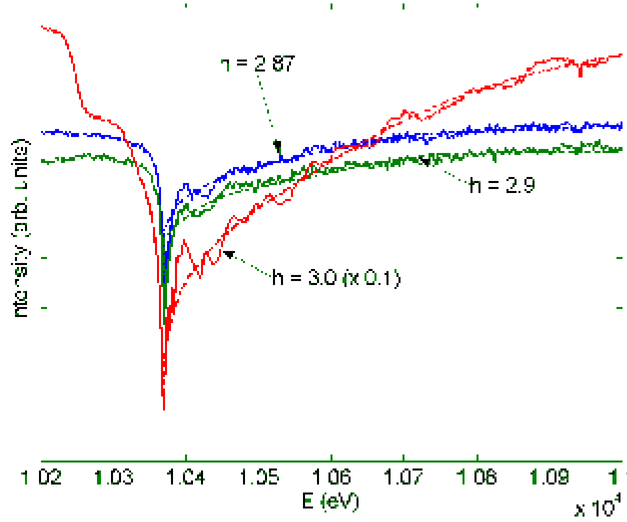
DAFS measurement around the GaK edge, the anomalous atoms (Ga) are everywhere ☹



The DAFS scans were measured in the points denoted by arrows

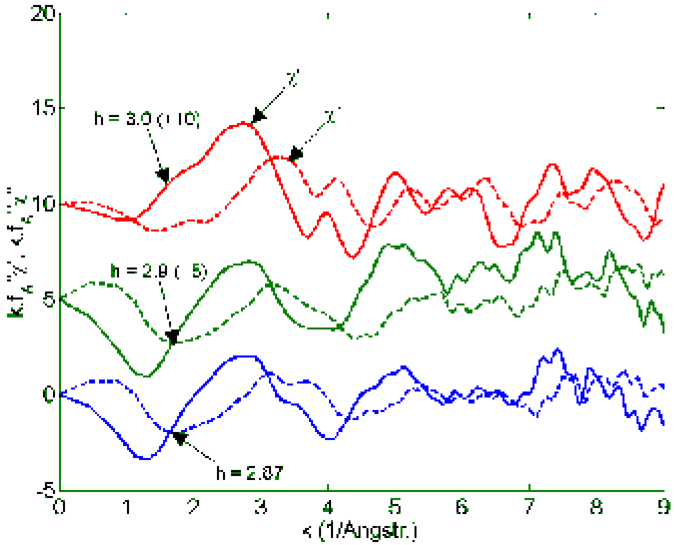




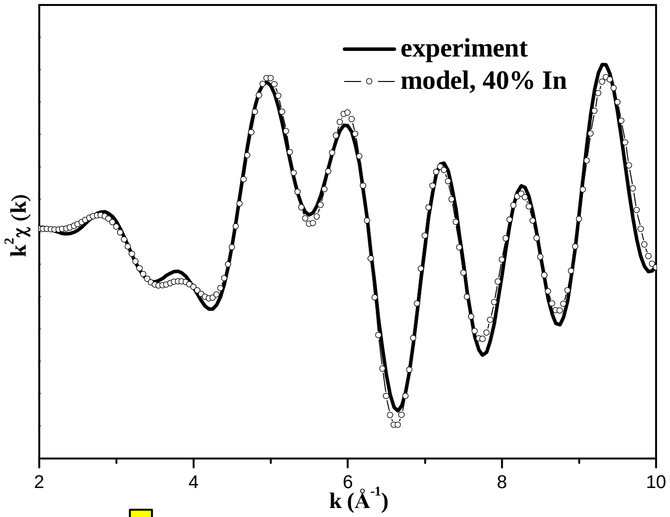


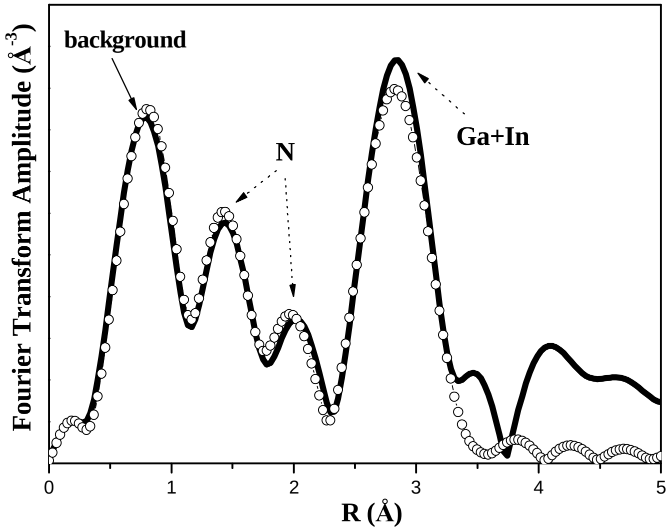
After fluorescence subtraction and fitting by I_{smooth}

Resulting profiles of the oscillatory parts of the Ga scattering factors



Fitting to the $\chi''(E)$ profile to ab-initio simulation results





close to the dot base (h=3.0) the lattice is strained and it contains (20±5)% In
at the dot apex (h=2.9) the lattice is relaxed and it contains (40±5)% In

E. Piskorska et al., phys. stat. sol. (c) 3, 1662 (2006).

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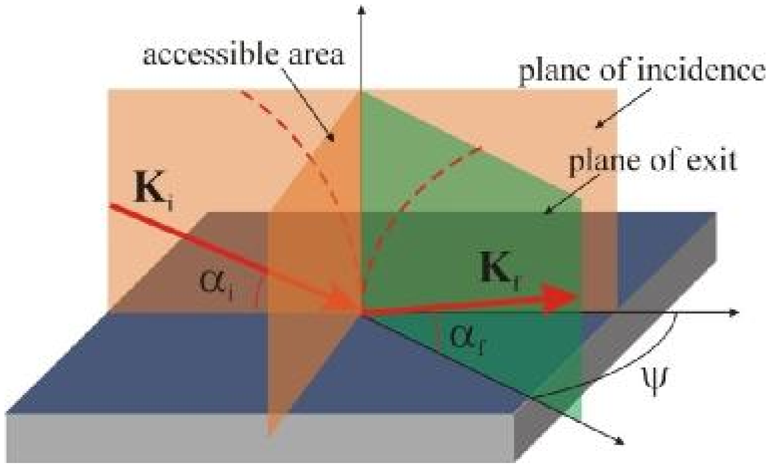
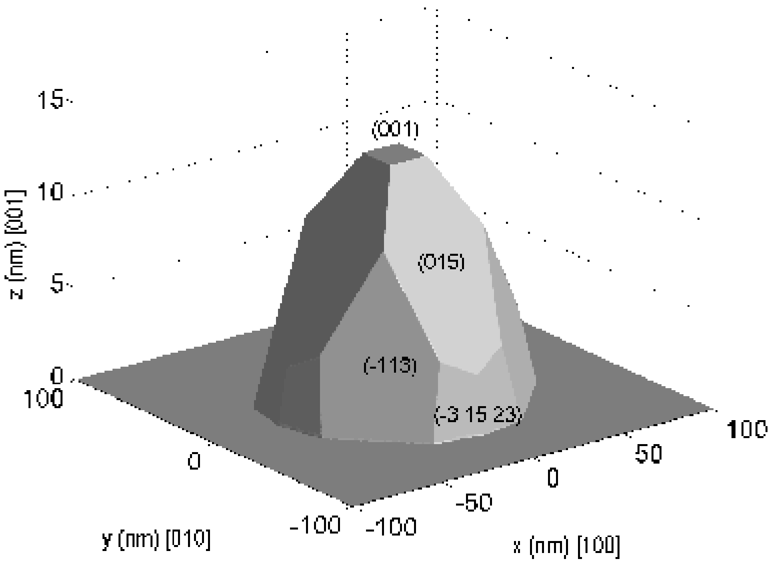
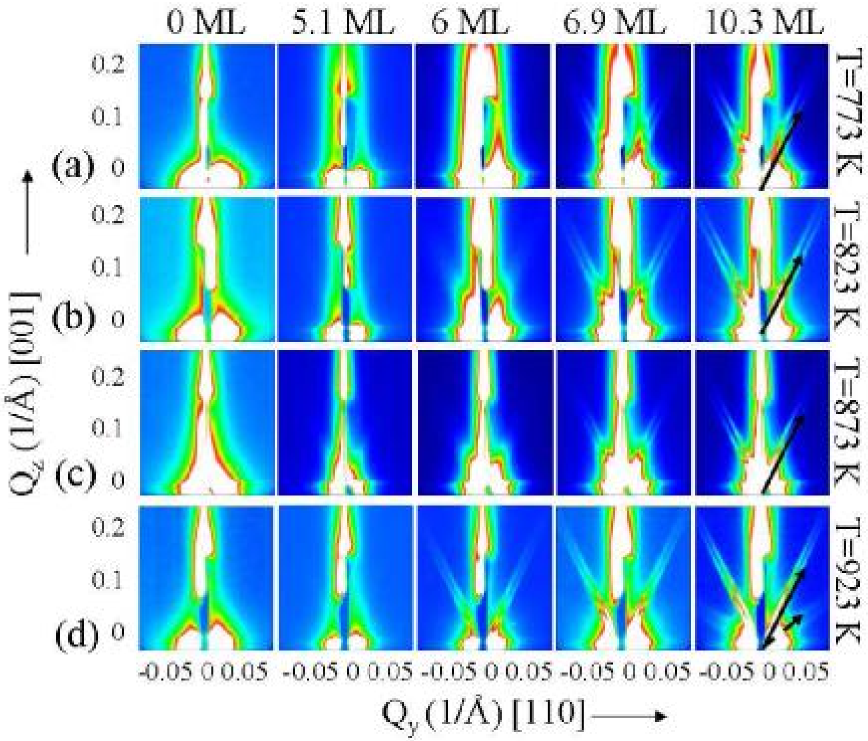
MBE growth chamber attached to the BM32 beamline at ESRF

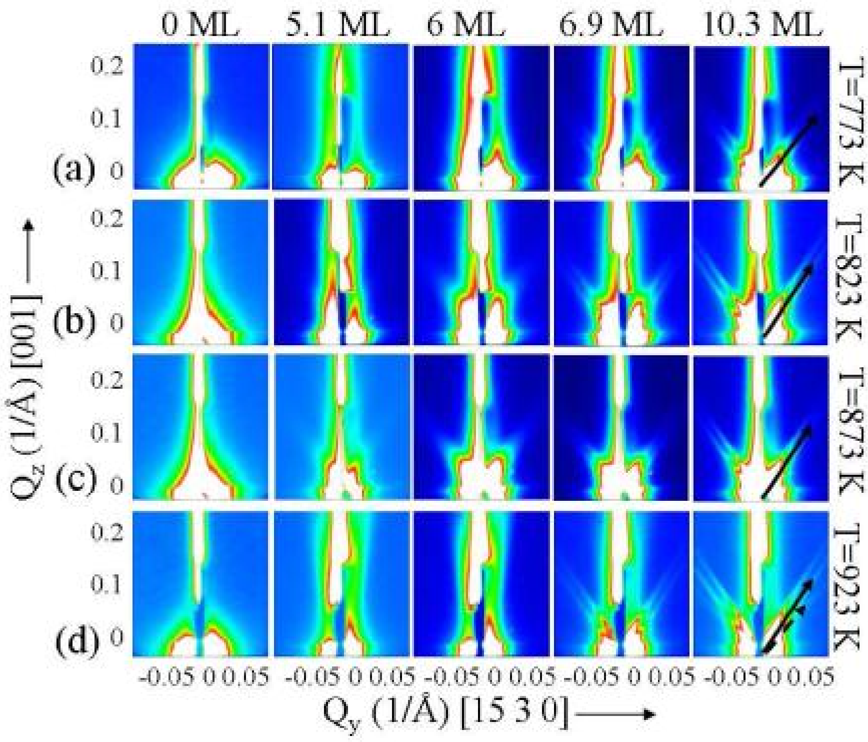
In-situ GISAXS study of the growth of Ge domes on Si(001)

measurement M.-I. Richard, T. Schuelli, G. Renaud







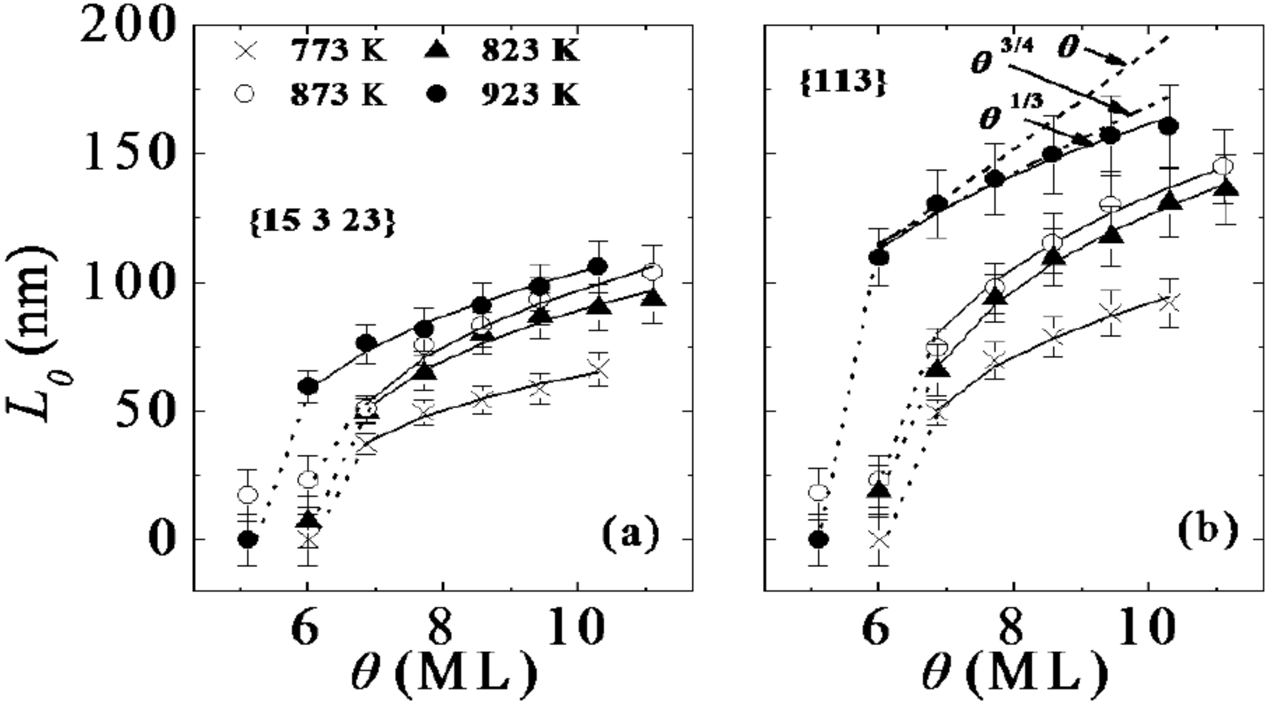


→ {113}

⇢ {111}

→ {15 3 23}

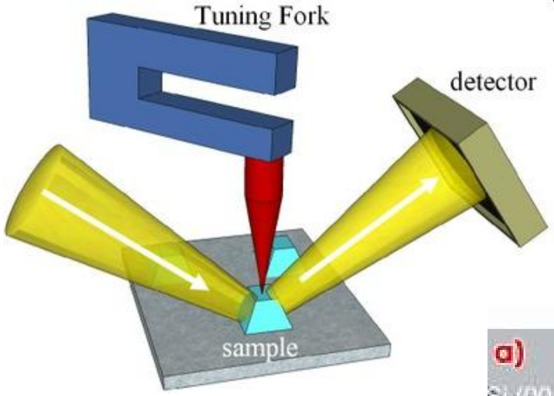
⇢ {20 4 23}



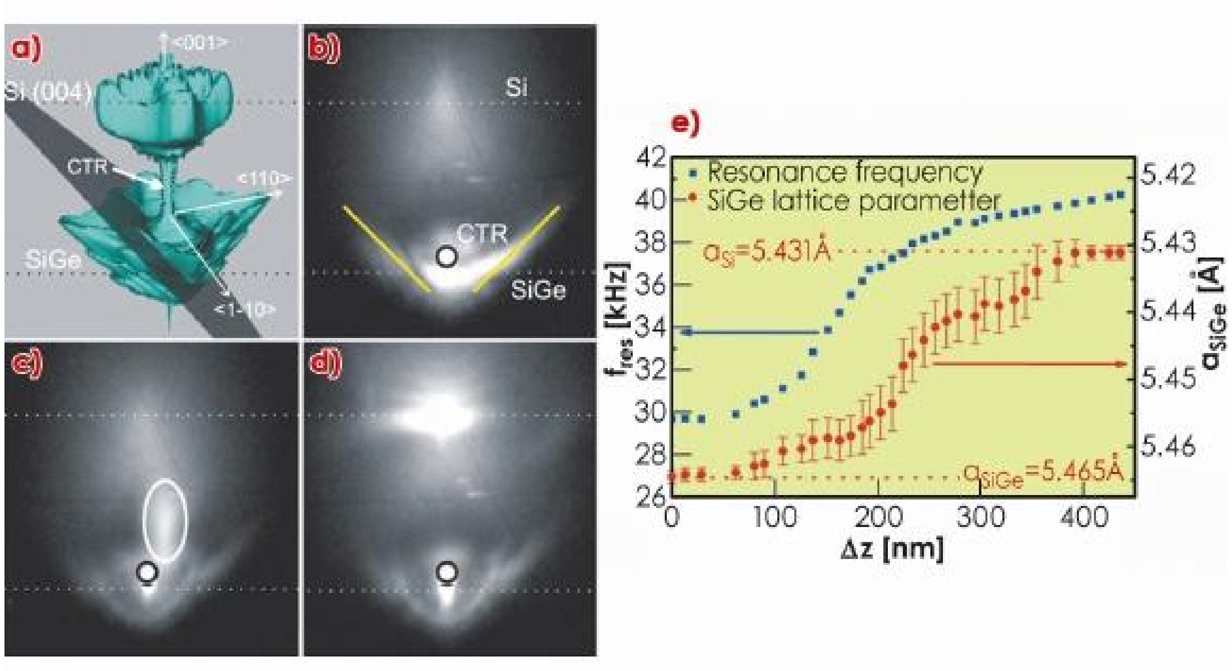
the dependence on the mean facet size on the Ge coverage θ

M.-I. Richard, T. U. Schülli, G. Renaud, E. Wintersberger, G. Chen, G. Bauer, and V. Holý, Phys. Rev. B 80, 045313 (2009).

Combining x-ray diffraction and AFM in-situ (ID01 at ESRF)



T. Scheler et al., Appl. Phys. Lett. 94, 023109 (2009)



From ESRF Highlights

1. What can be done by x-ray scattering?
2. Standard applications – rather boring
3. Iso-strain method & anomalous diffraction
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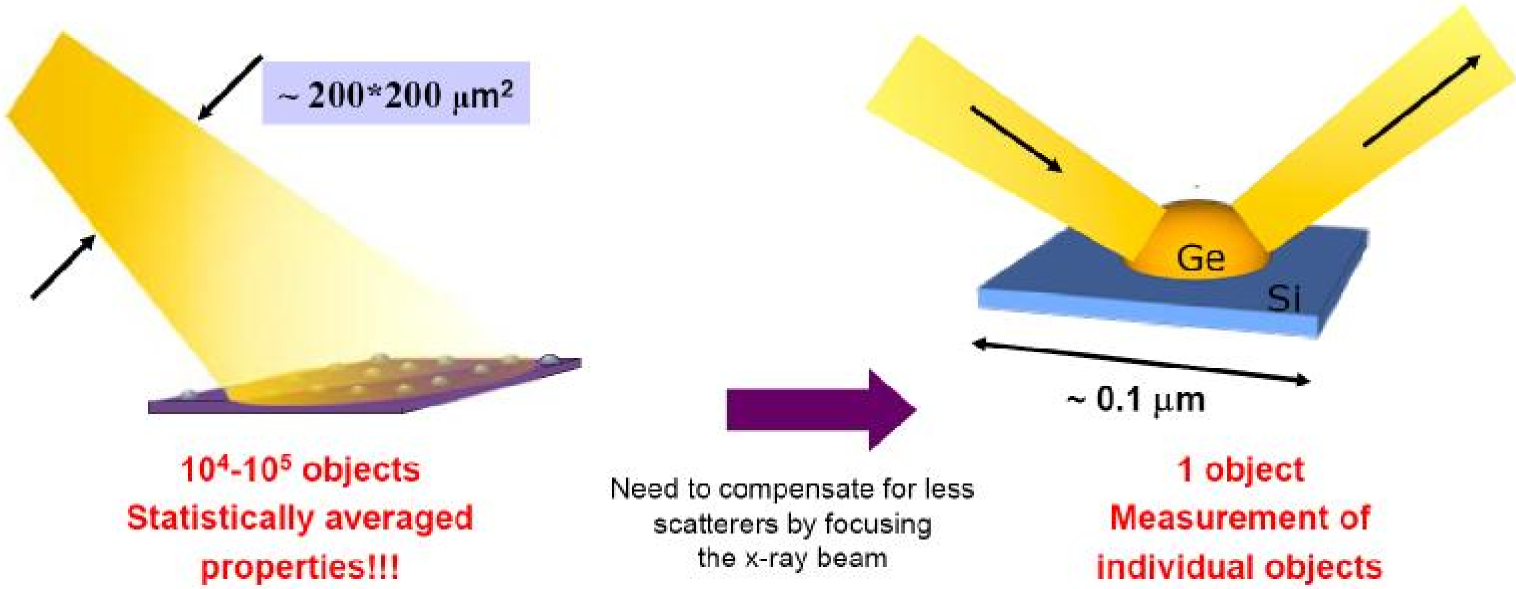
In previous examples, the measured intensity was an incoherent superposition of intensities scattered from a very large numbers of nano-objects

☺ the measured signal has a good statistical relevance

☹ details of the intensity distributions are smeared out, since the objects are not completely identical

To irradiate only one single object



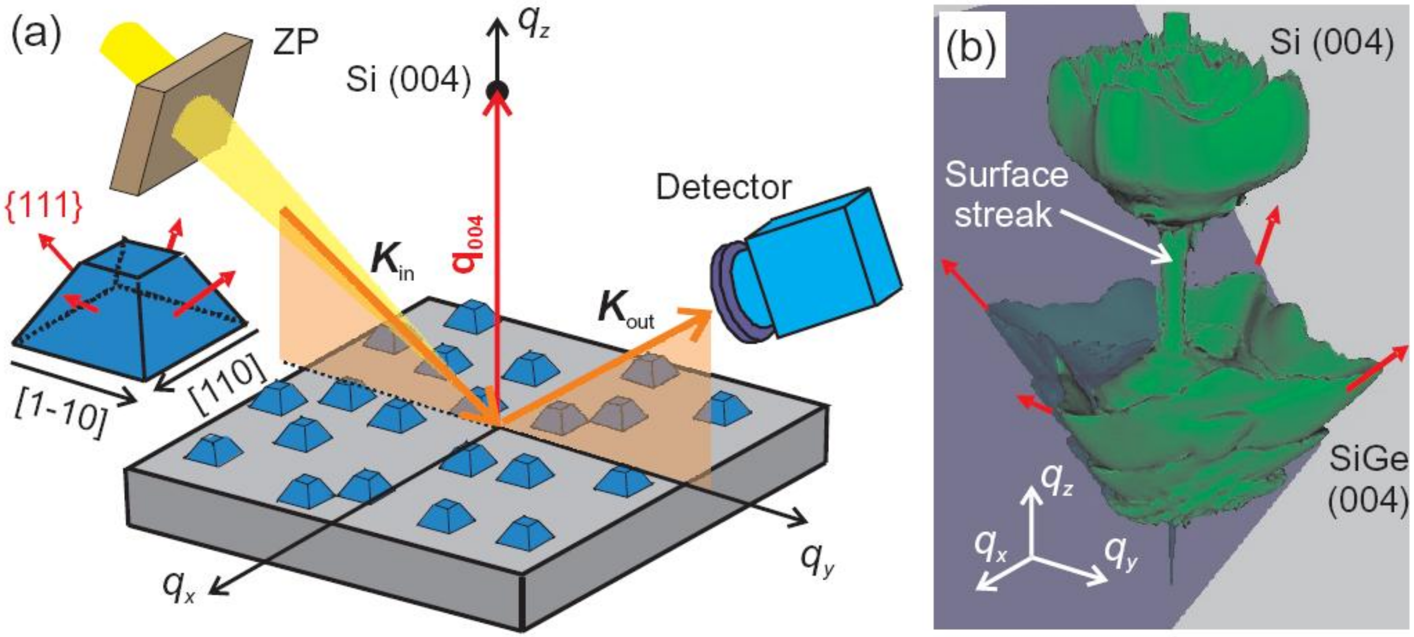
courtesy of Cristian Mocuta, ESRF

Then, the nano-object can be completely coherently irradiated

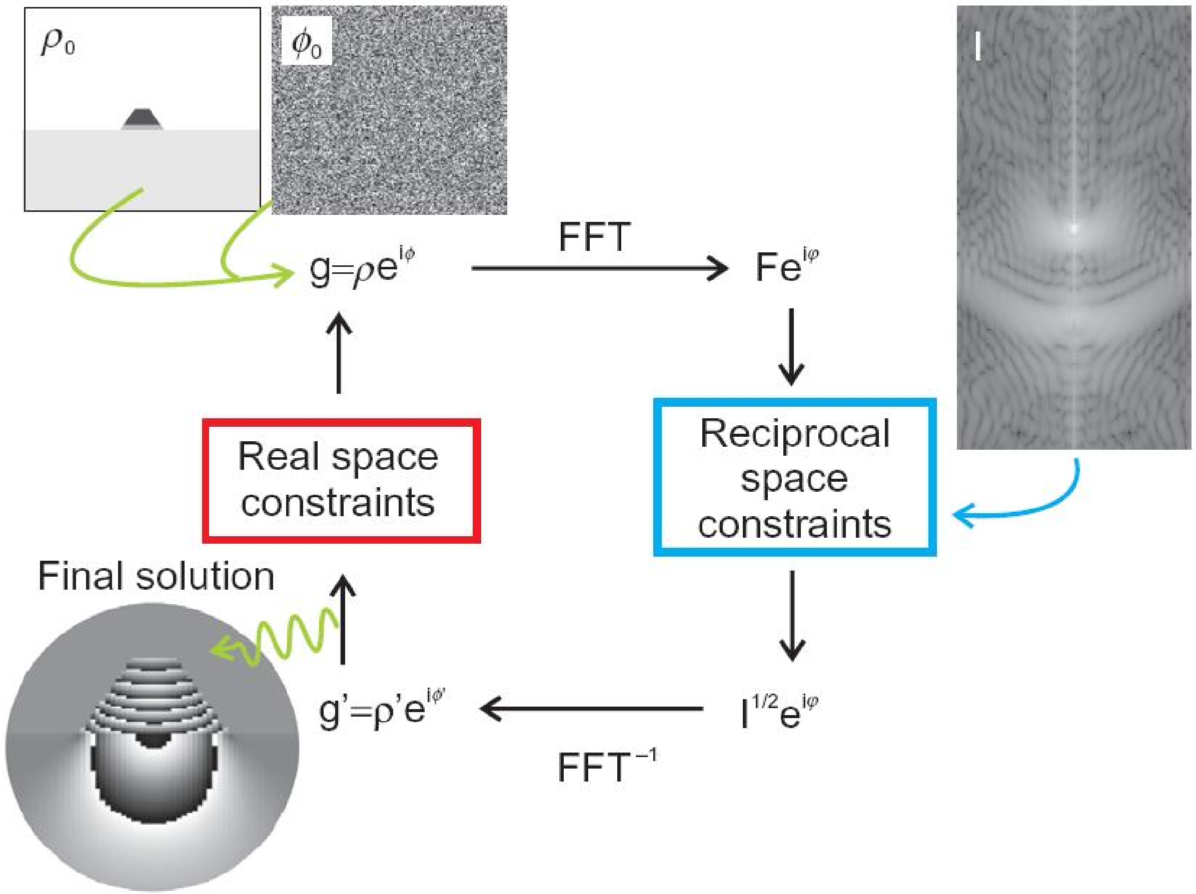
The phase-retrieval method makes it possible to reconstruct the shape and strain field directly without any structure model

Ge dots on Si(001)

Ana Diaz et al., New J. Phys. 12 035006, (2010)

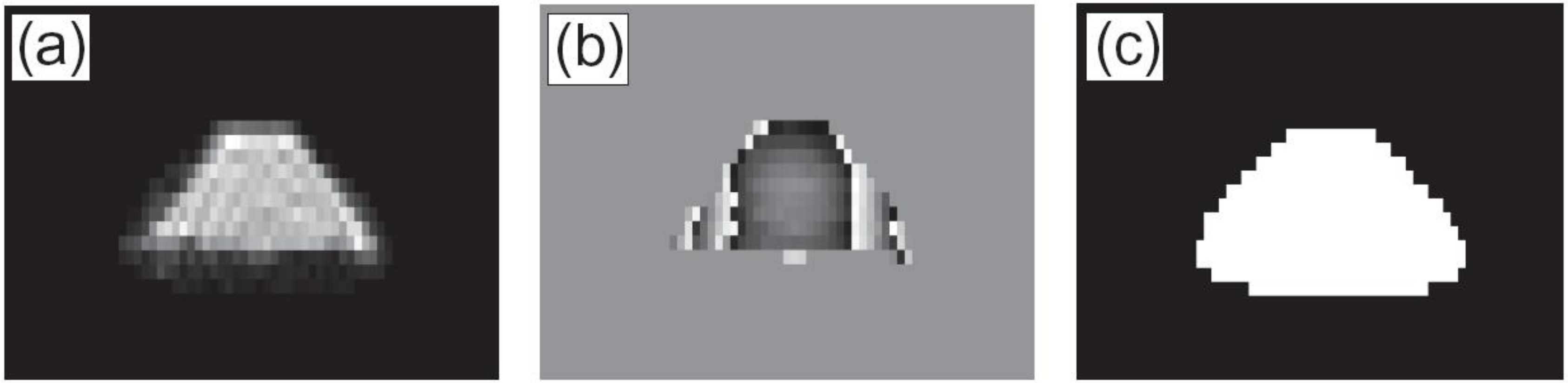


courtesy of A. Diaz, ESRF



courtesy of A. Diaz, ESRF

reconstructed amplitude | reconstructed phase | optimized real-space constraint

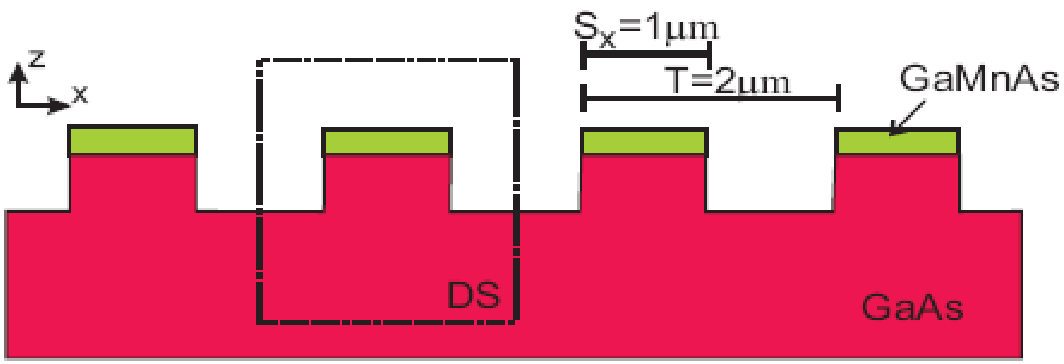


courtesy of A. Diaz, ESRF

GaMnAs wires in GaMnAs/GaAs(001) layers

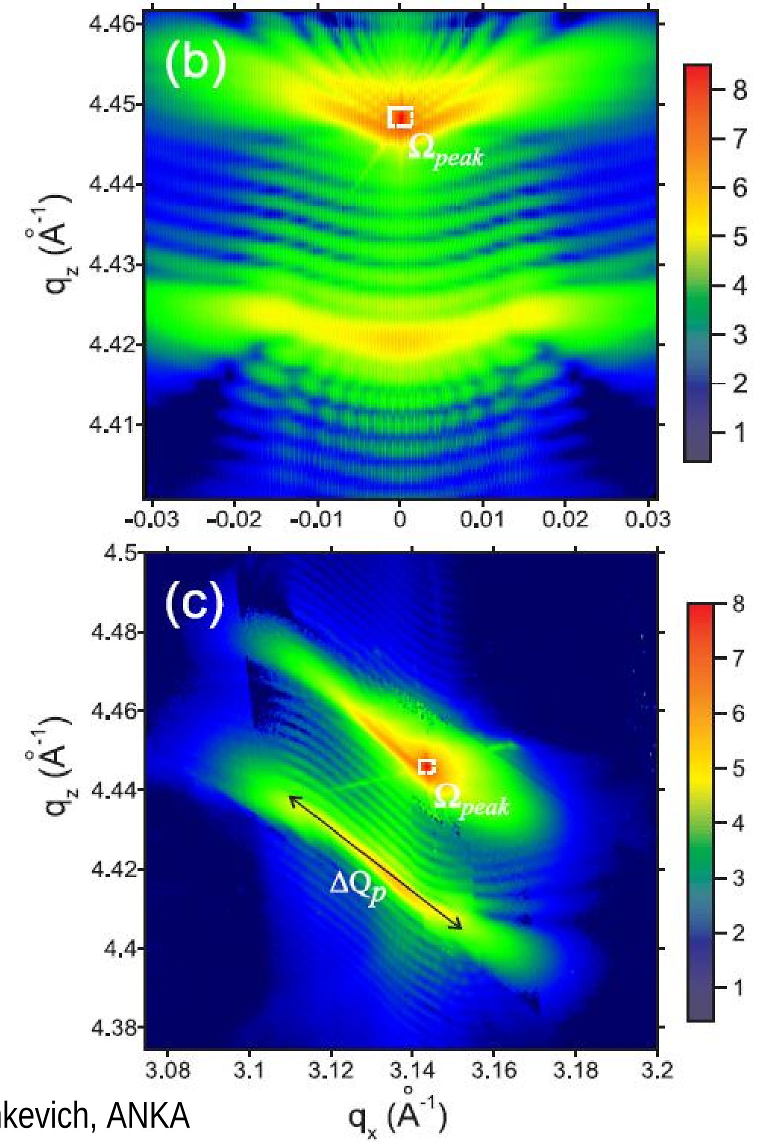
Samples grown by MBE at Institute of Physics, ASCR Prague and patterned at University of Cambridge, UK
ID10@ESRF, phase retrieval algorithm by A. Minkevich, ANKA Karlsruhe

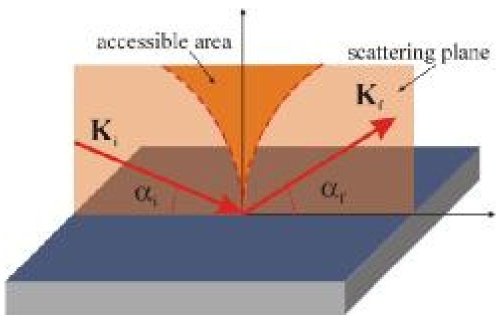
sample cross-section



courtesy of A. Minkevich, ANKA

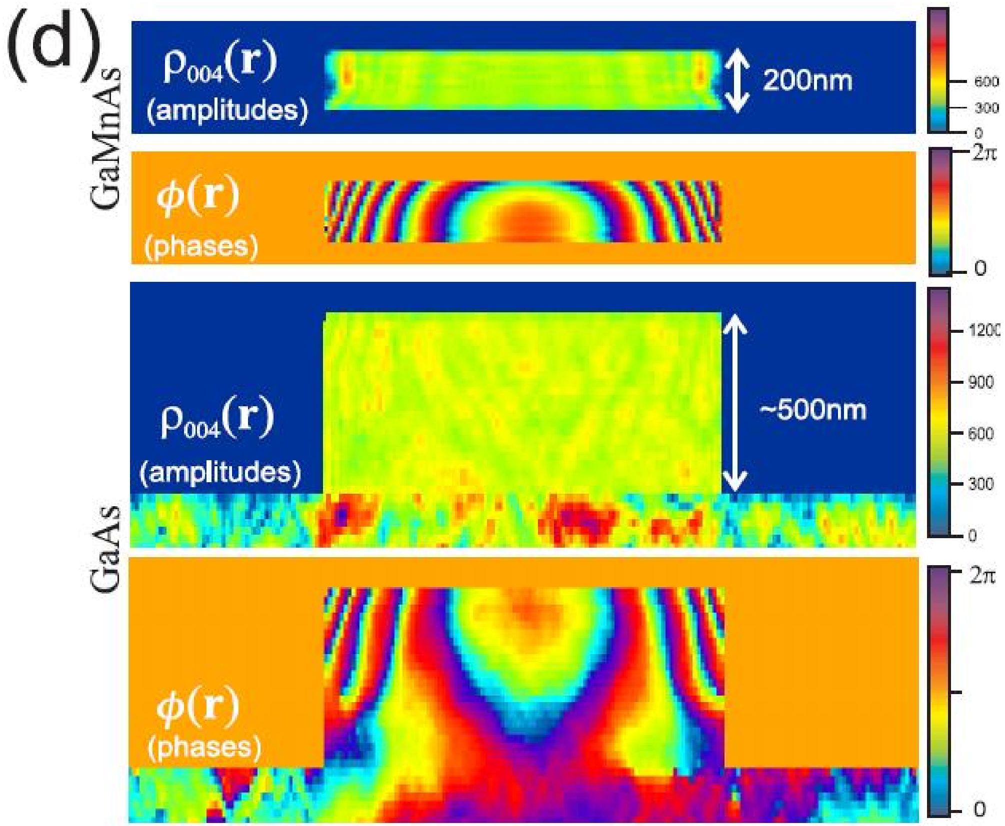
measured intensity distribution, coplanar symmetric diffraction 004 (upper panel) and 224 (lower panel)





courtesy of A. Minkevich, ANKA

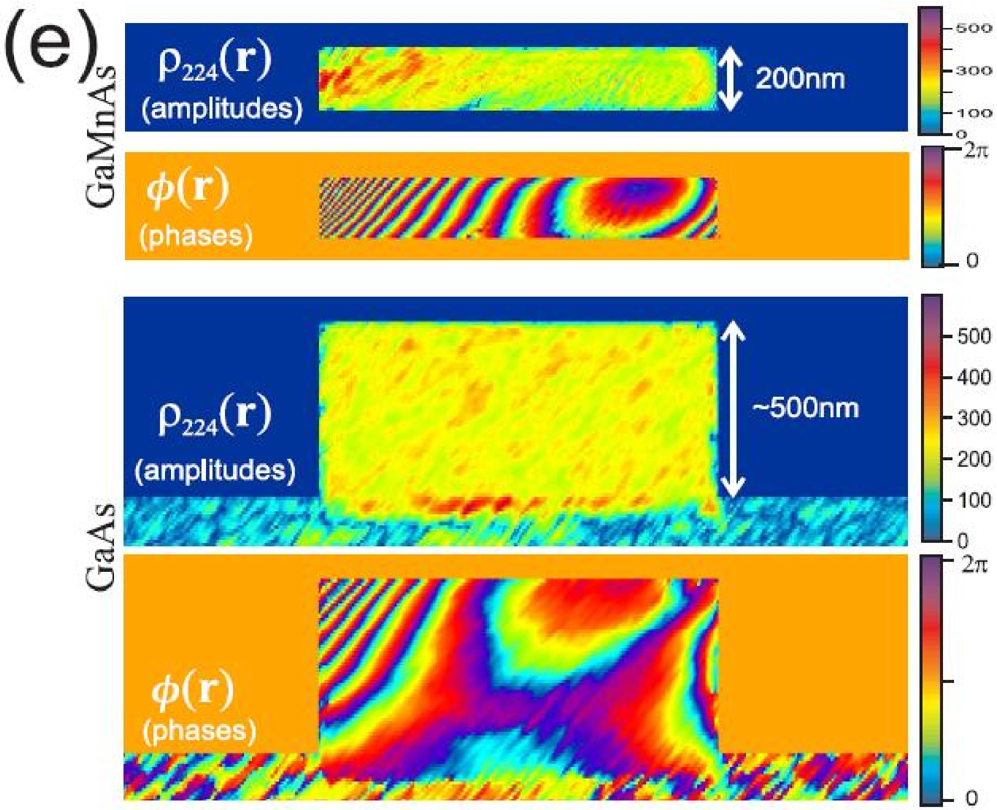
Amplitudes and phases of the field diffracted from the GaMnAs and GaAs volumes, diffraction 004



Amplitude ~ mean electron density
phase= *h.u*

courtesy of A. Minkevich, ANKA

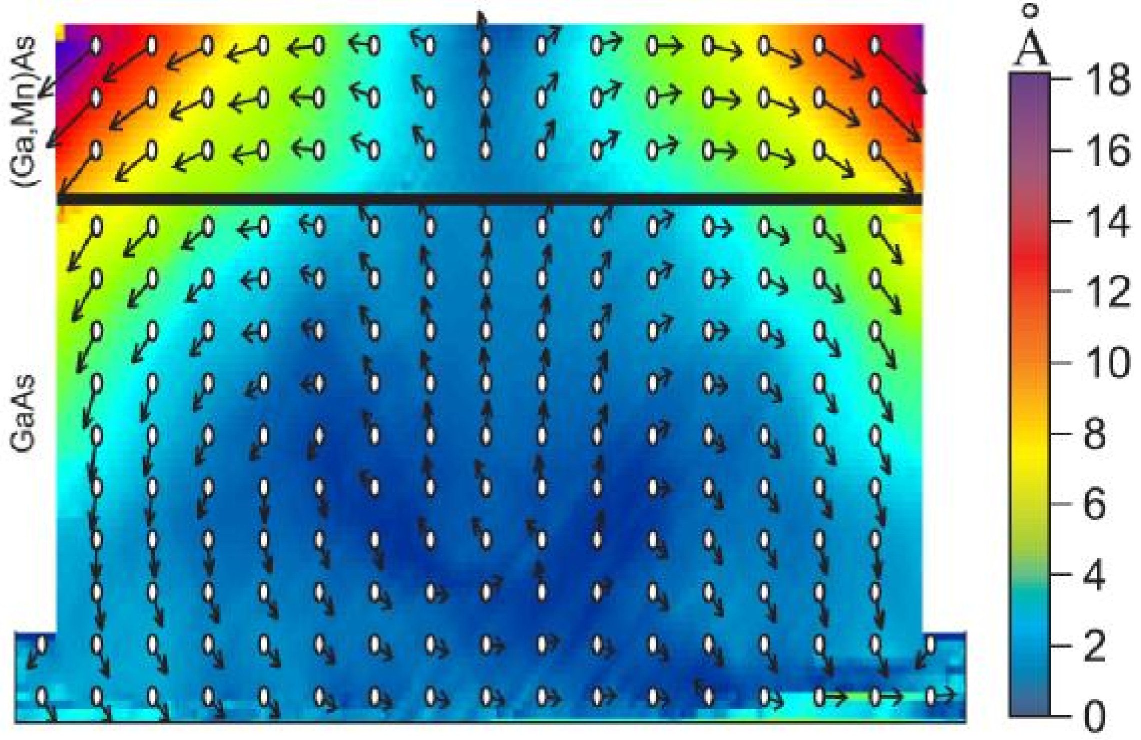
Amplitudes and phases of the field diffracted from the GaMnAs and GaAs volumes, diffraction 224



Amplitude ~ mean electron density
phase= *h.u*

courtesy of A. Minkevich, ANKA

Displacement field reconstructed from the x-ray data



A. Minkevich et al., submitted

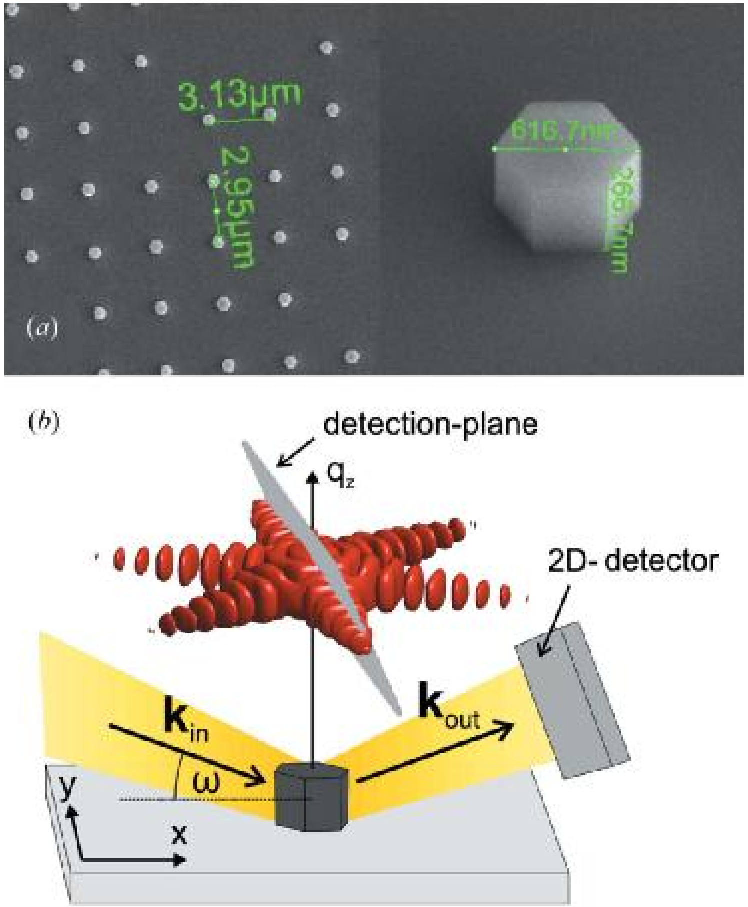
courtesy of A. Minkevich, ANKA

Another example: coherent diffraction from single nanorods

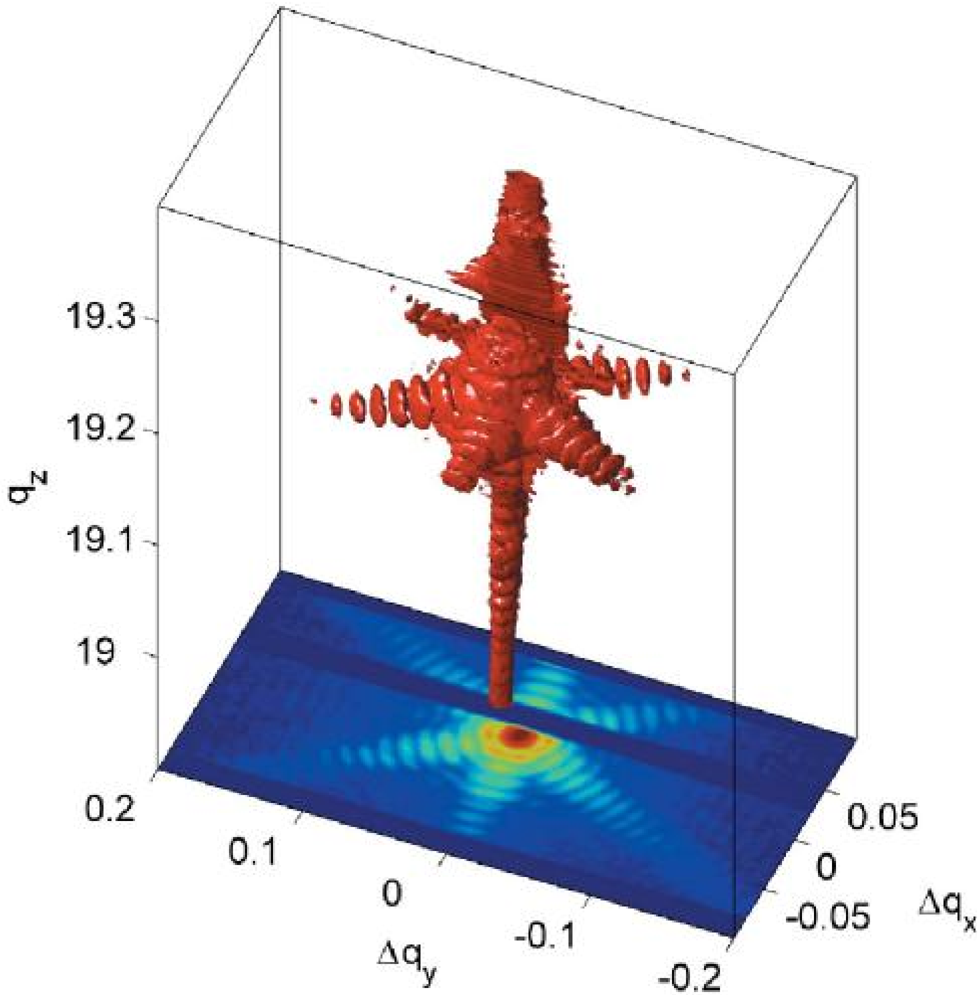
A. Biermanns et al., J. Synchr. Rad. 16, 796 (2009)

V. Favre-Nicolin et al., New Journal of Physics 12, 035013 (2010).

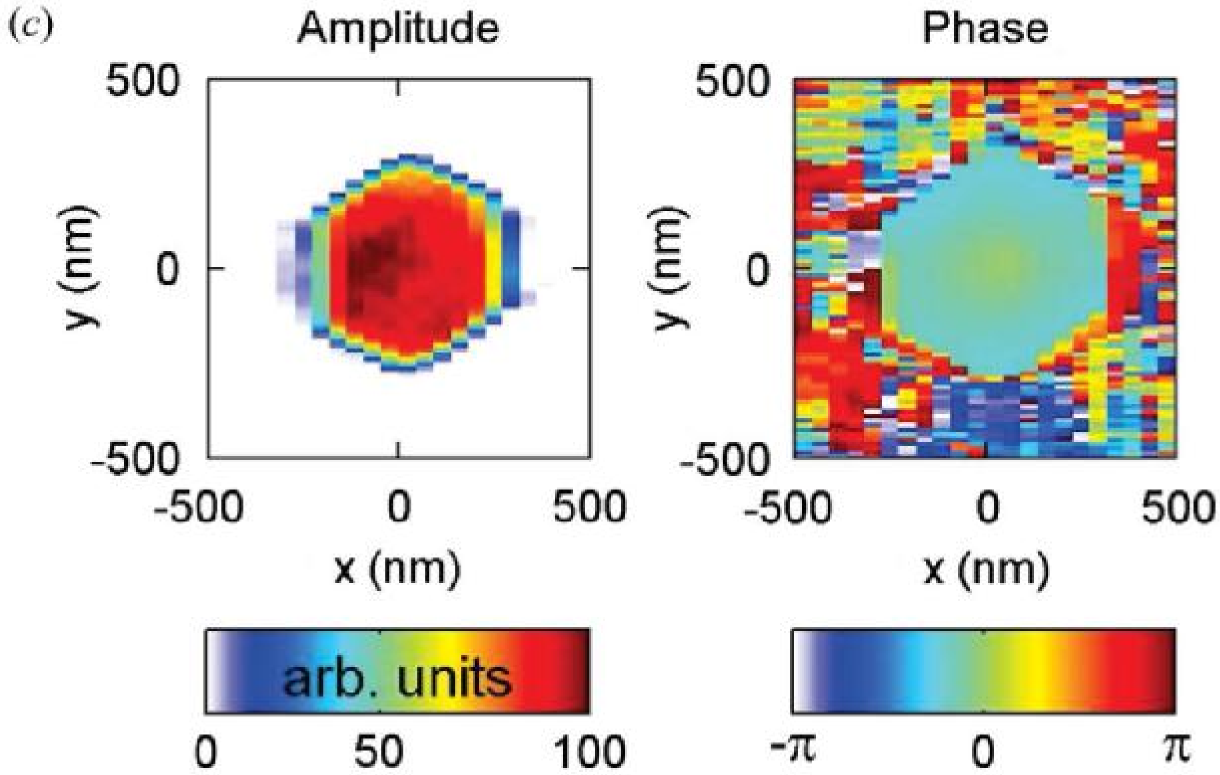
A. Biermanns et al.:



3D distribution of coherently scattered intensity in reciprocal space



Results of the phase-retrieval algorithm



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Recent trends using synchrotron radiation:

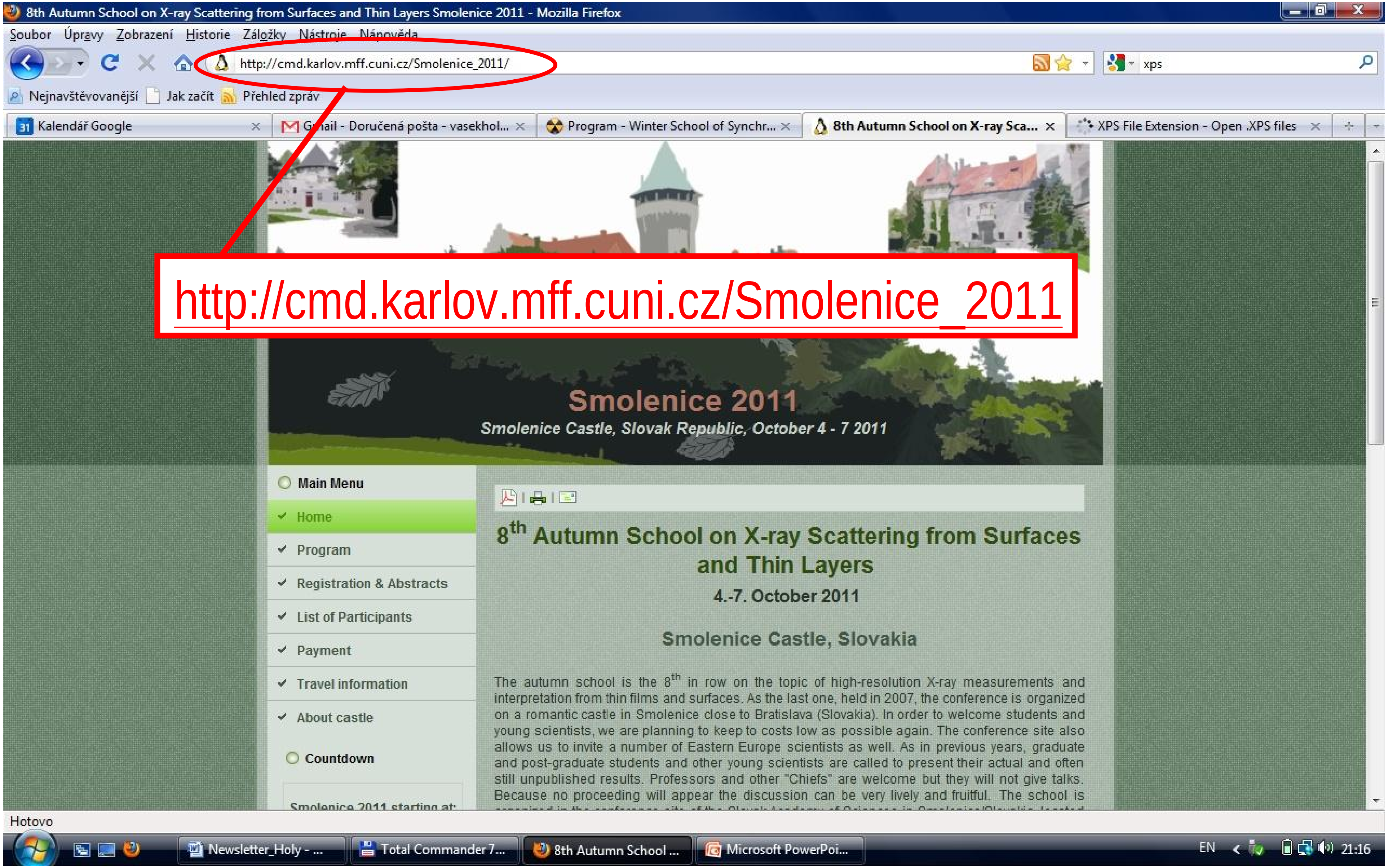
1. Multiwave anomalous diffraction (MAD) method
2. In-situ studies (during deposition, annealing, oxidation, elastic deformation etc)
3. Investigation of a single nanoobject
4. Phase-retrieval method

Substantial progress in laboratory instrumentation:

1 **THANK YOU FOR YOUR KIND ATTENTION**

2. In-situ measurements (annealing, oxidation etc.)
3. Better detectors (2D detectors with extreme dynamical range, very low noise but for a very high price ☹)

Projects: KAN400100652, EU project 214499 NAMASTE


8th Autumn School on X-ray Scattering from Surfaces and Thin Layers Smolenice 2011 - Mozilla Firefox
http://cmd.karlov.mff.cuni.cz/Smolenice_2011/
http://cmd.karlov.mff.cuni.cz/Smolenice_2011
Smolenice 2011
Smolenice Castle, Slovak Republic, October 4 - 7 2011
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8th Autumn School on X-ray Scattering from Surfaces and Thin Layers
4.-7. October 2011
Smolenice Castle, Slovakia
The autumn school is the 8th in row on the topic of high-resolution X-ray measurements and interpretation from thin films and surfaces. As the last one, held in 2007, the conference is organized on a romantic castle in Smolenice close to Bratislava (Slovakia). In order to welcome students and young scientists, we are planning to keep to costs low as possible again. The conference site also allows us to invite a number of Eastern Europe scientists as well. As in previous years, graduate and post-graduate students and other young scientists are called to present their actual and often still unpublished results. Professors and other "Chiefs" are welcome but they will not give talks. Because no proceeding will appear the discussion can be very lively and fruitful. The school is