Mean Inner Potential MIP = $\Phi_o = V_o$

- Why?

Refraction
Thickness & Shape
Composition
Work function & Fermi energy
Bond energy

- How?

Interferometry / Holography

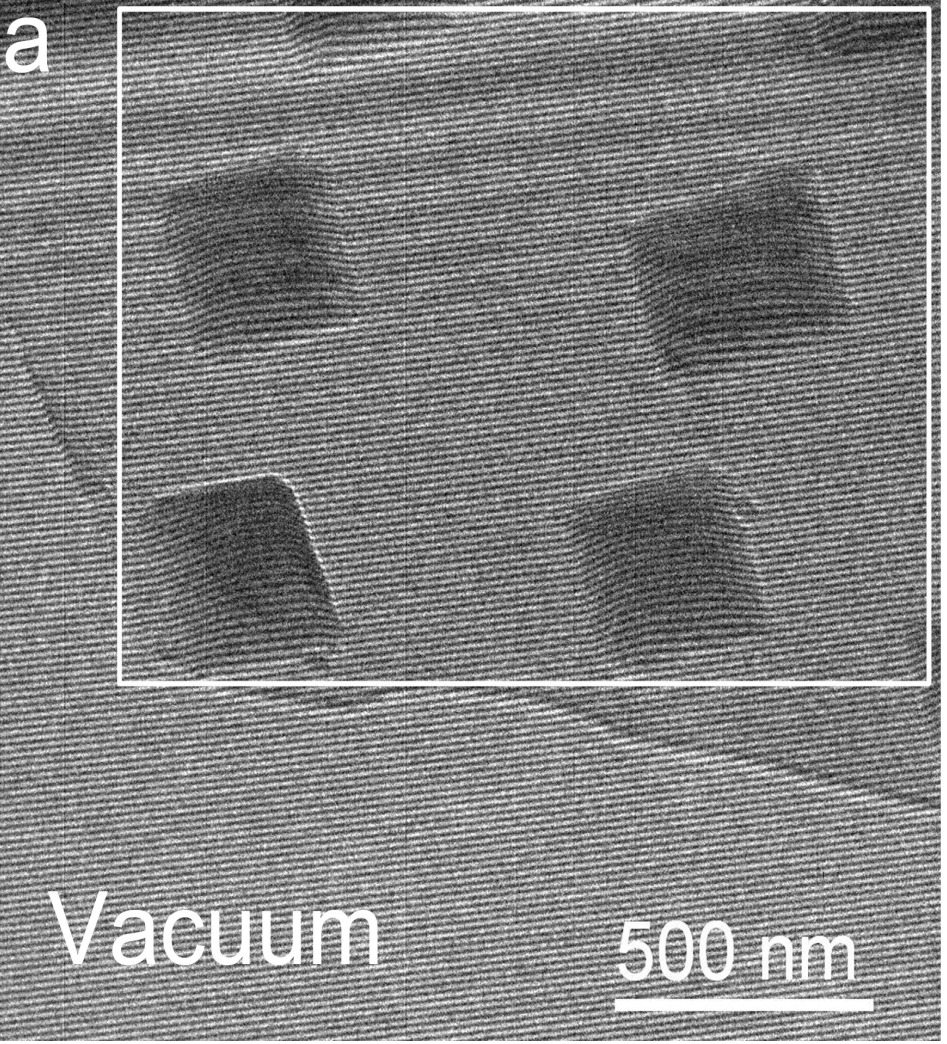
$$\varphi = \sigma V_0 t$$

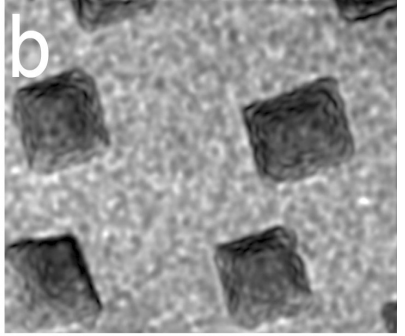
- Faq?

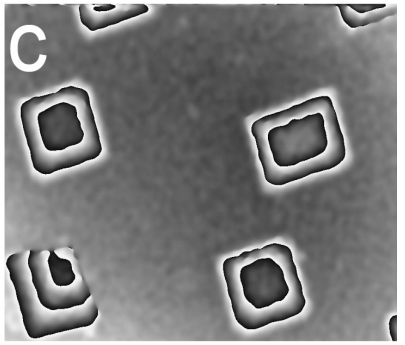
Inverse problem: Uniqueness
Strain / Stress / Defects

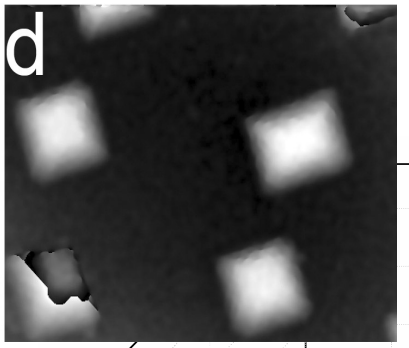
$\Psi \sim e^{i\pi At} \sim e^{iV_o t} \cdot e^{-\mu t} \cdot (1 + i\pi A' t + \ldots)$

Reconstruction of (Si,Ge) islands

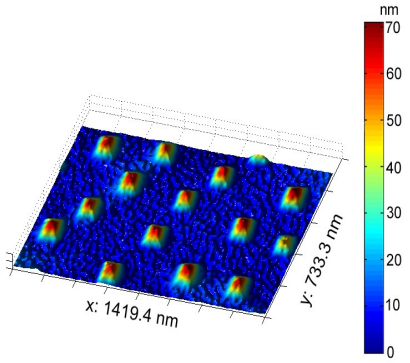


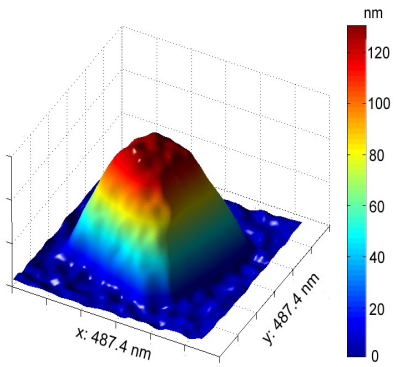






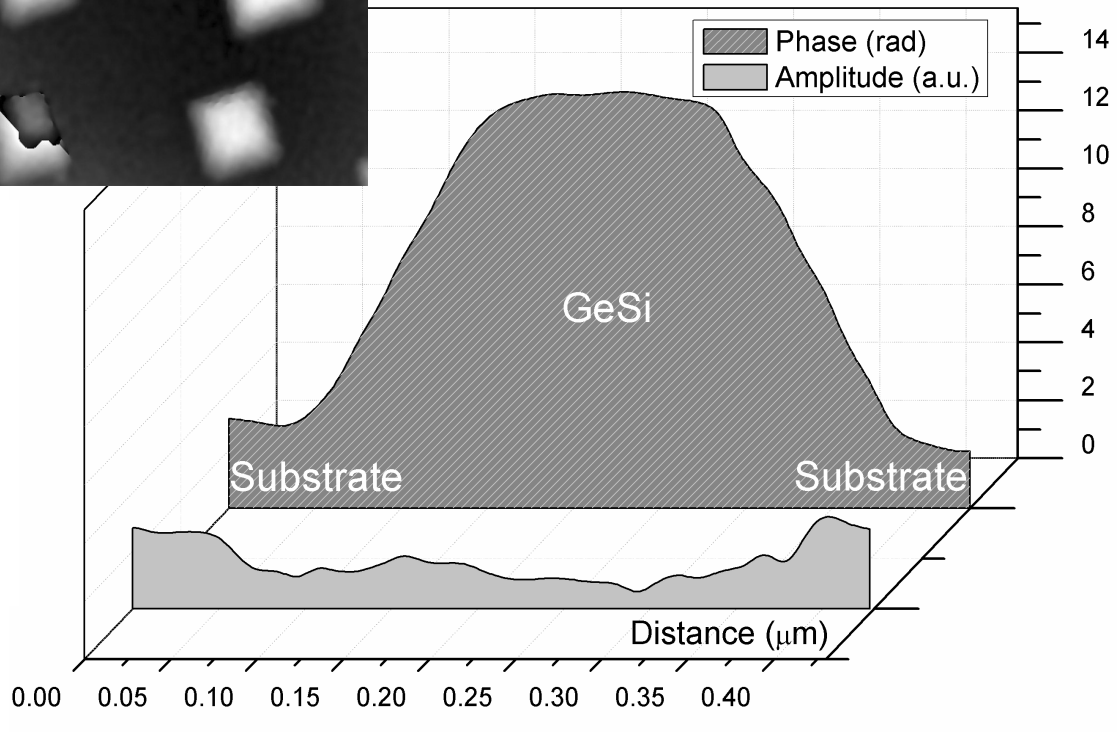
a hologram
b amplitude
c phase
d unwrapped phase





Sample A: h~ 66 nm

Sample B: h ~ 130 nm



Max von Laue, Materiewellen und ihre Interferenzen, Geest & Portig 1948

Die dynamische Theorie der Elektronen-Interferenzen.

§ 18. Das mittlere Potential im Raumgitter[1]).

Wir können nämlich zeigen, daß es für ein bis an seine Ränder ungestörtes Raumgitter im allgemeinen kein solches Potential gibt.

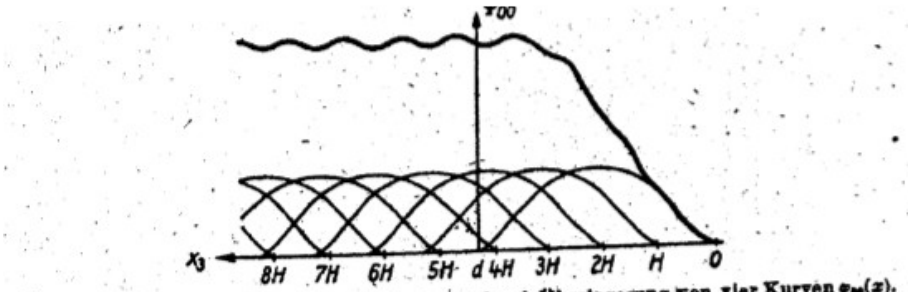


Fig. 108. Entstehung des Flächenmittels $\Phi_{00}(x)$ durch Überlagerung von vier Kurven $\varphi_{00}(x)$.

Gleichgültig, wie viele Kurven $\varphi_{00}(\xi)$ sich überlagern, zwischen welchen Werten also die ganze Zahl m laufen muß, immer gilt danach

$$\Phi_{000} = \frac{1}{H}\int_0^d \varphi_{00}(x_3)\,dx_3\,; \qquad (18.14)$$

M. O'KEEFFE AND J. C. H. SPENCE *Acta Cryst.* (1994). A**50**, 33–45

On the Average Coulomb Potential (Φ_0) and Constraints on the Electron Density in Crystals

In this paper, we argue that the average potential in a finite crystal is a well defined quantity, but that extending the concept to an *infinite* crystal can lead to incorrect conclusions.

cell
. However, the evaluation of V_0 using this expression ignores any surface effects, the ambiguity about the point of zero potential in an infinite crystal and leaves open the question of whether $V(\mathbf{r})$ can be obtained from the knowledge of $\varrho(\mathbf{r})$ alone for an infinite crystal.

For the case of a finite crystal with boundary condition of zero potential at infinity, the potential is completely defined by the charge distribution $\varrho(\mathbf{r})$ using Poisson equation. The correct procedure for defining V_0 is then to average the potential of the finite crystal according to [11,12], which is given by

$$V_0 \equiv -\frac{1}{\Omega_{\text{cryst}}}\iint \frac{\varrho(\mathbf{r})}{|\mathbf{r}-\mathbf{r}'|}\,d\mathbf{r}'\,d\mathbf{r} = \frac{2\pi}{3\Omega_{\text{cryst}}}\int r'^2\varrho(\mathbf{r}')\,d\mathbf{r}'\,. \qquad (4)$$

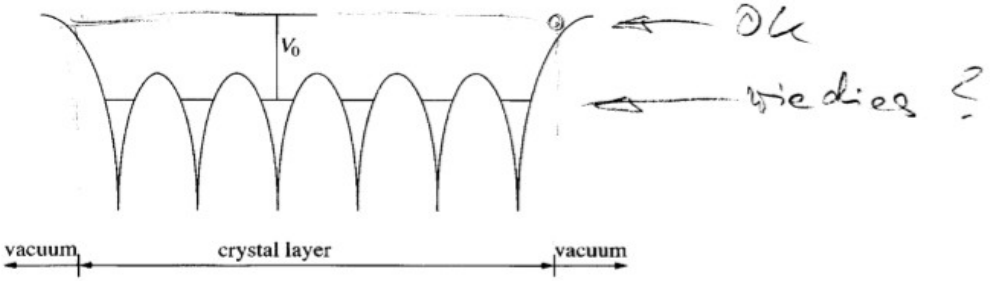


Fig. 1. Coulomb potential $V(z)$ for silicon (d) in Table 1 plotted as a function of distance through the slab. The method of defining V_0 is indicated. For silicon (110), the crystal layer width of a unit cell is 0.21 nm^2. The laterally-averaged potential falls to less than 0.02 V within a distance of 0.37 nm from the nuclei of the surface atoms, measured normal to the surface

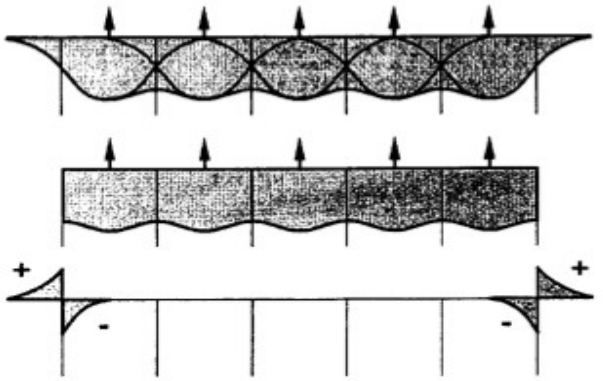
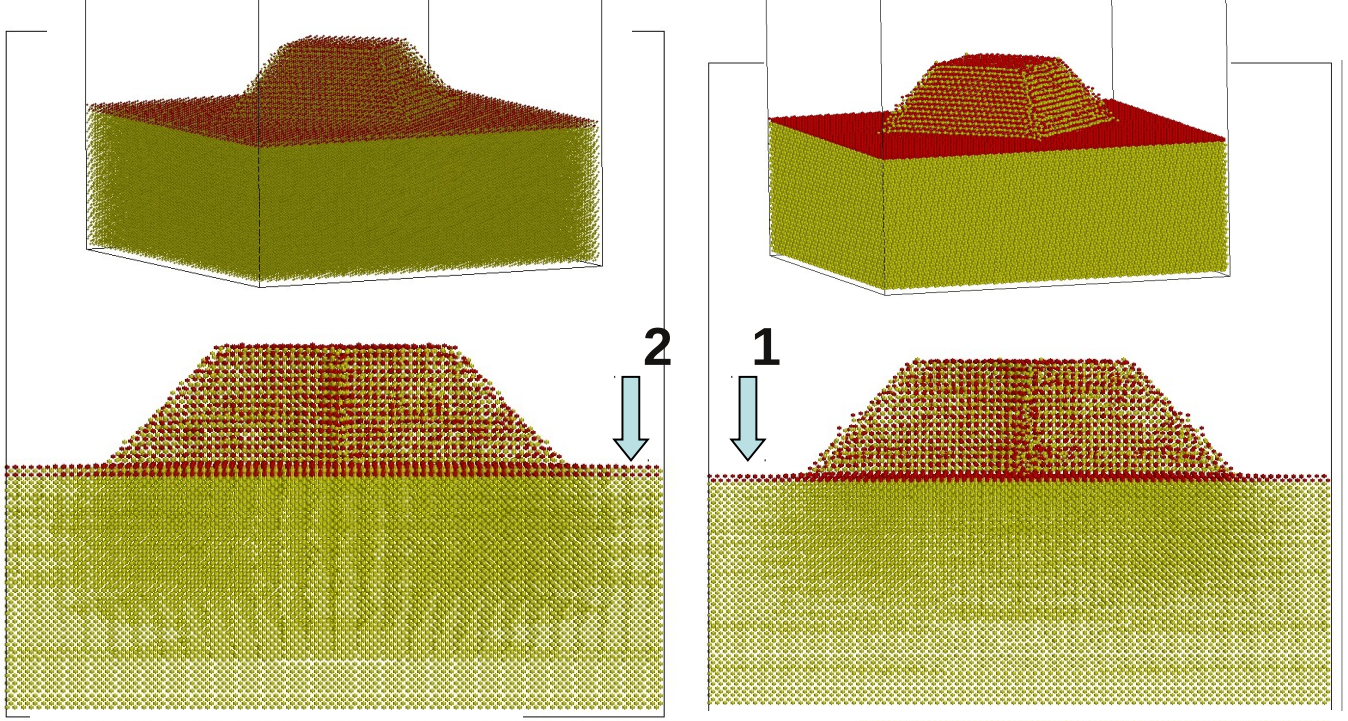


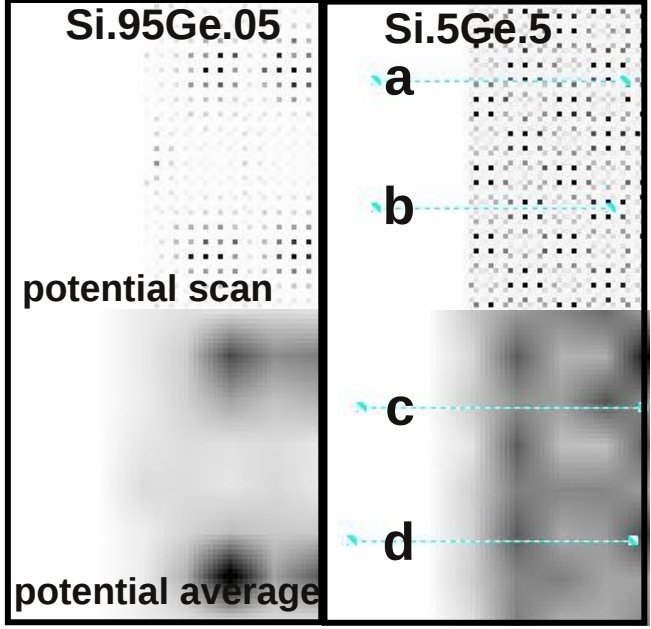
Fig. 7. Comparison of two ways of assembling a crystallite from bases. Top: charge density in a crystallite of pseudo-atoms. Middle: the charge density as an assembly of unit cells. Bottom: the difference showing the surface dipole layer. The arrows indicate the positions of the nuclei of the pseudo-atoms.

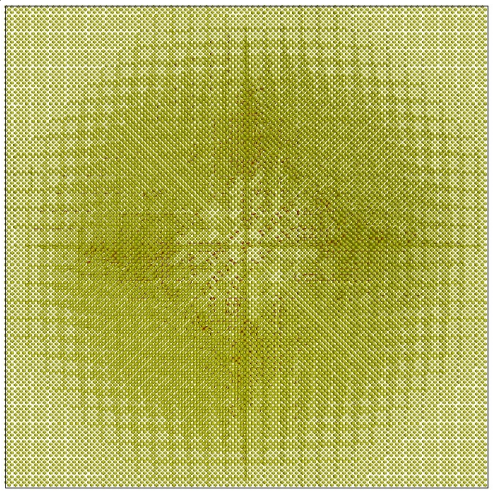
- SCF with ρ of isolated atoms insufficient
- DFT very extensive for larger structures
- MD with analytical bond order potential

$$E_{tot} = E_{rep} + E_{prom} + \sum H_{i\alpha,j\alpha}\Theta_{i\alpha,j\alpha}$$

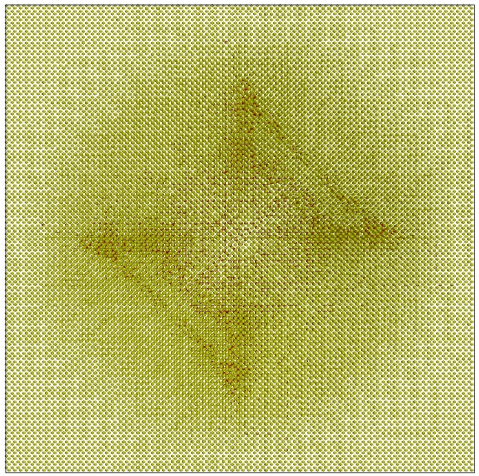
empirical s^2p^2->sp^3 band energy replaced by hopping integrals and bond order matrix

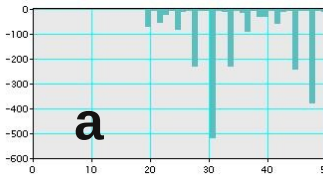


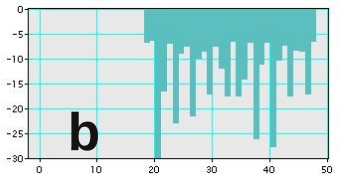


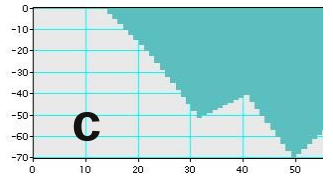


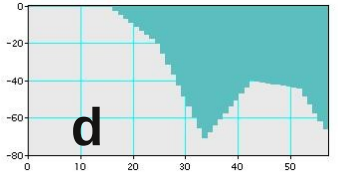
SiGeQD MDrelaxed & simulated exit wave

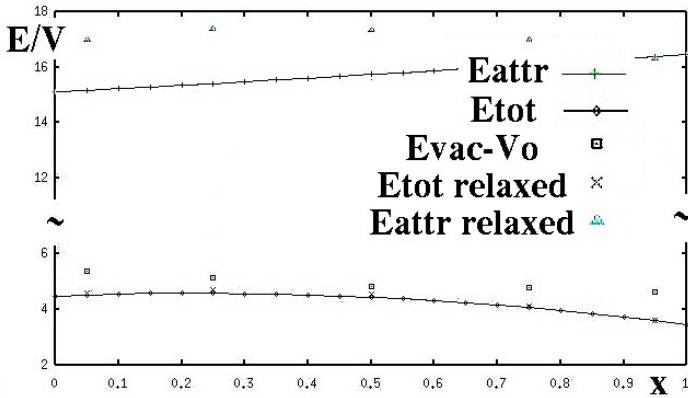




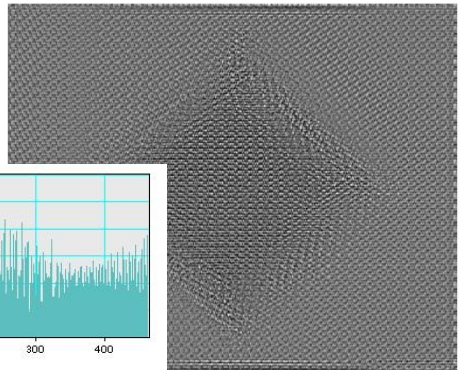


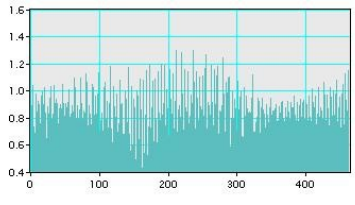




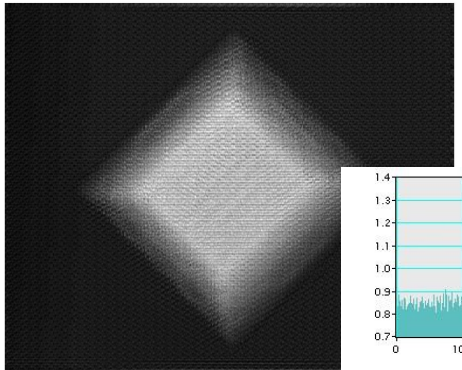


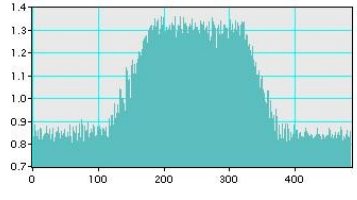
Amp

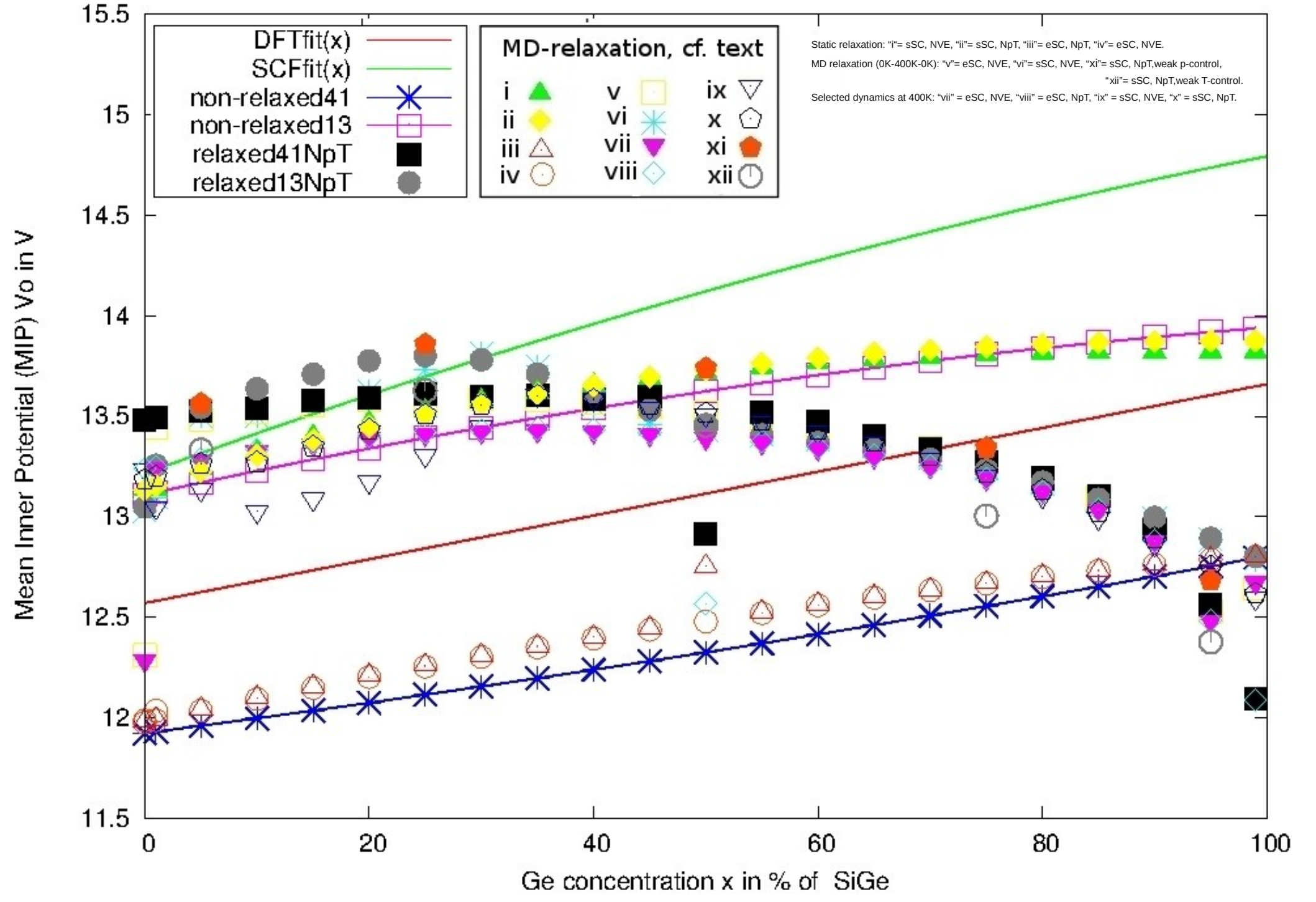




Phase

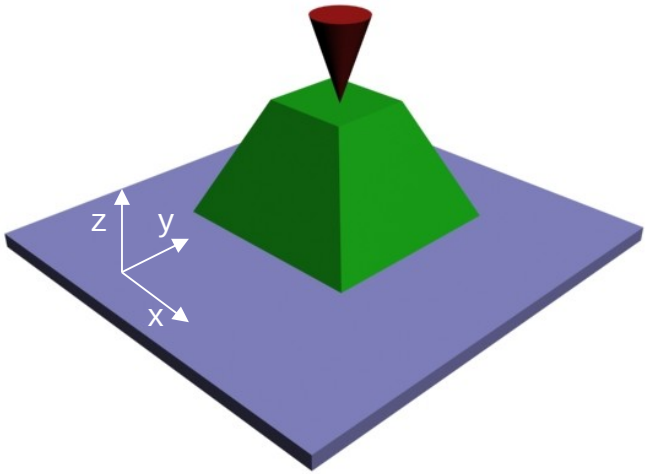




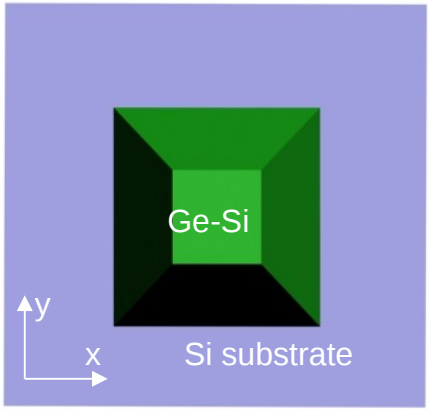

DFTfit(x)
SCFfit(x)
non-relaxed41
non-relaxed13
relaxed41NpT
relaxed13NpT
MD-relaxation, cf. text
i
ii
iii
iv
v
vi
vii
viii
ix
x
xi
xii
Static relaxation: “i”= sSC, NVE, “ii”= sSC, NpT, “iii”= eSC, NpT, “iv”= eSC, NVE.
MD relaxation (0K-400K-0K): “v”= eSC, NVE, “vi”= sSC, NVE, “xi”= sSC, NpT,weak p-control,
“xii”= sSC, NpT,weak T-control.
Selected dynamics at 400K: “vii” = eSC, NVE, “viii” = eSC, NpT, “ix” = sSC, NVE, “x” = sSC, NpT.
Mean Inner Potential (MIP) Vo in V
15.5
15
14.5
14
13.5
13
12.5
12
11.5
0
20
40
60
80
100
Ge concentration x in % of SiGe

Goal: Analyzing 3D (Si,Ge) nanostructures

STM and AFM



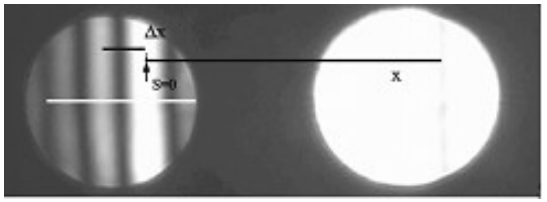
(C)TEM



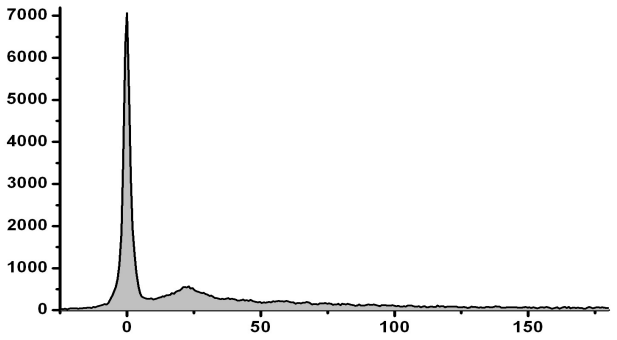
Composition, structure, strain, morphology of (Si,Ge) nanostructures

CTEM: two dimensional projection
dynamical diffraction complexity

CBED



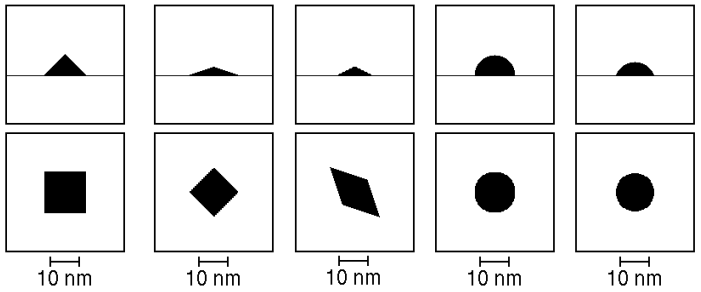
EELS



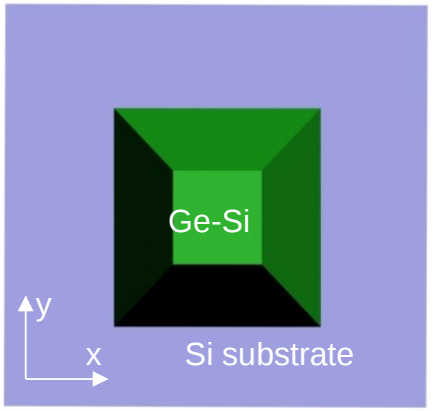
Physical properties (confinement)

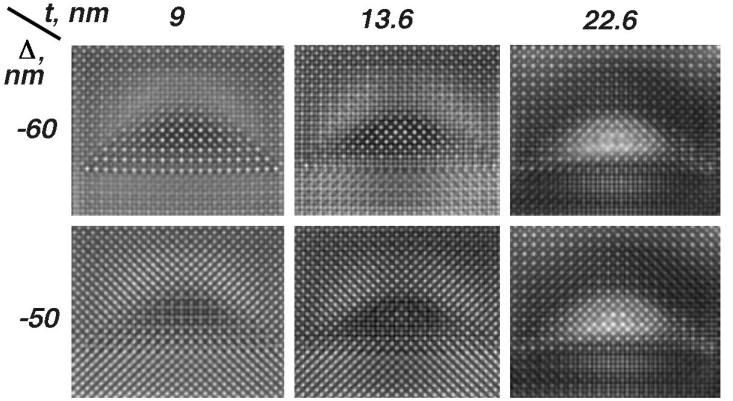
Applicability electronic / photonic devices

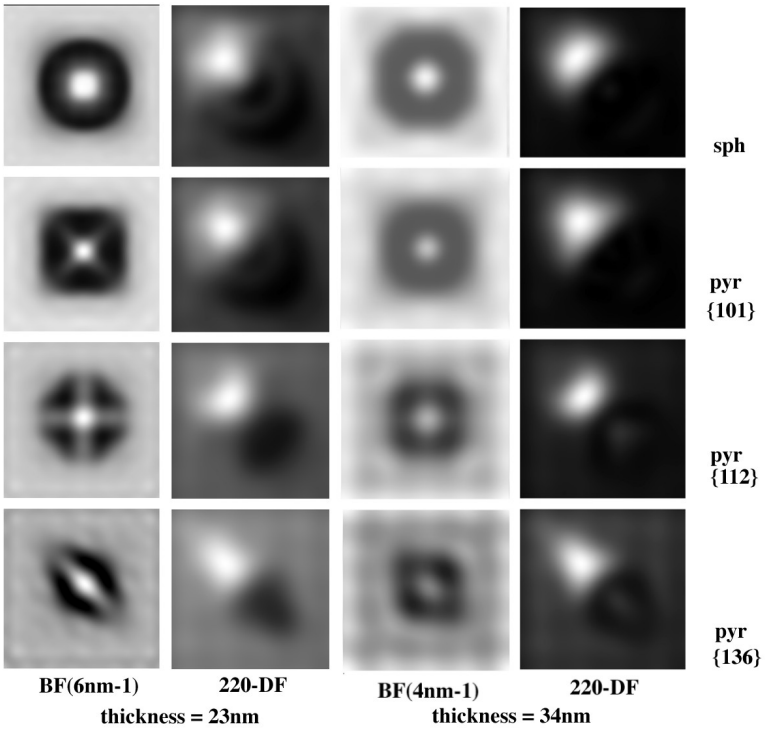
Analyzing TEM of 3D (Si,Ge) nanostructures

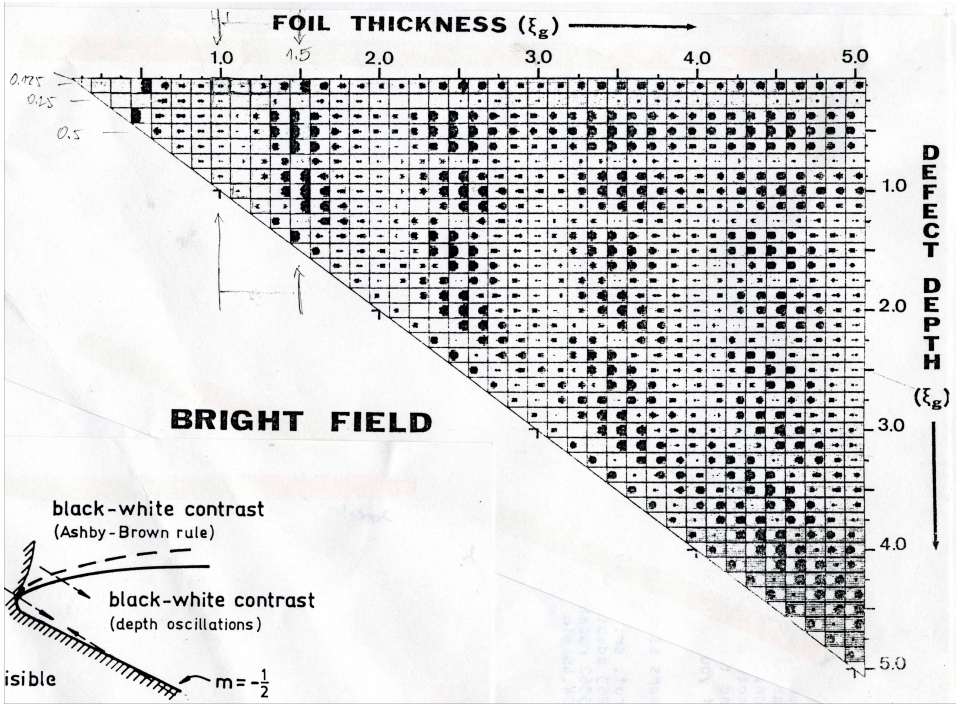


(C)TEM

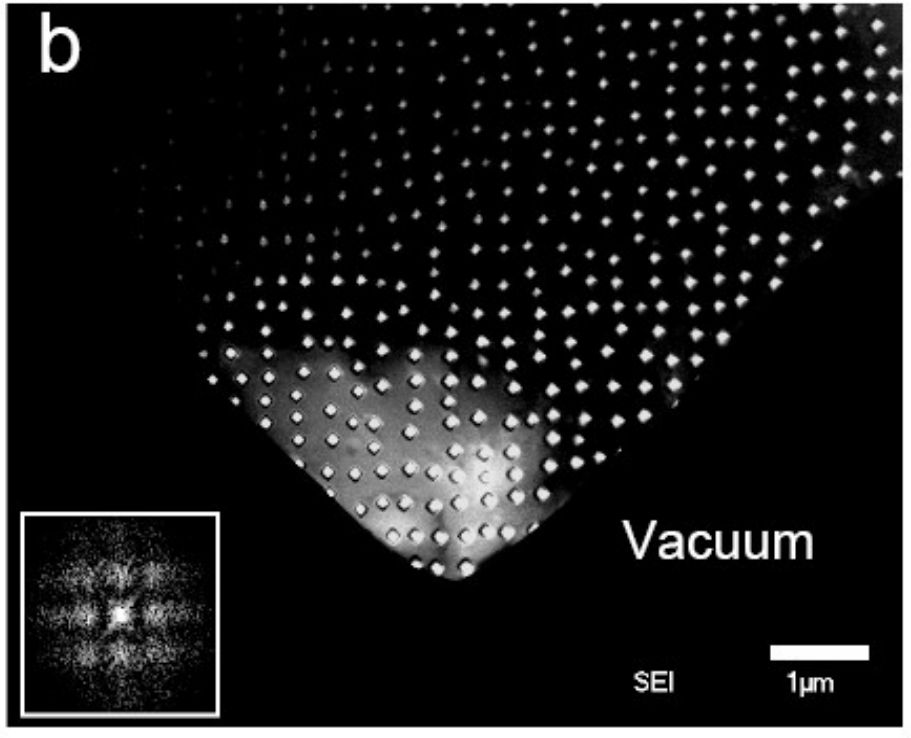






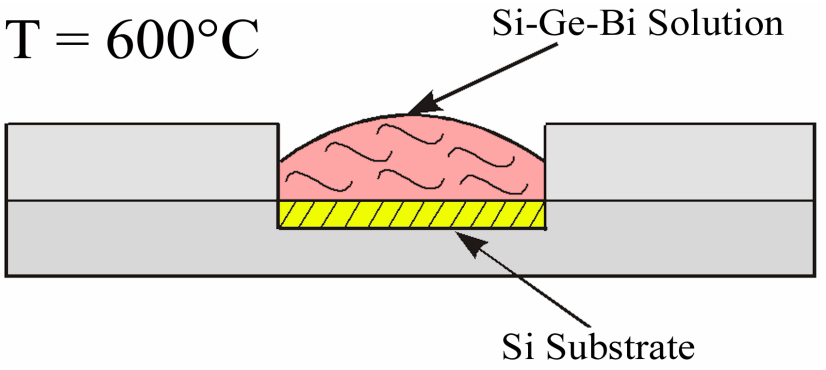


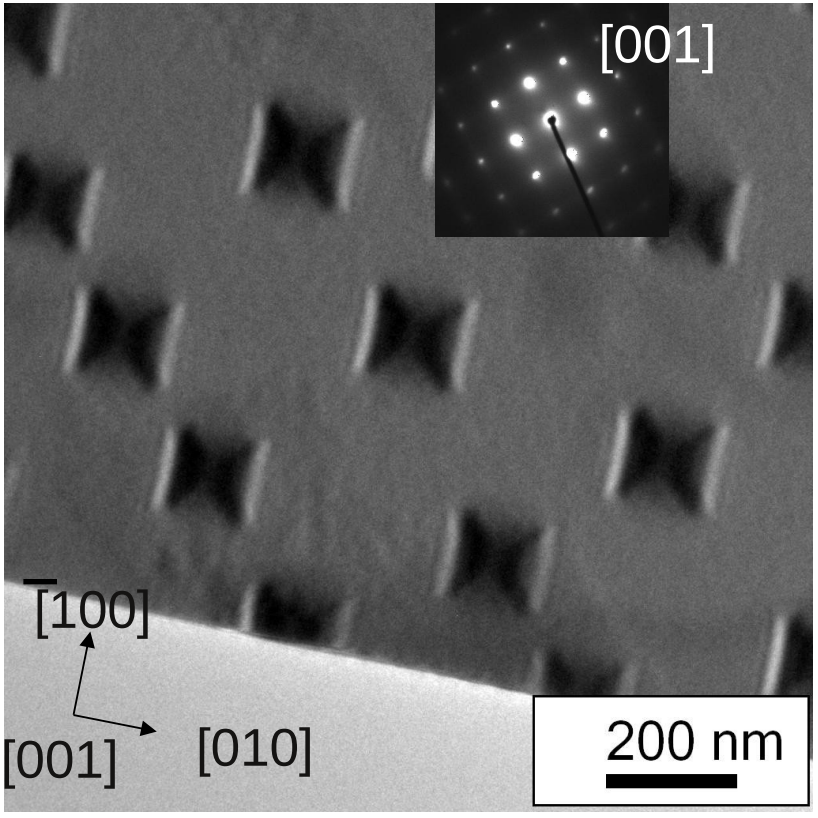
STEM-HAADF and BF-TEM of (Si,Ge) islands:



STEM-HAADF image

Liquid phase epitaxy growth



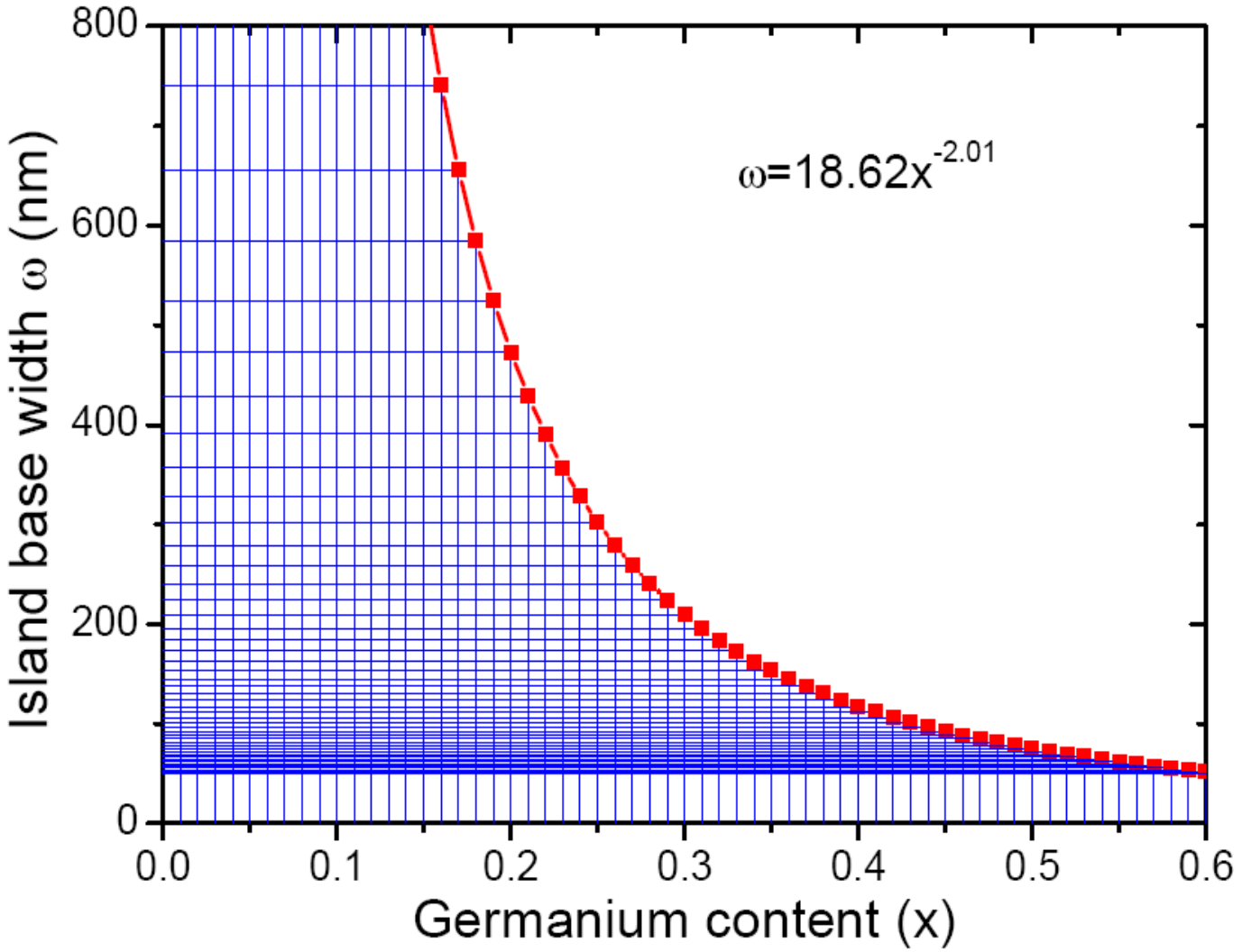


CTEM image

Sample A

LPE grown $Si_{1-x}Ge_x$ islands

Ge concentration x vs. base plane width ω

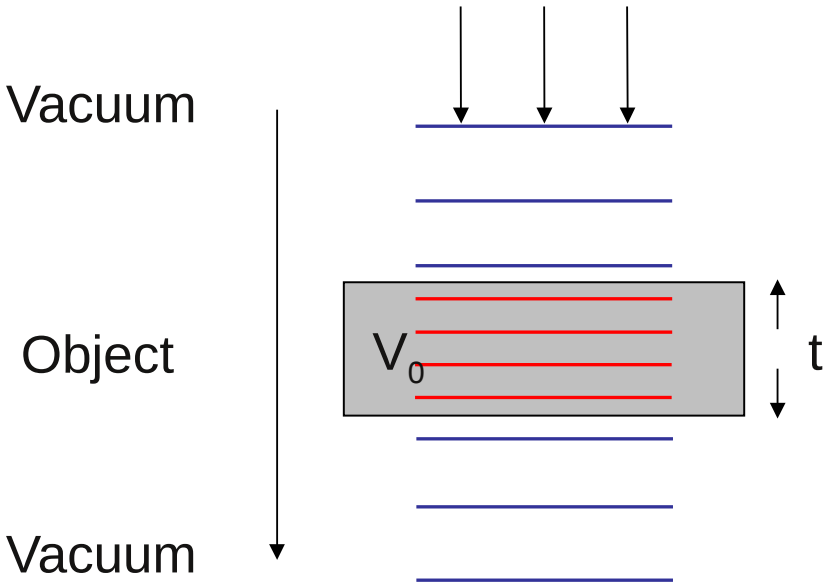


Sample A:
ω=130 nm
Ge: ~ 38%

Sample B:
ω=350 nm
Ge: ~23%

W. Dorsch et al. Appl. Phys. Lett. 72 (1998) 179 and J. Crystal Growth 183 (1998) 305

High energy electrons: Phase grating approximation

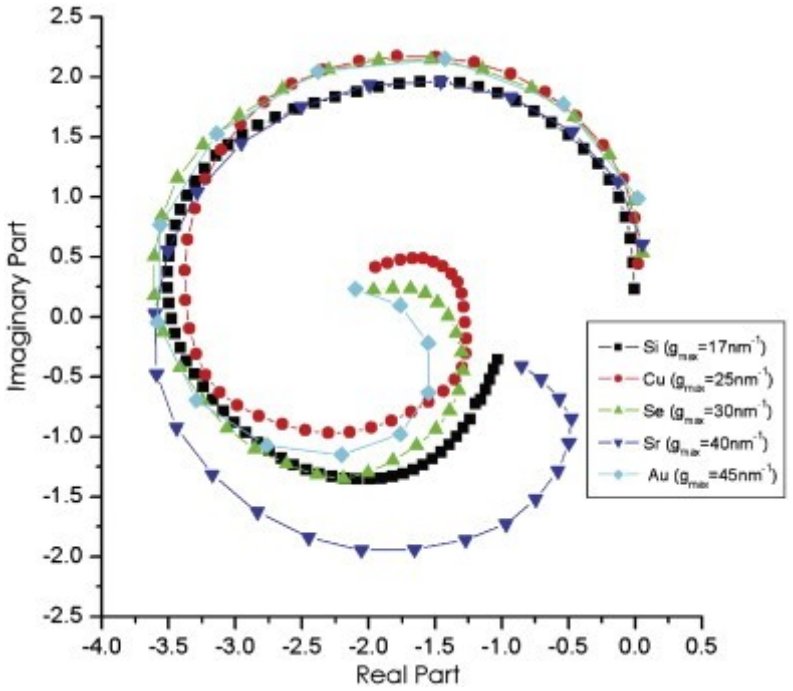


Phase shift $\varphi = \sigma V_0 t$

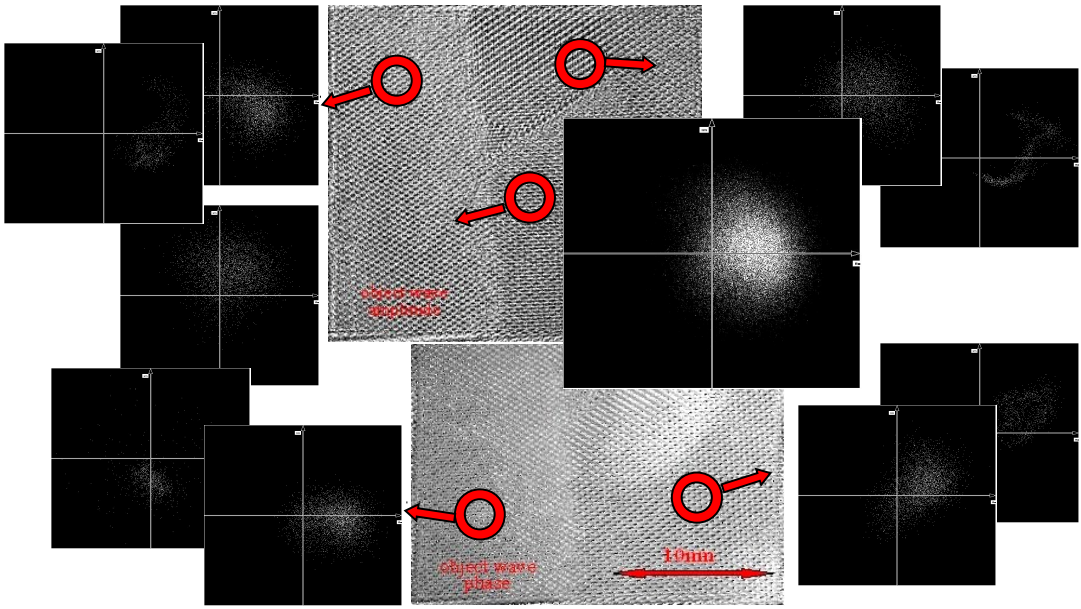
V_0: Mean inner Coulomb potential

t: Object thickness

$$\sigma = \frac{2\pi}{\lambda V}\left(\frac{m_0c^2 + eV}{2m_0c^2 + eV}\right) = \frac{2\pi me\lambda}{h^2}$$

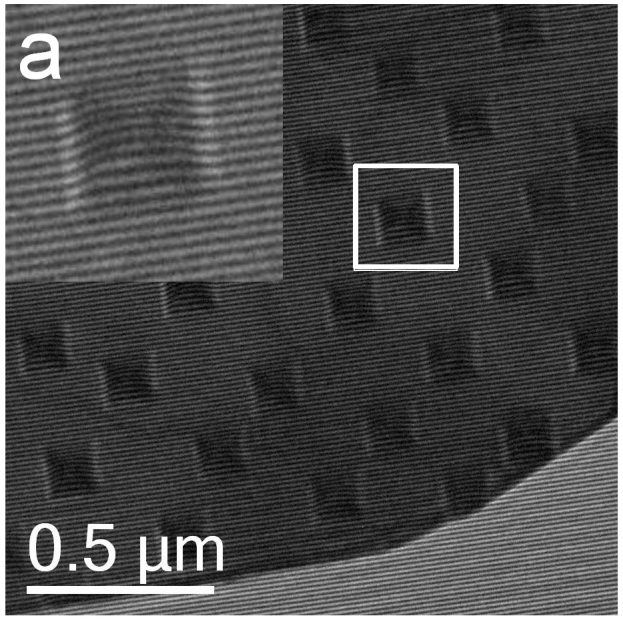


Multi-slice simulated Argand plots for [1 0 0] zone axis orientation, absorption potential = 0.1 real potential. A. Wang et al., UM 110 (2010) 527

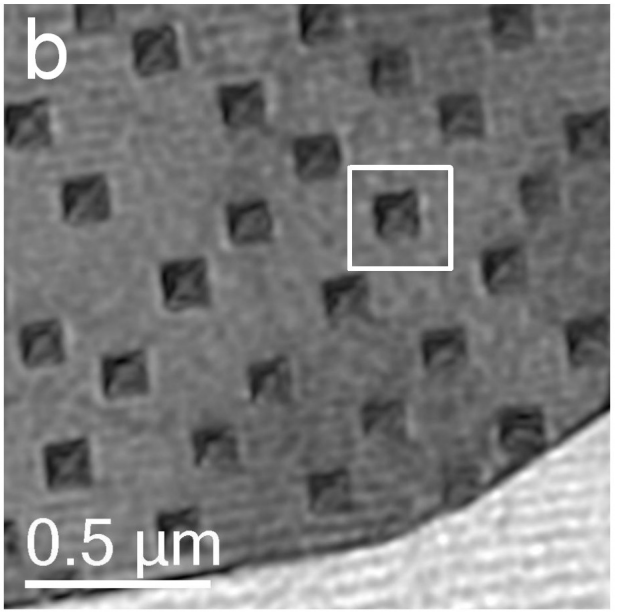


Argand plots for selected regions of the reconstructed GeCdTe 300kV exit wave (D. Smith, M. Lehmann)

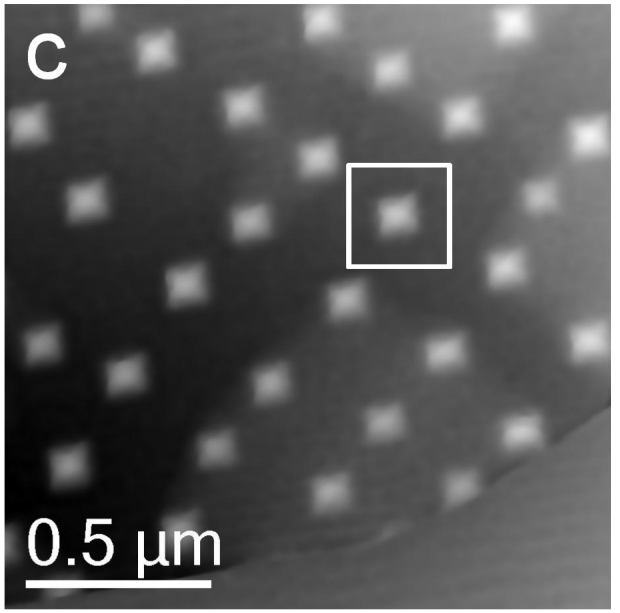
Electron holography of (Si,Ge) islands



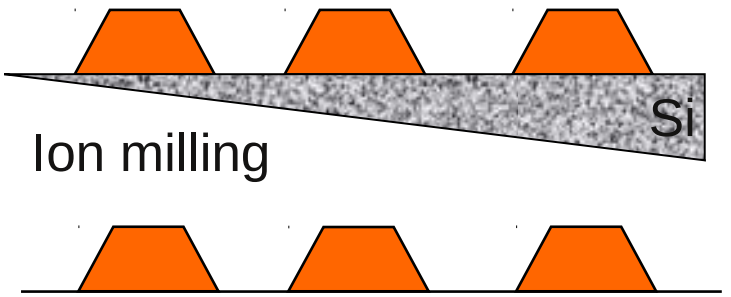
Electron hologram



Amplitude

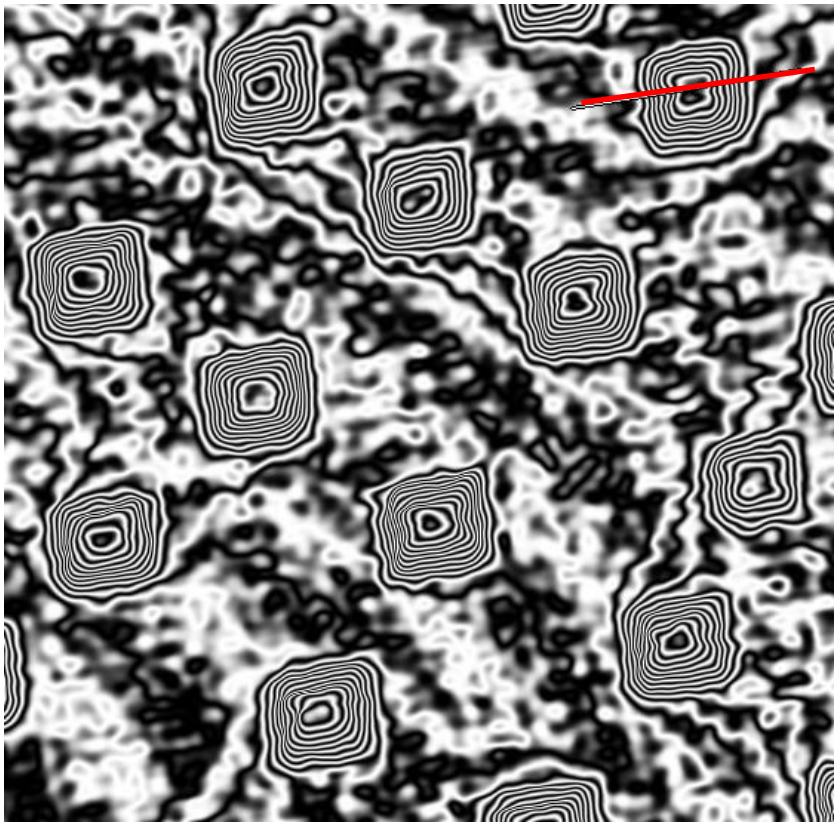


Unwrapped phase

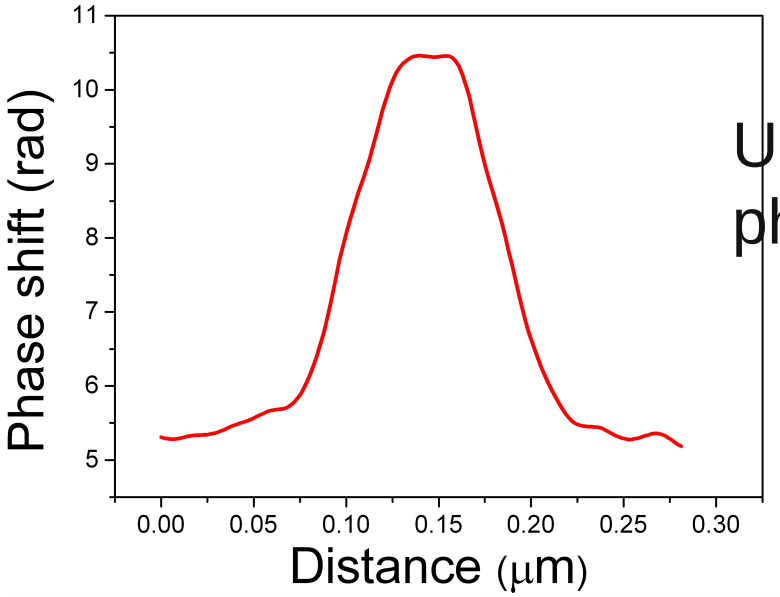


Sample A

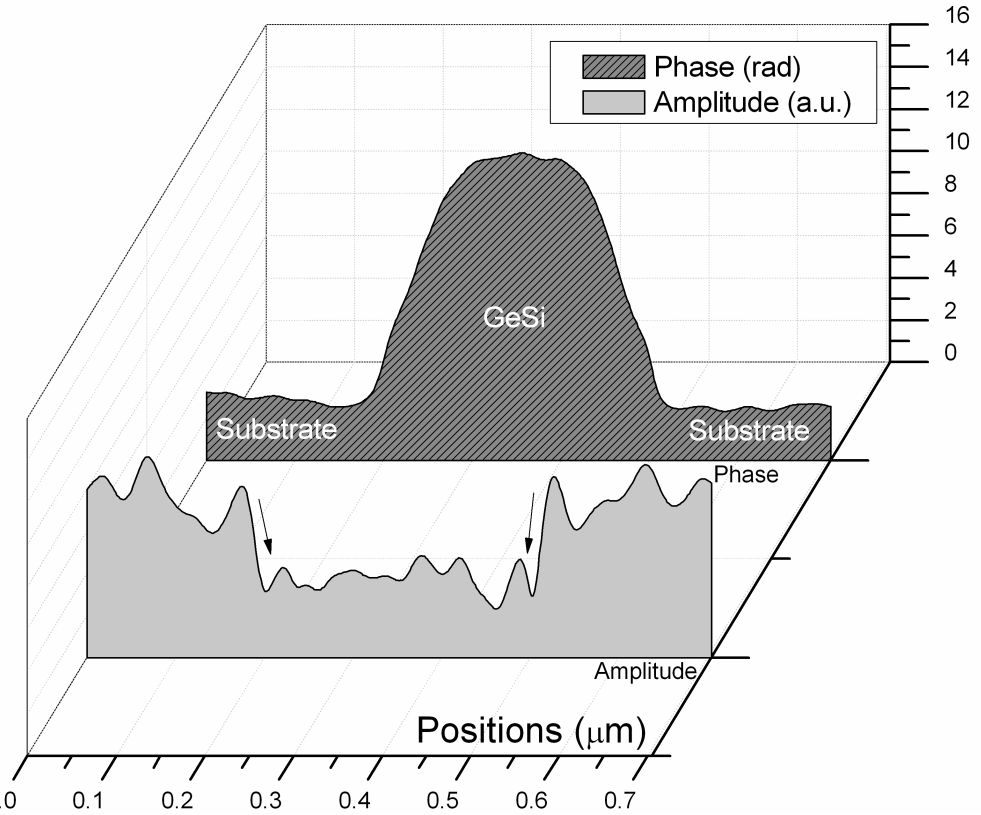
Amplified phase image of (Si,Ge) islands



10 × amplified phase image



Unwrapped phase profile

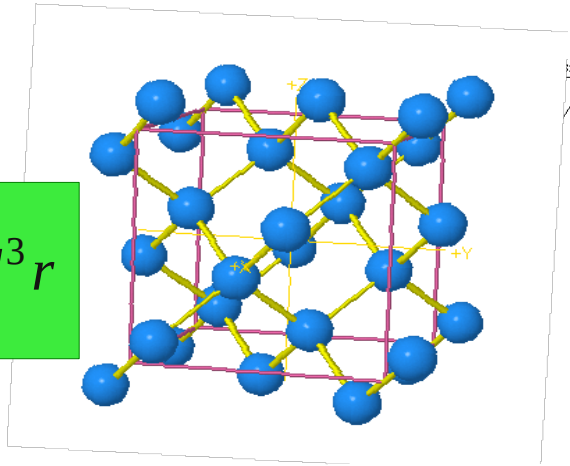


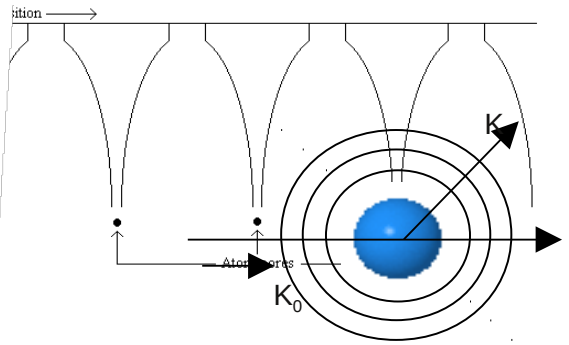
Sample A

Mean inner potential (MIP) & atomic scattering factors

MIP

$$V_0 = {}^{-1}\!/_{\Omega} \int V(r)\, d^3 r$$





atomic scattering & structure factor

$$f(0) = -\frac{m\Omega}{2\pi h^2} V_0$$

Ge_xSi_{1-x}

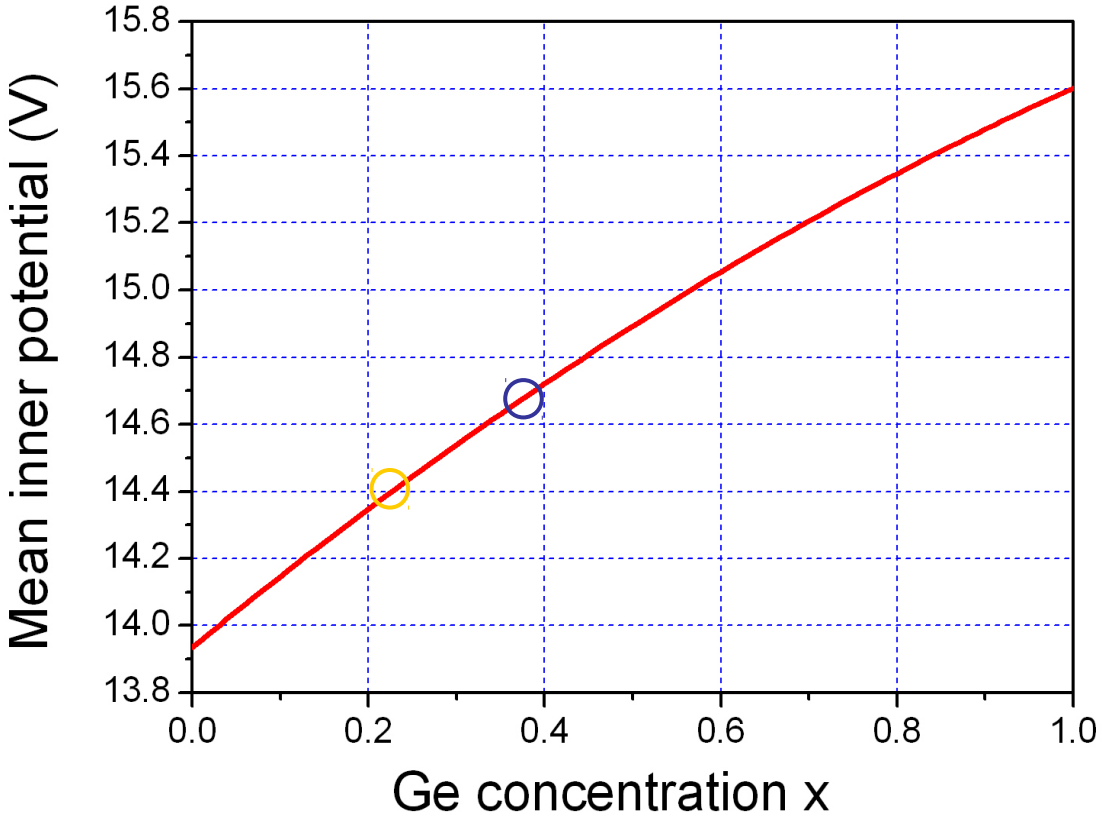
$f_{si}(0)$=5.828Å $f_{Ge}(0)$=7.378Å

$$V_0 = \frac{h^2}{2\pi me\Omega} \sum_i n_j f_{el}^j$$

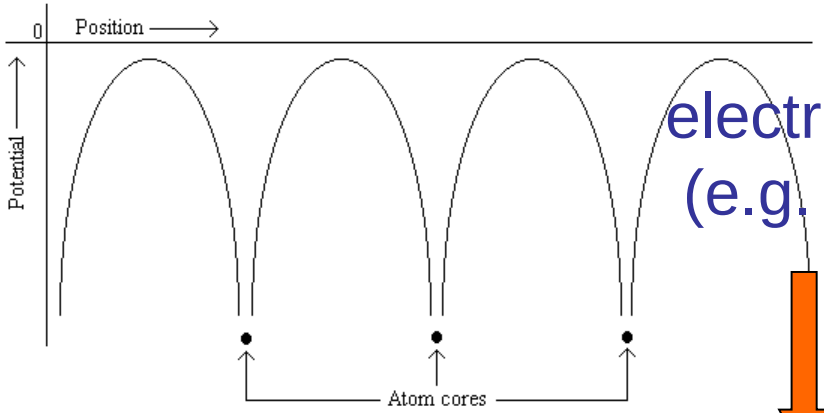
Lattice parameter:

a=(5.431 + 0.20*x* + 0.027*x*²) Å

J.P.Dismukes, et a.. *J. Appl. Phys.* **35**(1964)2899.



bonding & electron interaction effects

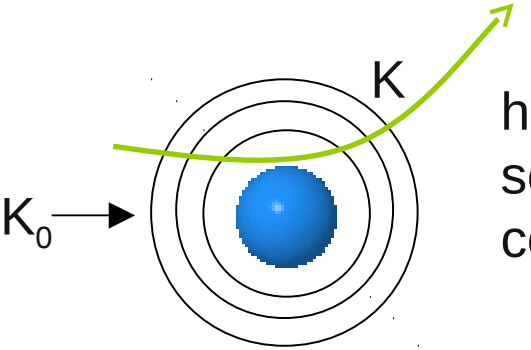


electron density
(e.g. via DFT)

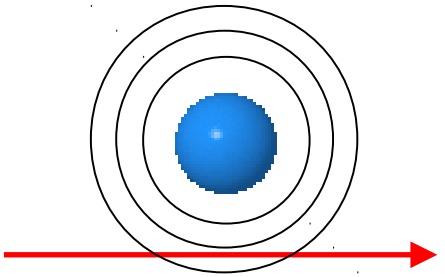
Poisson equation

$$\nabla^2\varphi = -\frac{\rho}{\varepsilon_0}$$

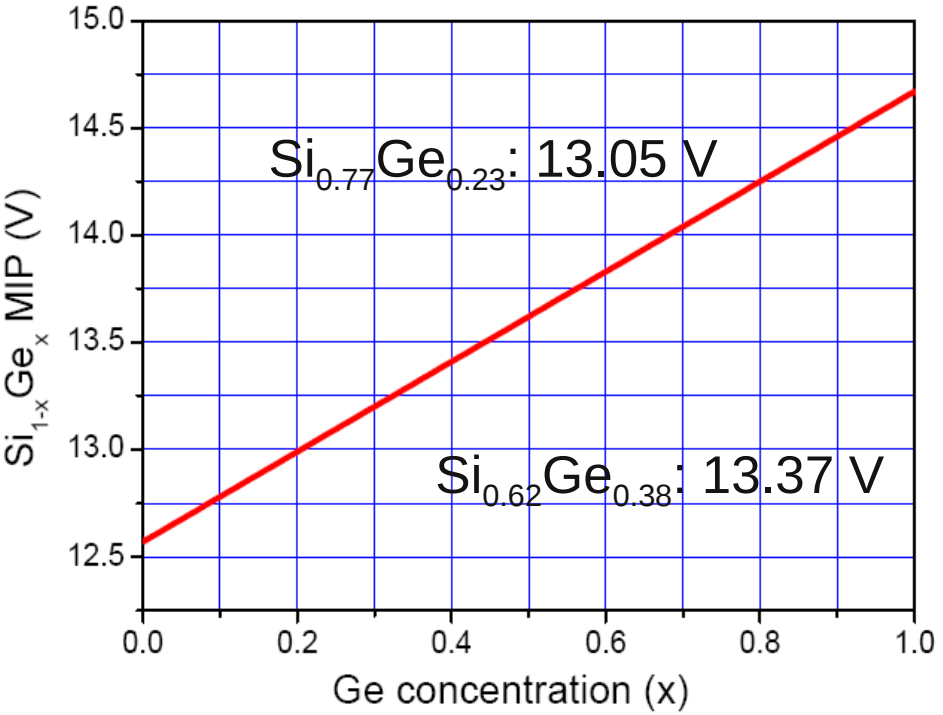
electric potential

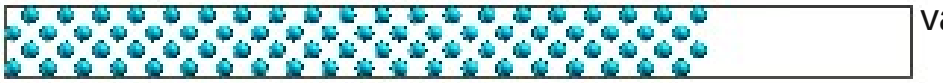


high angle scattering: core part



low angle scattering: mainly electron density





electron density from unit to super cell

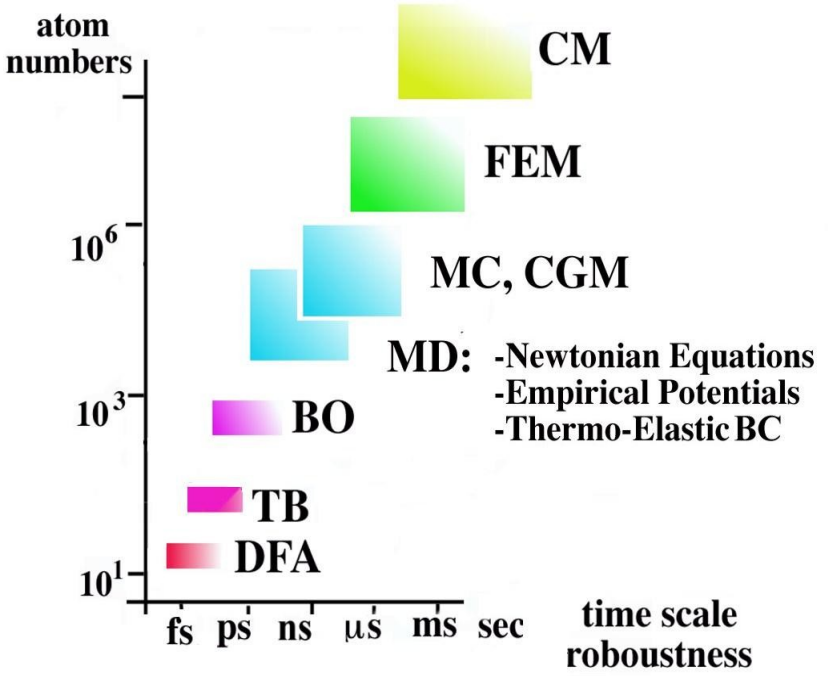
	DFT(V)	No-bonding (V)
Si	12.57	13.91
Ge	14.67	15.58

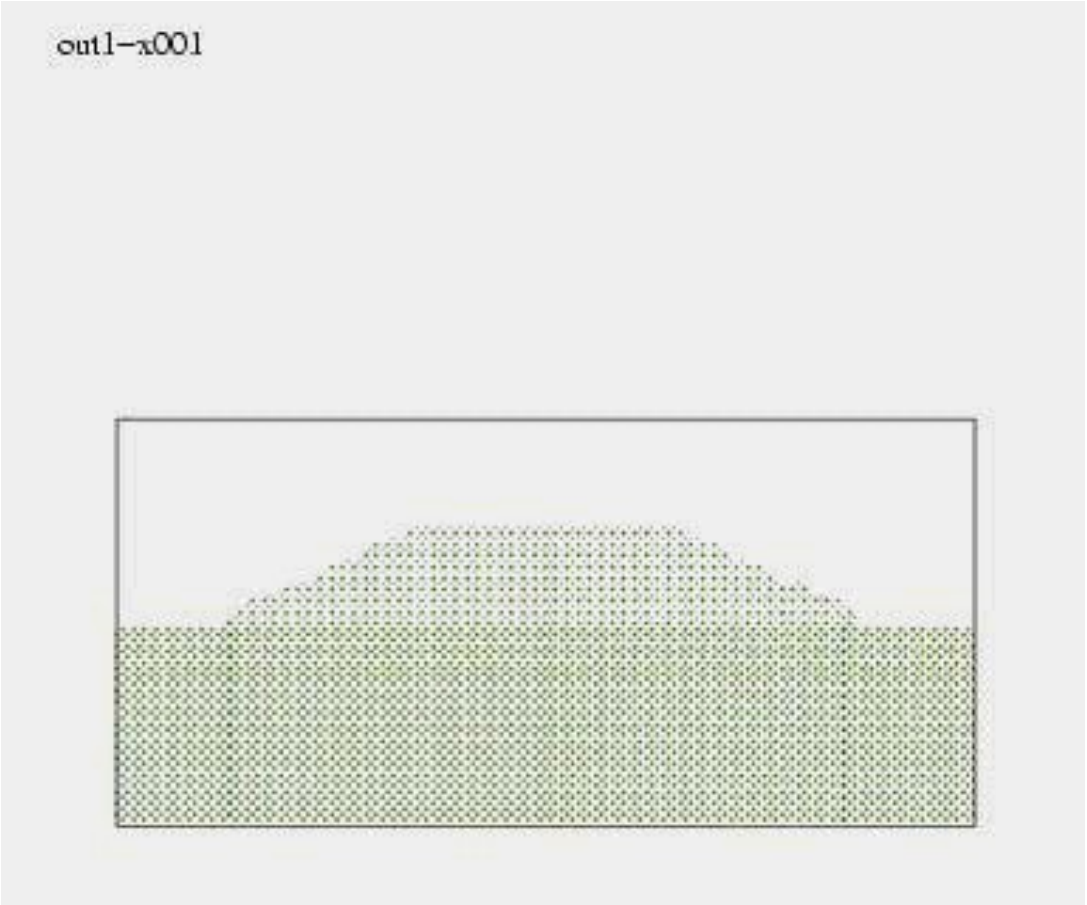
P. Kruse et al. Ultramicroscopy 106 (2006) 105

Classical molecular dynamics

time-integration (fs-steps) up to relaxation (< 1ns)
simultaneously 3N Newtonian equations of motion
with V= empirical interaction potential

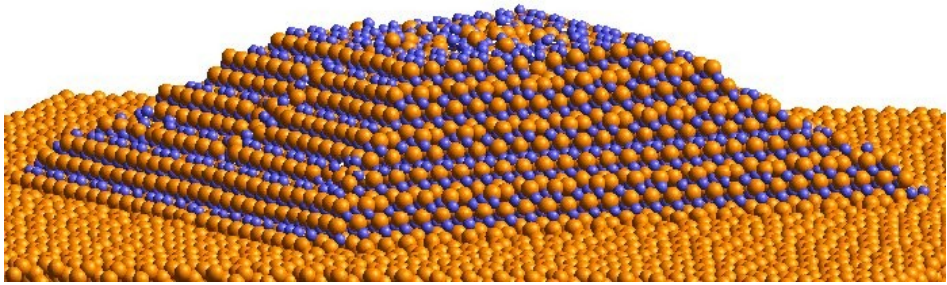
$$m_j \frac{d^2 r_j}{dt^2} = F_j = -\frac{\partial V}{\partial r_j}$$





Relaxation of an uncapped
(100)-SiGe/Si pyramid
$\Delta\Delta T$ = 20K/ns up/down

Δt = 0.1fs
T_{max} = 900K
Δt_{equil} = 1ns
Δt_{frame} = 0.1ns



BOP

embedded bonds instead atoms – two-center orthogonal TB
density matrix instead diagonalisation – Lanczos recursion

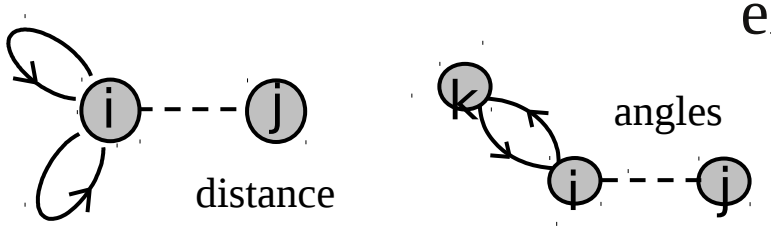
$E_{tot} = E_{rep} + E_{prom} + E_{band}(k)$

↓ ↓ ↓

empirical $s^2p^2 \rightarrow sp^3$ $\Sigma \mathbf{H}_{i\alpha,j\alpha}\mathbf{\Theta}_{i\alpha,j\alpha}$

↓ ↓

hopping integrals BOmatrix Pettifor-Aoki
Slater-Koster ssσ, spσ, ppσ...
electronic hopping in closed loops

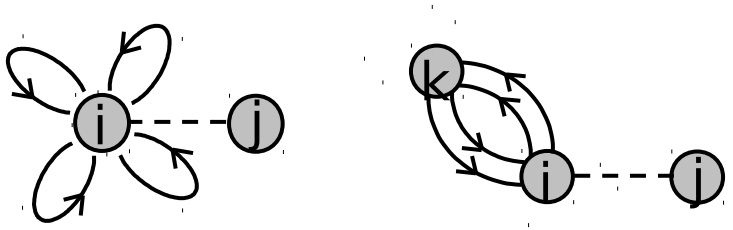


$$\Phi^i_{2\sigma} = \hat{\delta}^2_i + \sum_{k(i)} \hat{\beta}^2_{\sigma,ik}(g^2_{\sigma,jik} + g^2_{\pi,jik})$$

$$\delta_i = E_p - E_s$$

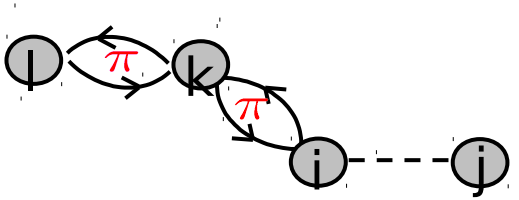
$$g_{\sigma,jik} = 1 + (\cos\theta_{jik} - 1)p_{\sigma,i}$$

$$g_{\varphi,jl} = p_{\pi,ik}\sqrt{p_{\sigma,i}p_{\sigma,k}}\cos\varphi_{jl}\sin\theta_{jik}\sin\theta_{ikl}$$



$$\Phi^i_{4\sigma} = \hat{\delta}^4_i + \hat{\beta}^4_{\sigma,ik}g^2_{\sigma,jik} + \text{.....} + \hat{\beta}^2_{\sigma,ik}\hat{\beta}^2_{\sigma,kl}\left[2g_{\sigma,jik}g_{\sigma,ikl} + g_{\varphi,jl}\right]g_{\varphi,jl}$$

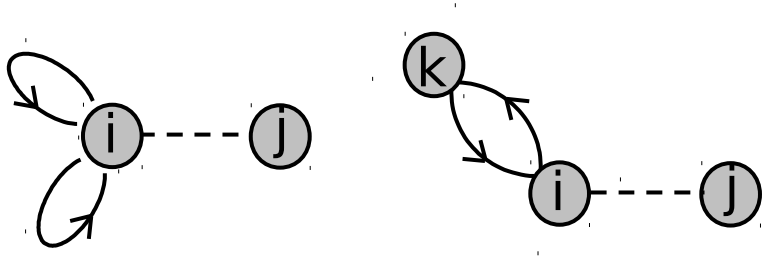
dihedral ,,,, torsion



Bond Order Potential

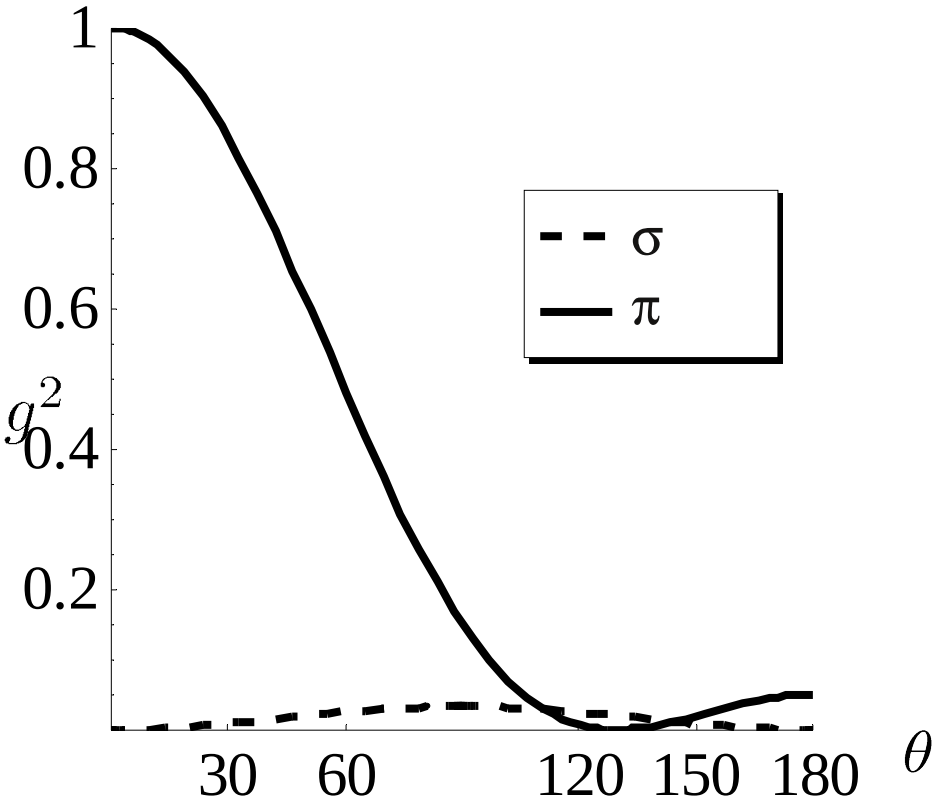
- Slater-Koster products = electronic hoppings in closed loops

$$\Phi^i_{n\sigma}(a_i,b_i) \Rightarrow \sum_{\alpha_1,\ldots,\alpha_{n-1}} \langle i\sigma|\hat{H}\,|\alpha_1\rangle\,\langle\alpha_1|\,\hat{H}\,|\alpha_2\rangle\,\ldots\,\langle\alpha_{n-1}|\,\hat{H}|i\sigma\rangle$$



$$\Phi^i_{2\sigma} \propto \langle i\sigma|\,\hat{H}\,|i\sigma_0\rangle^2 + \langle i\sigma|\,\hat{H}\,|k\rangle^2$$

On-Site Inter-Site



$$\Phi^i_{2\sigma} = \hat{\delta}^2_i + \sum_{k(i)} \hat{\beta}^2_{\sigma,ik}(g^2_{\sigma,jik} + g^2_{\pi,jik})$$

$$\delta_i = E_\mathrm{p} - E_\mathrm{s}$$

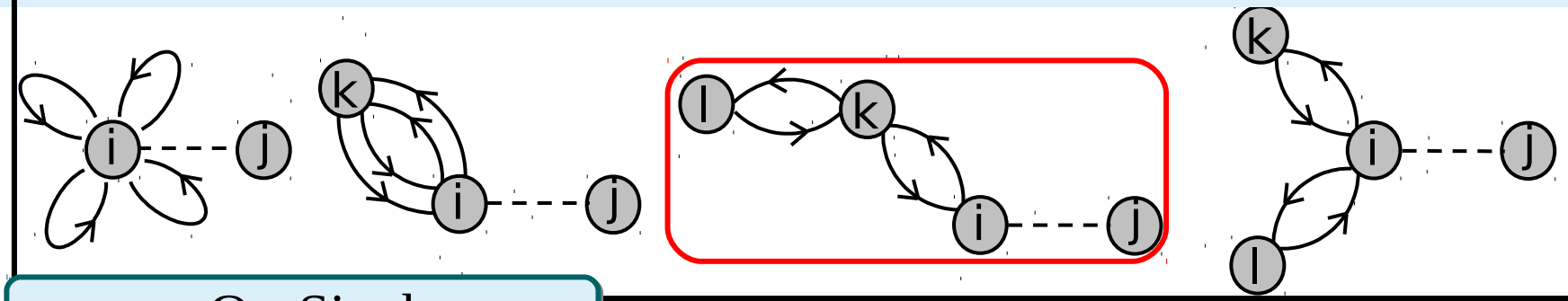
separation

$$g_{\sigma,jik} = 1 + (\cos\theta_{jik} - 1)p_{\sigma,i}$$

angular functions

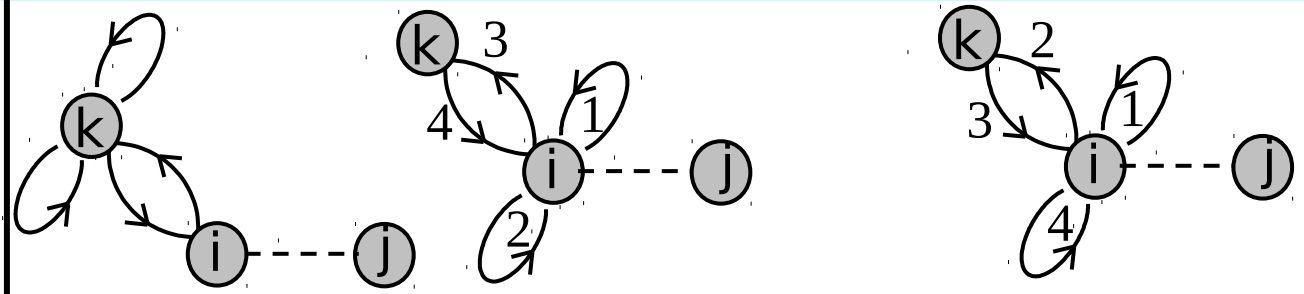
σ & On-Site hops

$$\Phi^i_{4\sigma} = \hat{\delta}^4_i + \hat{\beta}^4_{\sigma,ik} g^2_{\sigma,jik} + \hat{\beta}^2_{\sigma ik}\hat{\beta}^2_{\sigma kl} g^2_{\sigma,jik}\, g^2_{\sigma,ikl} + \hat{\beta}^2_{\sigma,ik}\hat{\beta}^2_{\sigma,il} g_{\sigma,jik} g_{\sigma,kil} g_{\sigma,jil}$$



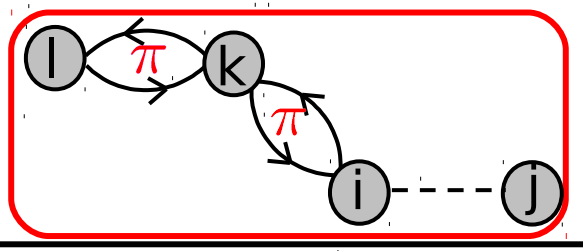
new On-Site hops

$$+ \hat{\beta}^2_{\sigma,ik} g^2_{\sigma,jik} (\hat{\delta}^2_k + 2\hat{\delta}^2_i) + \hat{\beta}^2_{\sigma,ik} p_{\sigma,i}(1-p_{\sigma,i})(1-\cos\theta_{jik})^2 \hat{\delta}^2_i$$



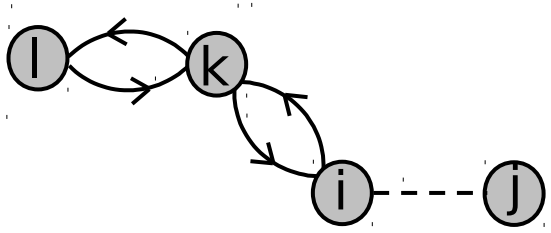
new π hops

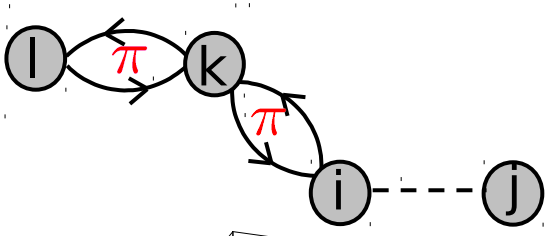
$$+ \hat{\beta}^2_{\sigma,ik}\hat{\beta}^2_{\sigma,kl} \left[2 g_{\sigma,jik} g_{\sigma,ikl} + g_{\varphi,jl}\right] g_{\varphi,jl}$$

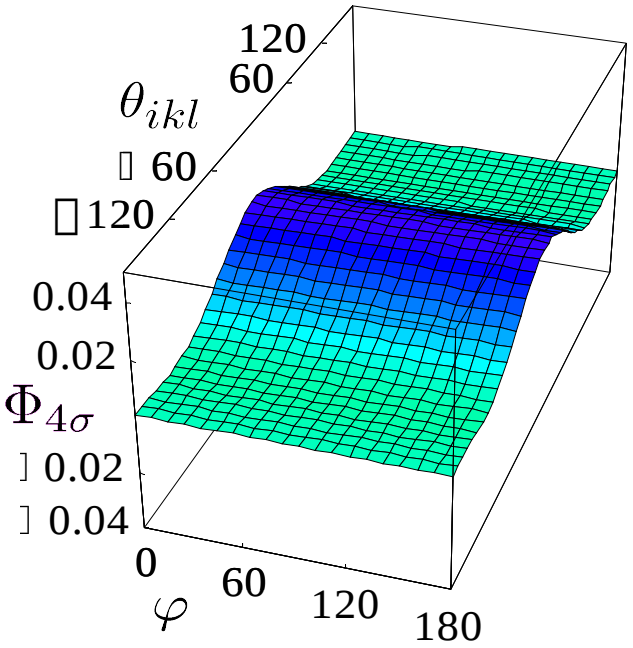


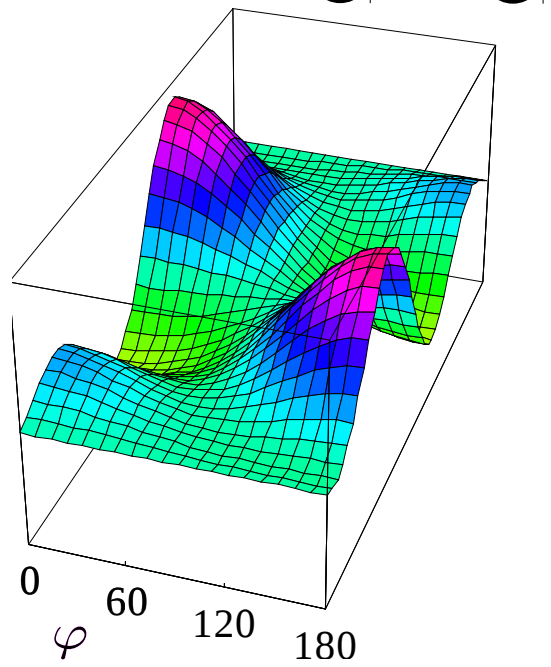
torsion included: $g_{\varphi,jl} = p_{\pi,ik}\sqrt{p_{\sigma,i} p_{\sigma,k}}\cos\varphi_{jl}\sin\theta_{jik}\sin\theta_{ikl}$

Comparison of π–and σ–contributions

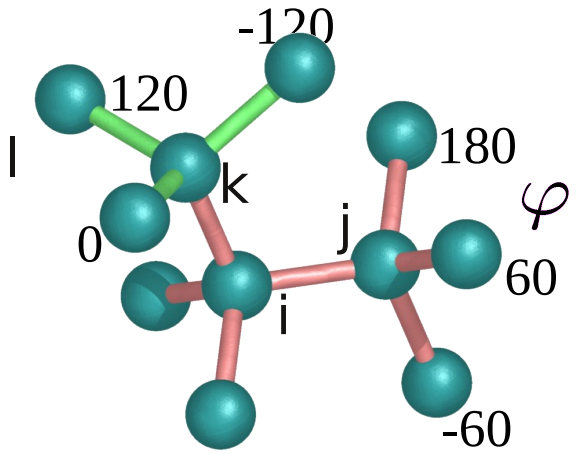






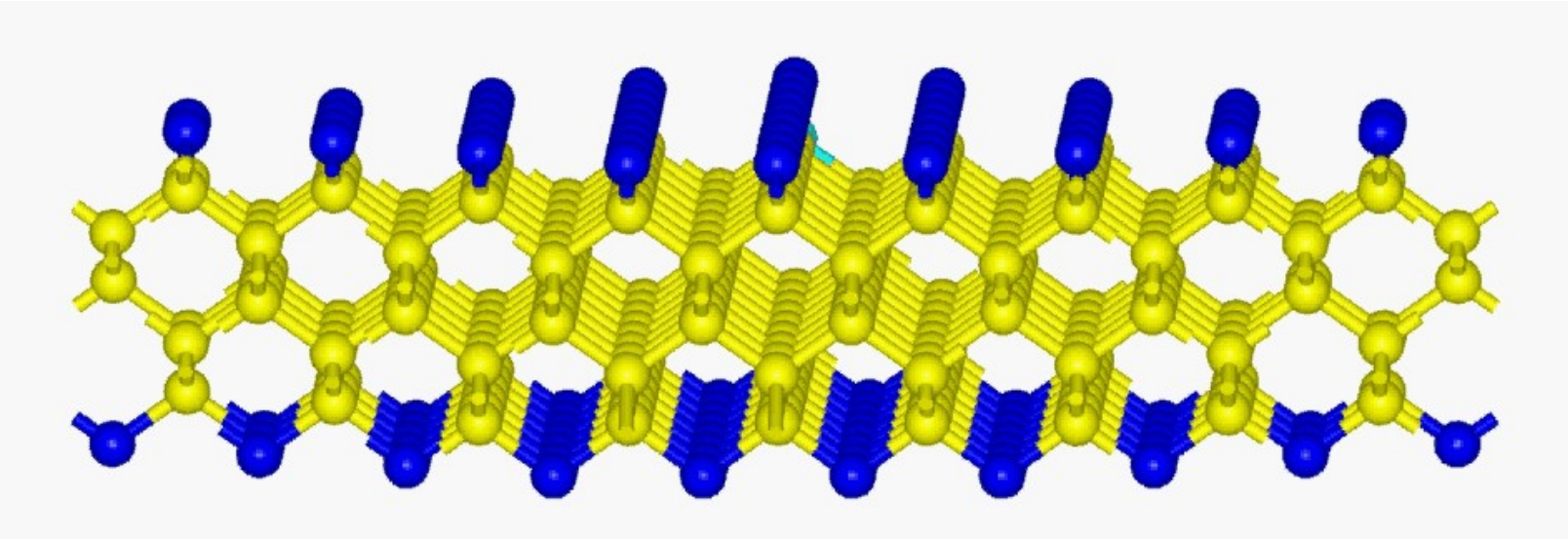


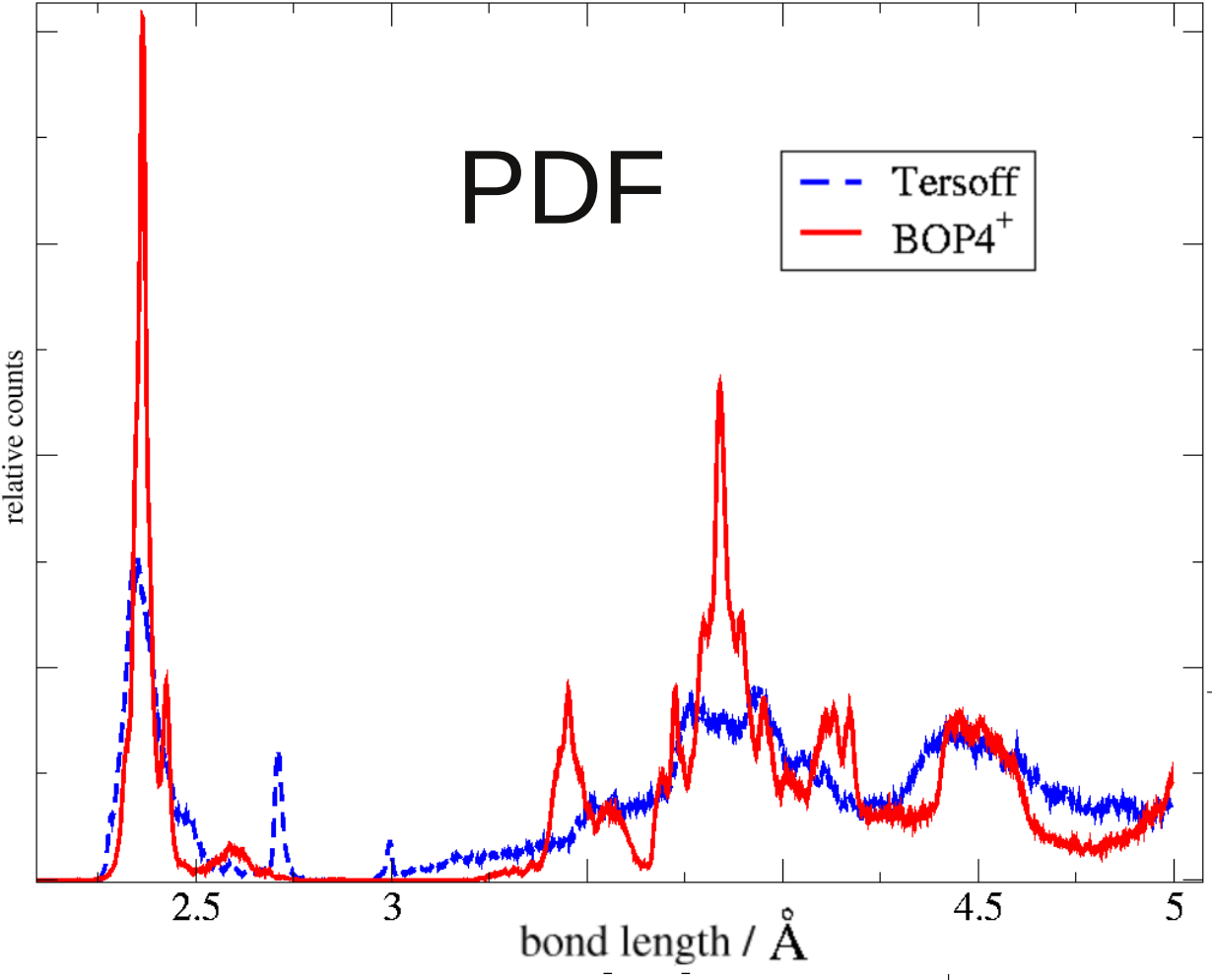
$$\cos\theta_{jik} = -1/3$$



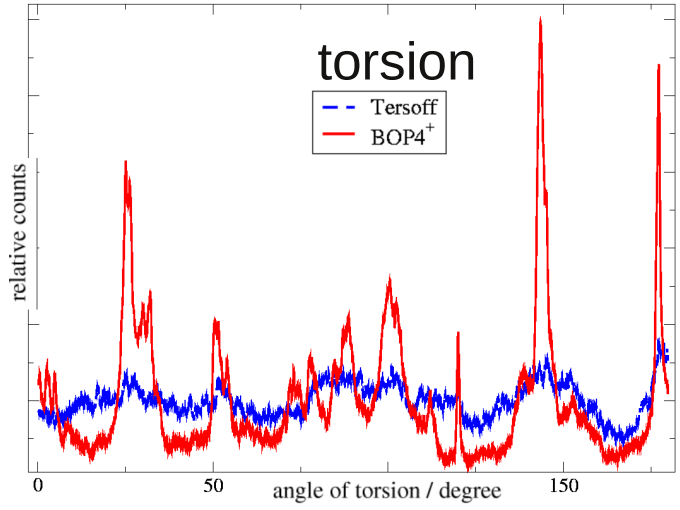
⇨ increased stiffness

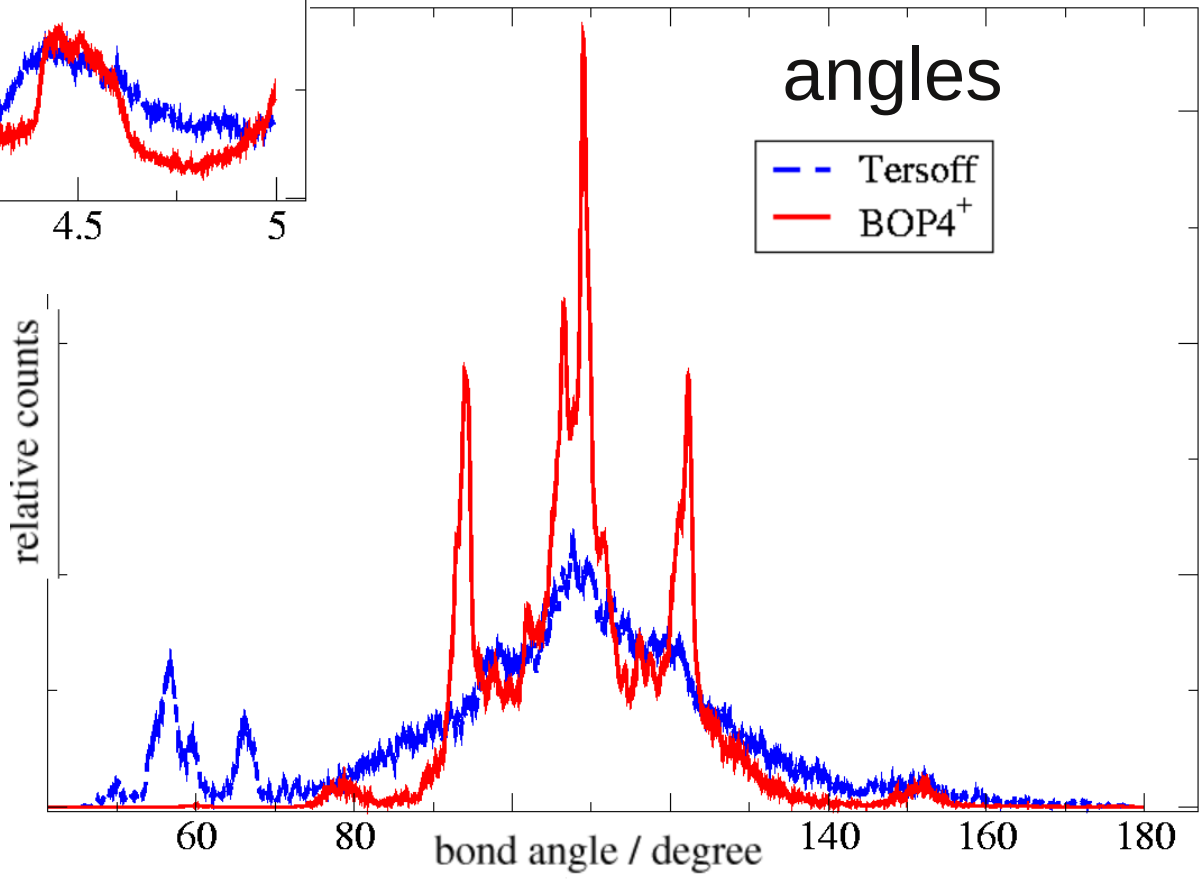
Si (110) surface dimerization 0K->600K->0K

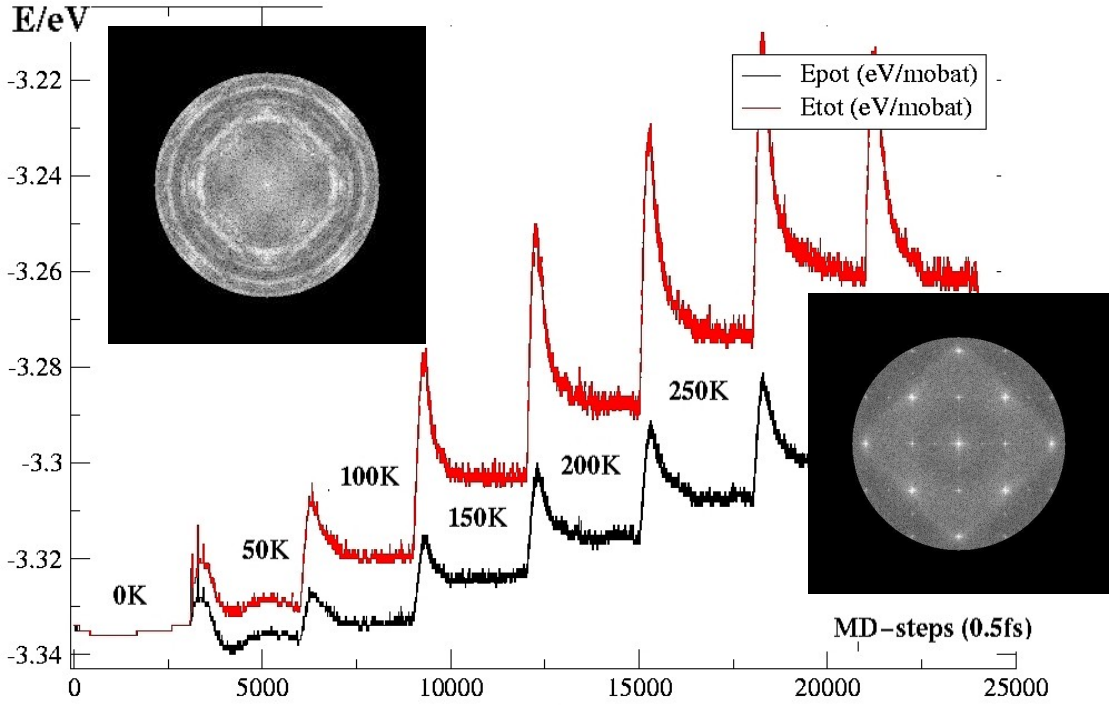


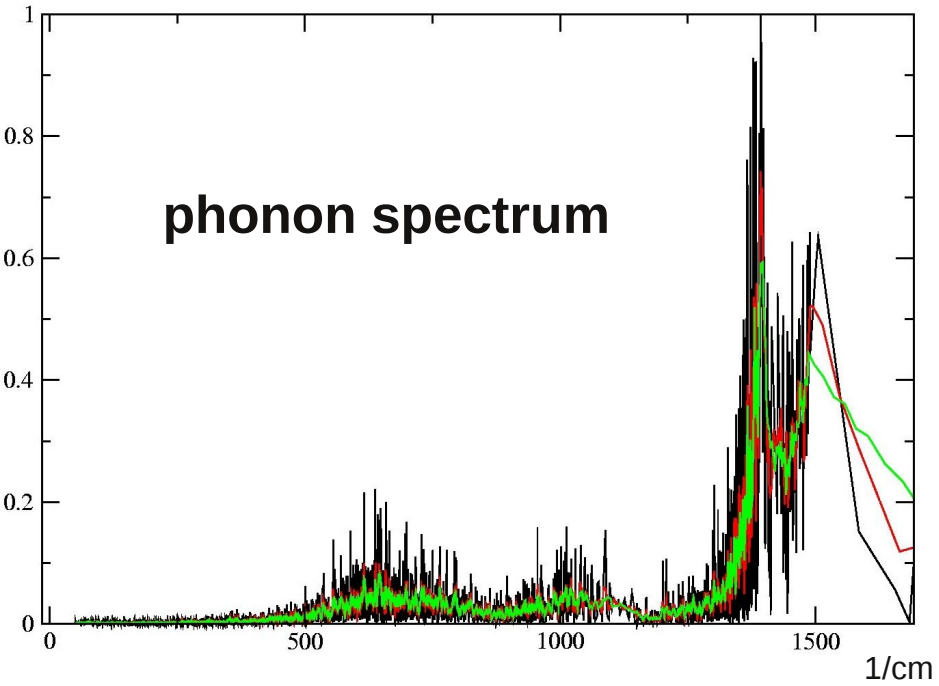


Distribution of pair distances, angles and torsions at the 1.4° twist bonded Si-001 interface comparing Tersoff and BOP4+ potentials after annealing at 900K

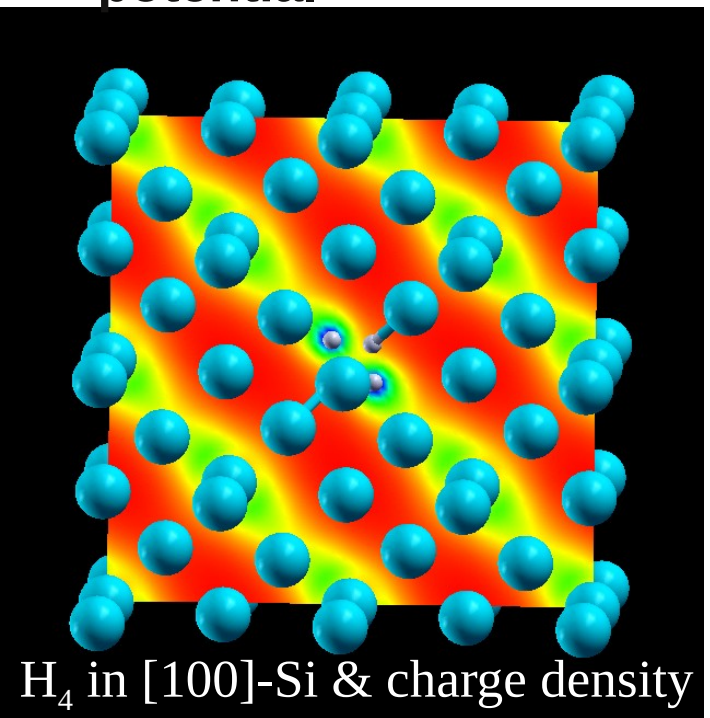


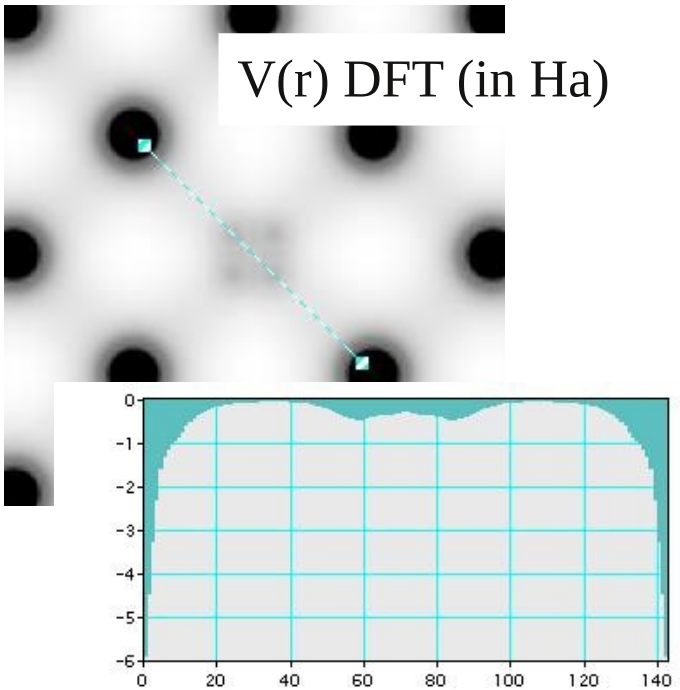


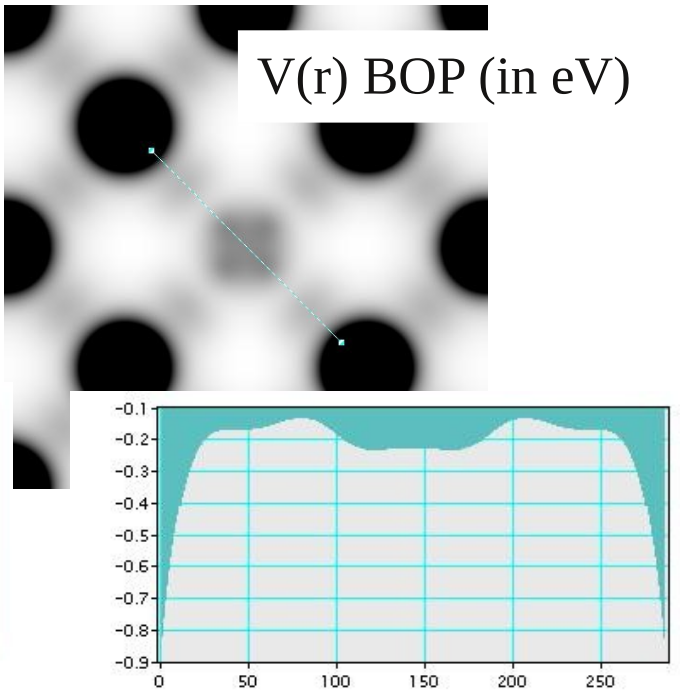


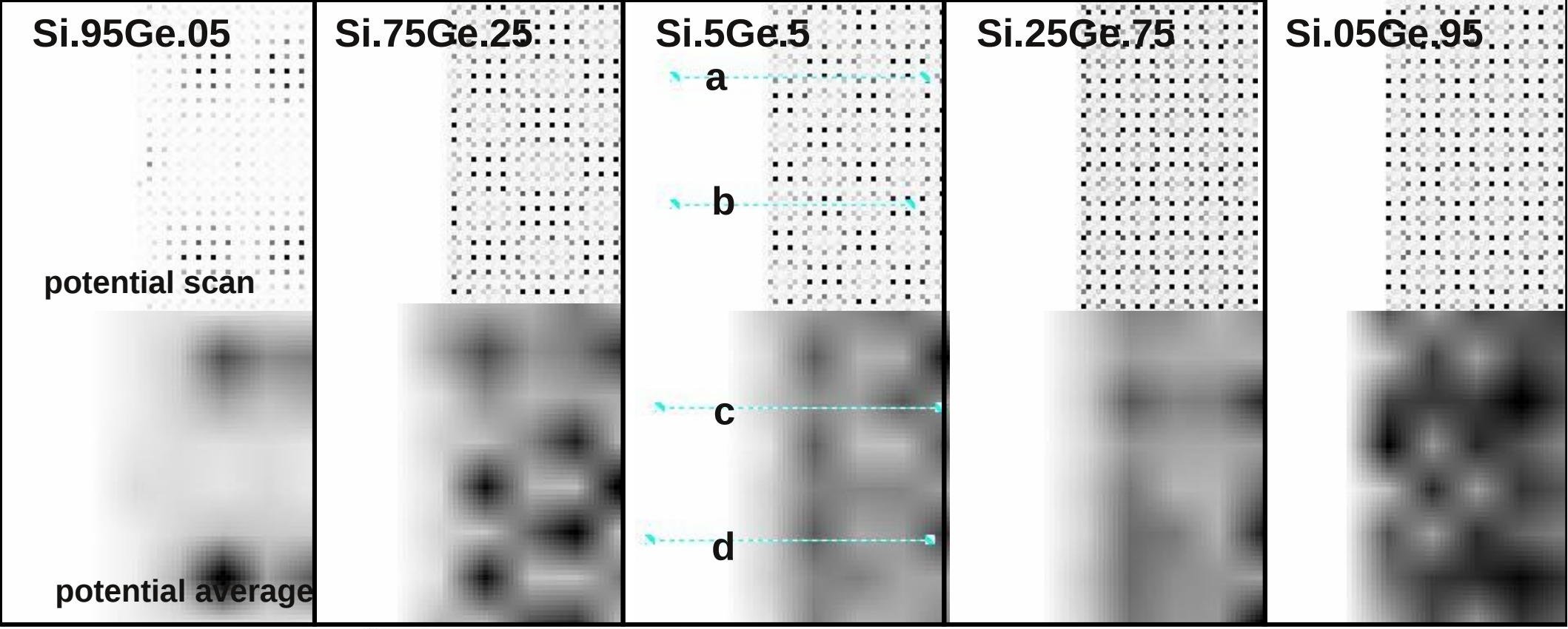


MD using BOP: TDS (Frozen Lattice) – charge density & scattering potential

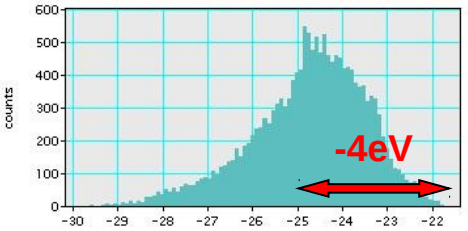


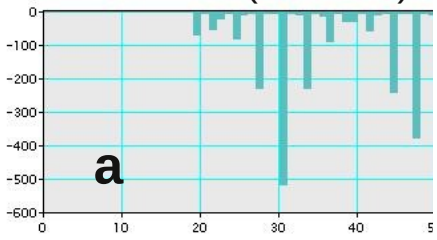


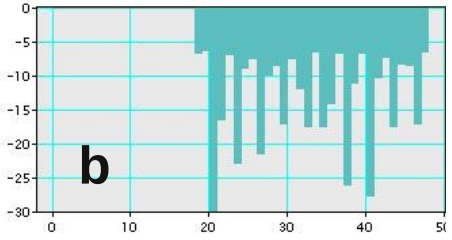


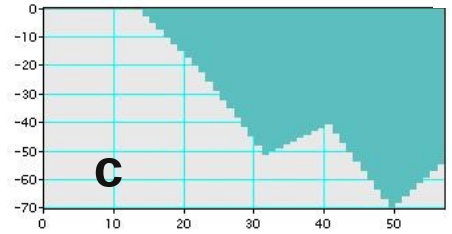


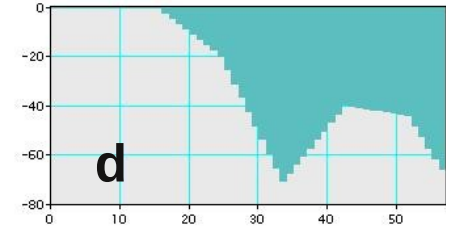
Si.5Ge.5
background
histogram
(counts/eV)
& linescans (Vo in eV)

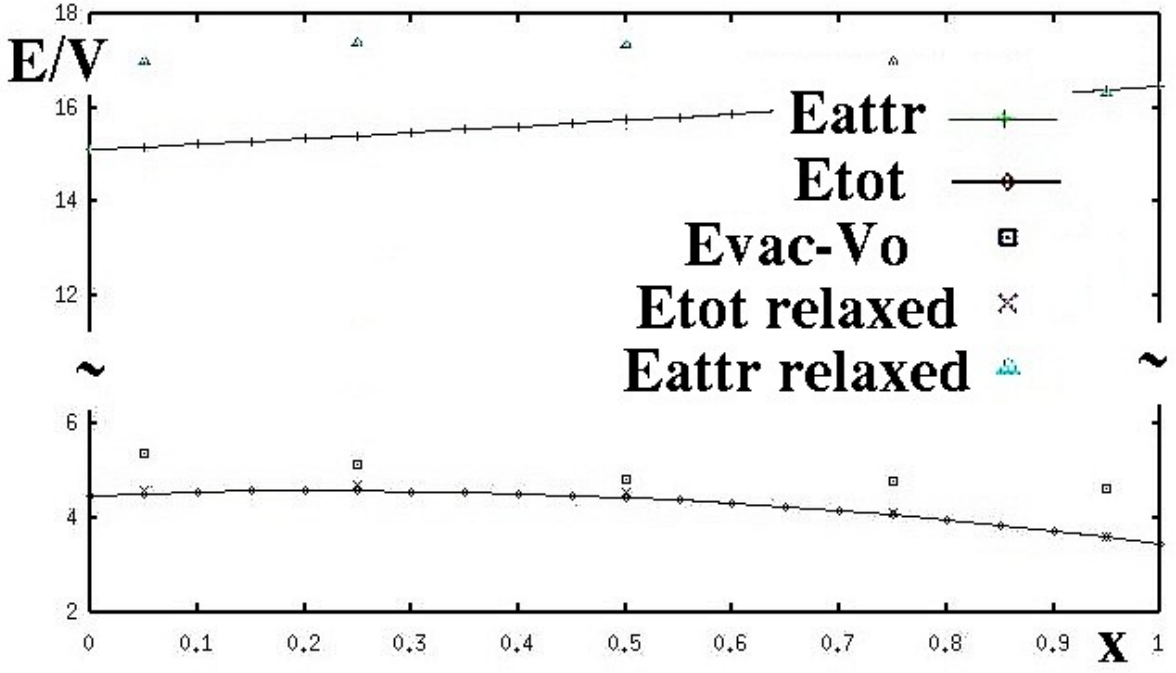


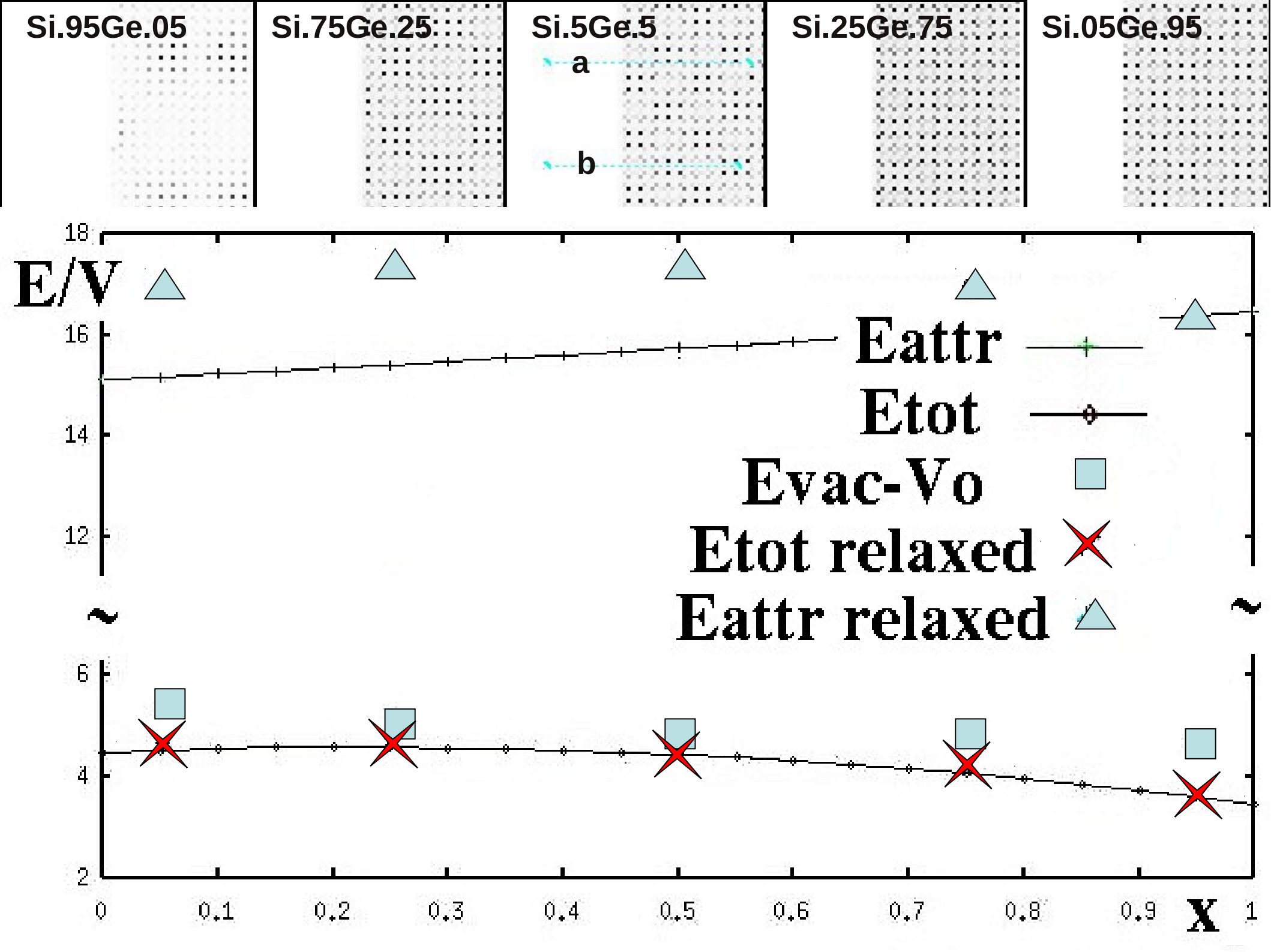


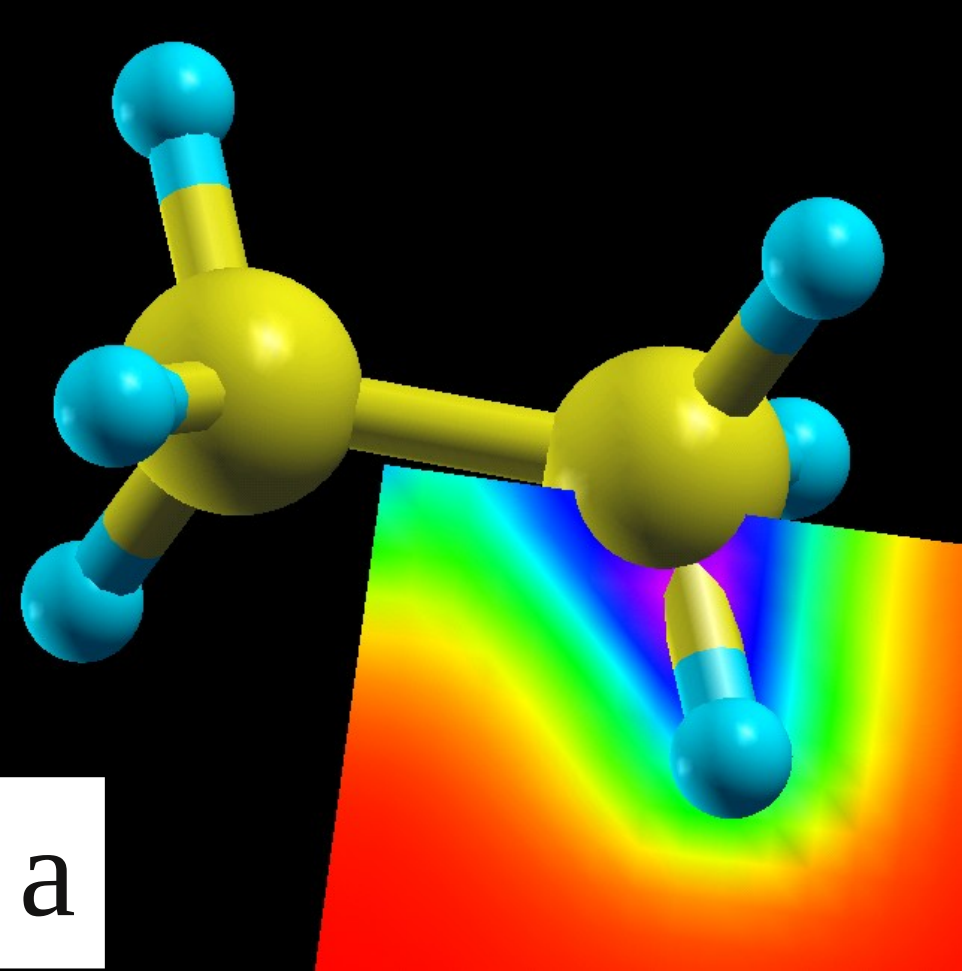




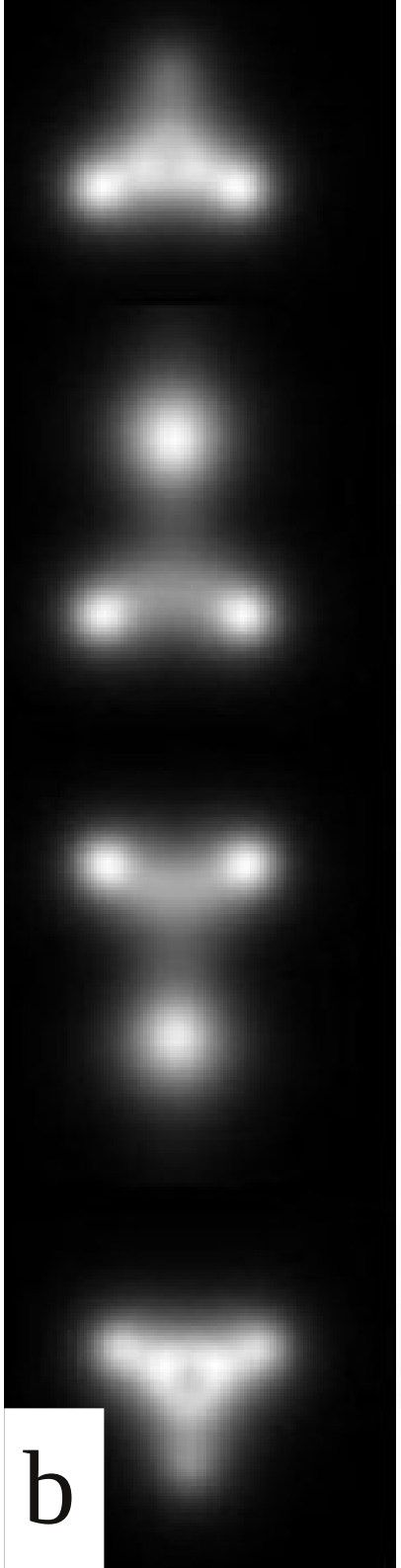


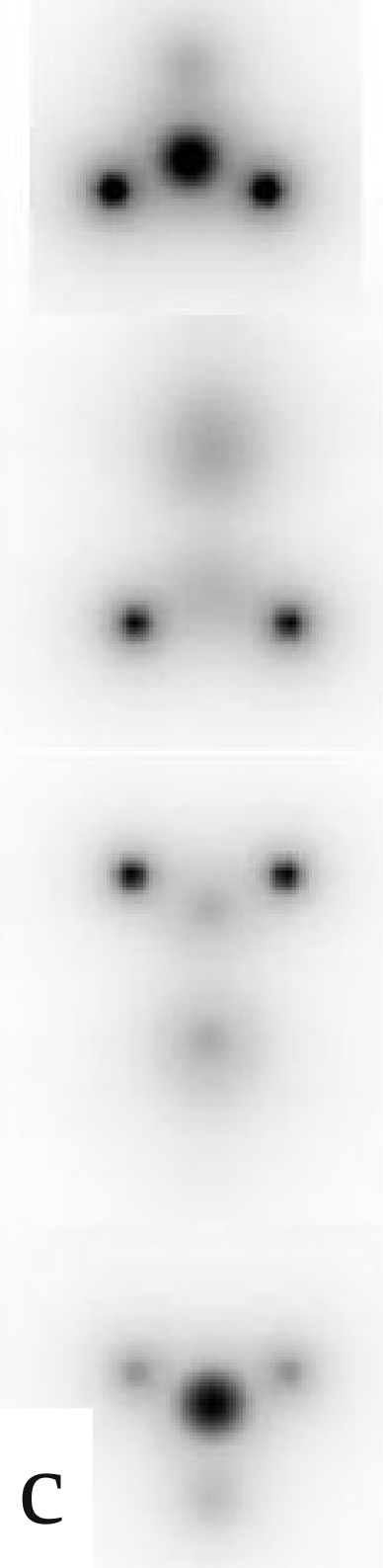


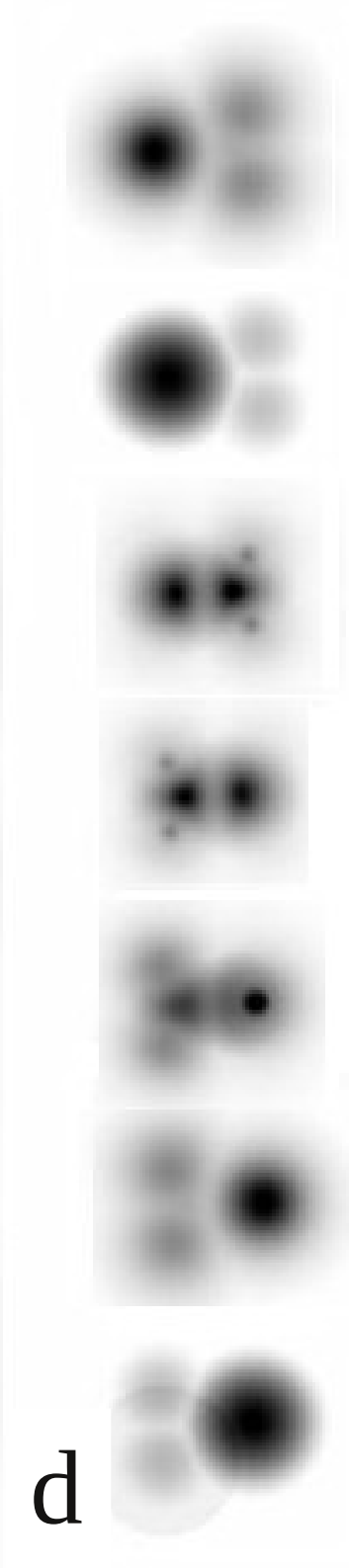

Si.95Ge.05
Si.75Ge.25
Si.5Ge.5
a
b
Si.25Ge.75
Si.05Ge.95
E/V
18
16
14
12
6
4
2
0
0.1
0.2
0.3
0.4
0.5
0.6
0.7
0.8
0.9
X
1
Eattr
Etot
Evac-Vo
Etot relaxed
Eattr relaxed



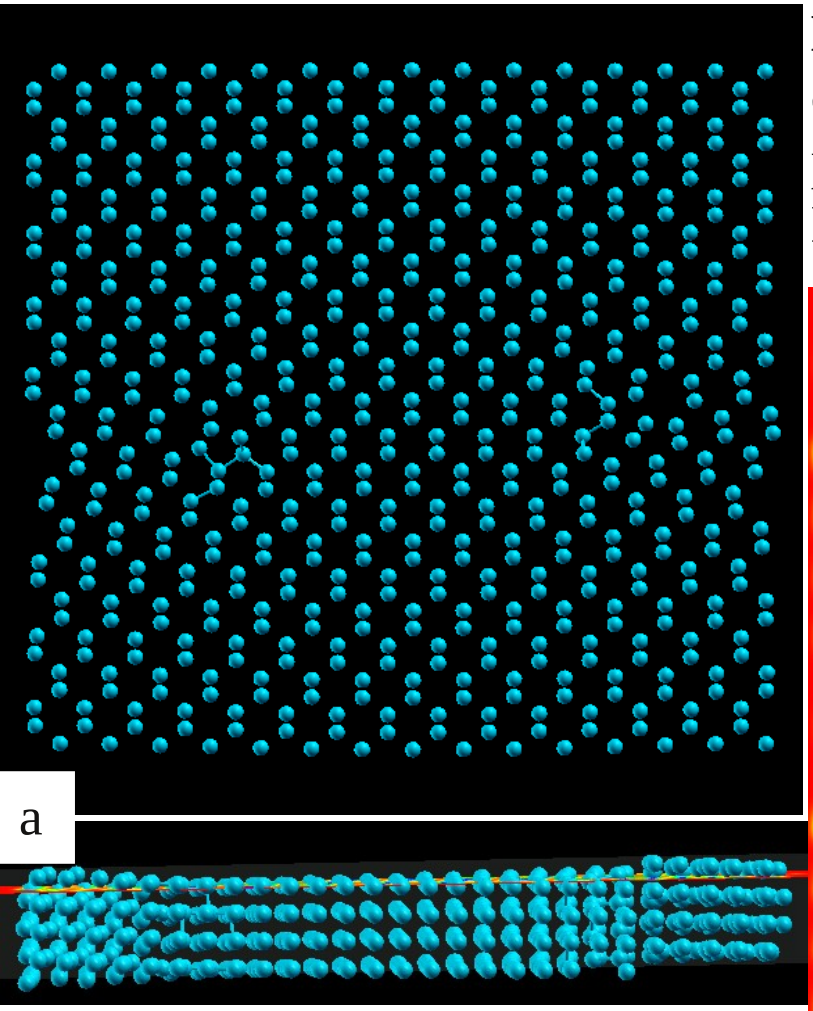
C_2H_6: DFT-optimized structure and charge density (a), selected cuts of density (b), potential (c), and sampled BOP (d)

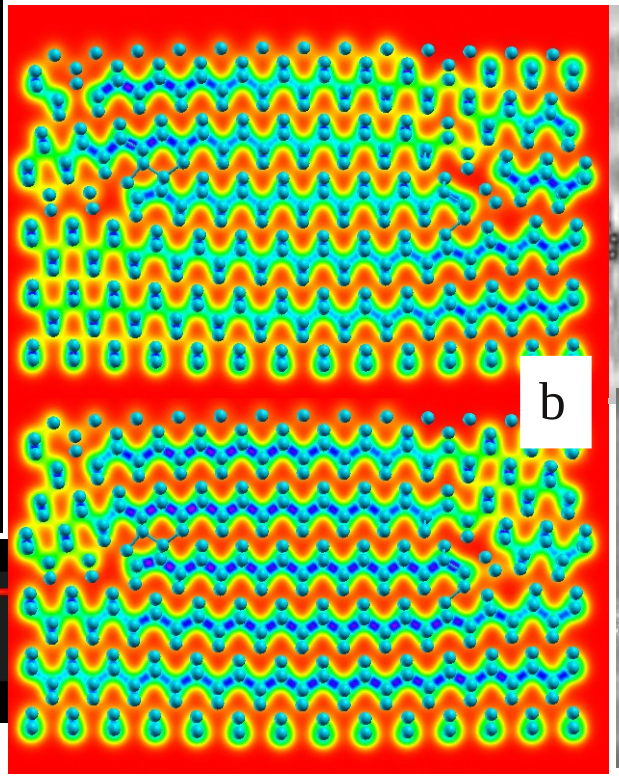


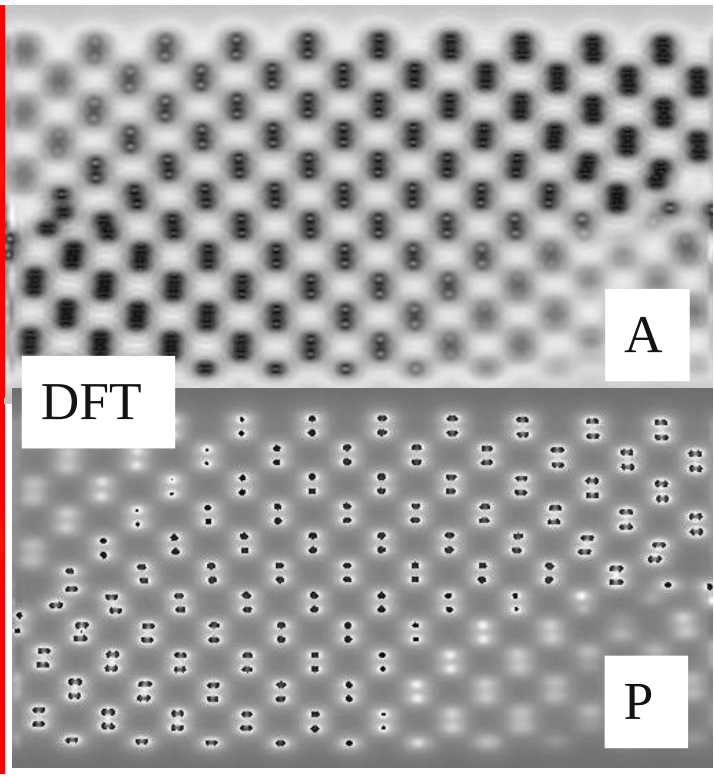


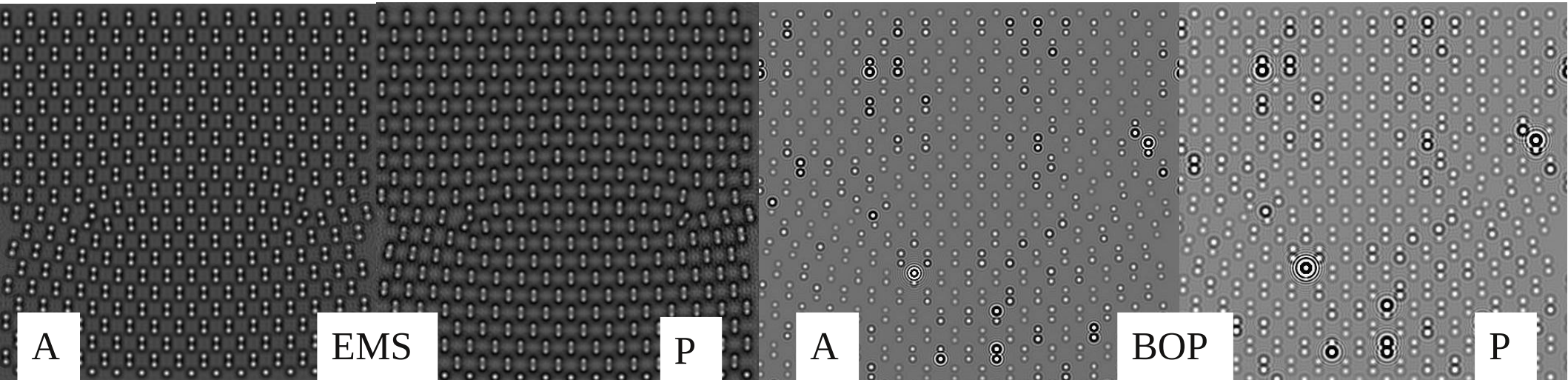


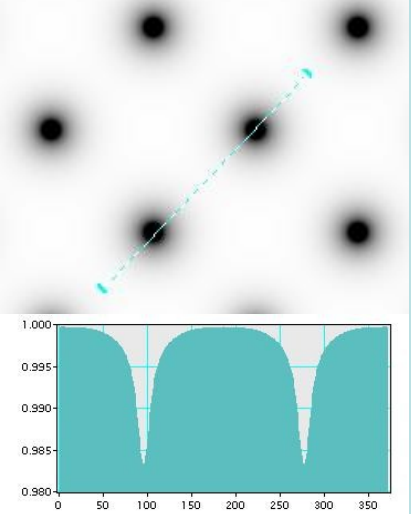
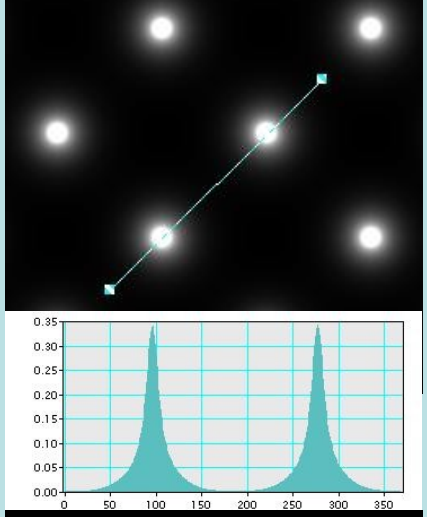
Dislocation dipole in 110-Si: BOP optimized structure (a), charge density (b), and simulated exit waves (amplitude A, phase P) for DFT, standard EMS potentials, and sampled BOP of 16 subslices (200kV, thickness=18.4nm).

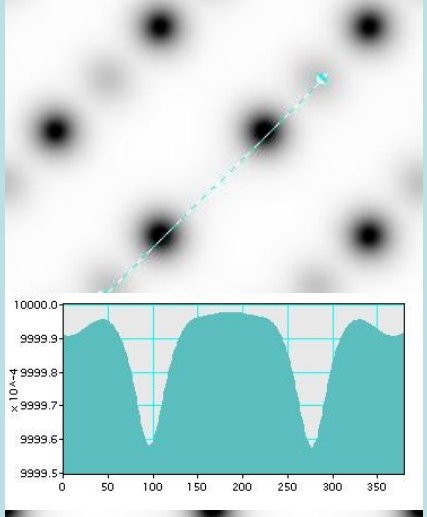
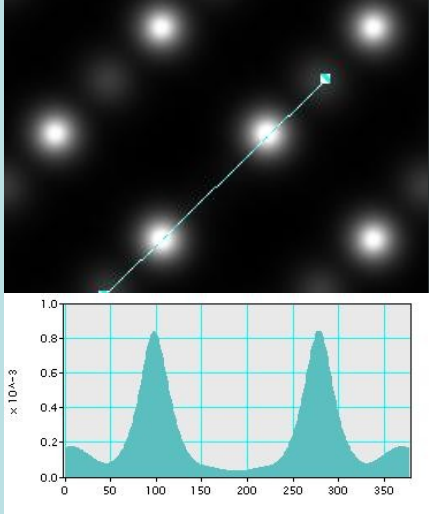






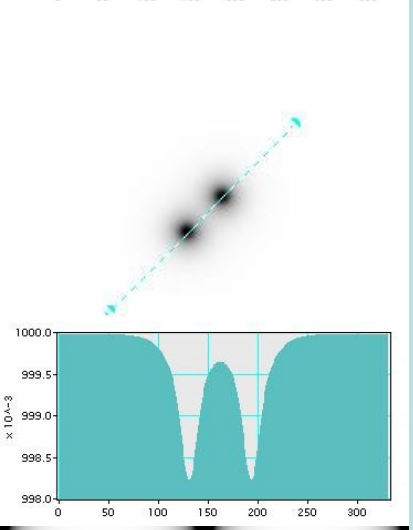
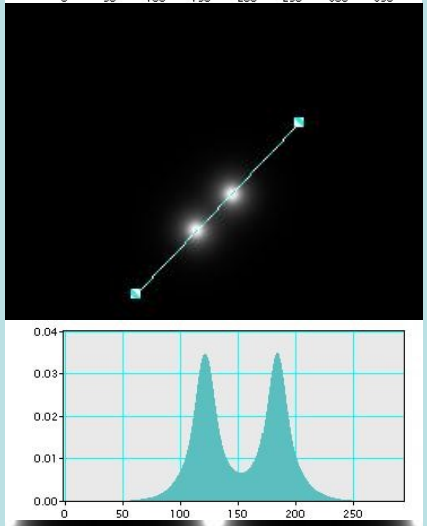


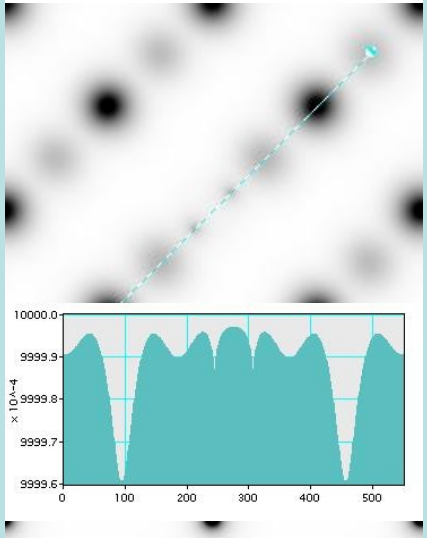
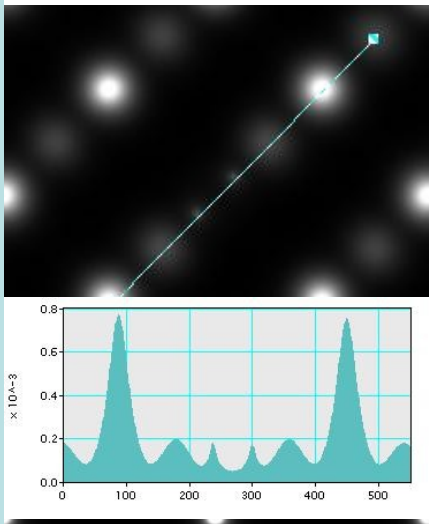



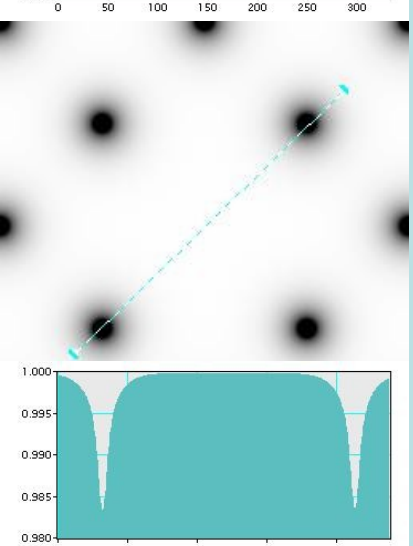
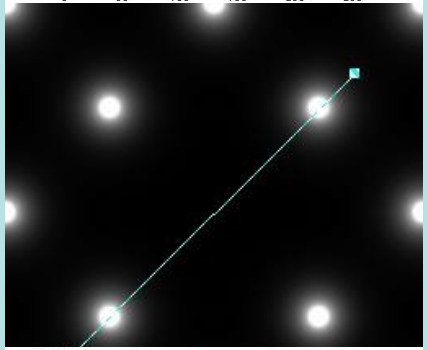
EMS **BOP**

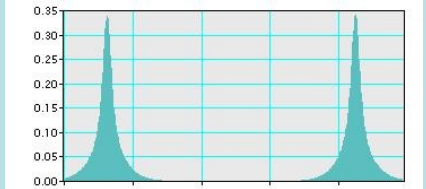
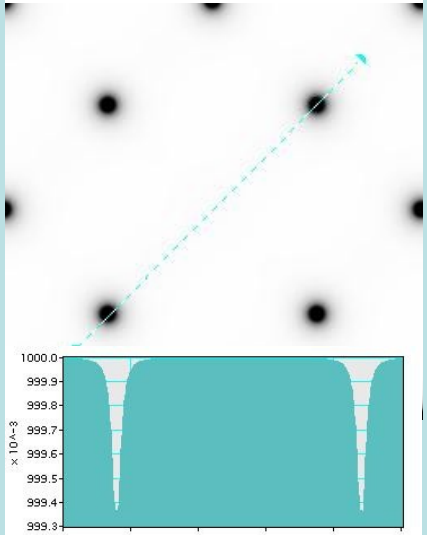
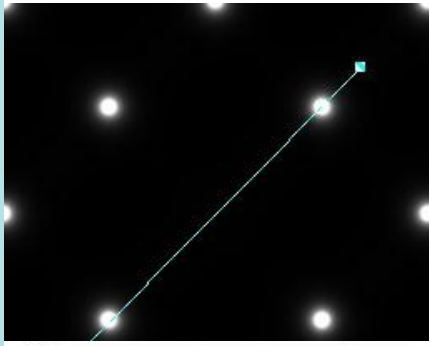
14/32 **14/32**

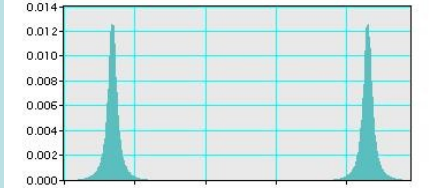



17/32 **15/32**



18/32 **16/32**

Comparison of scattering functions for sliced H4 in Si

multi-slice inversion
(van Dyck, Griblyuk, Lentzen,
Allen, Spargo, Koch)
Pade-inversion (Spence)
non-Convex sets (Spence)
local linearization

parameter
& potential

deviations from
reference structures:
displacement field (Head)
algebraic discretization

Inversion ?

no iteration
same ambiguities
additional instabilities

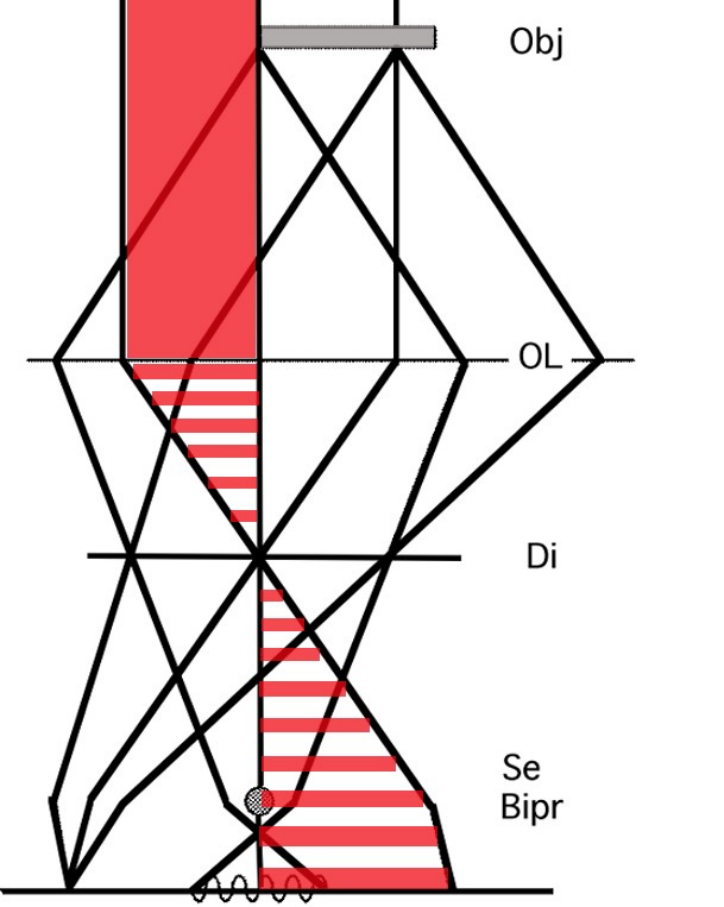
exit object
wave

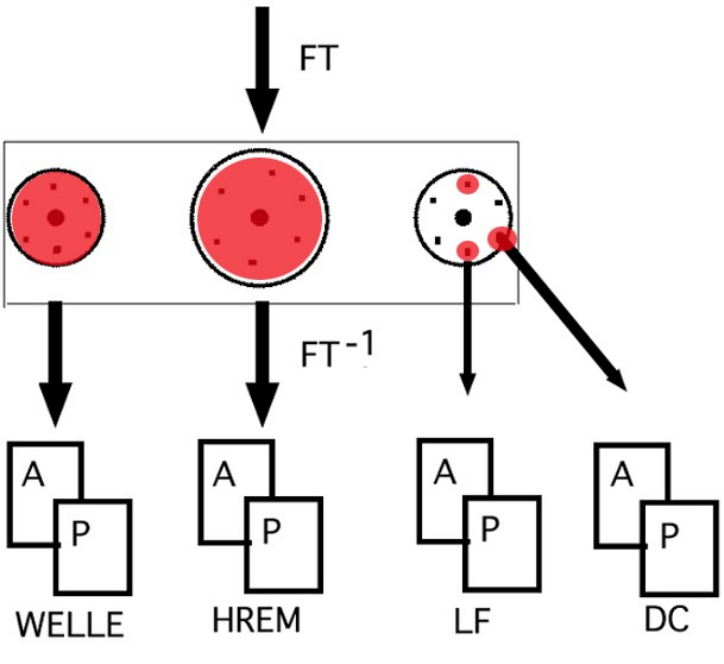
atomic
displacements

image

reference beam (holography)
defocus series (Kirkland, van Dyck …)
Gerchberg-Saxton (Jansson)
tilt-series, voltage variation

direct interpretation
by data reduction:
Fourier filtering
QUANTITEM
Fuzzy & Neuro-Net
Srain analysis

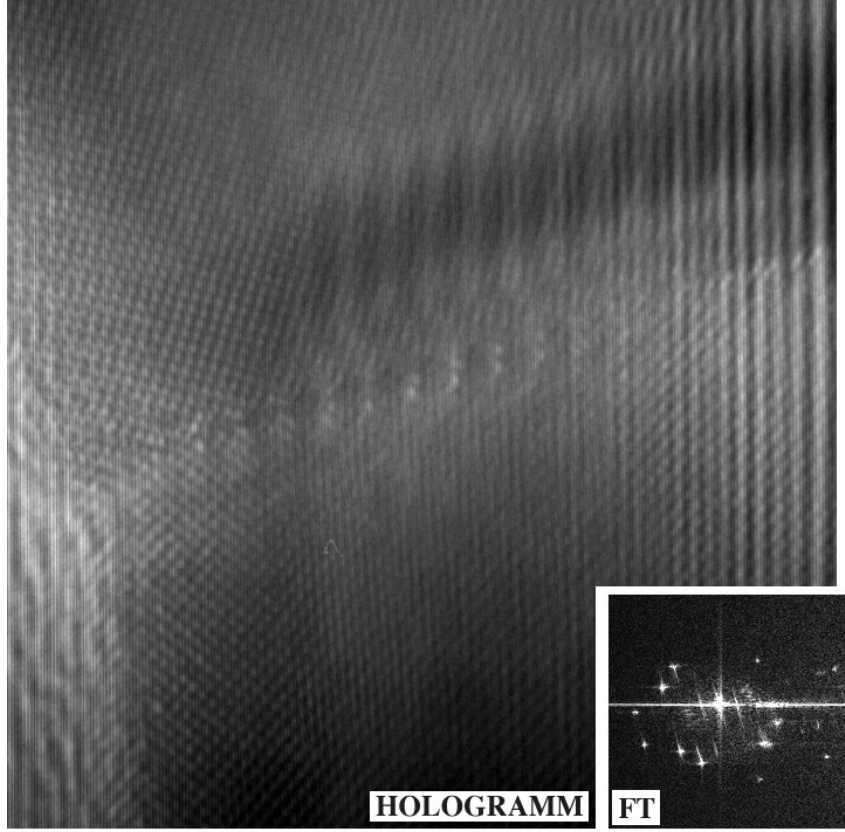


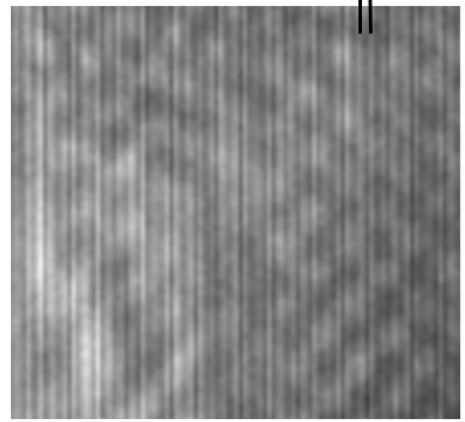


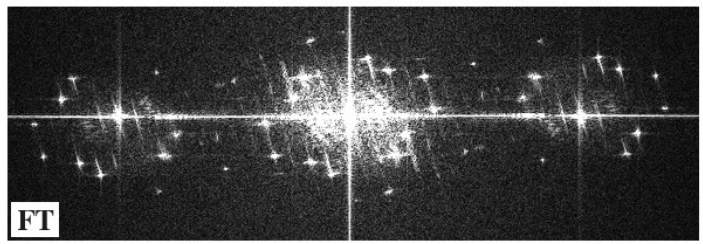
Sigma13-(001)-Au-Korngrenze

Präparation: R.Scholz, MPI Halle
Hologramm: A.Orchowski, Uni-Tübingen

300kV .05 nm

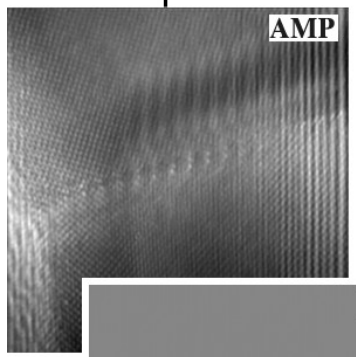




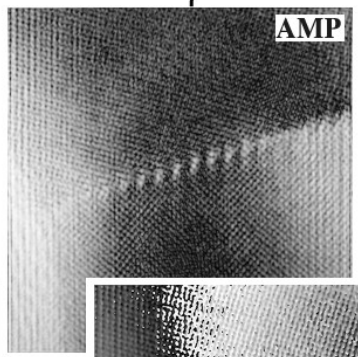


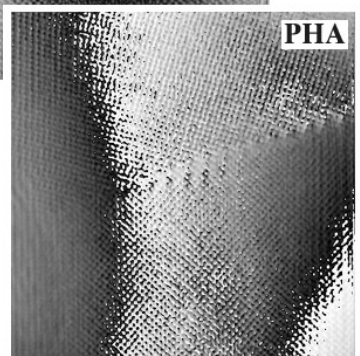
Autokorrelation | Seitenband | Reflexe

HREM

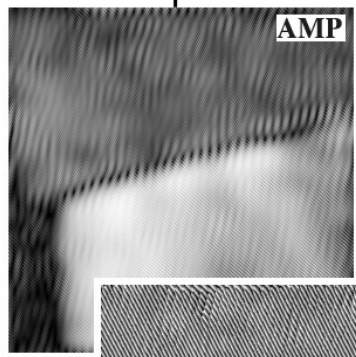


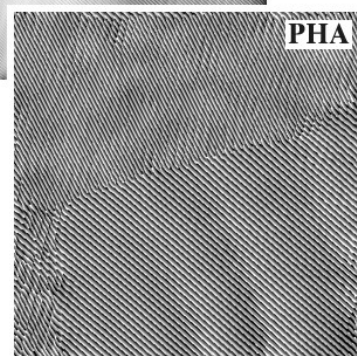
WELLE



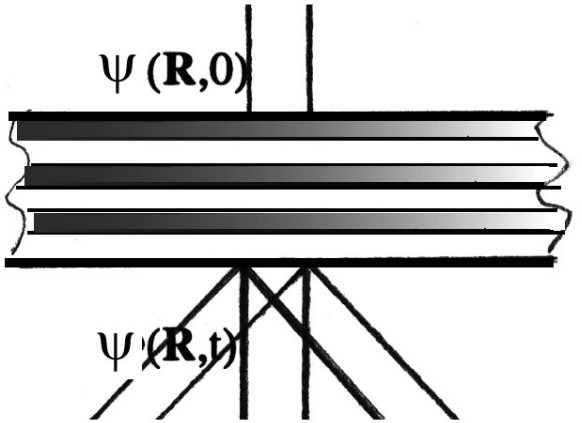


NL-LF





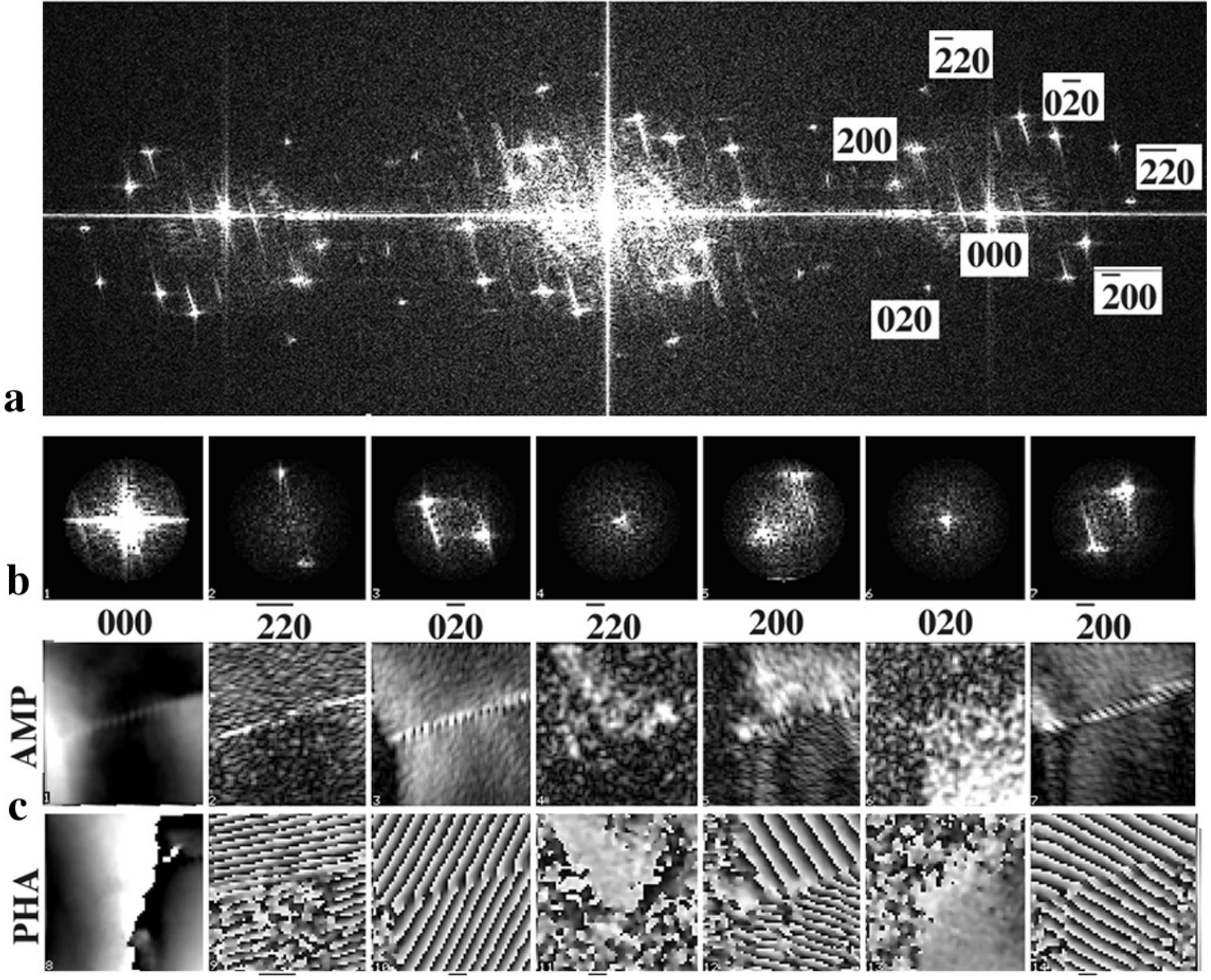
$$\psi = M(X)\,\psi_0$$

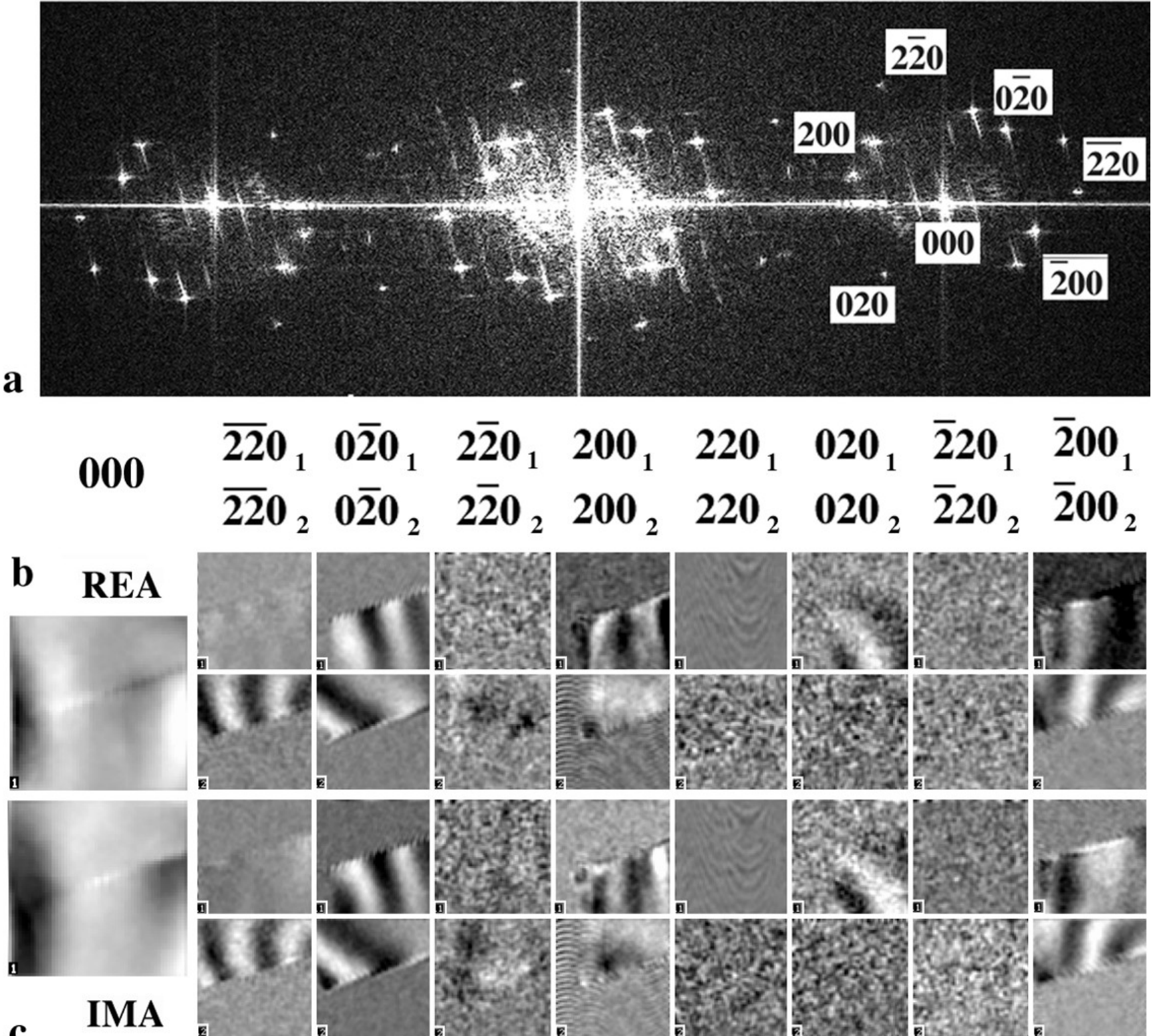


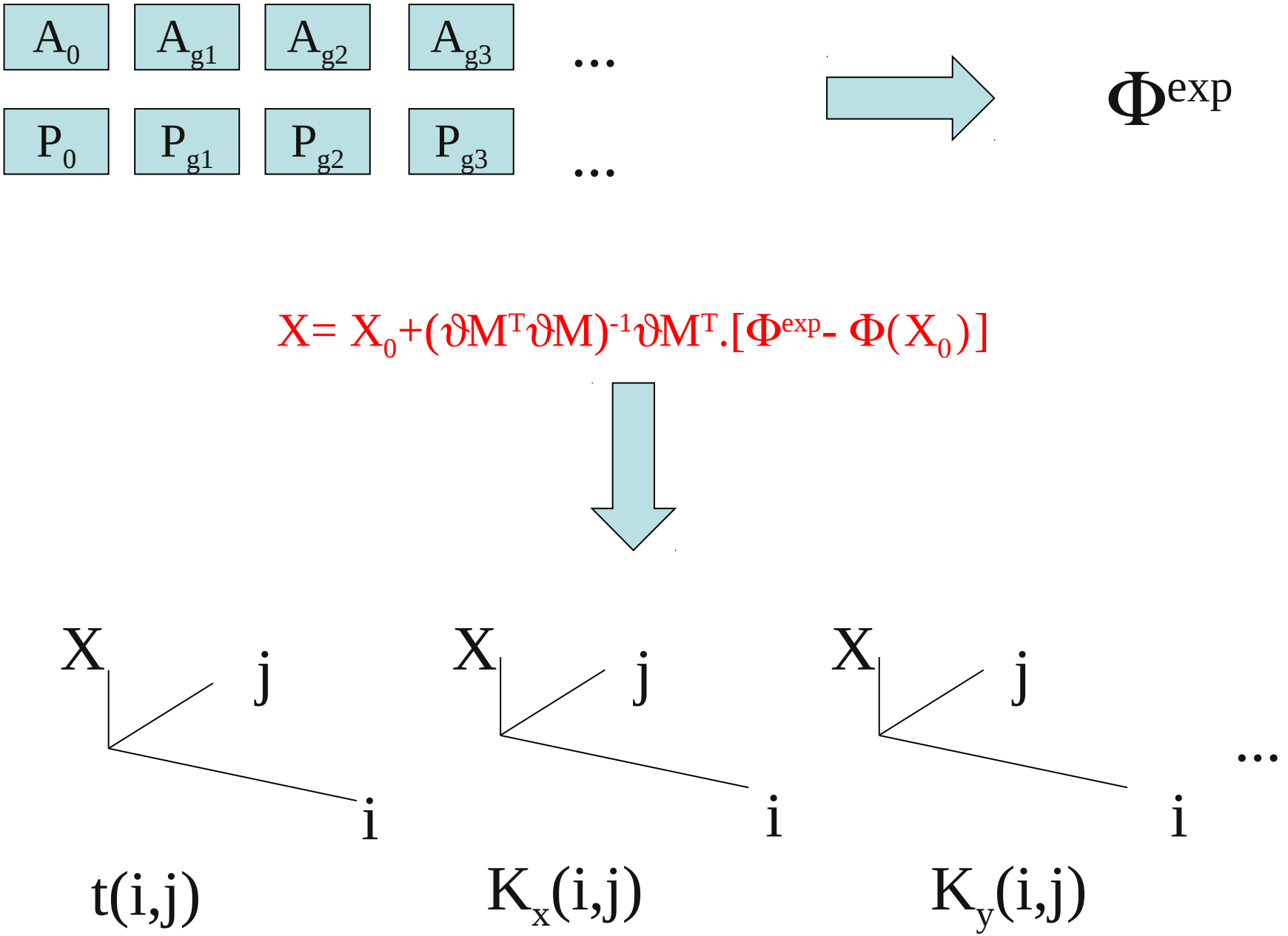
Assumptions:

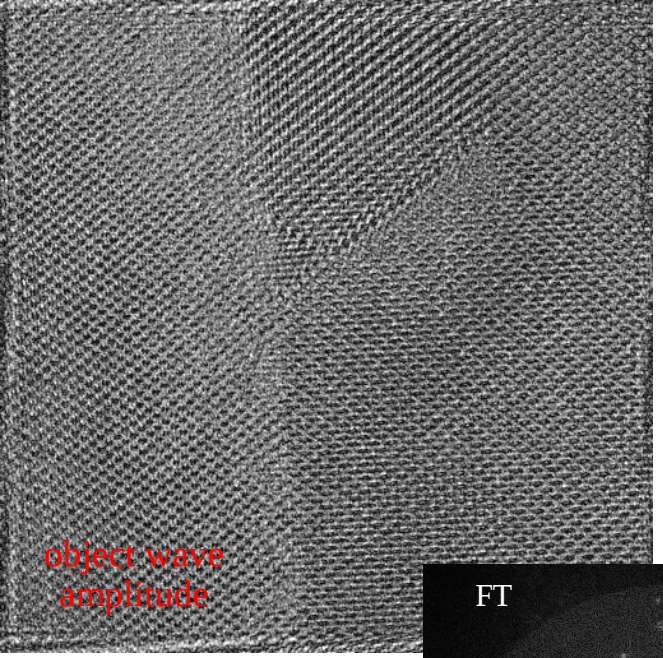
- object: weakly distorted crystal
- described by unknown parameter set $X=\{t, K, V_g, u\}$
- approximations of t_0, K_0 a priori known

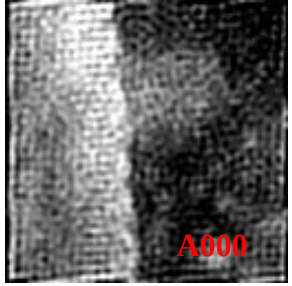
$$\psi = M(X_0)\,\psi_0 + \vartheta M(X_0)(X-X_0)\,\psi_0$$

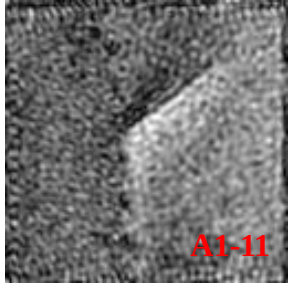

a
$\bar{2}20$
$0\bar{2}0$
200
$\bar{2}20$
000
$\bar{2}00$
020
b
000
$\bar{2}20$
$0\bar{2}0$
$\bar{2}20$
200
020
$\bar{2}00$
AMP
c
PHA

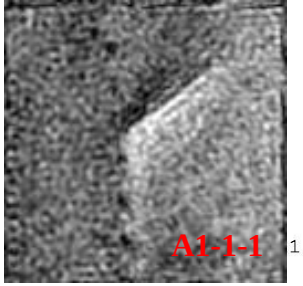

a
$2\bar{2}0$
$0\bar{2}0$
200
$\bar{2}20$
000
$\bar{2}00$
020
000
$\bar{2}\bar{2}0_1$ $0\bar{2}0_1$ $2\bar{2}0_1$ 200_1 220_1 020_1 $\bar{2}20_1$ $\bar{2}00_1$
$\bar{2}\bar{2}0_2$ $0\bar{2}0_2$ $2\bar{2}0_2$ 200_2 220_2 020_2 $\bar{2}20_2$ $\bar{2}00_2$
b
REA
IMA
c

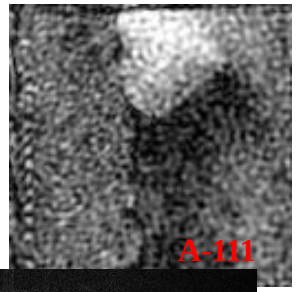

A_0
A_{g1}
A_{g2}
A_{g3}
...
P_0
P_{g1}
P_{g2}
P_{g3}
...
Φ^{exp}
$X = X_0 + (\vartheta M^T \vartheta M)^{-1} \vartheta M^T . [\Phi^{exp} - \Phi(X_0)]$
X
j
i
t(i,j)
X
j
i
$K_x(i,j)$
X
j
i
...
$K_y(i,j)$

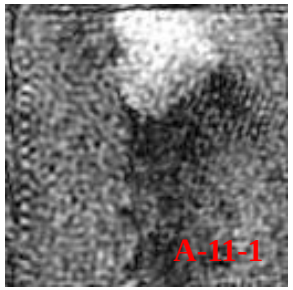


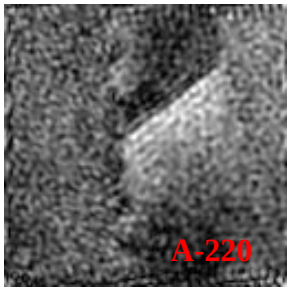


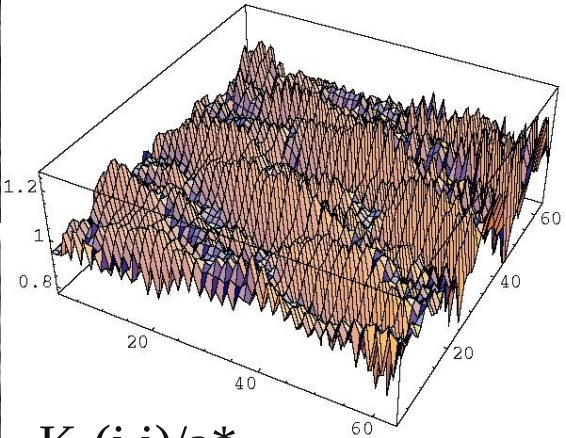




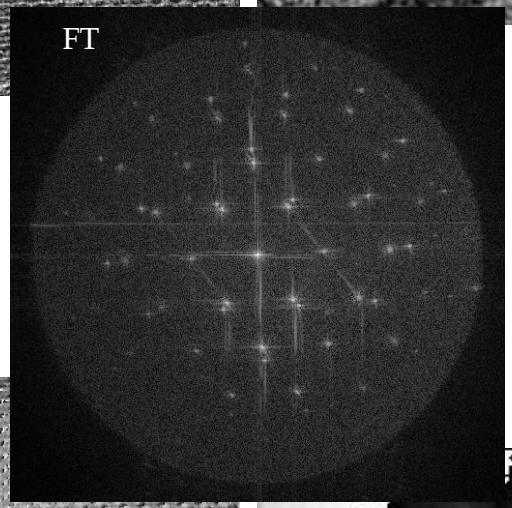








$K_x(i,j)/a^*$

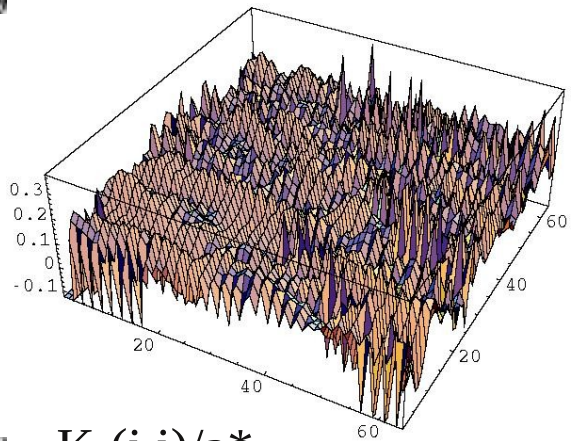


Ge-CdTe, 300kV
Sample: D. Smith
Holo: H. Lichte,
M.Lehmann

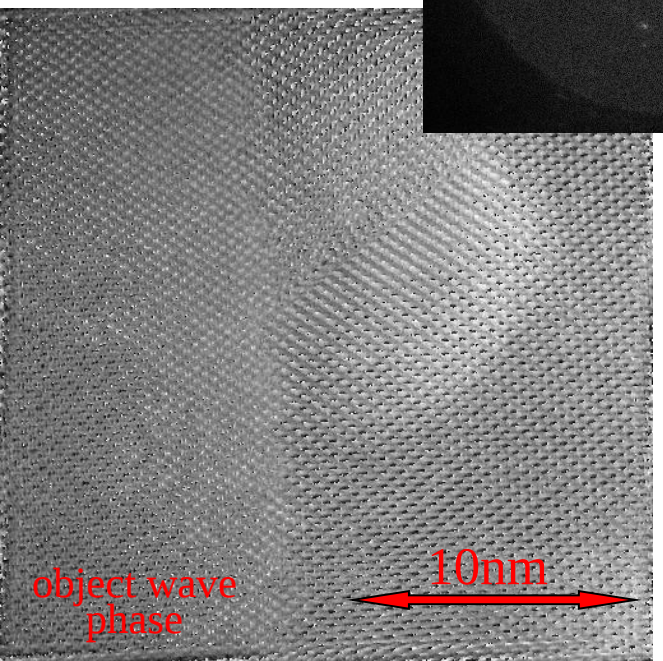
set 1: Ge
set 2: CdTe

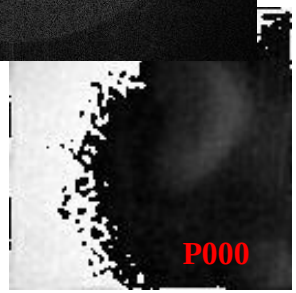
$dV_o/V_o = 0.02\%$

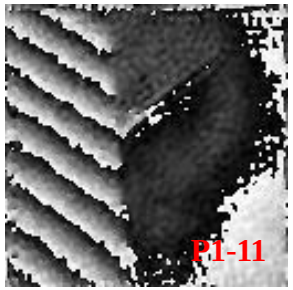
$dV'_o/V'_o = 0.8\%$

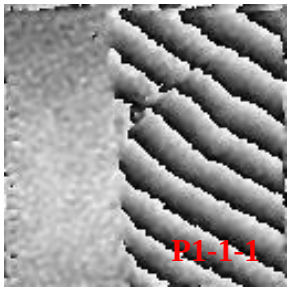


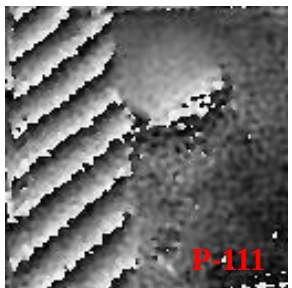
$K_y(i,j)/a^*$

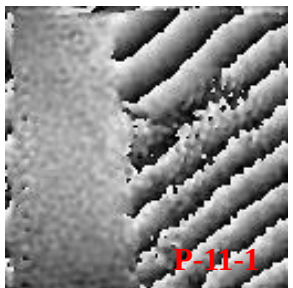


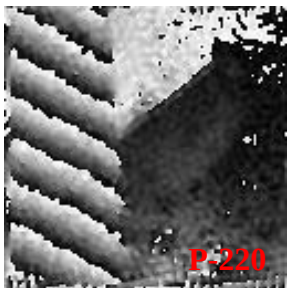


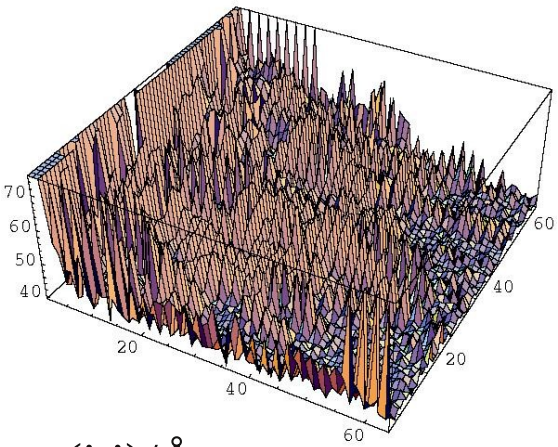












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