Computer Simulations (MD & EMS) For the Determination of the Imaging Conditions in the Study of *Solid-Liquid Interfaces* by HRTEM

Y. Kauffmann
W.D. Kaplan

Technion
Israel Institute of Technology
Department of Materials Engineering

Introduction:

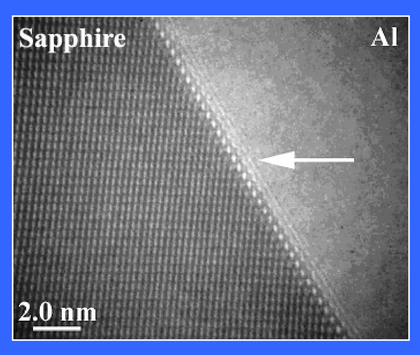
Understanding the nature of ***solid-liquid interfaces*** is very important for many **processes of technological interest**, such as:

- Solidification
- Epitaxial growth
- Wetting
- Liquid phase joining
- Crystal growth
- Lubrication

Characterization of the atomistic structure of an interface, and in particular interfaces between solids and liquids, **is a very challenging experimental problem.**

Molecular dynamics simulations [1] showed the presence of a **layering effect** in the liquid induced by the presence of a crystal. In some cases even **in-plane ordering** was observed.

The limited number of studies done by **HRTEM** [2,3] also point to the existence of a layering effect at the solid-liquid interface.



HRTEM micrograph of liquid Al in contact with sapphire. The contrast perturbations in the liquid, immediately adjacent and parallel to the (0001) sapphire surface, indicate ordering within the liquid Al. The contrast perturbations are not an artifact, and exist over a range of objective lens defocus .Data acquired by C. Scheu on the MPI ARM, at 750°C.

The main problem with direct HRTEM investigations: ***The imaging conditions have a significant influence on the contrast in the image, and may lead to inaccurate conclusions !!!***

The main controllable parameters that may affect the resulting lattice image acquired by HRTEM are:

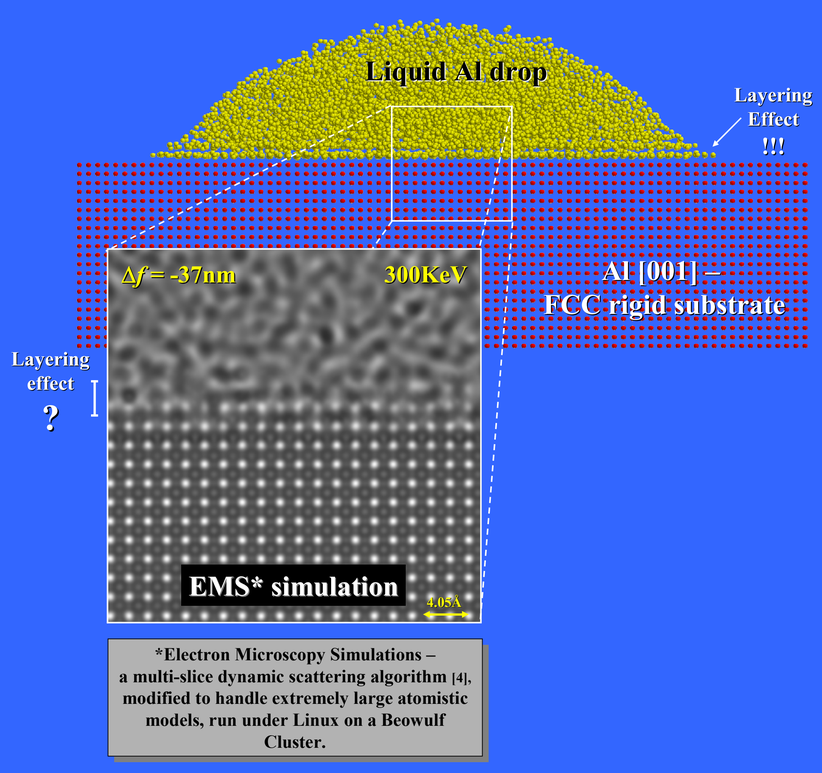
- Objective lens defocus (Δf).
- Astigmatism.
- The size of the objective aperture.
- Interface inclination.
- Z.A. deviation.

The aim of this work:

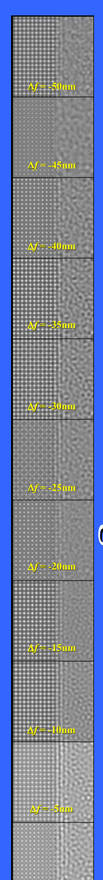
Define, by means of **computerized HRTEM image simulations (EMS)** combined with **molecular dynamics (MD) simulations,** the exact imaging conditions that will ***enable accurate analysis of the layering effect occurring at solid-liquid interfaces.***

The Simulated Specimens:

Simulated using molecular dynamics (EAM)

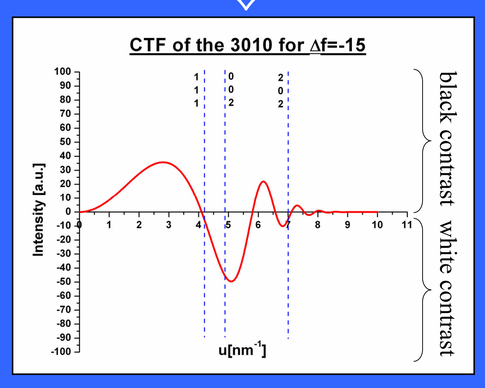


*Electron Microscopy Simulations – a multi-slice dynamic scattering algorithm [4], modified to handle extremely large atomistic models, run under Linux on a Beowulf Cluster.



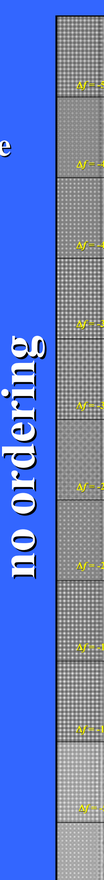
Best Defocus Determination:

If white spots = column positions ⇒ the image intensity is directly interpretable

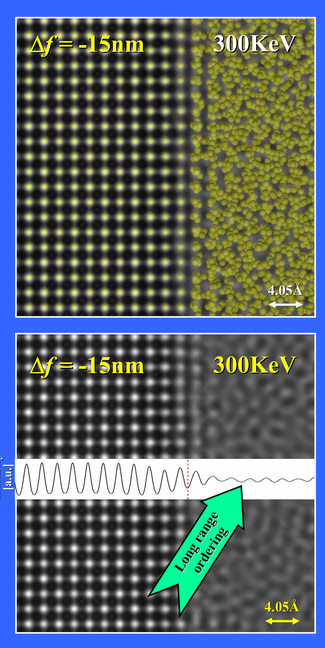


The intensities corresponding to the periodicity of the (002) planes are white.

best Δf = -15nm

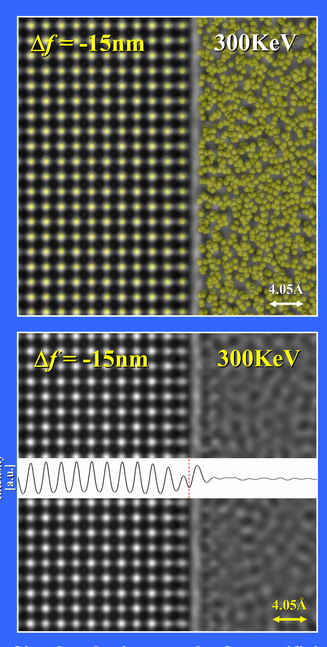


Ordering



Simulated micrograph of an interface with ordering, from MD simulations.

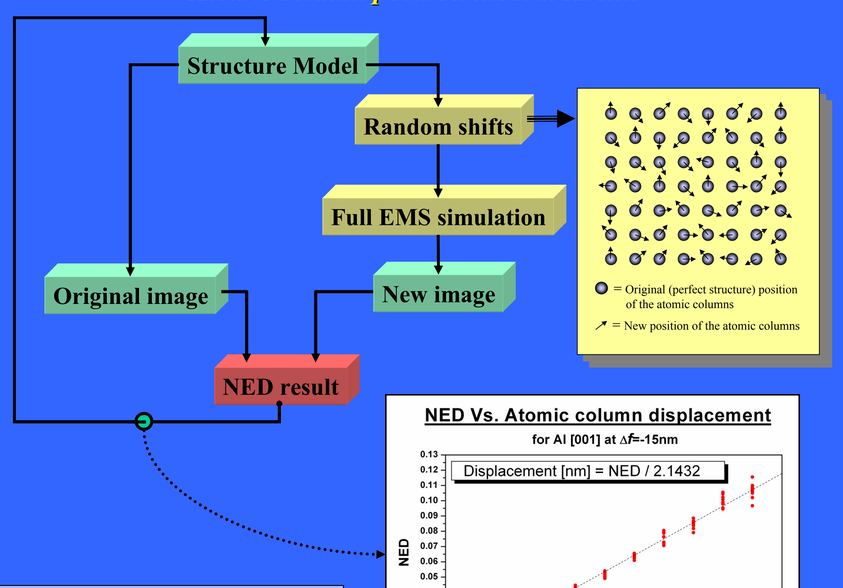
No ordering



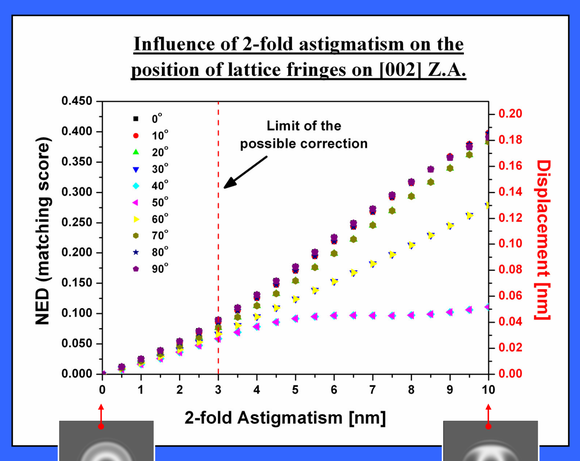
Simulated micrograph of an artificial interface with no ordering, produced by joining bulk liquid Al (MD) to crystalline Al.

Atomic Column Displacement Simulation

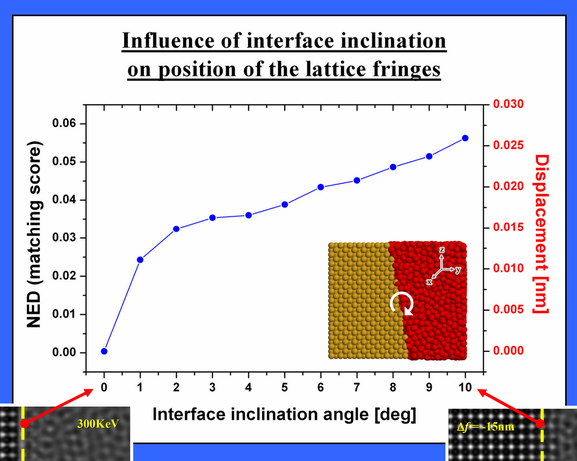
Translation of a metric (NED) to the precision of Atomic column position measurement.



$$NED(\text{image A, image B}) = \frac{\sqrt{\sum_{i,j=0}^{N}(a_{ij} - b_{ij})^2}}{\sqrt{\sum_{i,j=0}^{N} a_{ij}^2 \sum_{i,j=0}^{N} b_{ij}^2}}$$

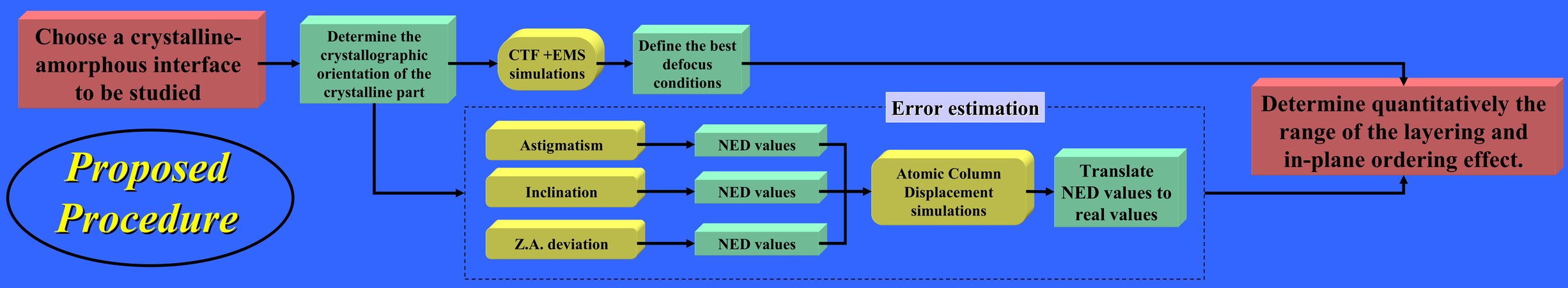


Very significant for large astigmatism !!!



Very significant for large inclination angles !!!

Proposed Procedure



Conclusions

- Scherzer defocus is not always the best option for the investigation of solid-liquid interfaces.
- Optimum defocus = f (microscope used & Z.A. of the observed crystalline zone).
- CTF of the microscope + EMS simulations of the specific crystalline orientation = BEST DEFOCUS.
- reasonable 2-fold Astigmatism and interface inclination may lead to errors of a few tenths of an Angstrom in the determination of the ordering effect range.

The Future

- Apply the procedure on real systems, such as equilibrium amorphous films, high temperature Cu-Al_2O_3 or Ni-Al_2O_3 interfaces.
- Define a procedure to quantify the in-plane ordering.

References:

1. A. Hashibon, J. Adler, M. Finnis and W.D. Kaplan, Computational Materials Science, 24:443, 2002.
2. W. J. Ma and J. R. Banavaf, J. of Chemical Physics, 97[l]:485, 1992.
3. J.M. Howe, Philosophical Magazine A, 74[3]:761, 1996.
4. P.A. Stadelmann, Ultramicroscopy, 21:131, 1987.

Acknowledgments:

- Dr. Adham Hashibon – MD simulations.
- Amir Avishai & Hila Sadan – Vibrant discussions.