Impact of Molecular Dynamics (MD) Simulations in Materials Research

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Contributions from D. K. Ward and M. E. Foster are greatly appreciated

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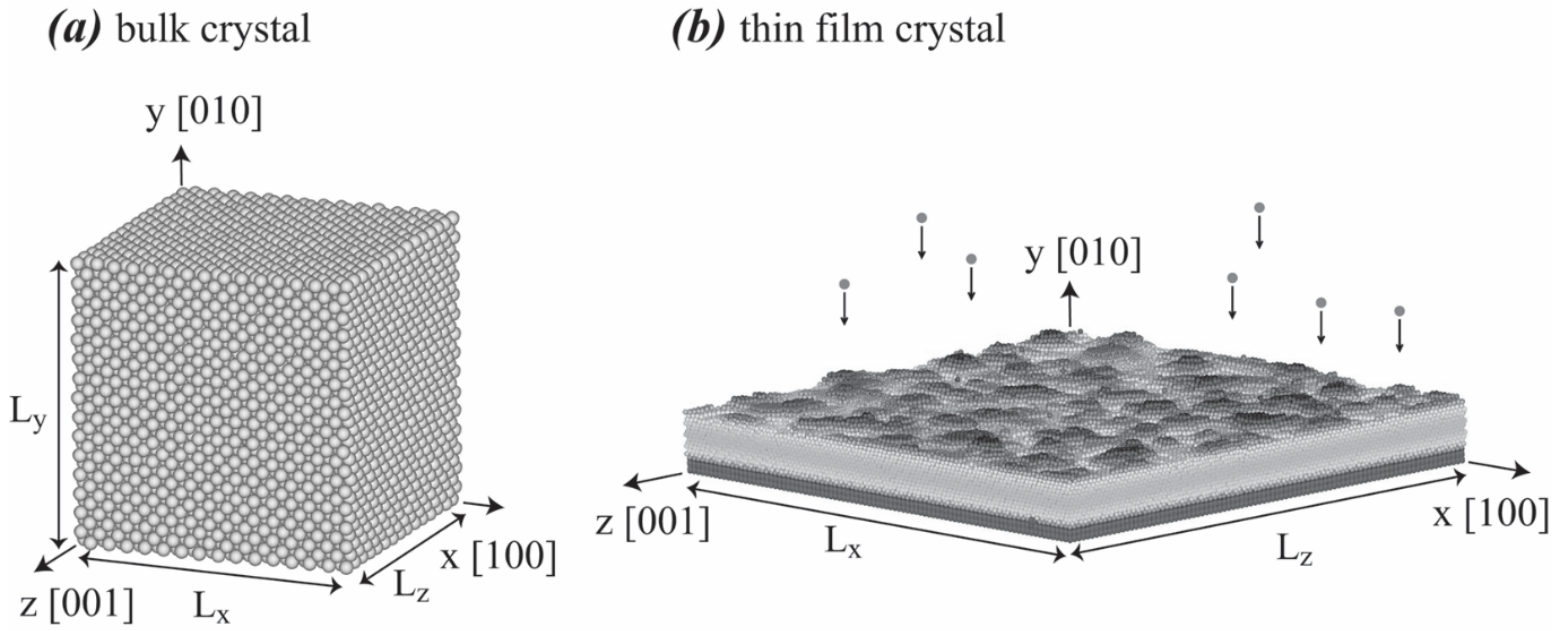
Outline

- Molecular dynamics simulation methods
- Graphene
- Giant magnetoresistive multilayers
- CdTe/CdS solar cell films
- $In_xGa_{1-x}N$/GaN solid state lighting films
- Misfit dislocation theory
- Al-Cu bond order potential
- $2H \Rightarrow H_2$ chemical reaction
- CdZnTe radiation detection materials

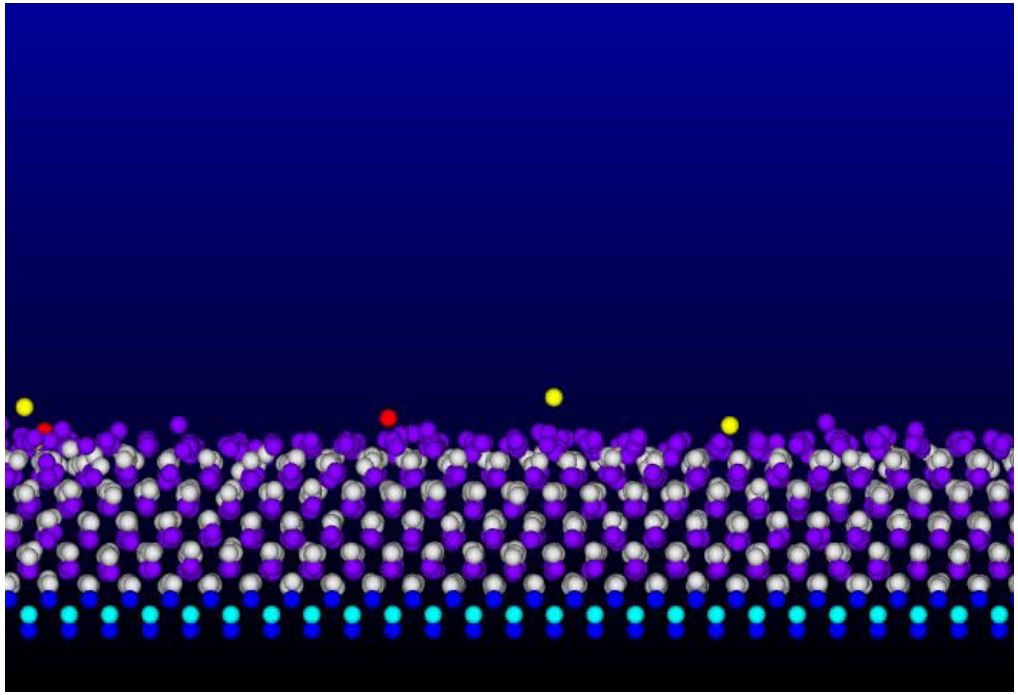
MD Simulation Methods



- Positions of atoms as a function of time from Newton's equations
- Thermal transport and phonon vibration calculations even without defects
- Computational mechanical testing simulations (elastic constants, strength)
- Defect properties (diffusion of point defects, dislocation energies, etc.)
- Structure from synthesis without any artificial assumptions

MD Growth Simulations

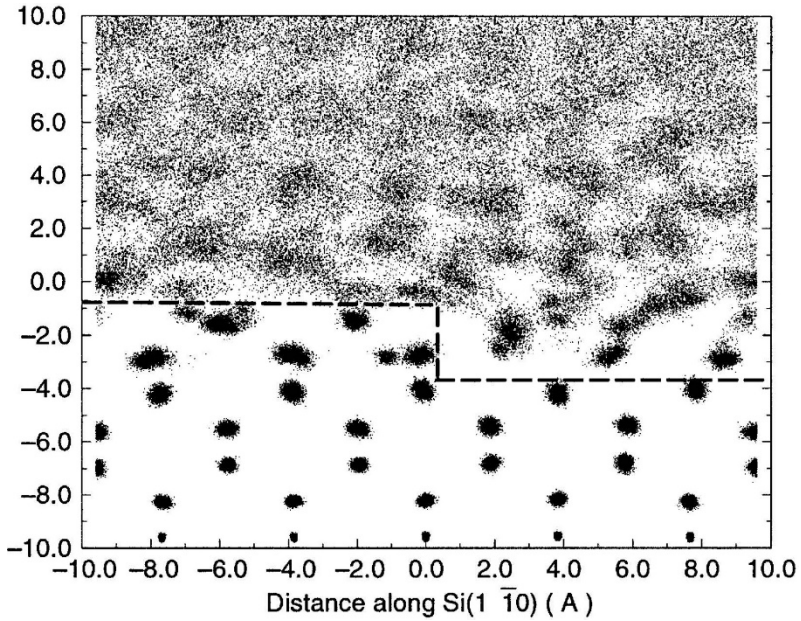
(CdTe on CdSe)



- Modern technologies rely on electronic devices based on thin film growth
- InGaN/GaN solid state lighting devices are currently limited by lattice mismatch defects
- CdTe/CdS photovoltaic devices are also limited by lattice mismatch defects
- These defects cannot be easily understood from experiments alone
- Growth simulations can reveal defect formation without any artificial assumptions
- Can rigorously test the model

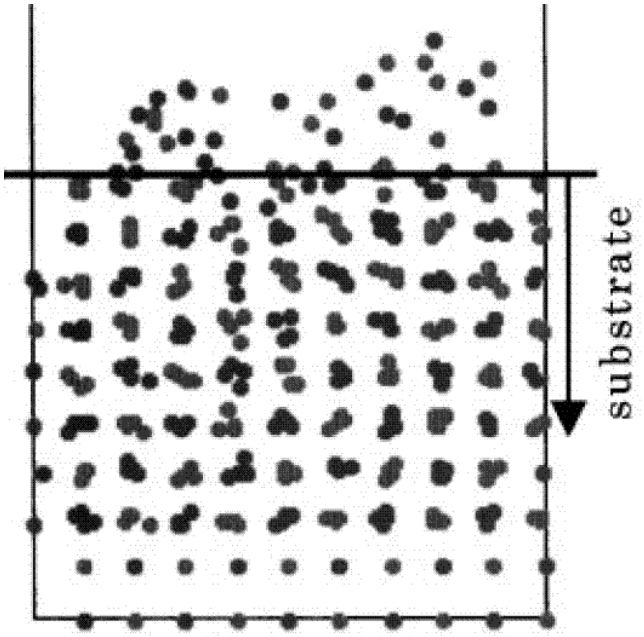
Challenges: Interatomic Potentials

CdTe on Si



J. Oh, and C. H. Grein, J. Cryst. Growth, 193, 241 (1998)

InAs on GaAs

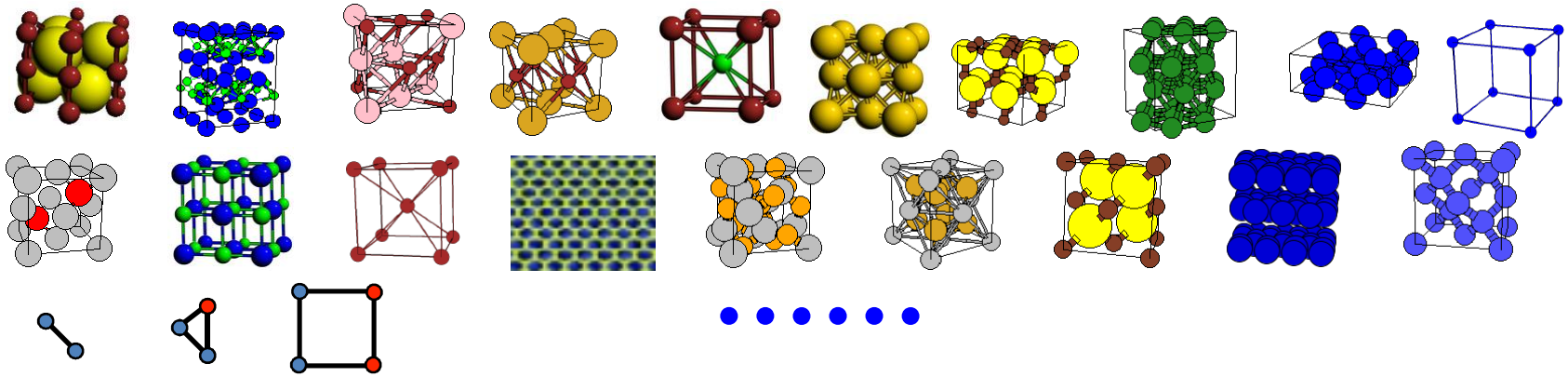


M. Nakamura, H. Fujioka, K. Ono, M. Takeuchi, T. Mitsui, and M. Oshima, J. Cryst. Growth, 209, 232(2000)

Because amorphous phase is the issue, fitting finite crystalline phases may never solve the problem

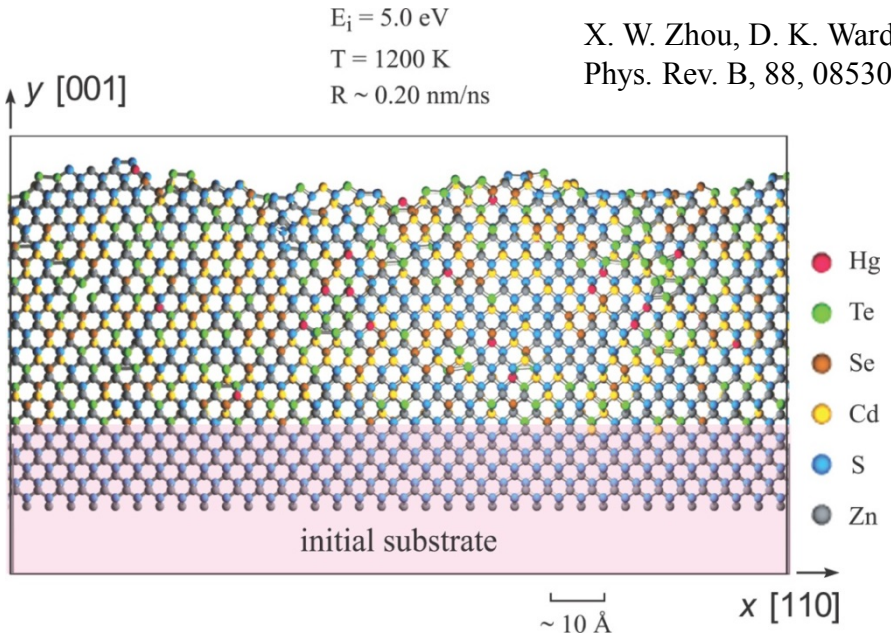
1. J. Tersoff, Phys. Rev. B, 39, 5566(1989) – for Si (amorphous growth, but can re-crystallize at 2200 K through bulk transformation)
2. P. A. Ashu, J. H. Jefferson, A. G. Cullis, W. E. Hagston, and C. R. Whitehouse, J. Cryst. Growth, 150, 176(1995). – for GaAs
3. R. Smith, Nucl. Instru. Meth. B, 67, 335(1992). – for GaAs

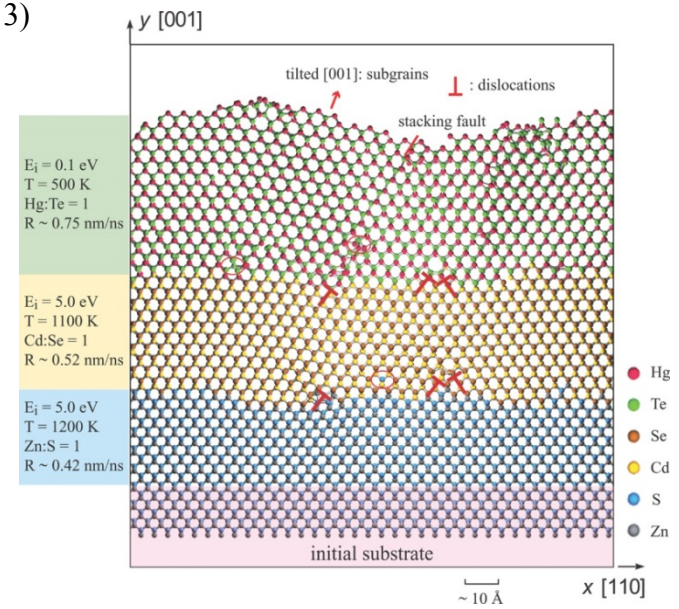
Towards Growth Simulation Enabling Potentials



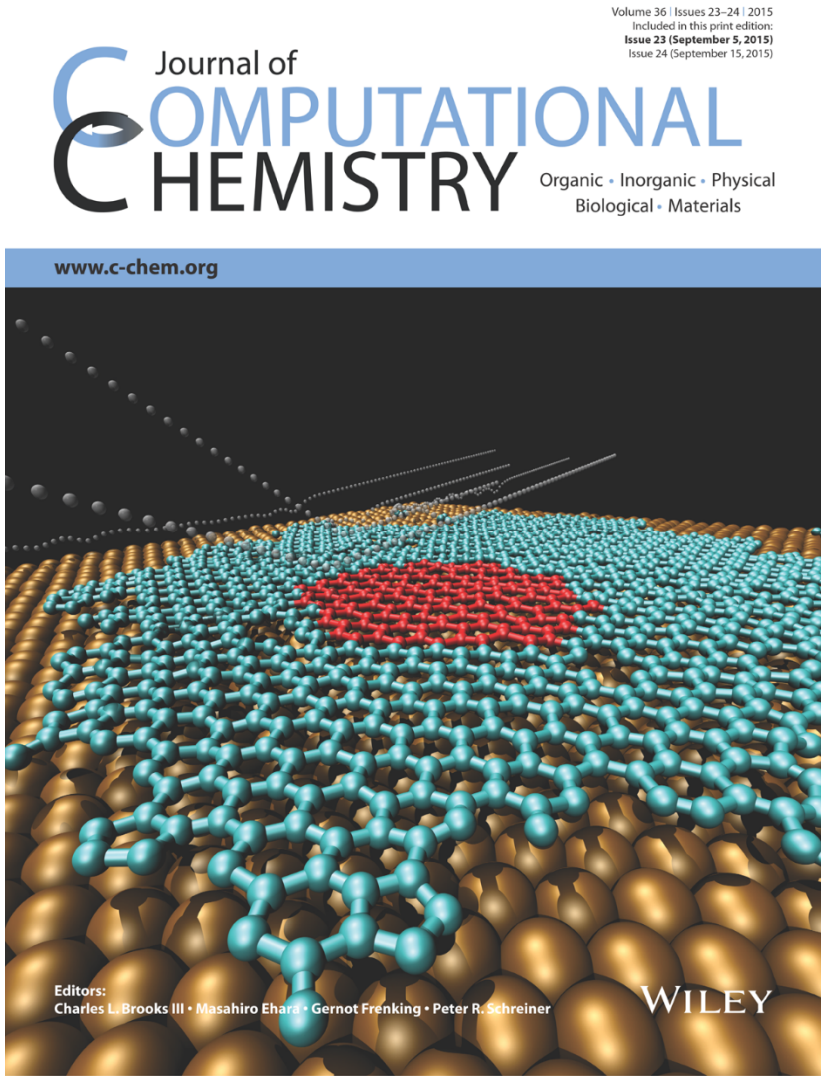
A Zn-Cd-Hg-S-Se-Te Potential we developed

X. W. Zhou, D. K. Ward, J. E. Martin, F. B. van Swol, J. L. Cruz-Campa, and D. Zubia, Phys. Rev. B, 88, 085309 (2013)

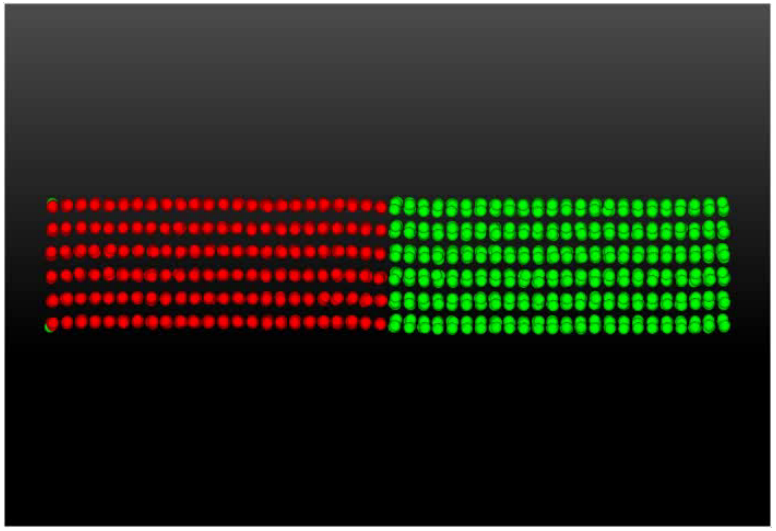




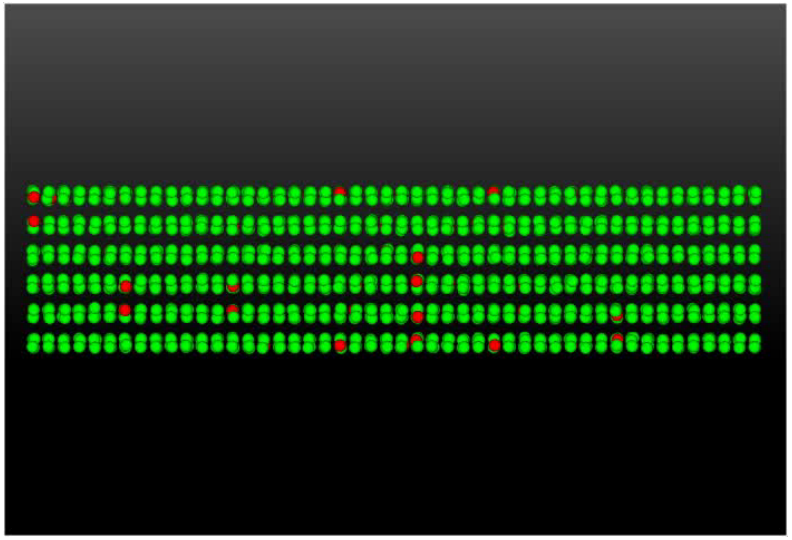
We Developed a Bond Order Potential to Enable Carbon Simulations



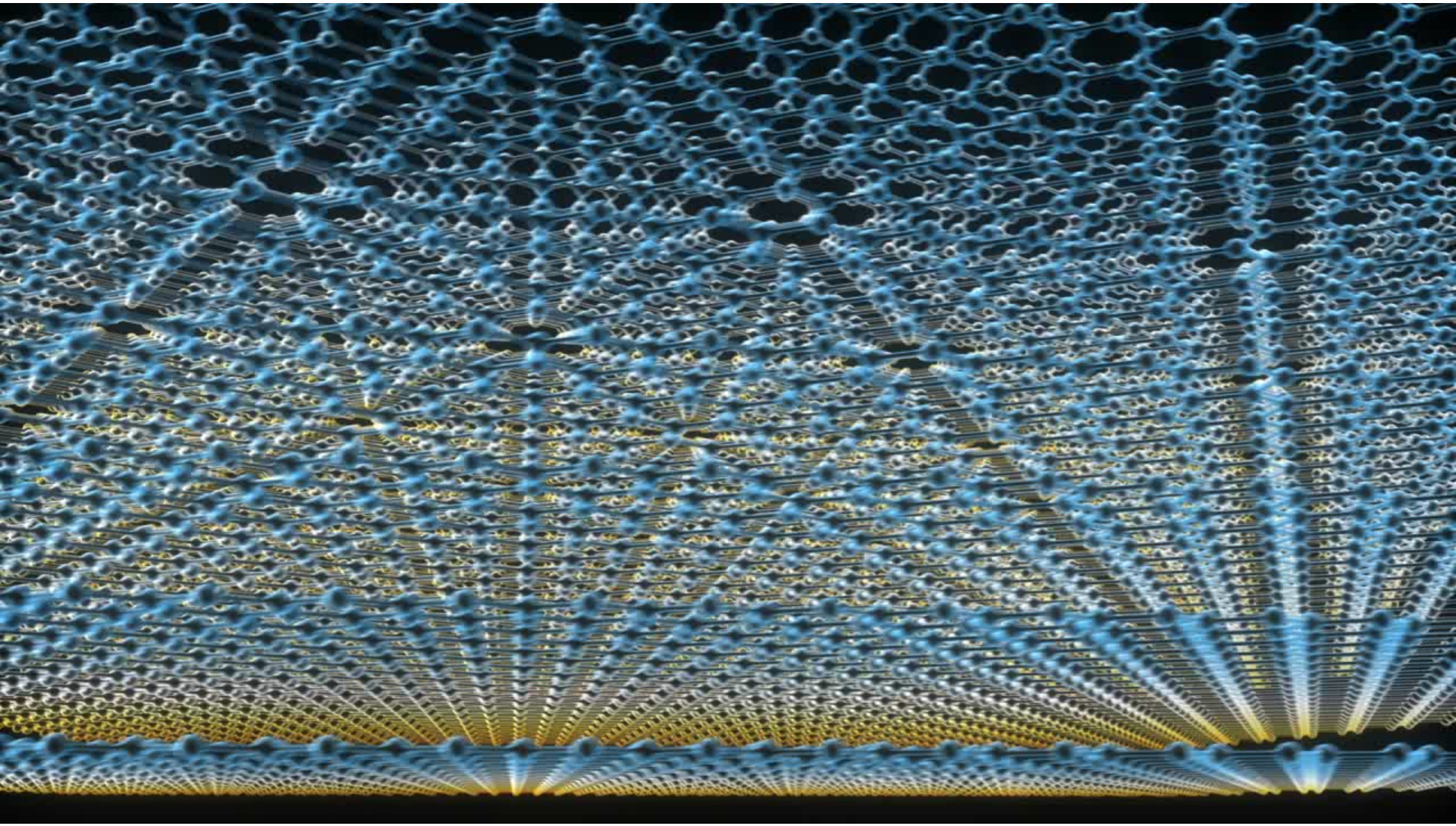
T = 2000 K, P = 0.6 Mbars (compressive)



T = 1600 K, P = -0.6 Mbars (tensile)

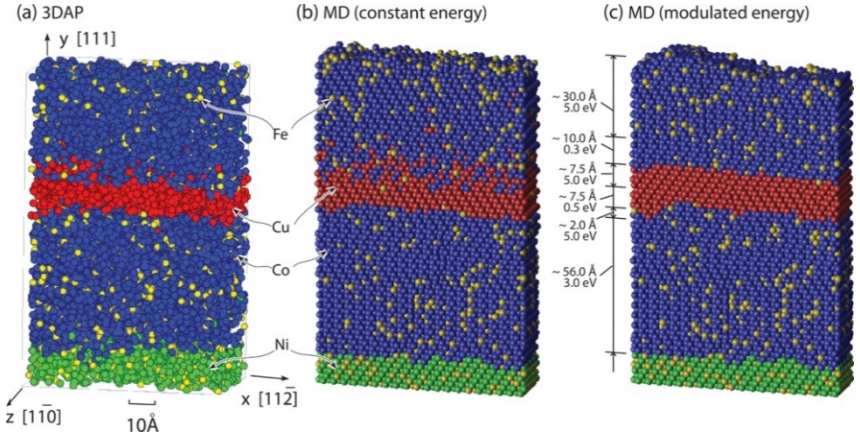


Graphene Growth Simulation

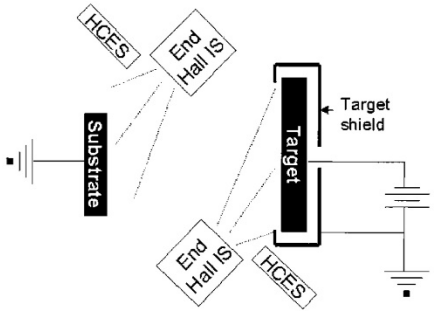


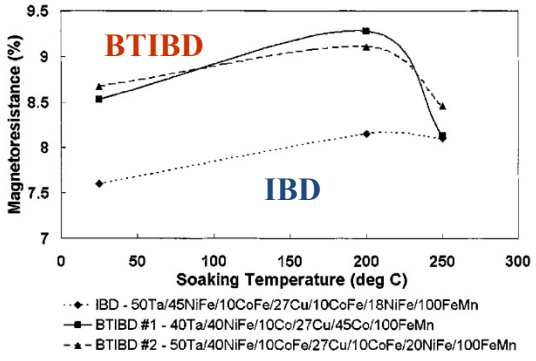
Real Word Impacts: Giant Magnetoresistive Multilayers

MD prediction of improved synthesis
(giant magnetoresistive spin valve)



MD leads to a new BTIBD growth method
(by CVC, Inc.)





- **X. W. Zhou et al, Acta Mater., 49, 4005 (2001)**
- **X. W. Zhou et al, J. Appl. Phys., 84, 2301 (1998)**
- **T. L. Hylton et al, IEEE Trans. Mag., 36, 2966 (2000)**

High-fidelity simulations enable us to:

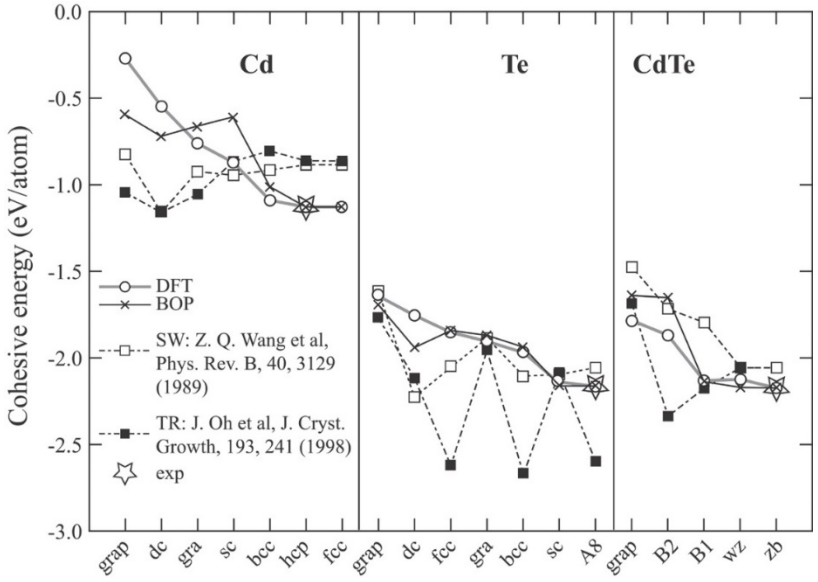
- ❑ Visualize strain relaxation defect formation in situ
- ❑ Thoroughly explore growth conditions
- ❑ Brainstorm novel synthesis (e.g., intra layer modulation of conditions)
- ❑ Design of defect-free nanostructures

Solar Cell Example: CdTe/CdS

- CdTe/CdS solar cells have the low cost compared to other photovoltaic technologies, defect reduction further reduces cost
- Misfit dislocations are one of the primary defects
- BOP-based MD models have predicted dislocation-free CdTe/CdS solar cell structures

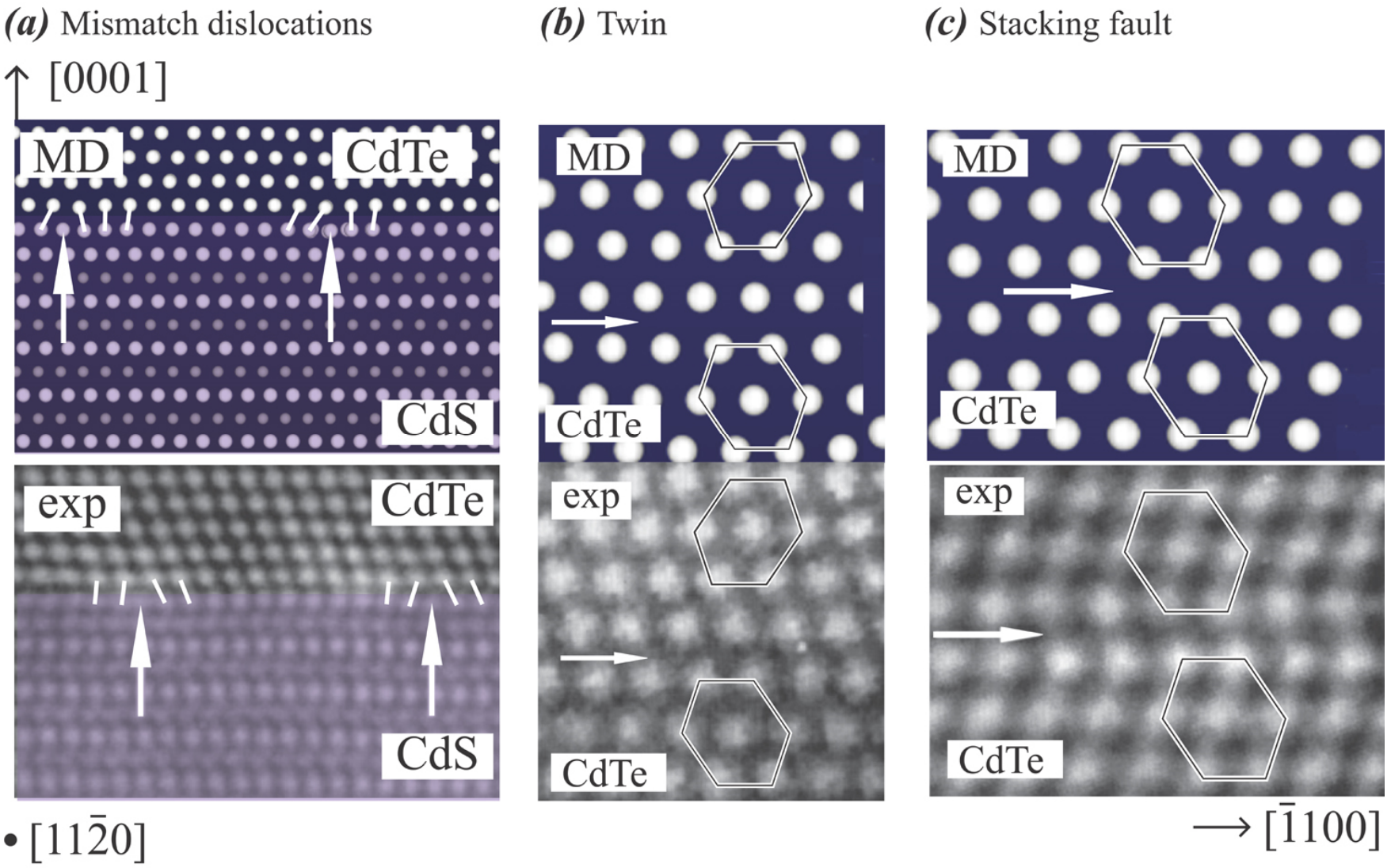
CdTe/CdS solar cells widely used

CdTe bond order potential captures DFT energy trends



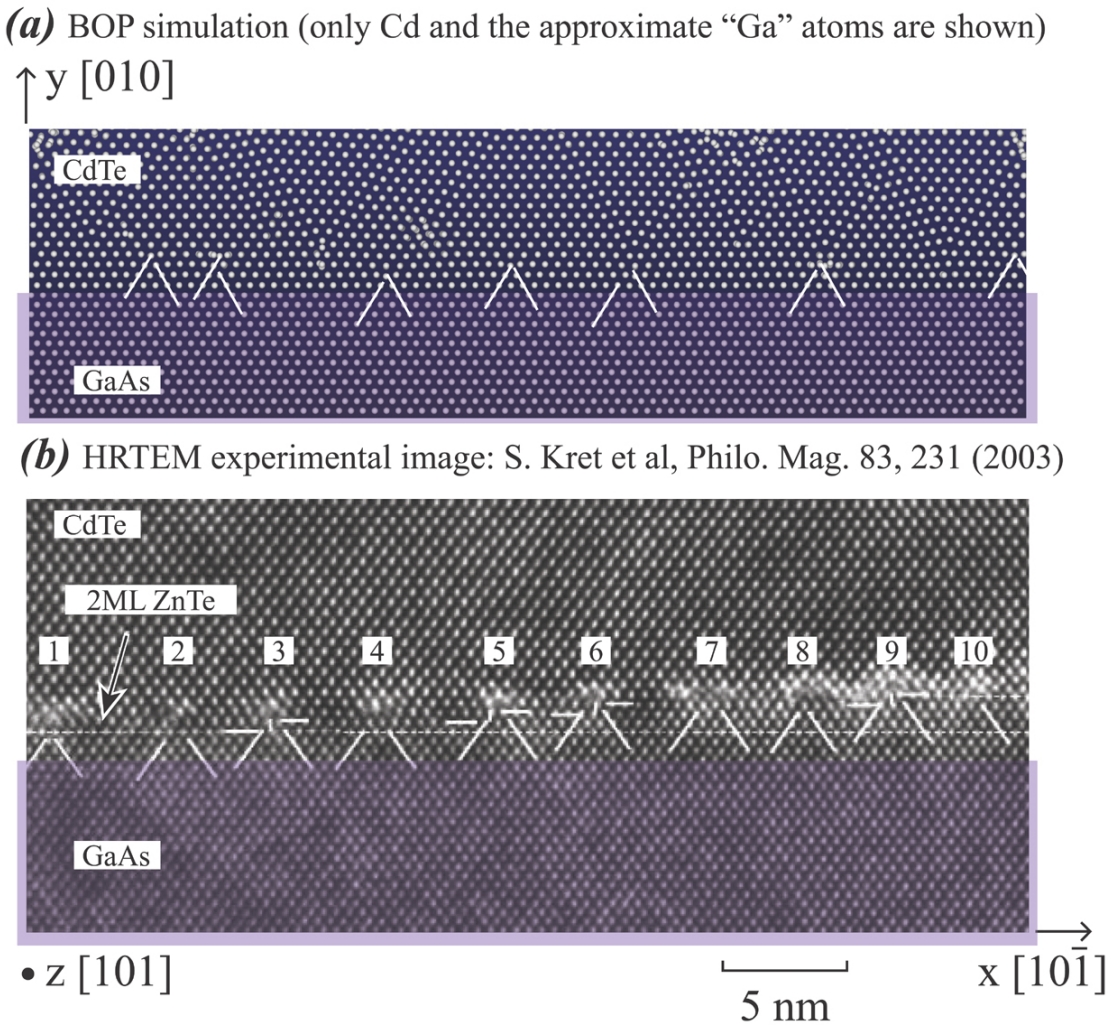
X. W. Zhou et al, Prog. Photovoltaics., 23, 1837 (2015)

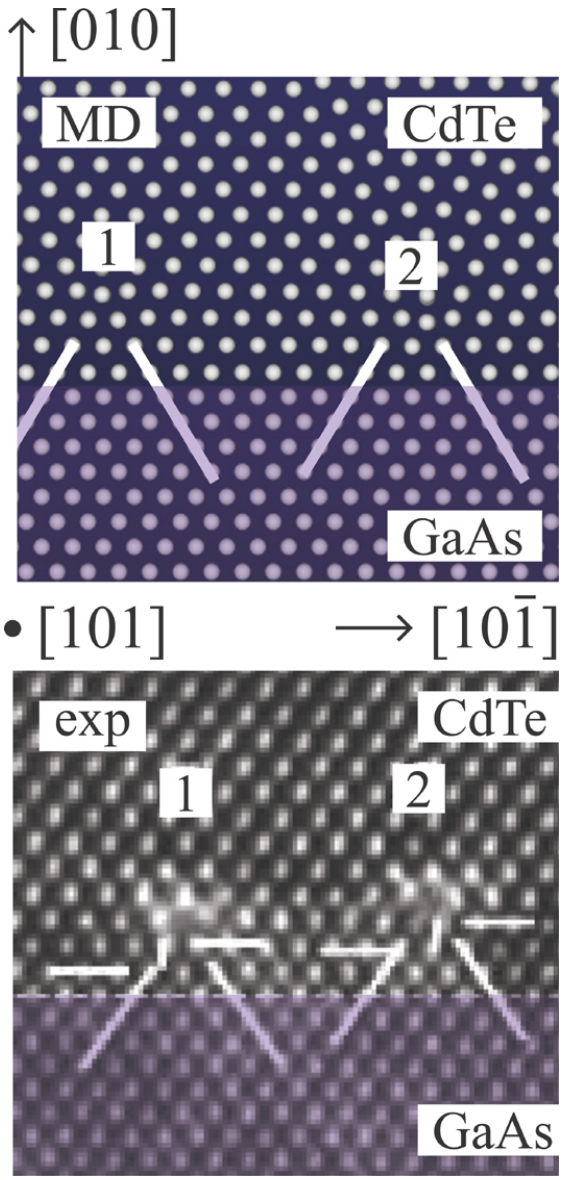
MD Validation I: CdTe/CdS Defects



HRTEM from Y. Yan, R. G. Dhere, K. M. Jones, and M. M. Al-Jassim, J. Appl. Phys. 89, 5844 (2001)

MD Validation II: CdTe/GaAs Defects

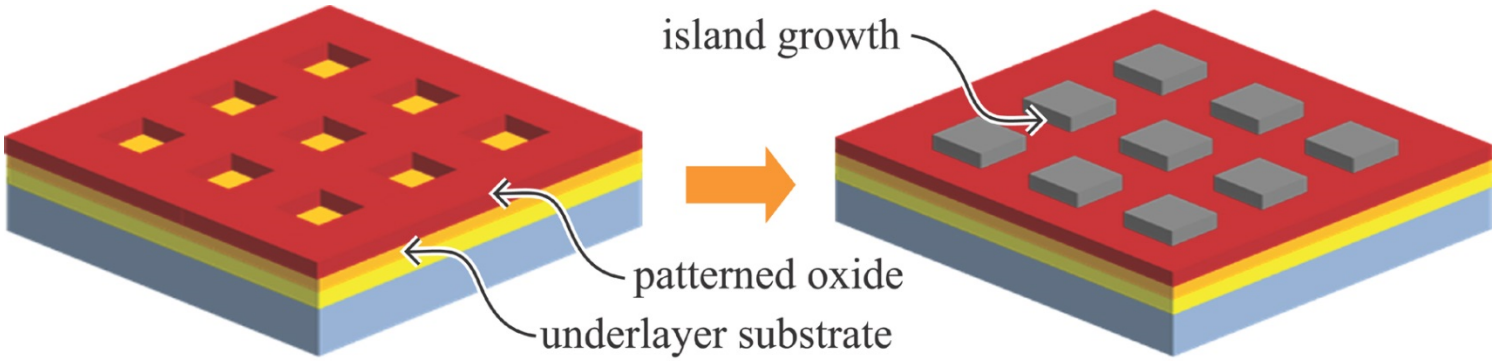




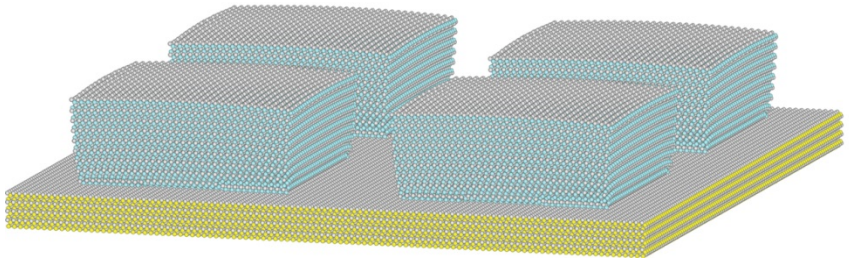
J. J. Chavez, D. K. Ward, B. M. Wong, F. P. Doty, J. L. Cruz-Campa, G. N. Nielson, V. P. Gupta, D. Zubia, J. McClure, and X. W. Zhou, Phys. Rev. B, 85, 245316 (2012)

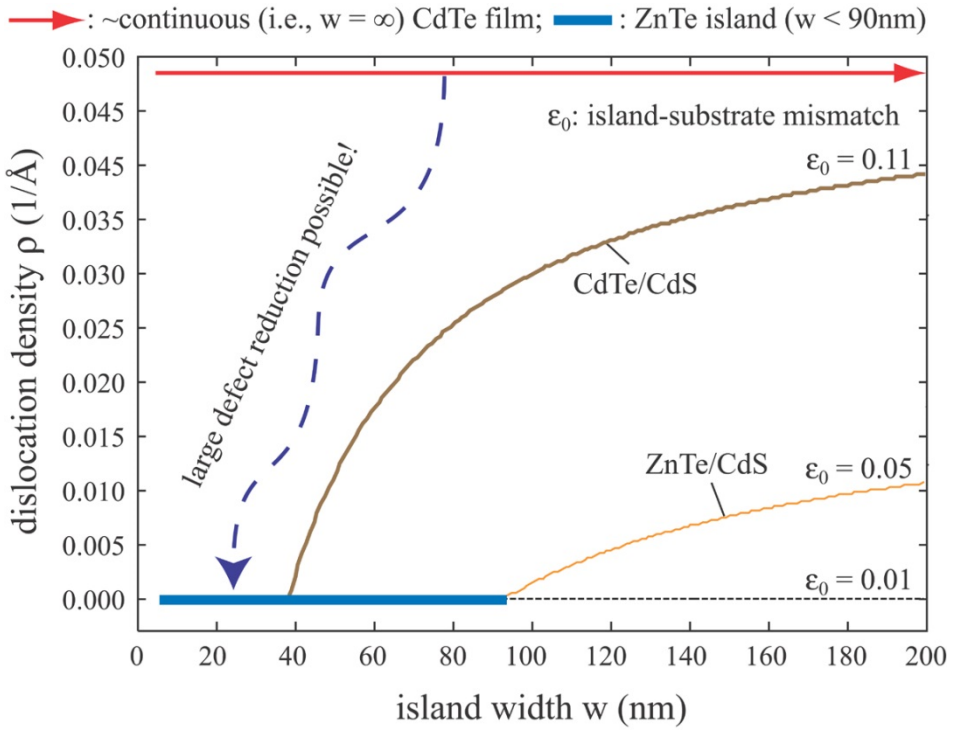
Dislocation-Free CdTe/CdS Solar Cells

Modern nano technology:



BOP simulation:

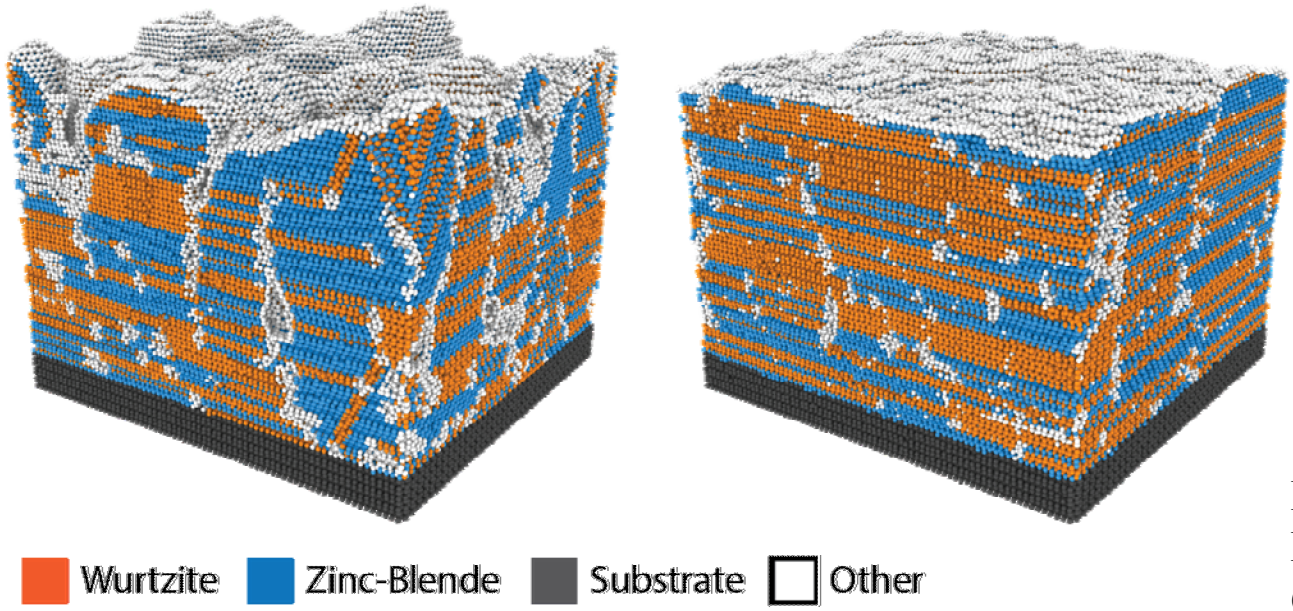


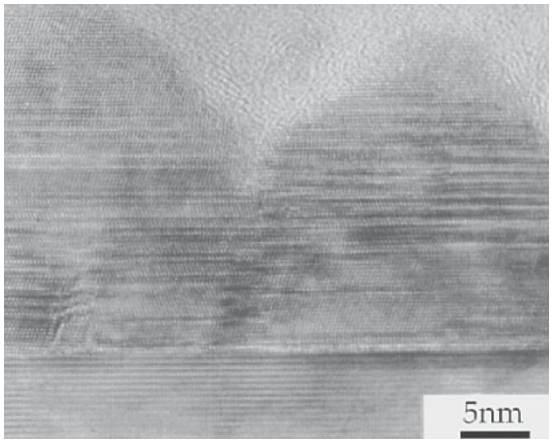


Solid State Lighting Example: GaN on (0001) GaN

(a) T = 2000 K (T/T_m = 0.56)

(b) T = 2800 K (T/T_m = 0.78)

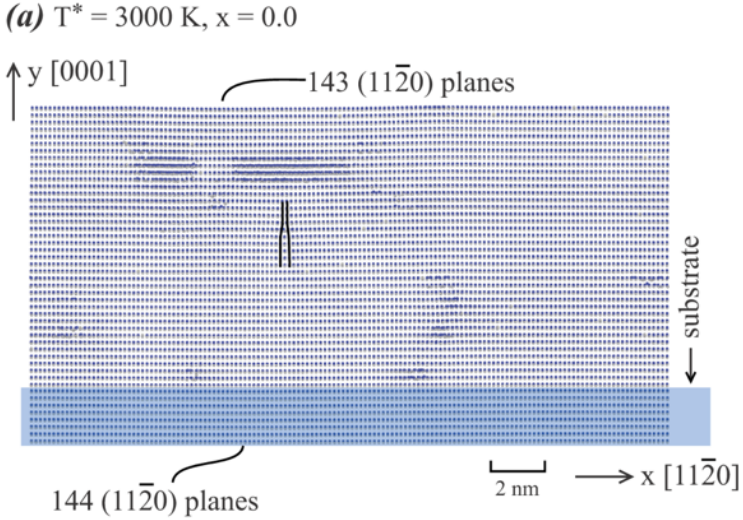


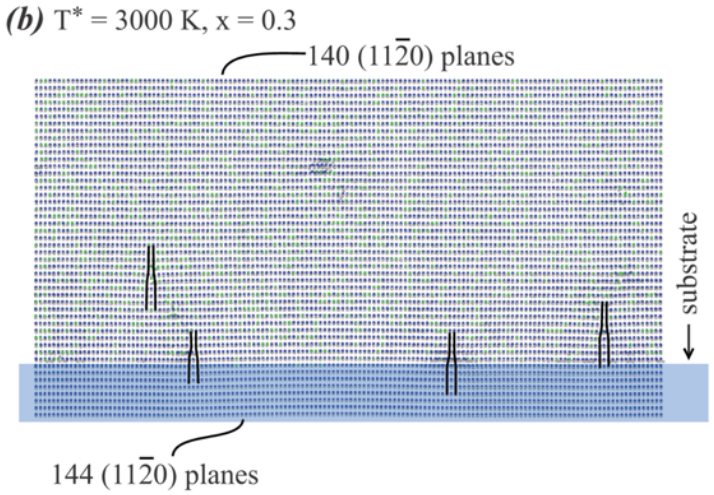


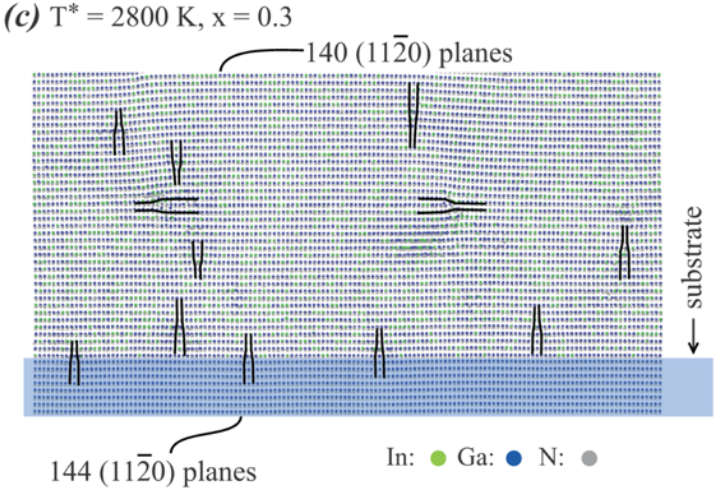
Experimental image from X. H. Wu et al, Appl. Phys. Lett., 68, 1372 (1996)

1. MD and experimental melting temperatures T_m are not the same. Use T/T_m
2. Significant polytypism is observed with the (0001) growth
3. Increasing temperature reduces surface roughness and associated defects

Misfit Dislocation in $In_xGa_{1-x}N/GaN$



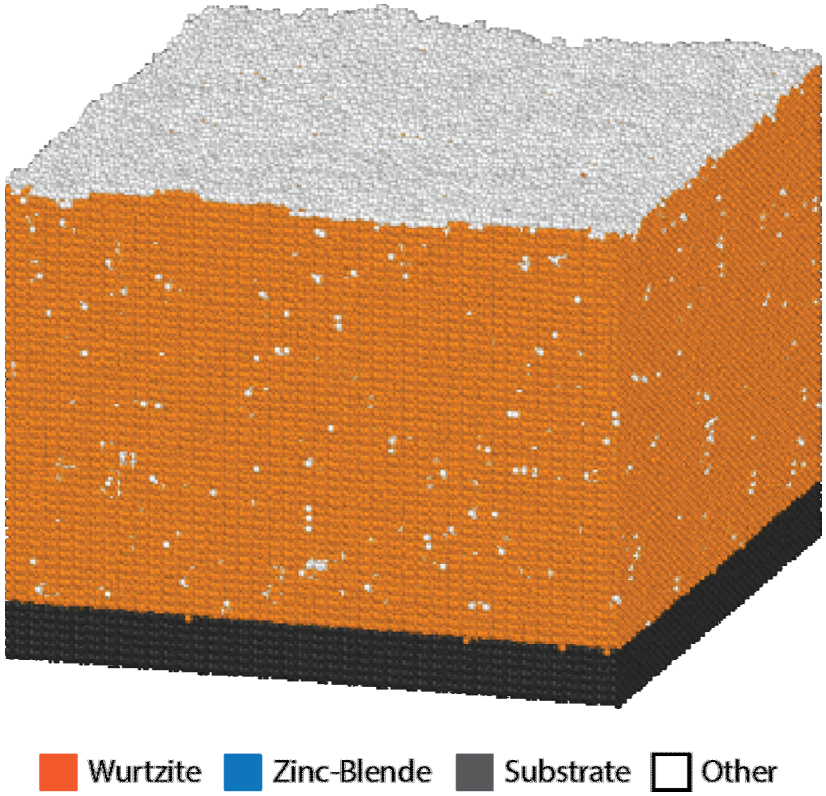




1. Misfit dislocations in $In_xGa_{1-x}N/GaN$ films at (a) x=0.0, T = 3000 K (T/T_m = 0.84); (b) x=0.3, T = 3000K (T/T_m = 0.90); and (c) x=0.3, T = 2800 (T/T_m = 0.84). Atoms are colored by species
2. Some dislocations are kinetically trapped. The number of misfit dislocations equals difference of number of planes between top and bottom surfaces

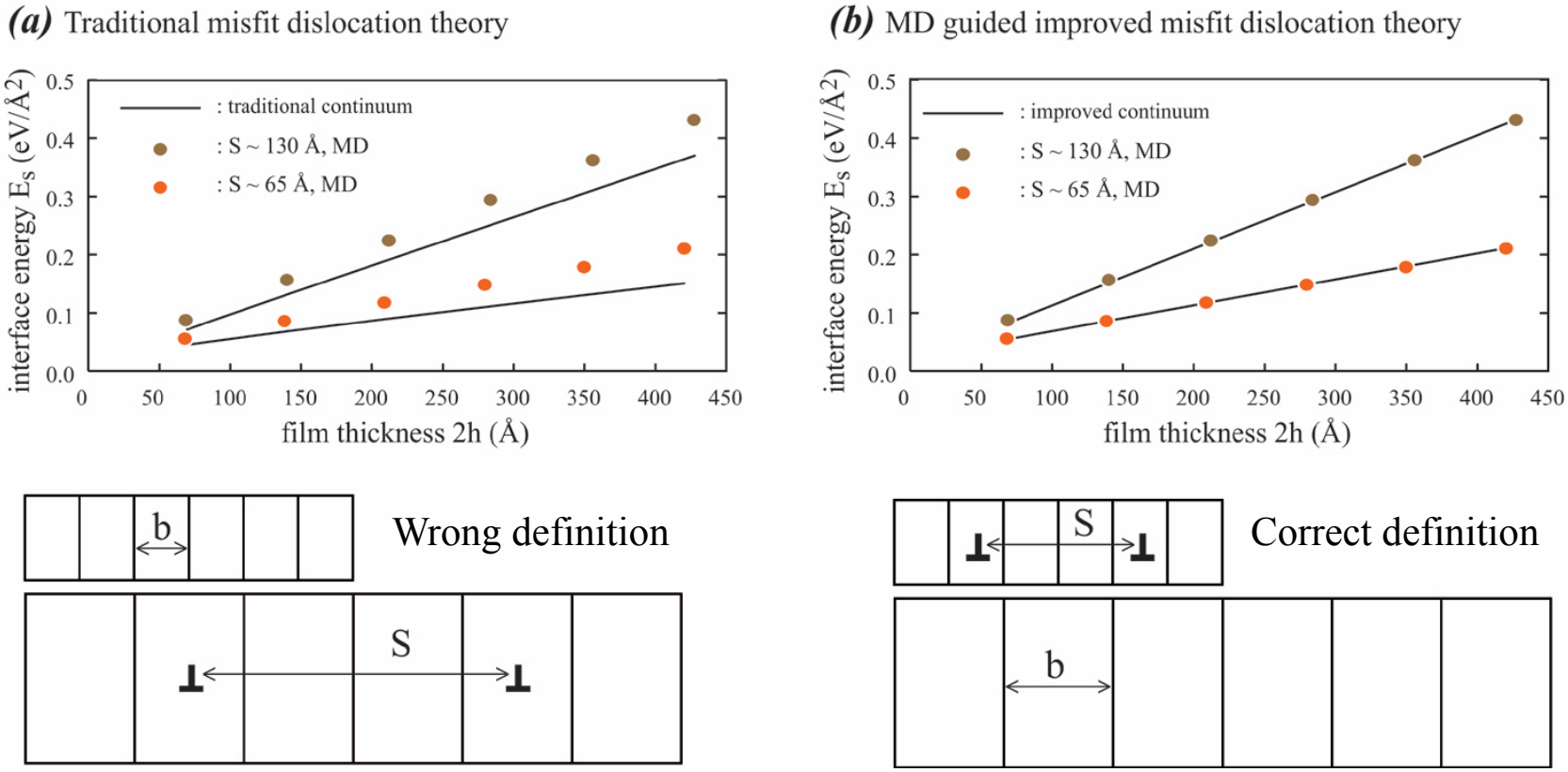
$In_{0.4}Ga_{0.6}N$ on (11-20) GaN

GaN on a (11-20) GaN at 2800 K (T/T_m = 0.78)



1. **Formation of polytypism is due to the ABCABC… stacking, where atoms can occupy either B or C sites on a given plane**
2. **To test the polytypism formation mechanism, growth is simulated on (11-20) plane where the stacking sequence is ABAB…**
3. **Indeed, the polytypism is eliminated on the (11-20) growth**

MD Validation of Misfit Dislocation Theory

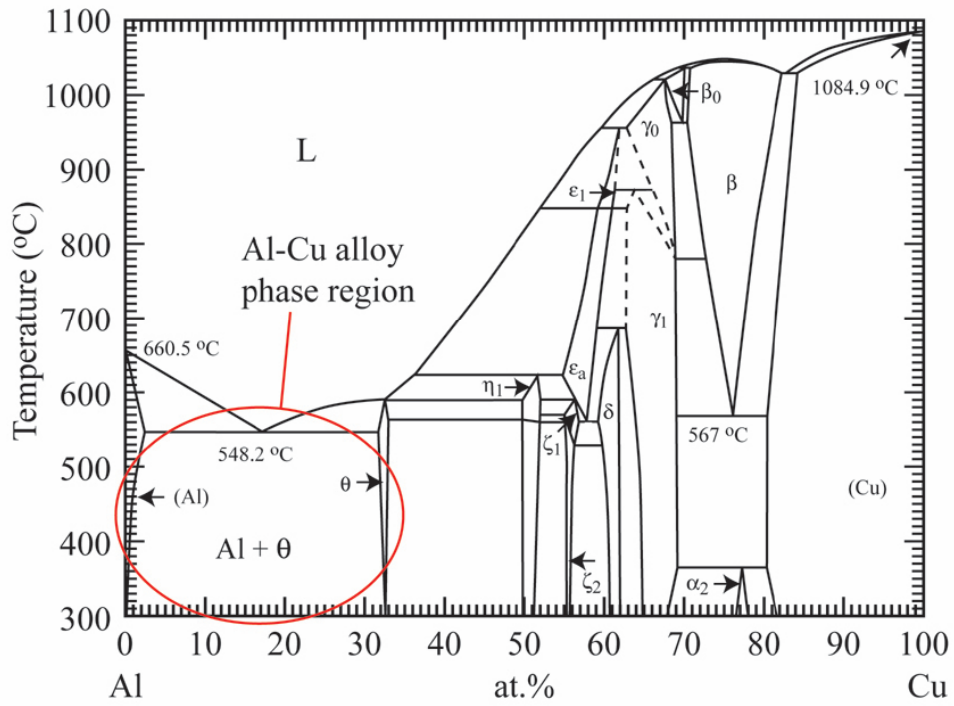


- **Continuum misfit dislocation theory has been widely used since 1980's**
- **MD simulations revealed that traditional continuum misfit dislocation theory is incorrect in the definition of dislocation Burgers vector and dislocation spacing**
- **This example indicates that MD can "validate" and improve continuum models**

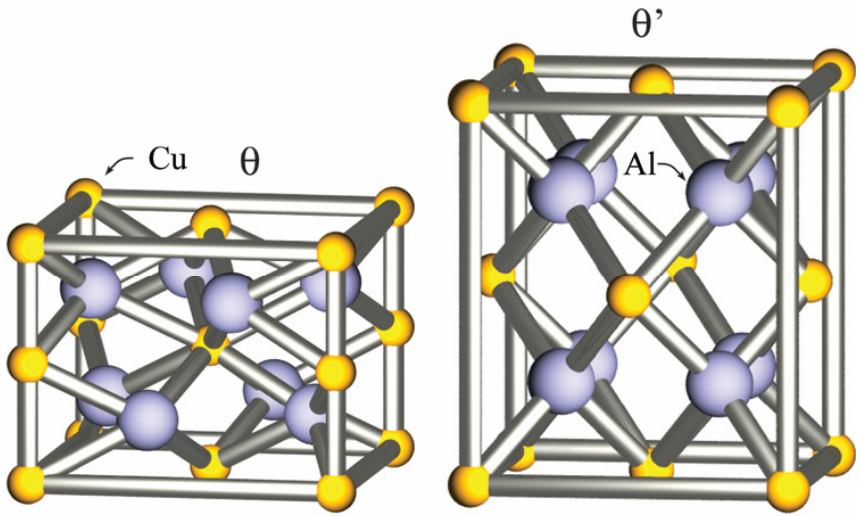
X. W. Zhou et al, J. Mech. Phys. Sol., 91, 265 (2016)

Al-Cu Bond Order Potential

(a) Al-Cu phase diagram

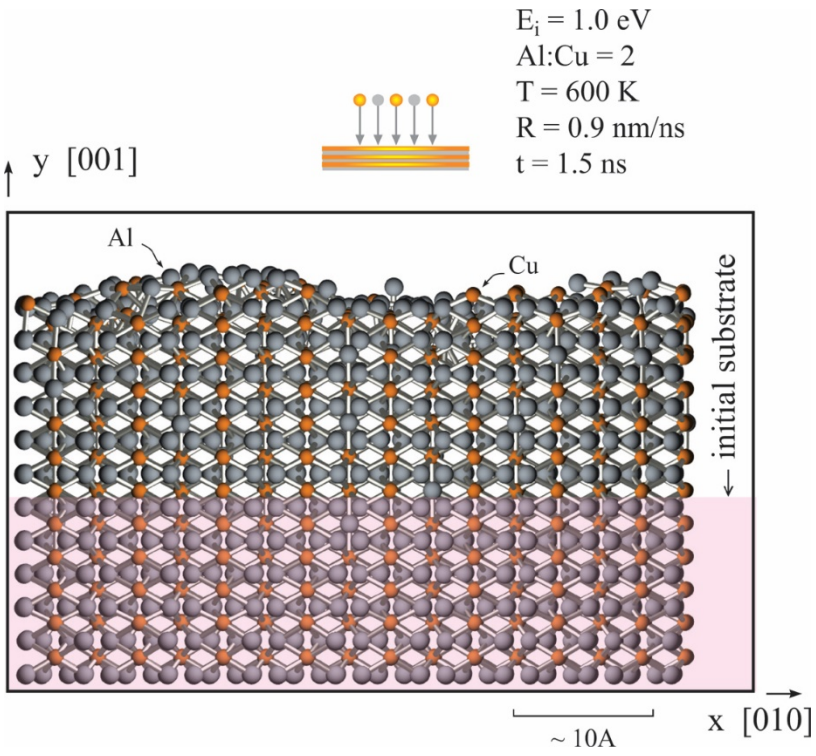


(b) crystal structure of the θ and θ' phases

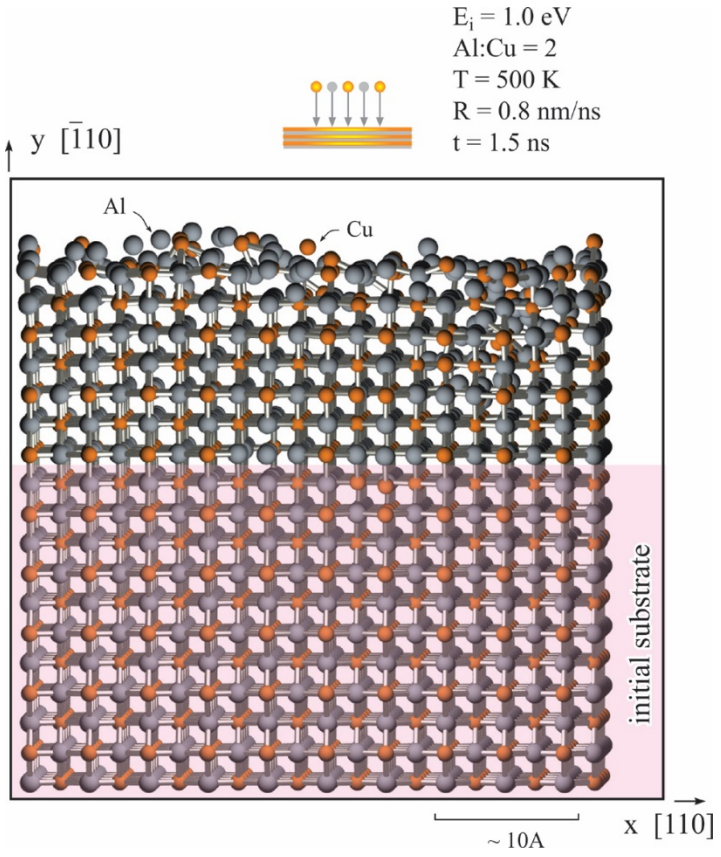


Growth Simulation Tests on BOP

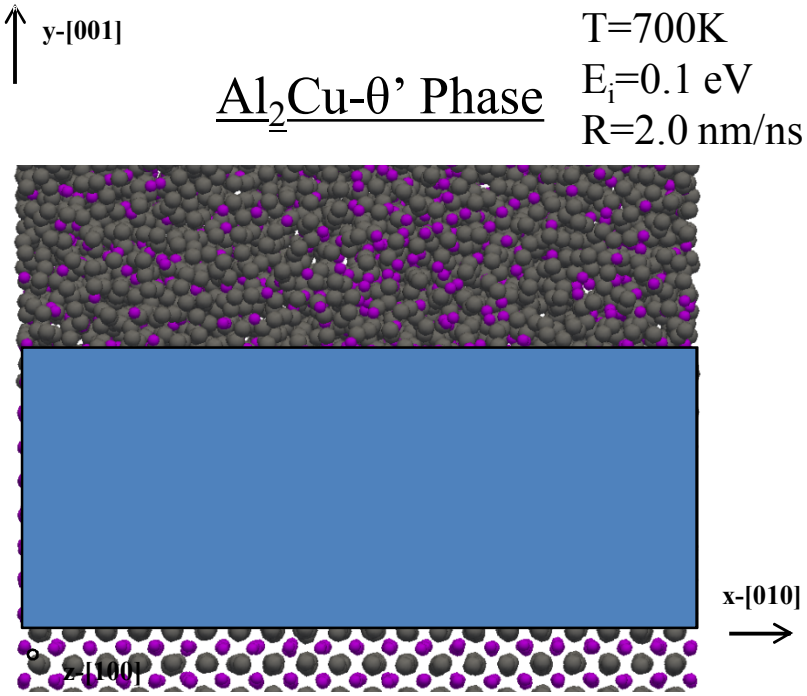
Crystalline growth of θ-Al_2Cu



Crystalline growth of θ'-Al_2Cu

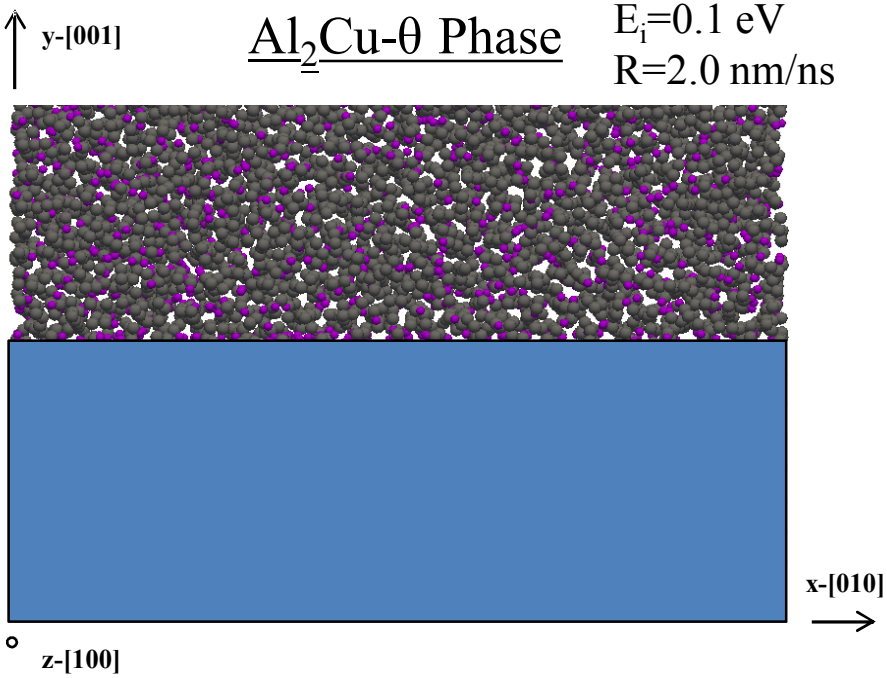


Growth Simulation Tests on the Previously Best Potential (Mishin et al)



Al_2Cu-θ' Phase

T=700K
E_i=0.1 eV
R=2.0 nm/ns



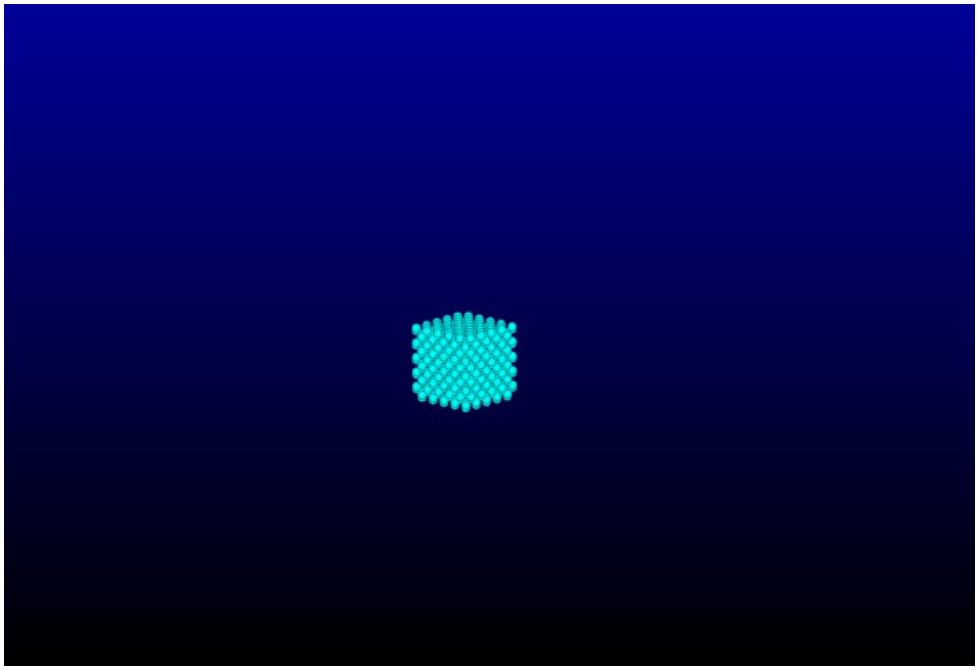
Al_2Cu-θ Phase

T=600K
E_i=0.1 eV
R=2.0 nm/ns

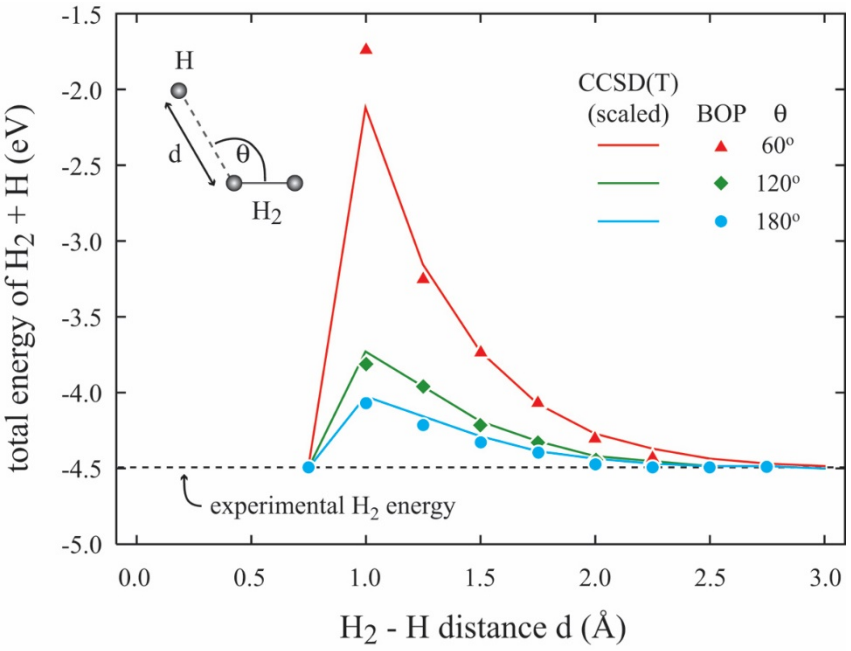
ADP: F. Apostol and Y. Mishin, PRB 83, 054116 (2011)

Chemical Reaction Simulations Using Our Bond Order Potential (BOP)

Hydrogen crystal to H_2 gas

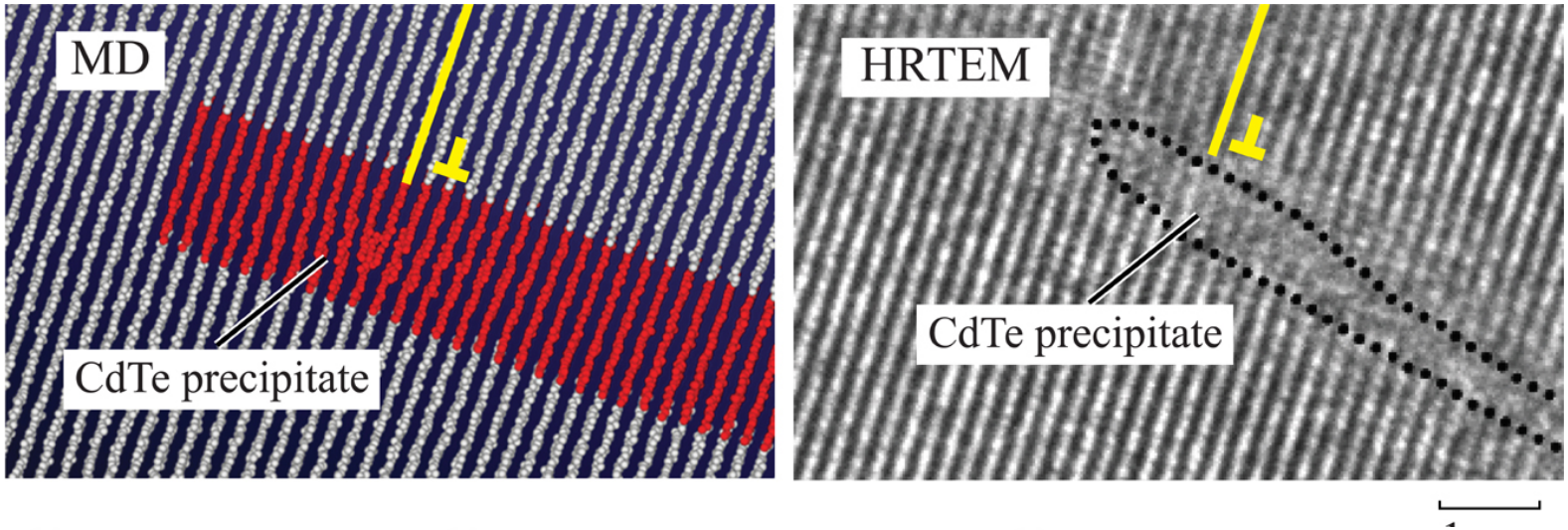


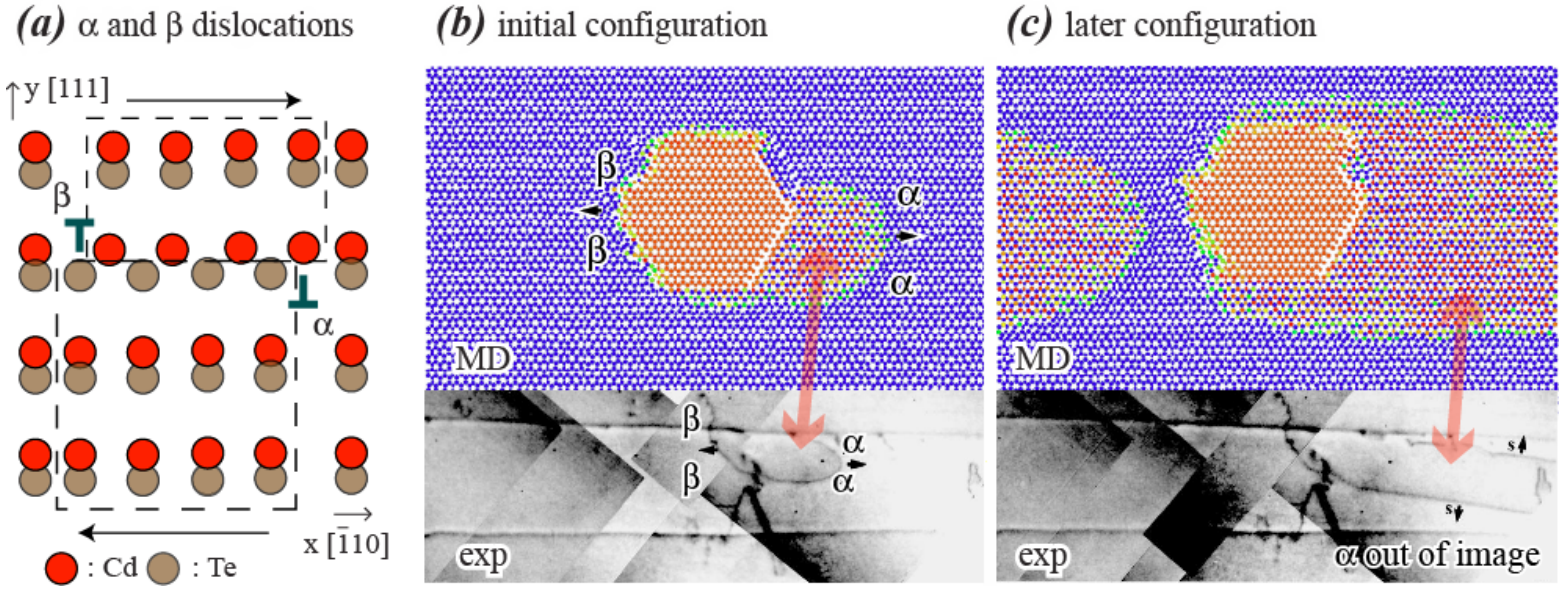
$H_2+H \rightarrow H+H_2$ energy profiles



The BOP we developed has captured the $H_2+H \rightarrow H+H_2$ reaction about 1-2 years ago, paper has been published: X. W. Zhou, D. K. Ward, M. Foster, J. A. Zimmerman, J. Mater. Sci., 50, 2859 (2015).

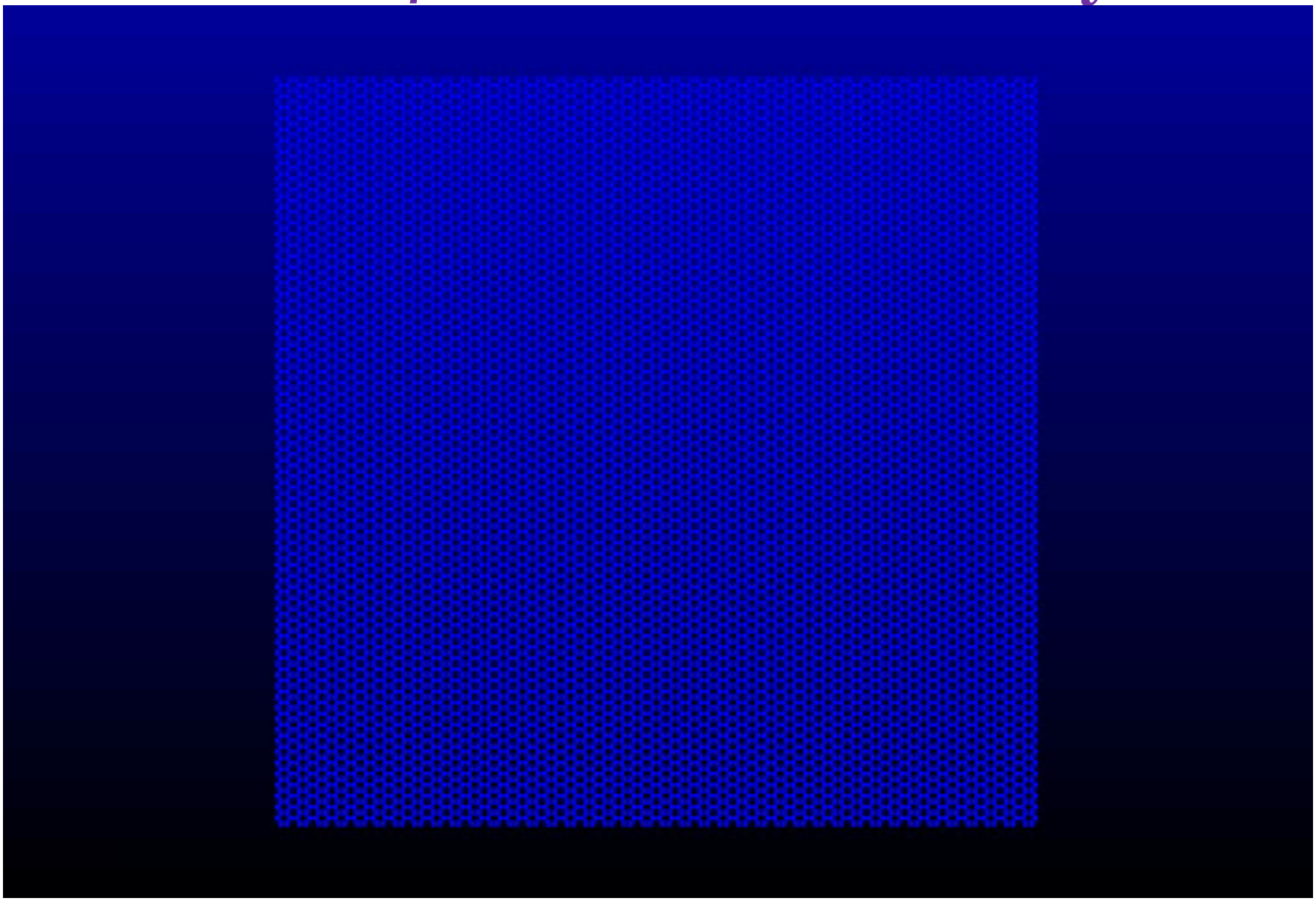
Dislocation Behavior in Semiconductor Compounds: Radiation Detecting $Cd_xZn_{1-x}Te$





X. Zhou, D. K. Ward, B. M. Wong, F. P. Doty, and J. A. Zimmerman, J. Phys. Chem., 116, 17563 (2012)

α and β Dislocation Mobility



Conclusions

MD simulations have impacted:

- Graphene growth
- Giant magnetoresistive multilayers
- CdTe/CdS solar cell films
- $In_xGa_{1-x}N$/GaN solid state lighting films
- Misfit dislocation theory
- Research on Al-Cu alloys
- Chemical reactions between H_2 and surfaces
- Radiation detecting $Cd_xZn_{1-x}Te$

Thank You …