A Perspective of Molecular Dynamics (MD) Simulations

SAND2015-1657PE

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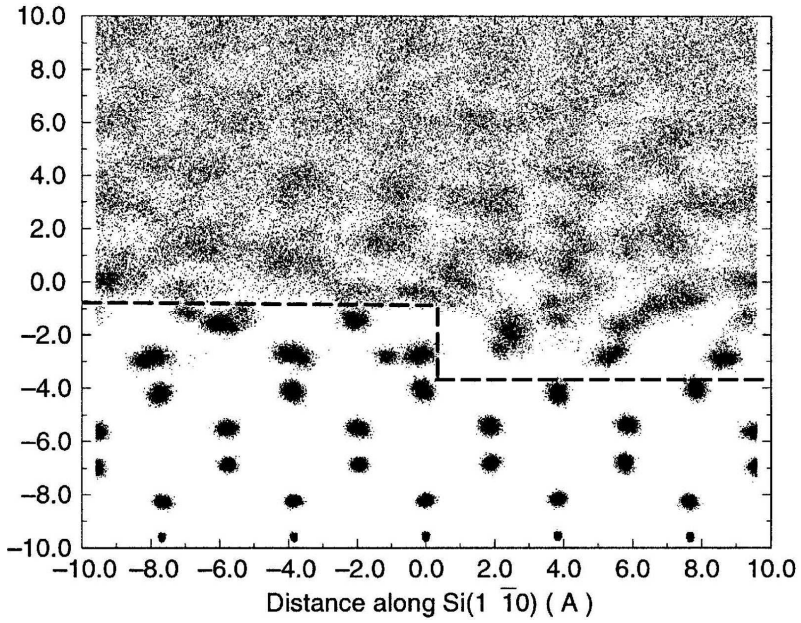
Contributions from Don Ward, Mike Foster, and Bryan Wong are greatly appreciated

1. The Limiting Factor - **Potentials**
   - Growth simulation tests
   - 2D materials
   - Chemical reactions
   - Beyond EAM for metals
   - Database potentials
   - Temperature dependent potential
   - State-of-the-art potentials
2. MD Derived **Continuum Rules**
   - Misfit dislocation theory
3. Some **Examples**
   - CdTe/CdS solar cells
   - Dislocation behavior in compound semiconductors
   - Melt growth of CdTe
   - Towards improved $LaBr_3$

1. The Limiting Factor – Potential

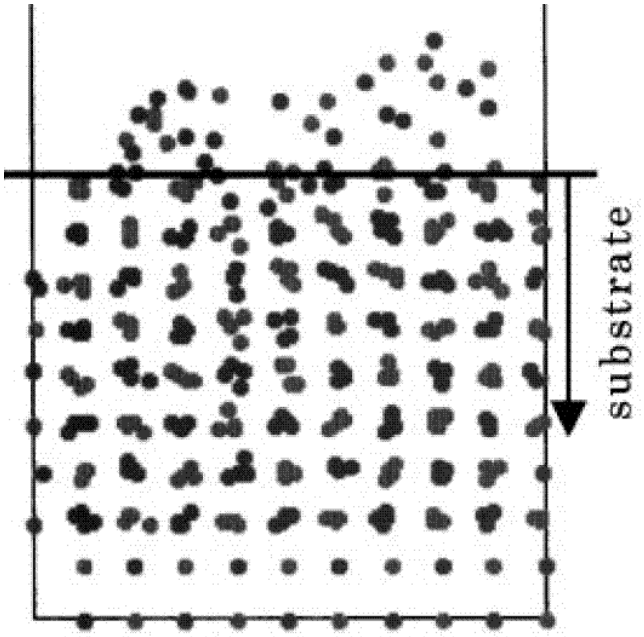
❑ Growth simulation tests

CdTe on Si



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

InAs on GaAs



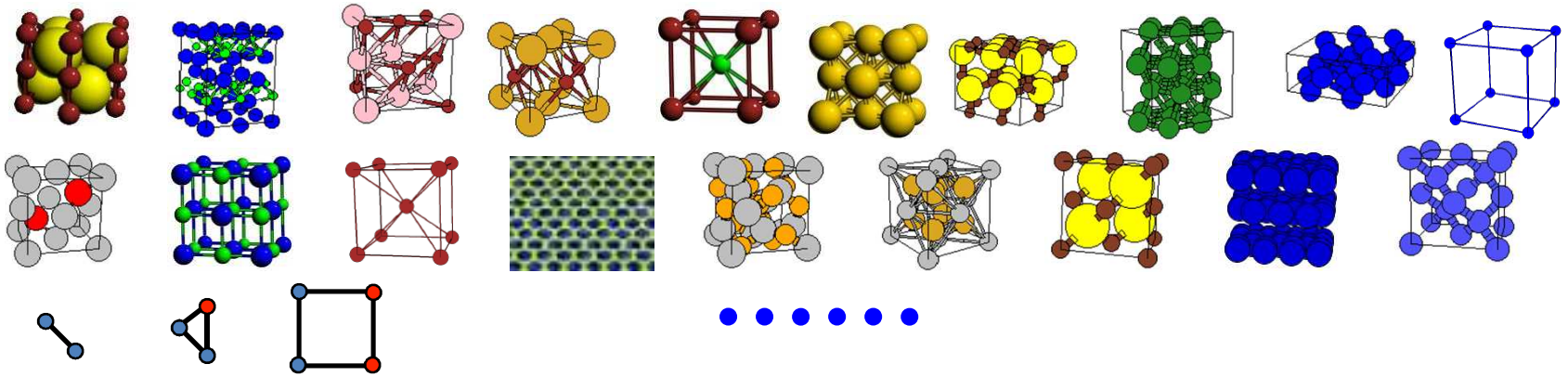
M. Nakamura, H. Fujioka, K. Ono, M. Takeuchi, T. Mitsui, and M. Oshima, J. Cryst. Growth, 209, 232(2000).

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.

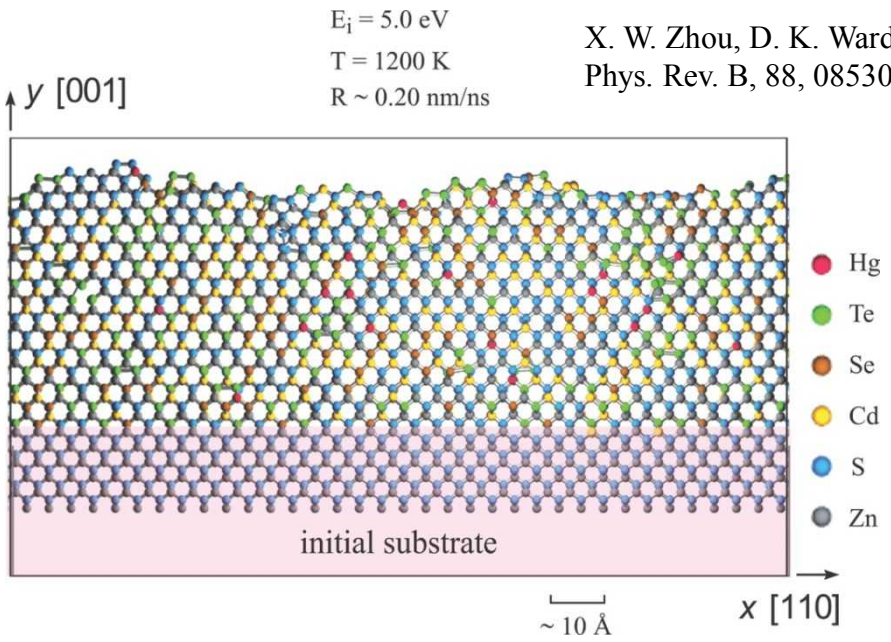
1. The Limiting Factor – Potential

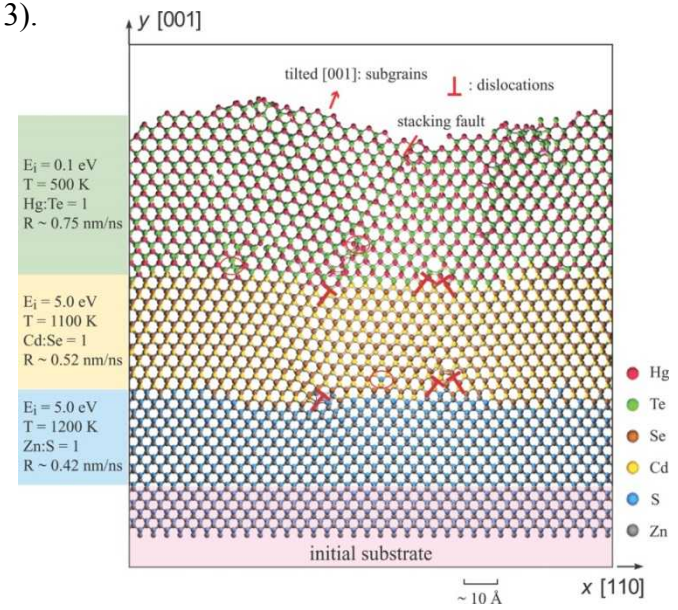
❑ Growth simulation tests



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

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).



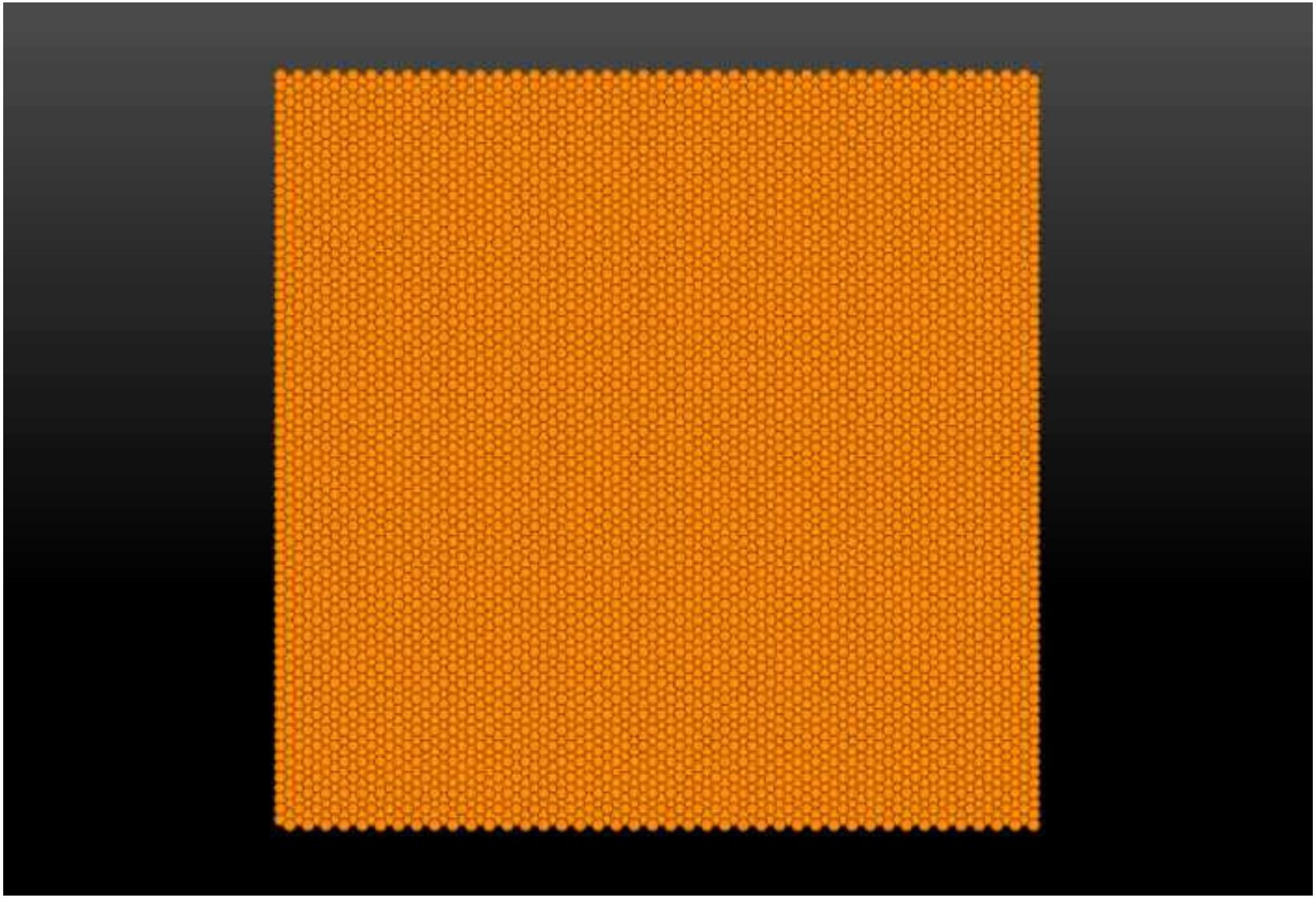


Current parameterization requires human involvement. Need new algorithm to replace that

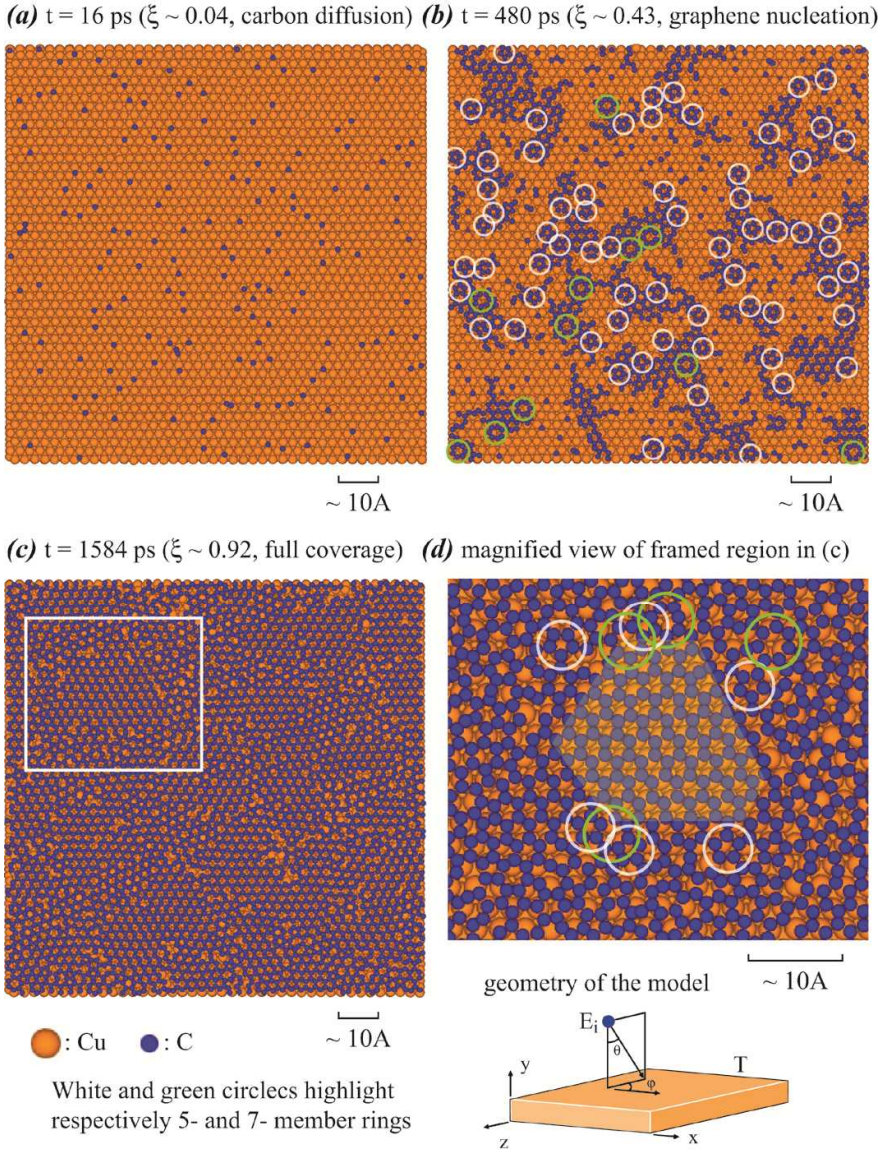
1. The Limiting Factor – Potential

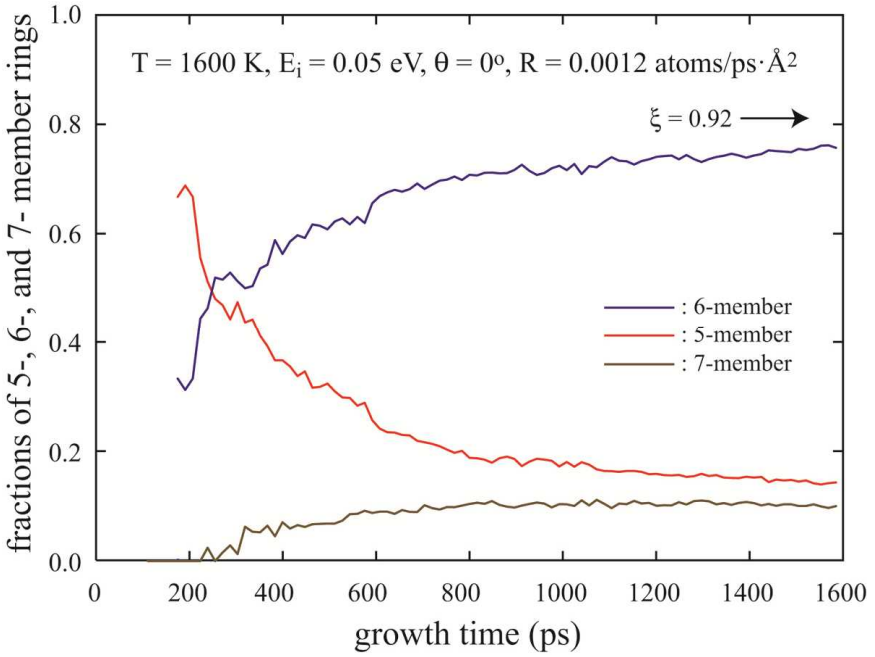
❑ 2D materials

We have developed a bond order potential to enable MD simulations of graphene growth on Cu(111)



Insights Gained from MD Simulations

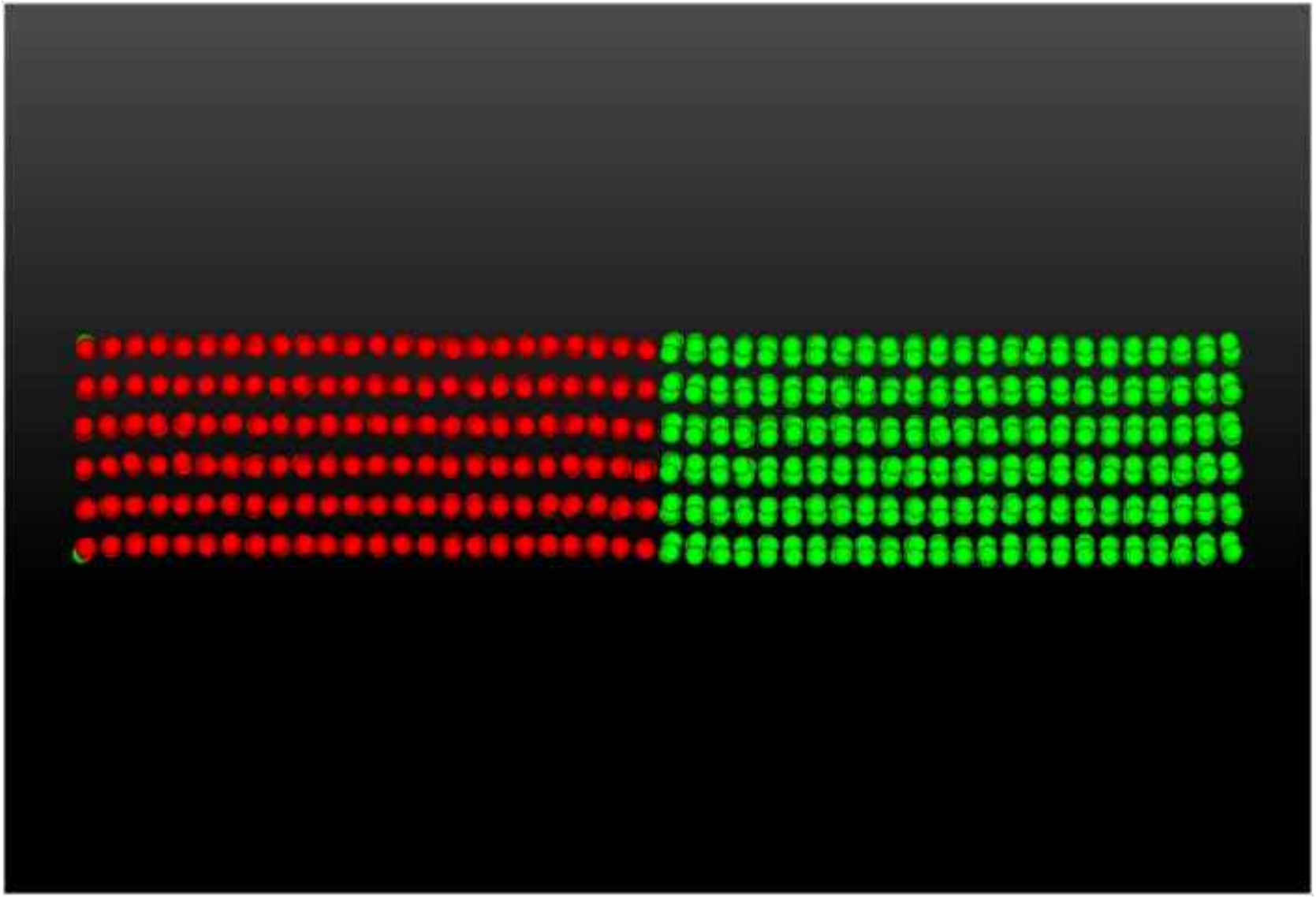




MD simulations indicate: defects form at the graphene edge throughout the growth, not when island impinge

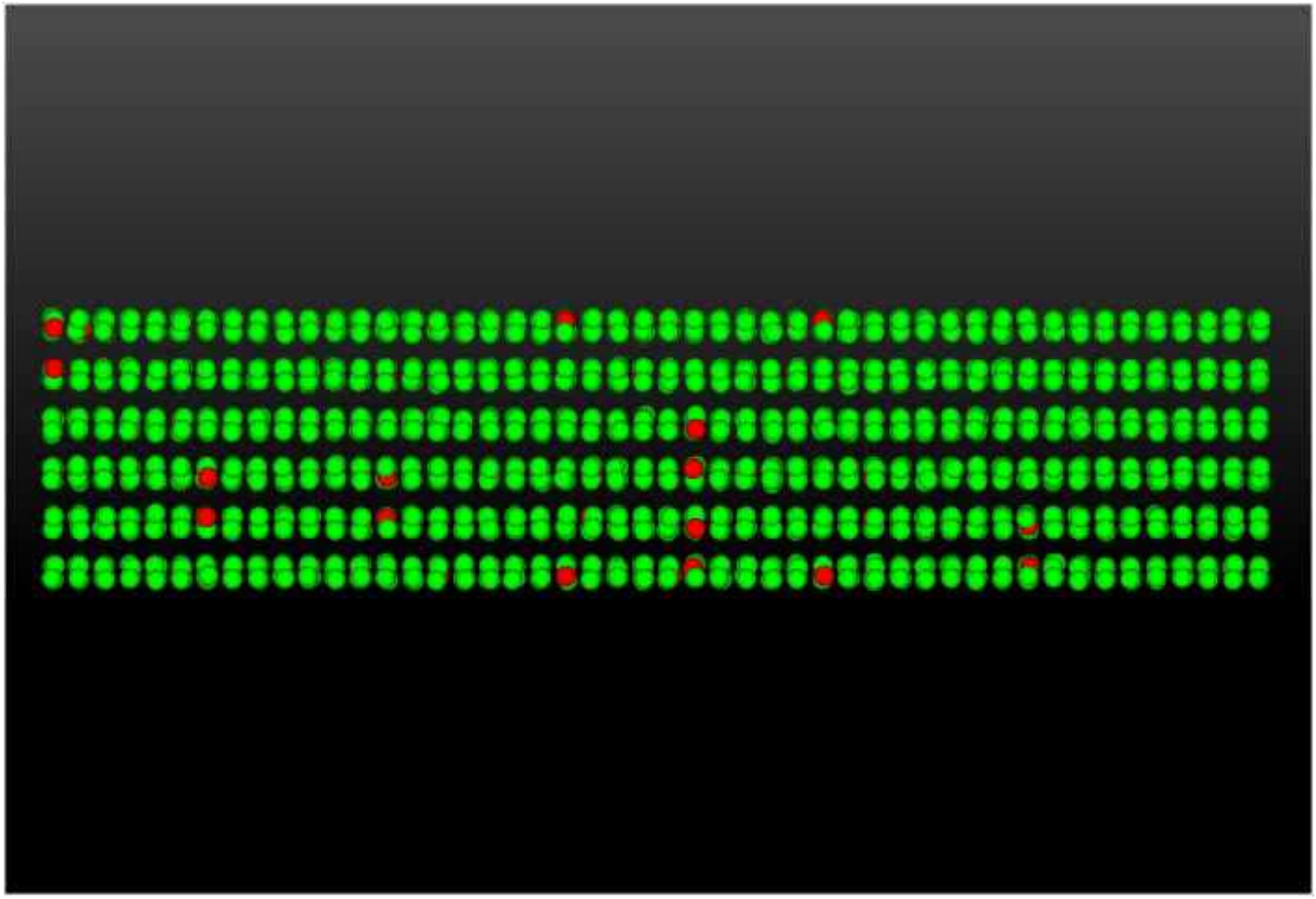
Graphite to Diamond Transformation

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

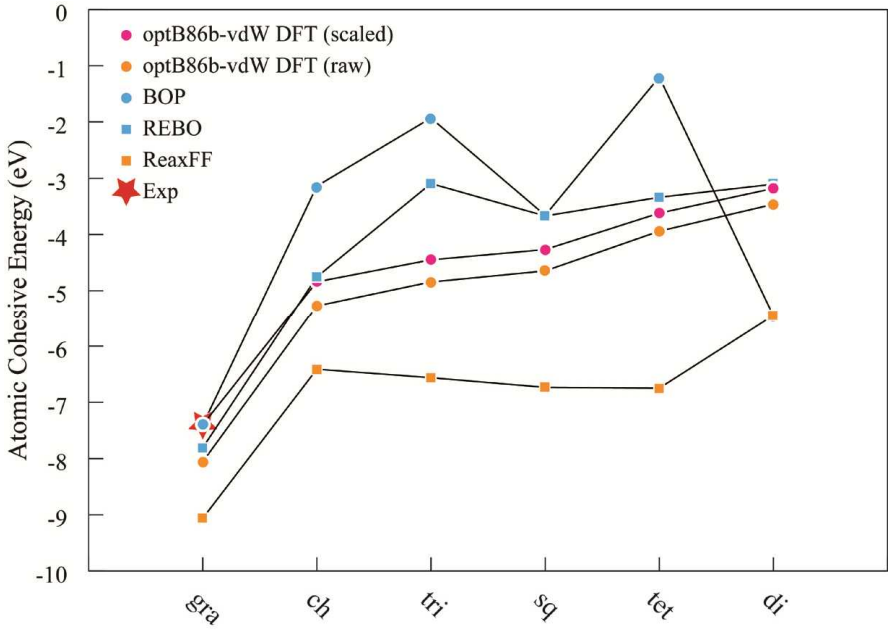


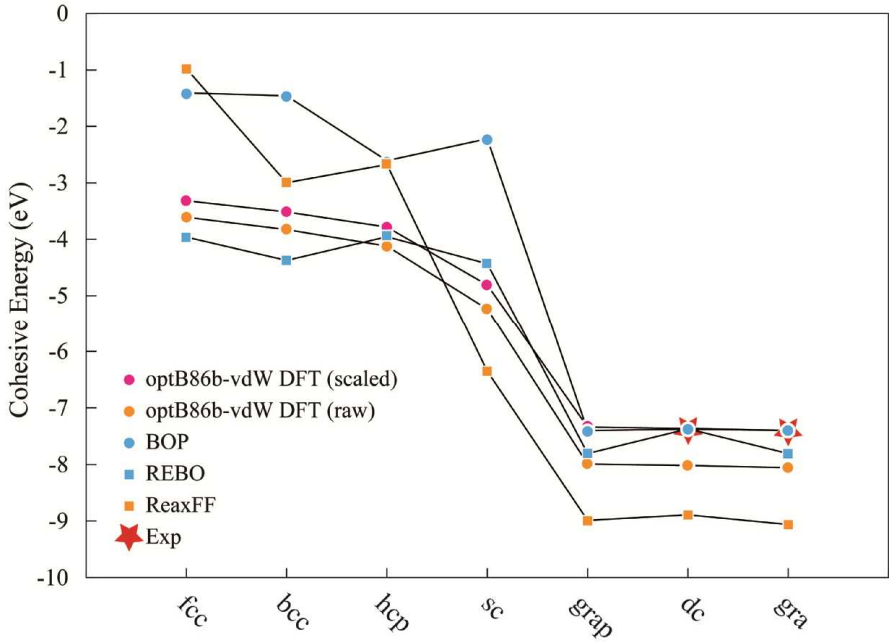
Diamond to Graphite Transformation

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



Top Carbon Potentials

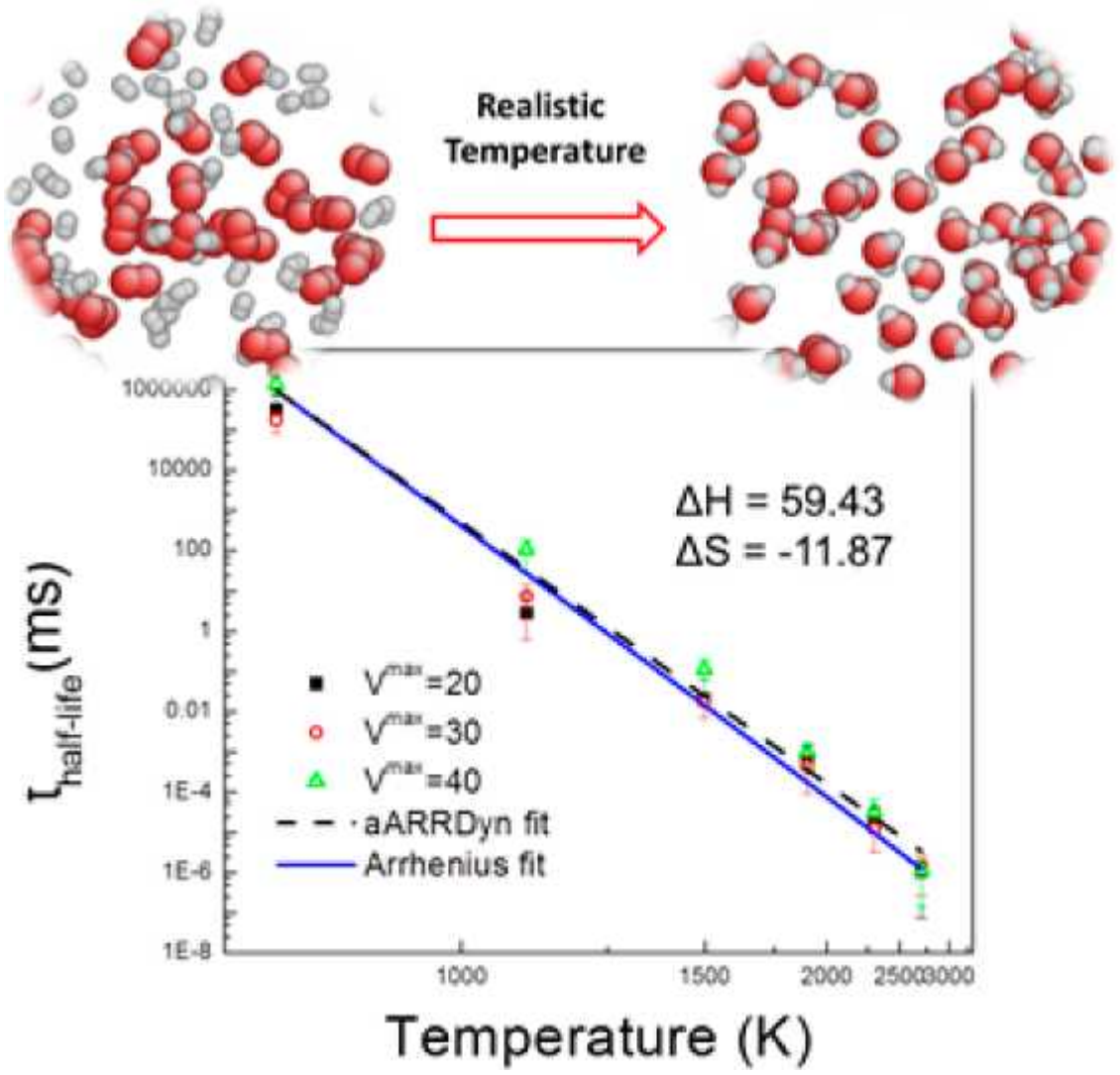




- **REBO** (Brenner et al), **ReaxFF** (van Duin and Goddard et al), and **EDIP** (Marks et al) are top literature carbon potentials
- **REBO** appears to capture better carbon structures than ReaxFF (which is optimized for C-H molecules)
- The growth simulation capability is unique to our **BOP** parameterization

1. The Limiting Factor – Potential

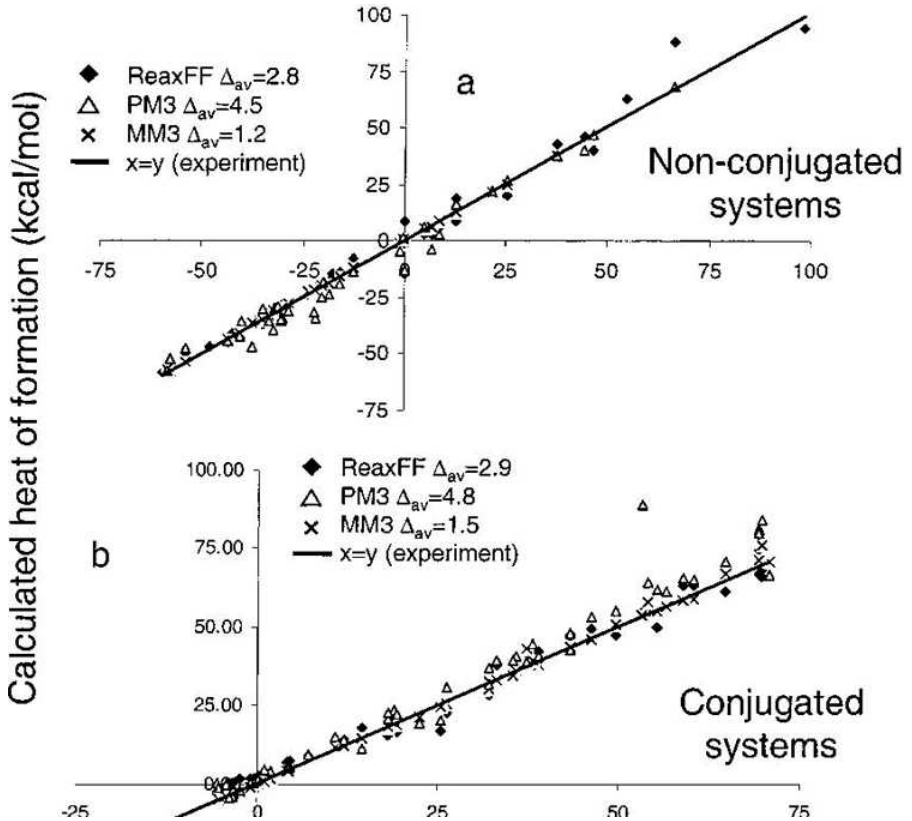
❑ Chemical reactions

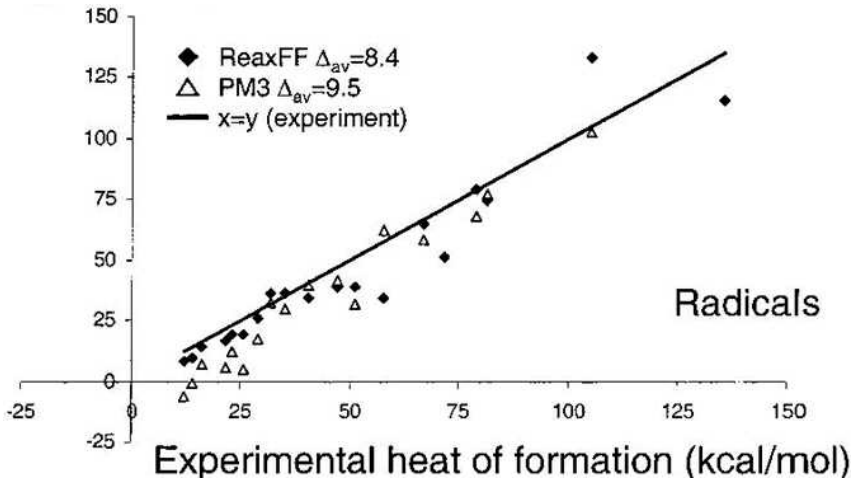


- **MD traditionally for physical processes;**
- **Transferrable potentials can predict chemical processes;**
- **$H_2 + ½O_2 \rightarrow H_2O$ combustion reaction was recently demonstrated by Caltech using ReaxFF;**
- **Fidelity of future potentials is expected to enable more and more chemical reaction simulations.**

T. Cheng, A. Jaramillo-Botero, W. A. Goddard, III, and H. Sun, JACS, 136, 9434 (2014).

Towards MD Simulations of Combustions Involving C-H-O-N





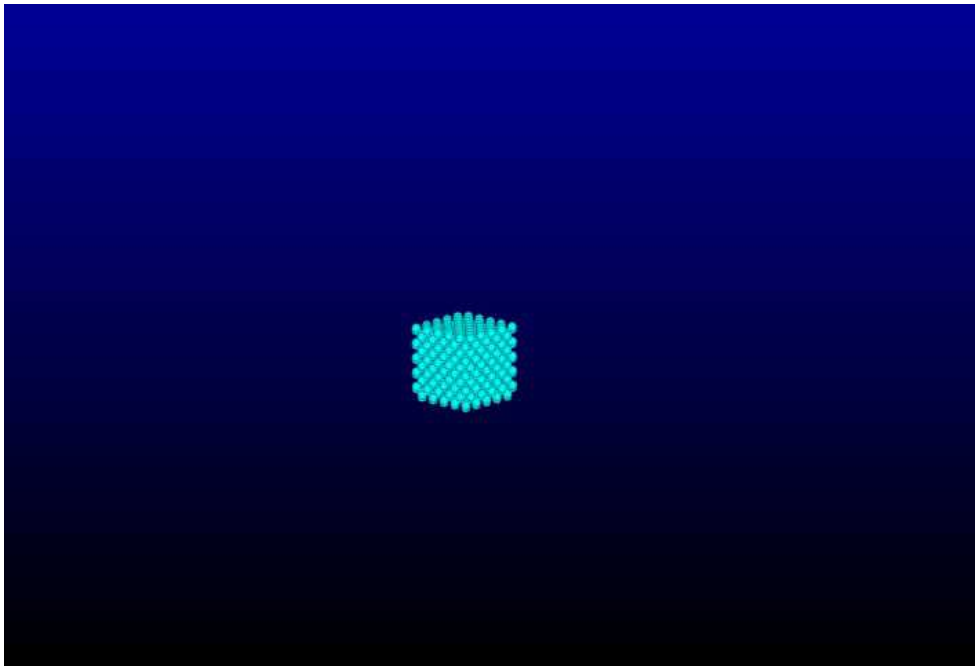
Potential from: A. C. T. van Duin, S. Dasgupta, F. Lorant, and W. A. Goddard III, J. Phys. Chem. A, 105, 9396 (2001).

~409 citations.

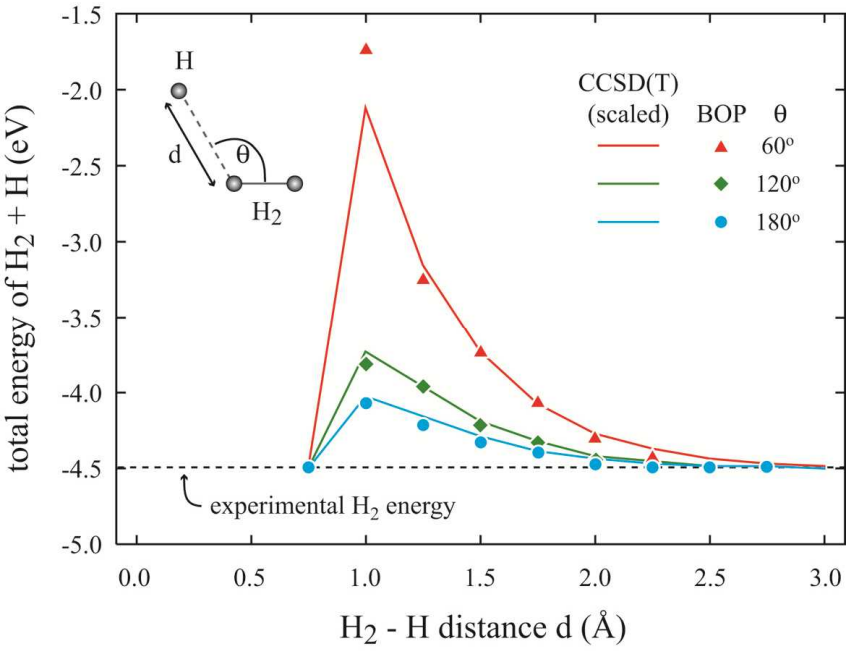
Progress by ReaxFF is to be commended. Still long way to go.

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).

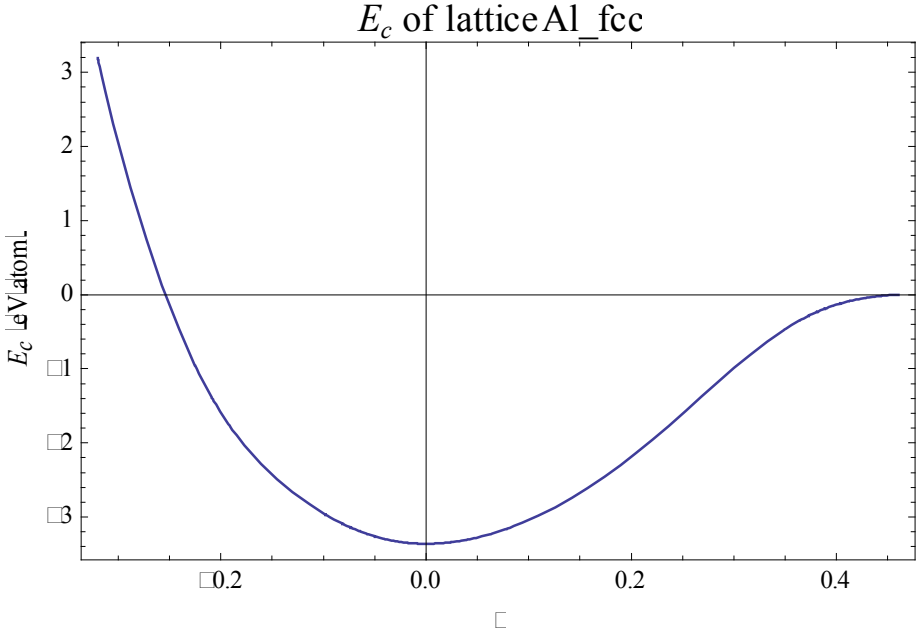
1. The Limiting Factor – Potential

❑ Beyond EAM for metals

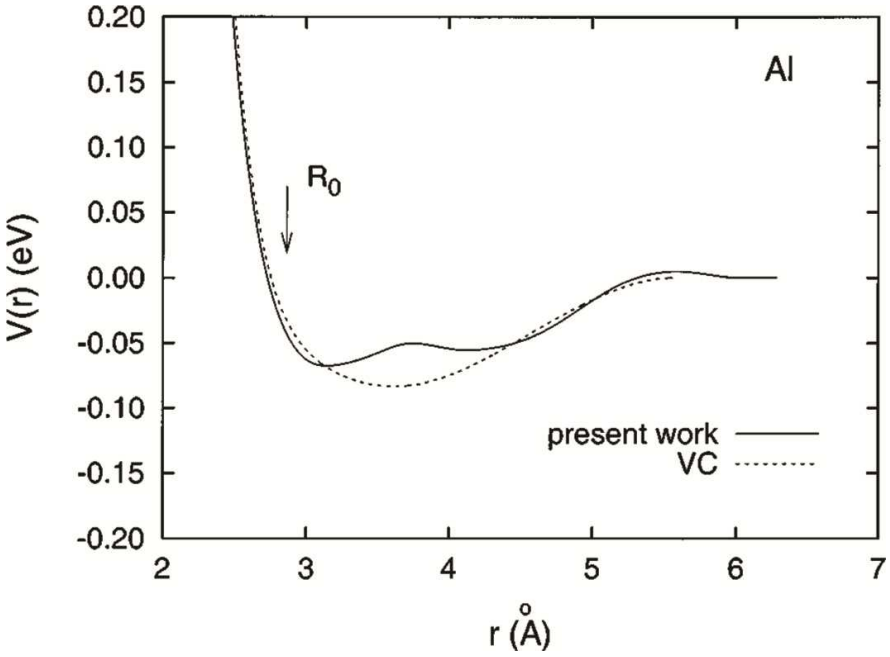
Al-Cu potential

Al has a high stacking fault energy (122-144 mJ/m^2) that cannot be captured by EAM

Our BOP
(γ_{sf} = 133 mJ/m^2)



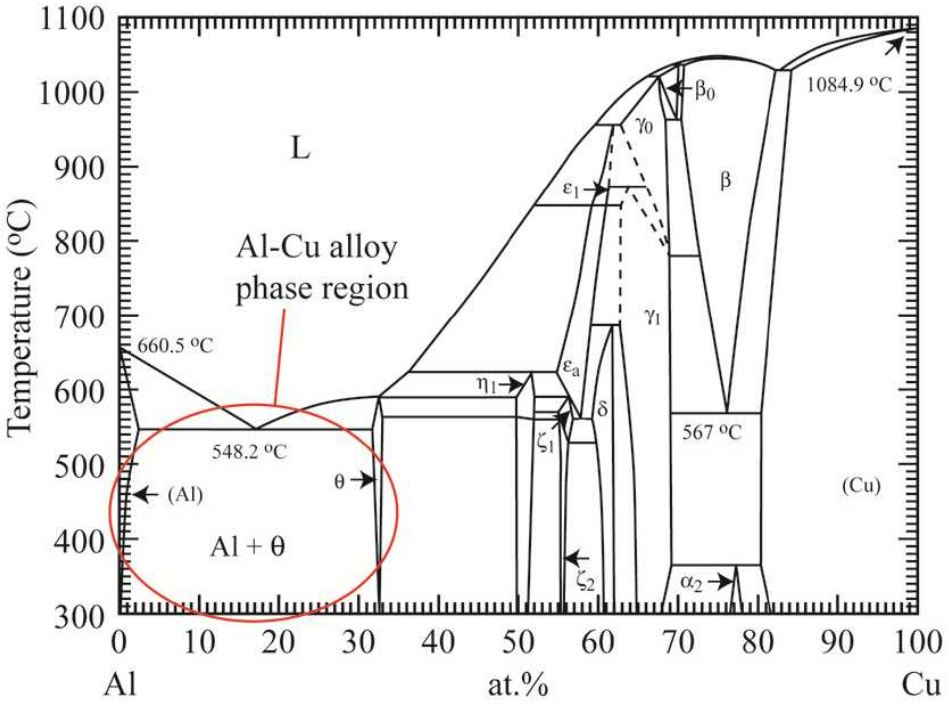
Mishin et al's EAM*
(γ_{sf} = 141 mJ/m^2)



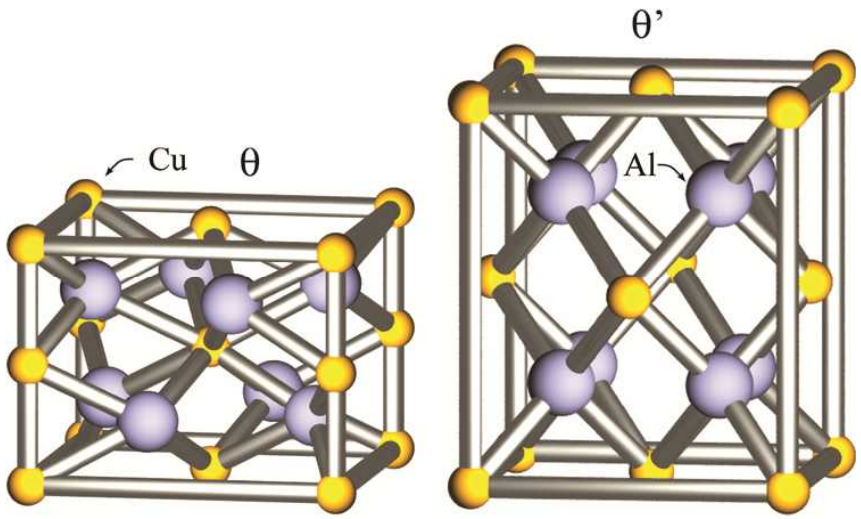
* Phys. Rev. B, 59, 3393 (1999); 83, 054116 (2011).

Incorrect Cu→Al Heat of Solution in Literature

(a) Al-Cu phase diagram



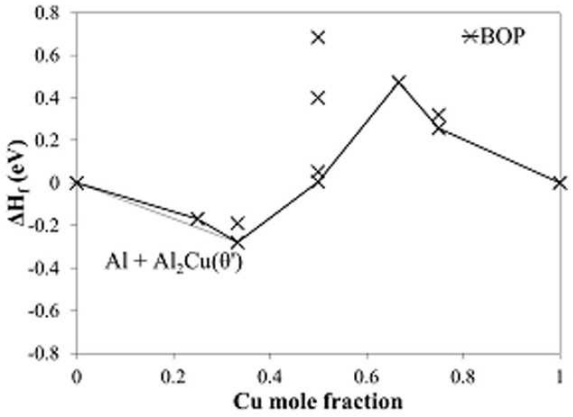
(b) crystal structure of the θ and θ' phases



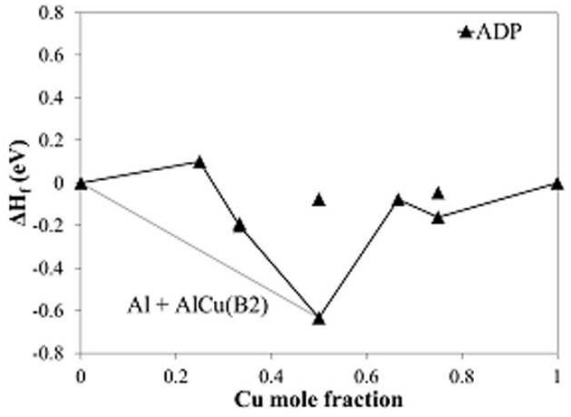
The Cu→Al heat of solution should be the energy change for dissolving 1 molecule of Al_2Cu in Al, rather than 1 atom of Cu in Al!

Two Problems of Literature Al-Cu Potentials

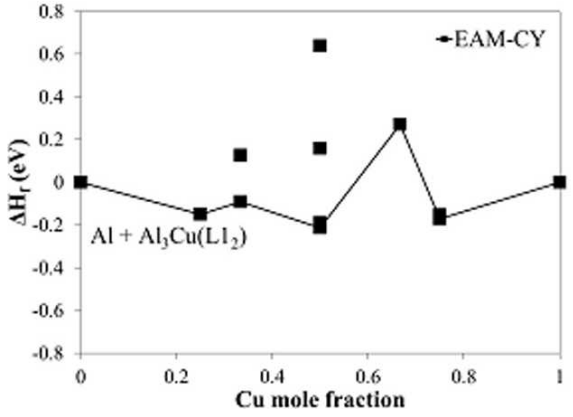
(a) BOP



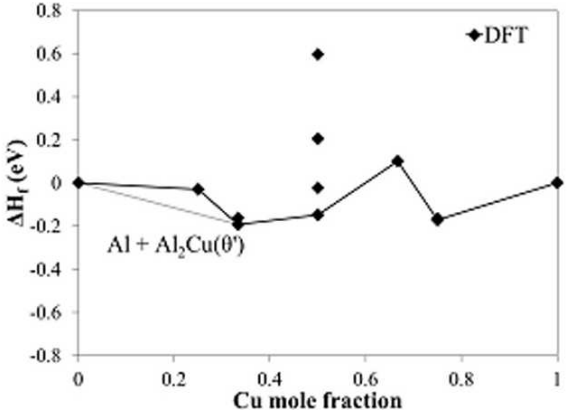
(b) ADP (Mishin et al's EAM)



(c) EAM-CY (by Cai and Ye)



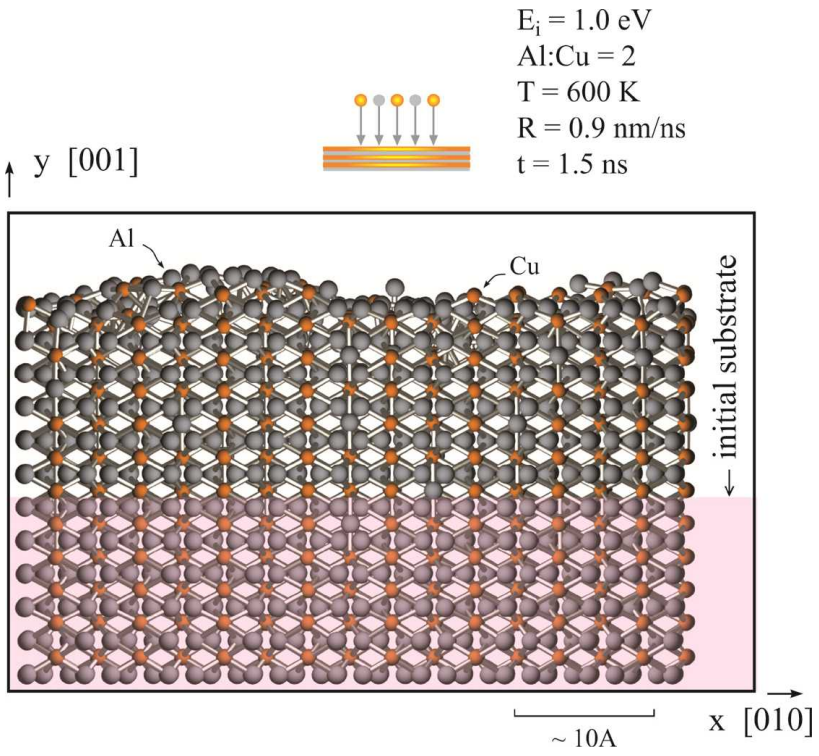
(d) DFT



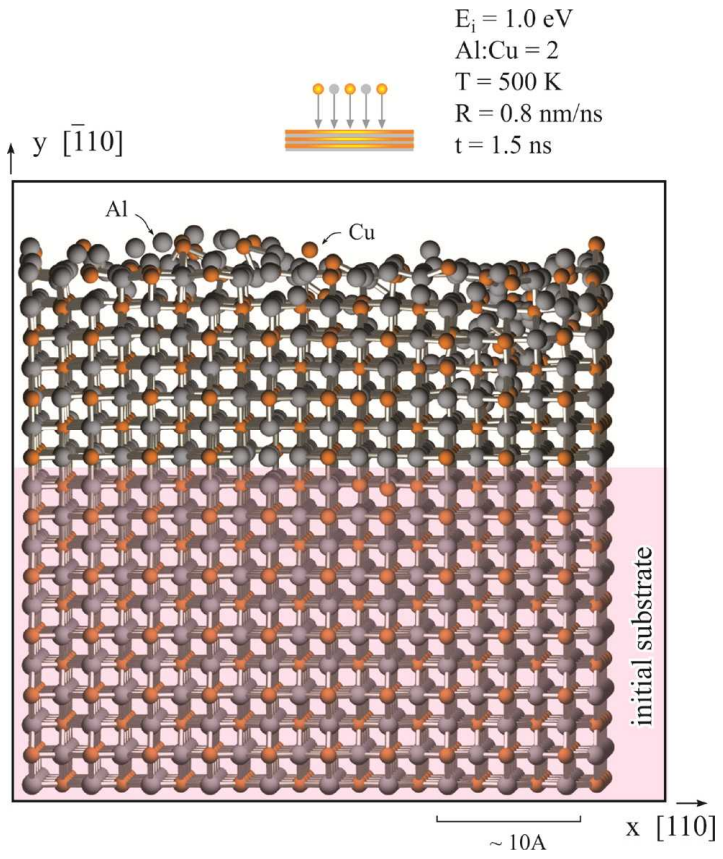
(1) without growth simulation (or simulated annealing) tests, there is no way to ensure the lowest energy compared with ANY other configurations; (2) it is the two-phase mixture, but not the Al_2Cu compound, that must have the lowest energy.

Growth Simulation Tests on BOP

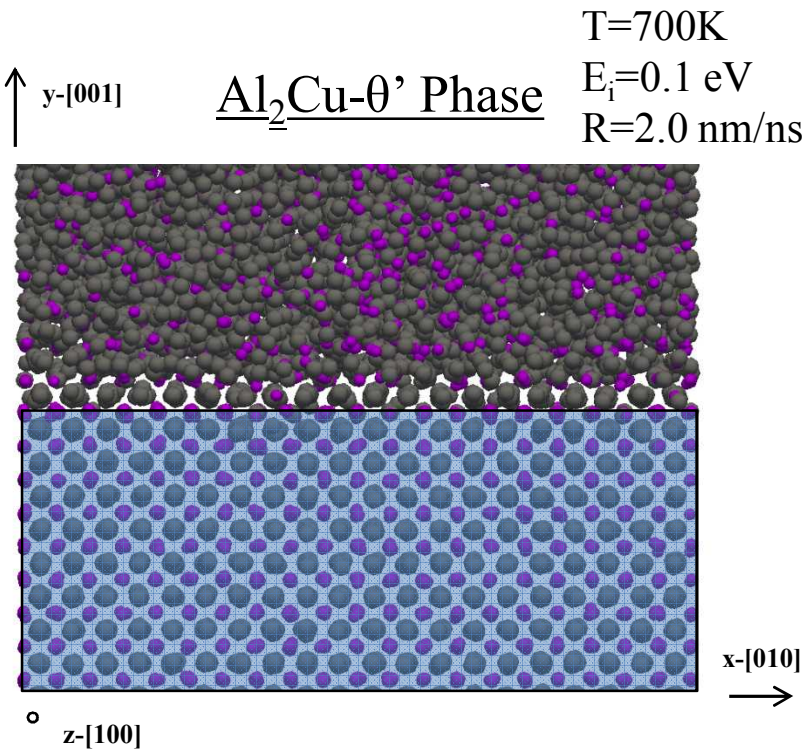
Crystalline growth of θ-Al_2Cu

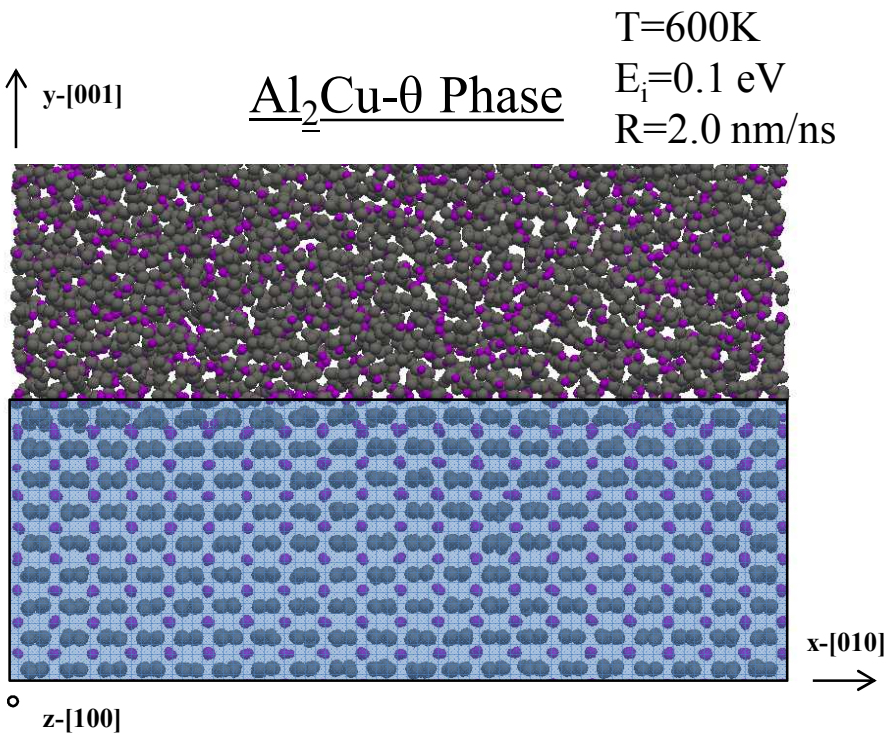


Crystalline growth of θ'-Al_2Cu



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





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

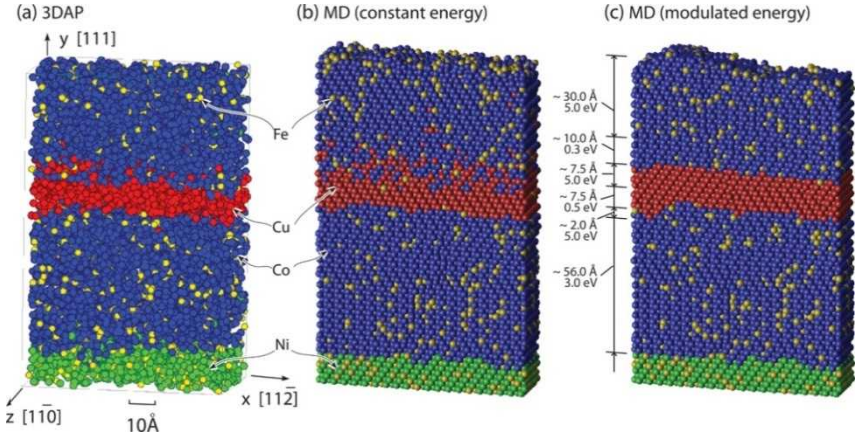
1. The Limiting Factor – Potential

❑ Database potentials

Two of the most cited articles[1,2] that I have co-authored contain the EAM database for Cu, Ag, Au, Ni, Pd, Pt, Al, Pb, Fe, Mo, Ta, W, Mg, Co, Ti, and Zr

1. X. W. Zhou, H. N. G. Wadley, R. A. Johnson et al, Acta Mater., 49, 4005 (2001).
2. X. W. Zhou, R. A. Johnson, and H. N. G. Wadley, Phys. Rev. B, 69, 144113 (2004).

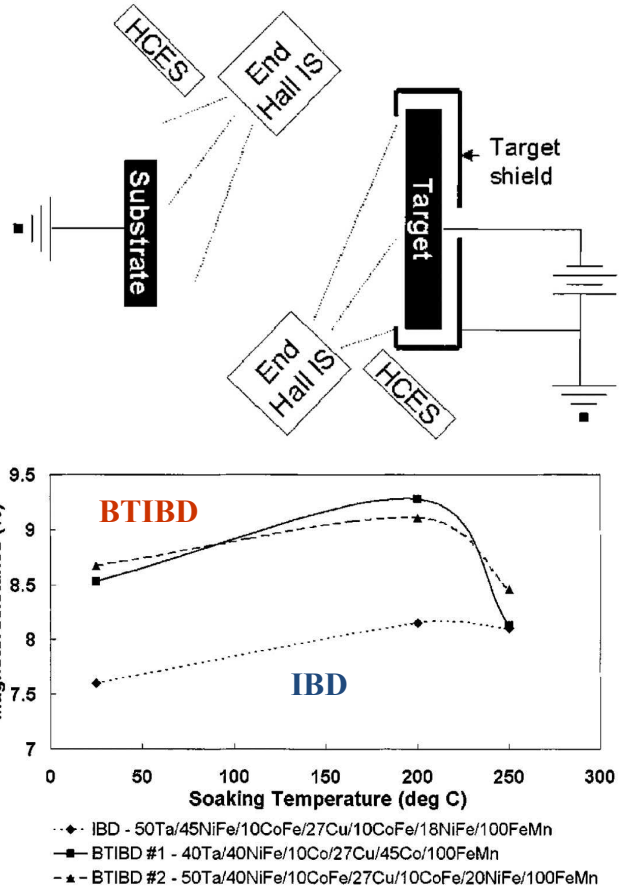
MD prediction of improved synthesis
(giant magnetoresistive spin valve)



X. W. Zhou, and H. N. G. Wadley, J. Appl. Phys., 84, 2301 (1998).

I have developed a growth simulation enabling Zn-Cd-Hg-S-Se-Te potential (Phys. Rev. B, 88, 085309, 2013). Is seeking to develop a similar database potential for III-V.

MD-based BTIBD growth method
(by CVC, Inc.)

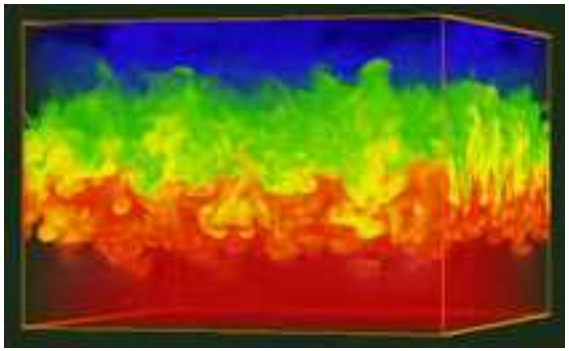


T. L. Hylton, et al, IEEE Trans. Mag., 36, 2966 (2000).

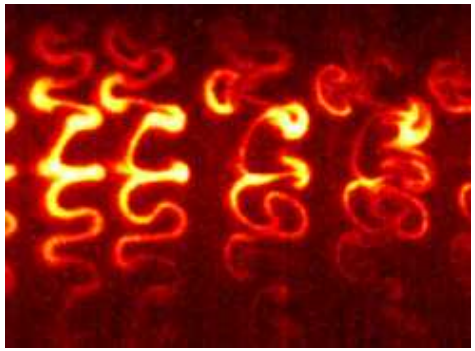
1. The Limiting Factor – Potential

❑ Temperature dependent potentials

Rayleigh-Taylor instability



Richtmyer–Meshkov instability



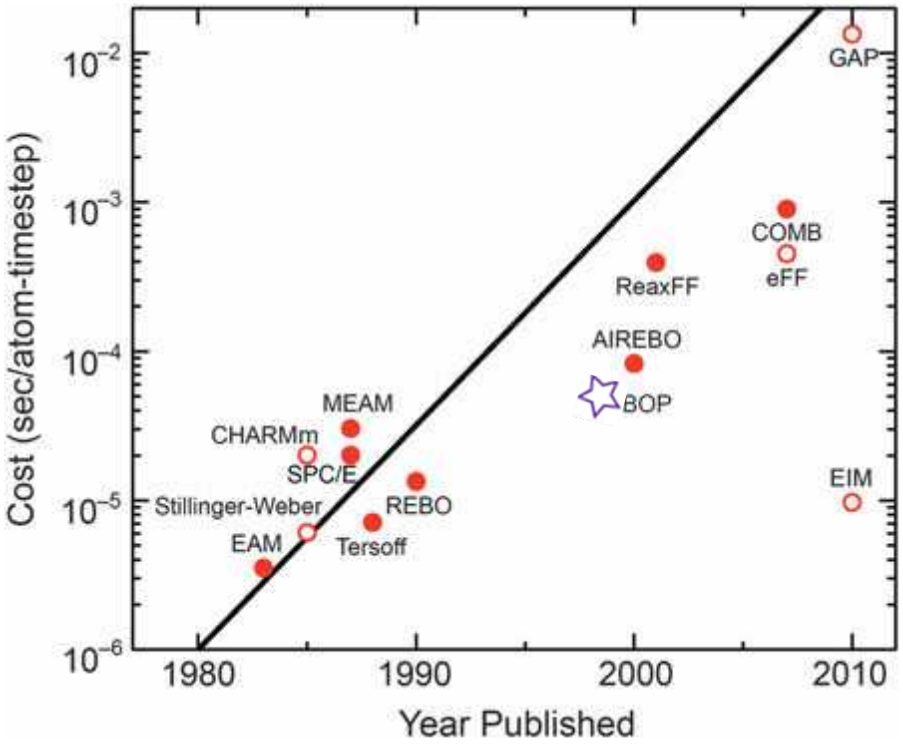
Kelvin–Helmholtz instability

The electron Force Field method recently developed by Caltech (Goddard's group) begins to address this: A. Jaramillo-Botero, J. Su, A. Qi, W. A. Goddard III, J. Comp. Chem., 32, 497, (2010)

I am seeking opportunities to work on this. The idea is to address atoms and electrons explicitly, allowing the temperature effect on interatomic potential to be incorporated without violating Newton's law

1. The Limiting Factor – Potential

❑ State-of-the-art potentials



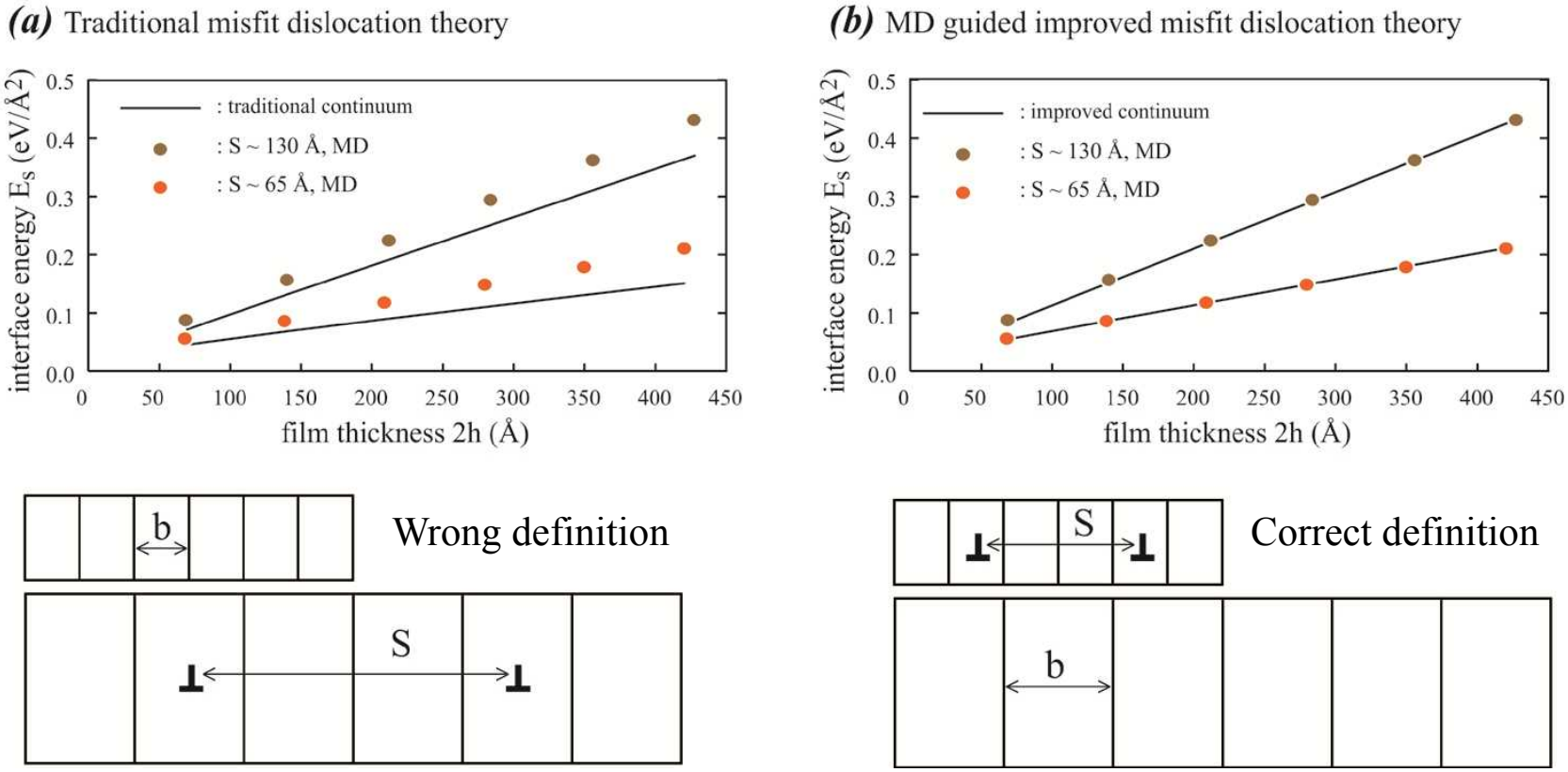
Single CPU cost.

1. **EAM (embedded-atom method);**
2. **MEAM (modified embedded-atom method);**
3. **REBO (reactive empirical bond order);**
4. **BOP (bond-order potential);**
5. **AIREBO (adaptive intermolecular REBO);**
6. **ReaxFF (reactive force field);**
7. **COMB (charge optimized many-body);**
8. **GAP (Gaussian approximation potential)/SNAP.**

S. J. Plimpton, and A. P. Thompson, MRS Bulletin, 37, 513 (2012)

2. MD Derived Continuum Rules

❑ Misfit dislocation theory



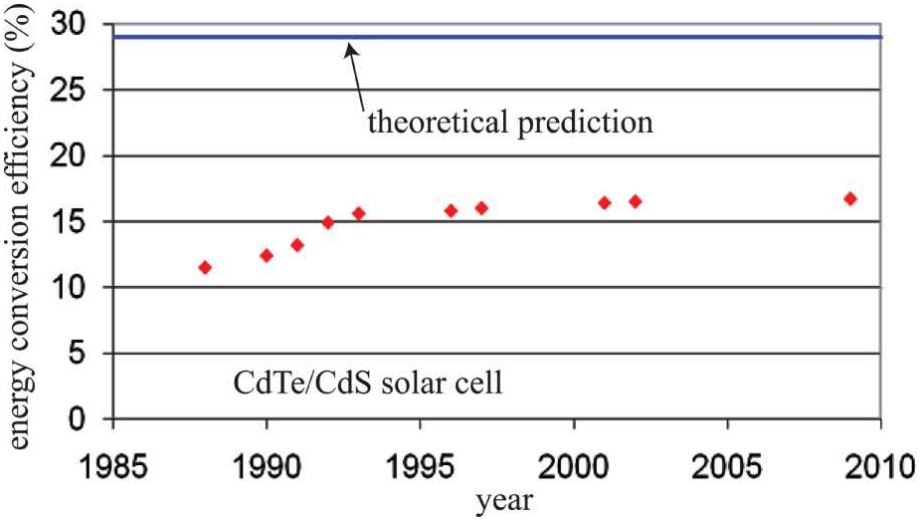
- **Continuum misfit dislocation theory has been widely used since 1980's**
- **MD simulations revealed that traditional continuum misfit dislocation theory is incorrect in the definition of dislocation Burgers vector and dislocation spacing**
- **This example indicates that MD can "validate" and improve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

3. Some Examples

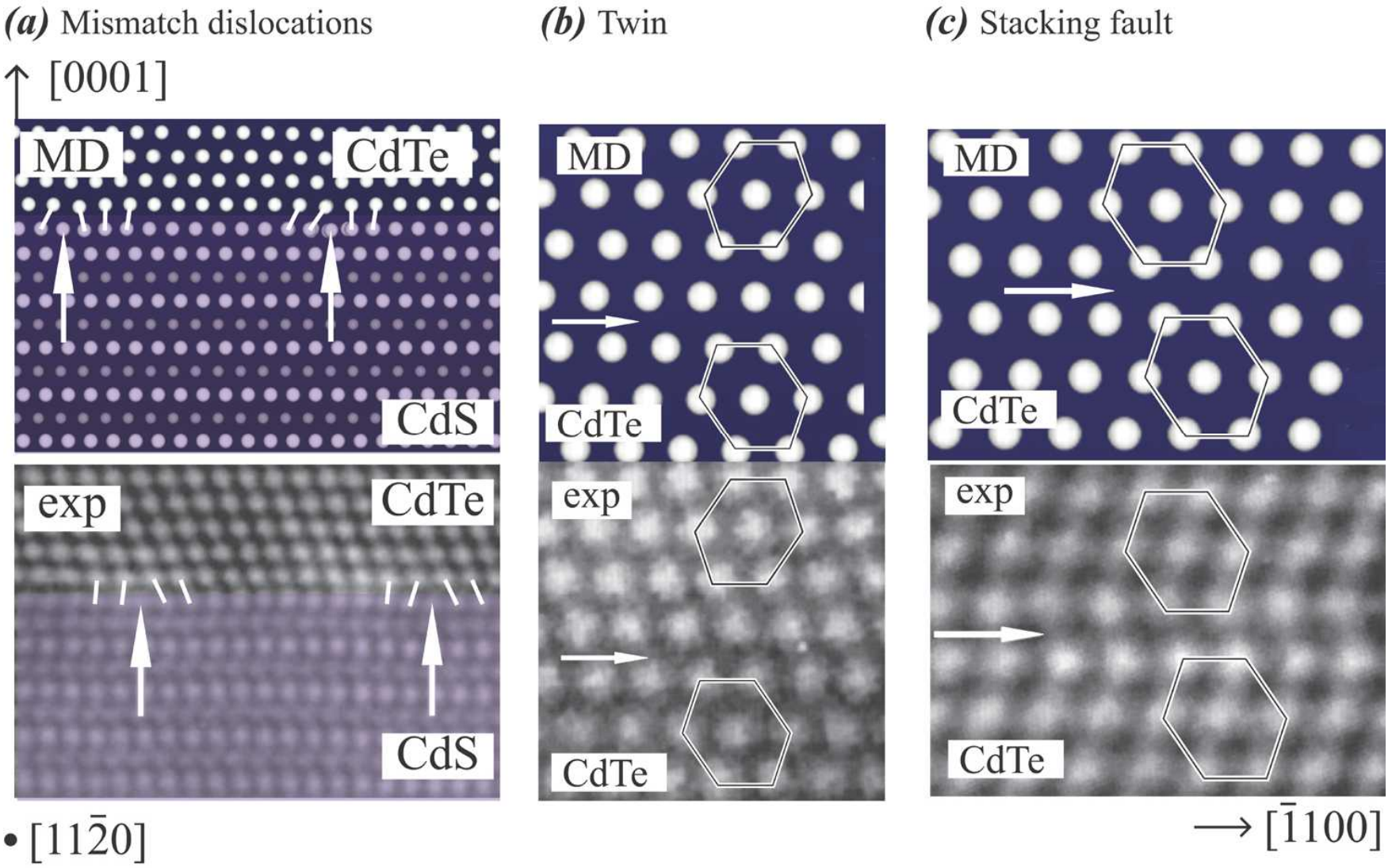
❑ CdTe/CdS solar cells

1. CdTe/CdS solar cells have the lowest cost among all photovoltaic technologies*;
2. Misfit dislocations are one of the primary defects;
3. BOP-based MD models have predicted dislocation-free CdTe/CdS solar cell structures.

* K. D. Dobson, I. Visoly-Fisher, G. Hodes, and D. Cahen, Solar Energy Mater. Solar Cells, 62, 295 (2000).

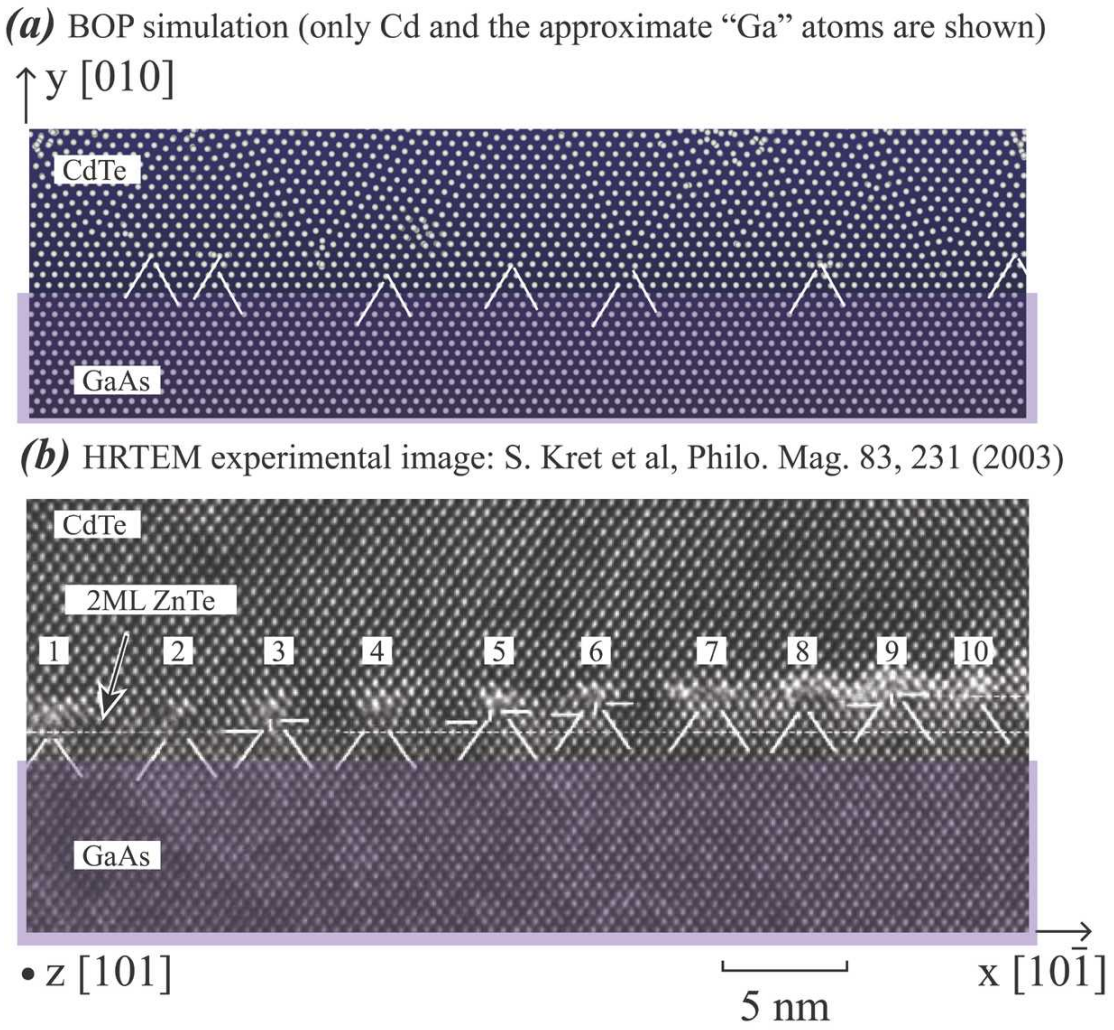


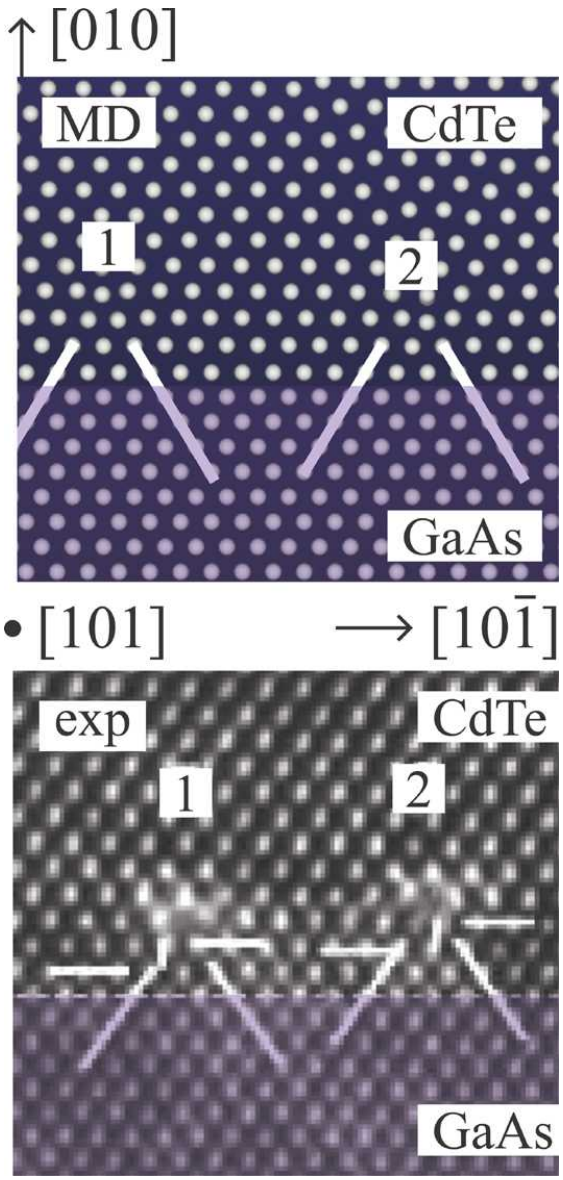
BOP validation I: CdTe/CdS defects



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

BOP validation II: CdTe/GaAs defects

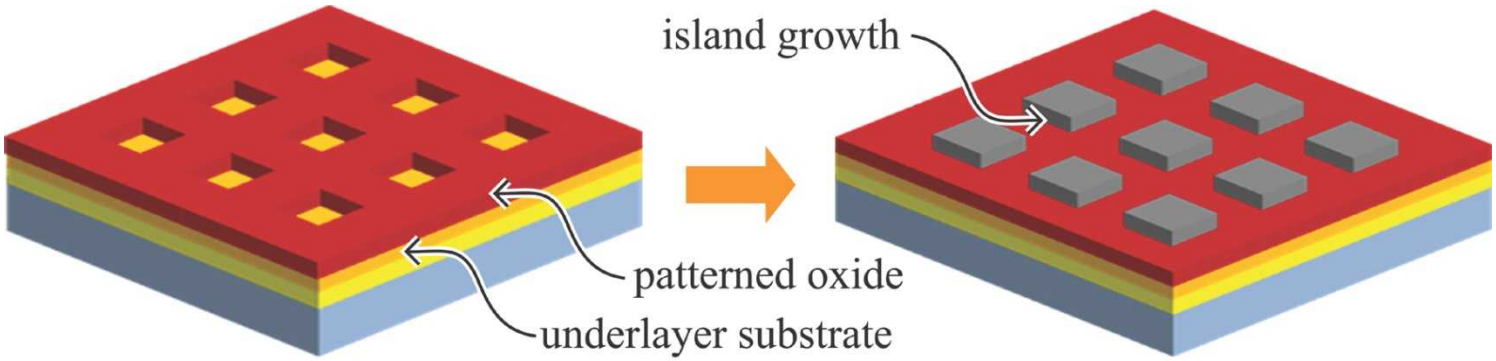




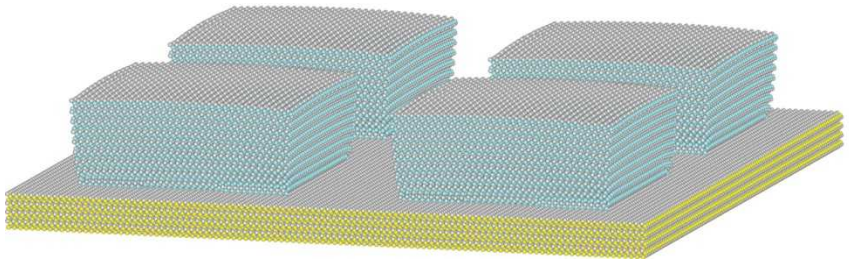
J. J. Chavez, D. K. Ward, B. M. Wong, F. P. Doty, J. L. Cruz-Campa, G. N. Nielson, V. P. Gupta, D. Zubia, J. McClure, and X. W. Zhou, Phys. Rev. B, 85, 245316 (2012).

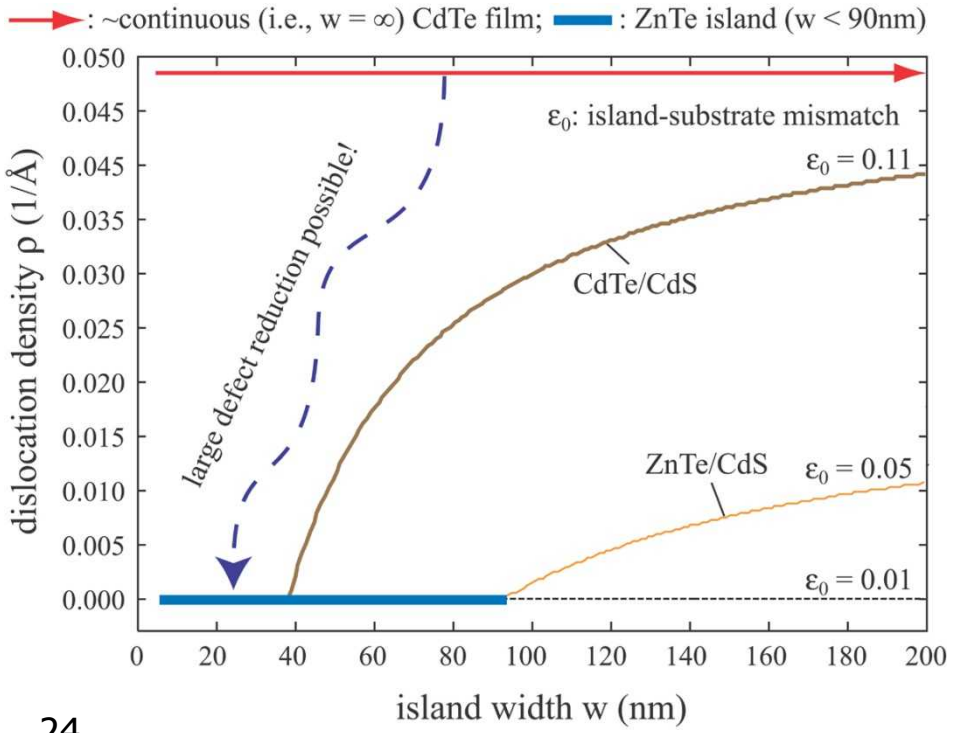
Dislocation-Free CdTe/CdS Solar Cells

Modern nano technology:



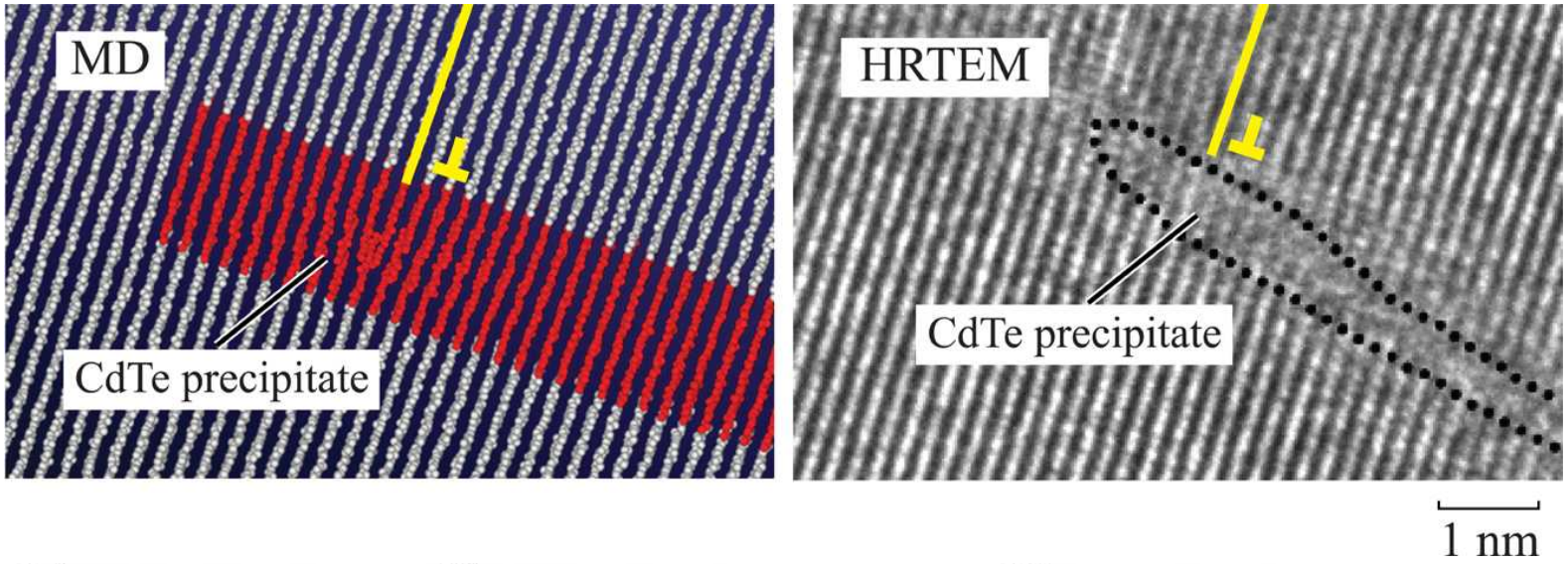
BOP simulation:

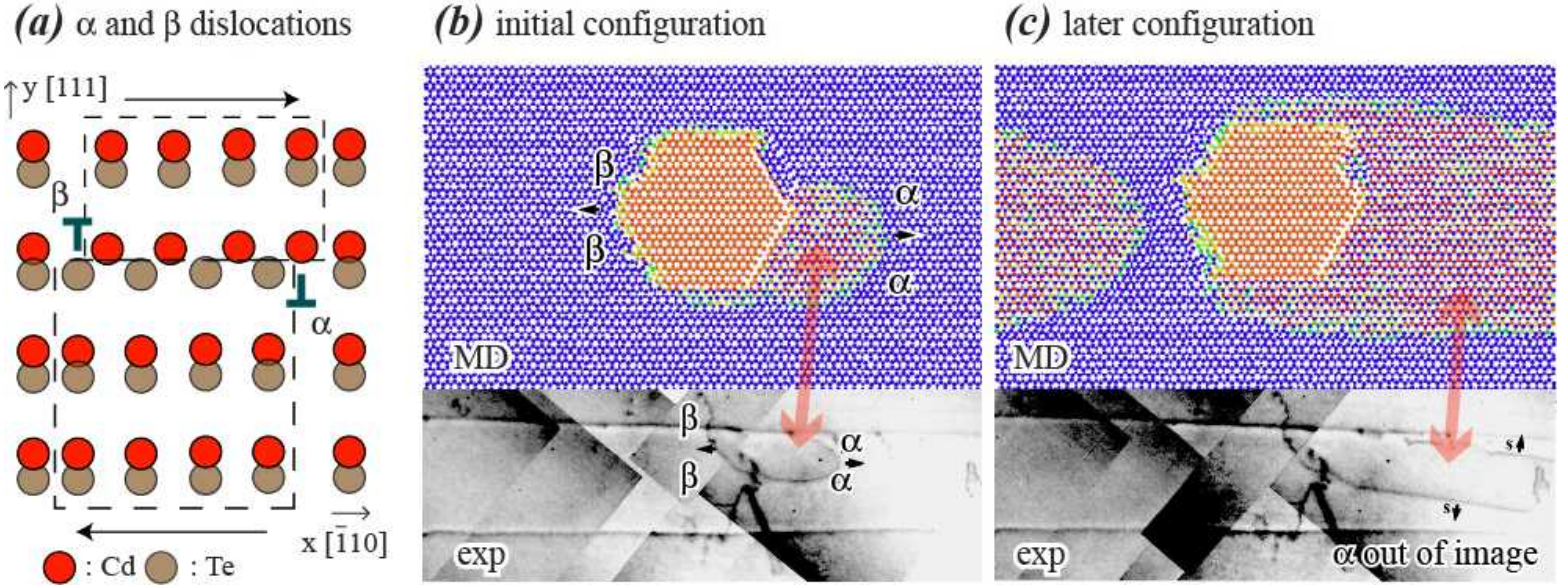




3. Some Examples

❑ Dislocation behavior in compound semiconductors

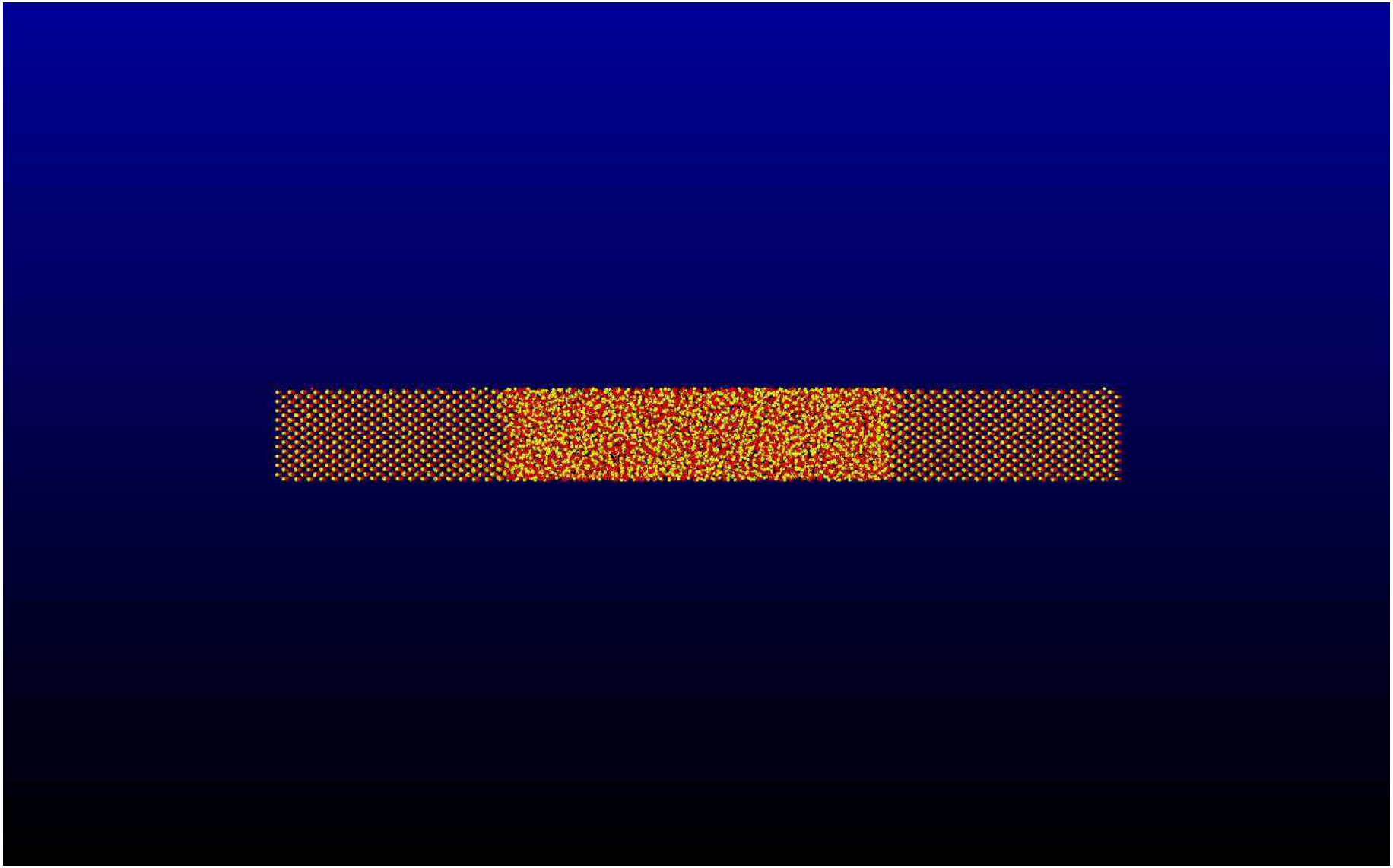




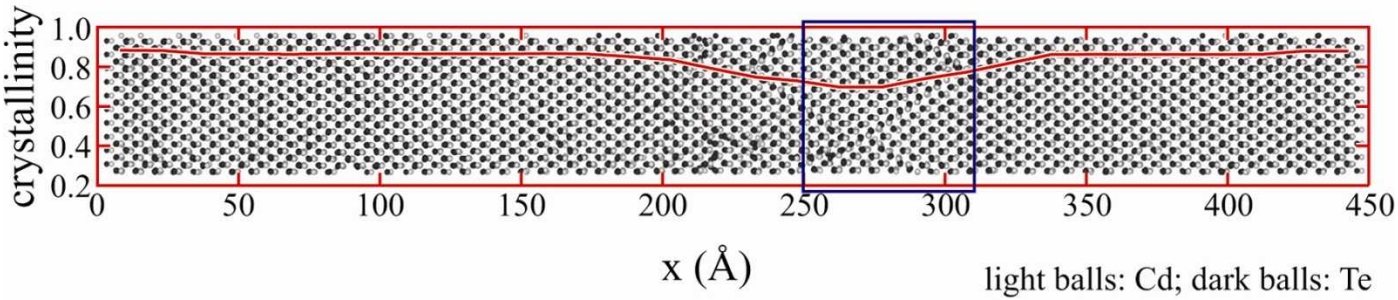
X. Zhou, D. K. Ward, B. M. Wong, F. P. Doty, and J. A. Zimmerman, J. Phys. Chem., 116, 17563 (2012)

3. Some examples

- Melt growth of CdTe

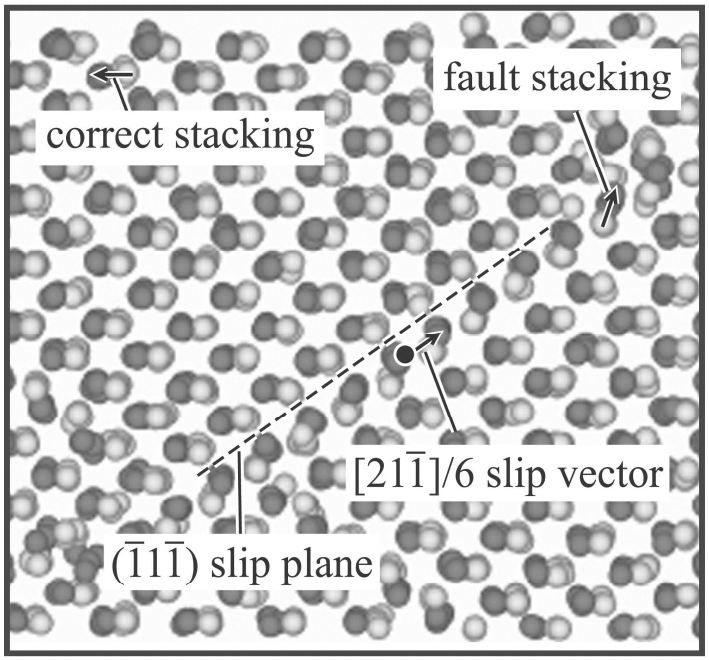


Melt growth defect formation in CdTe

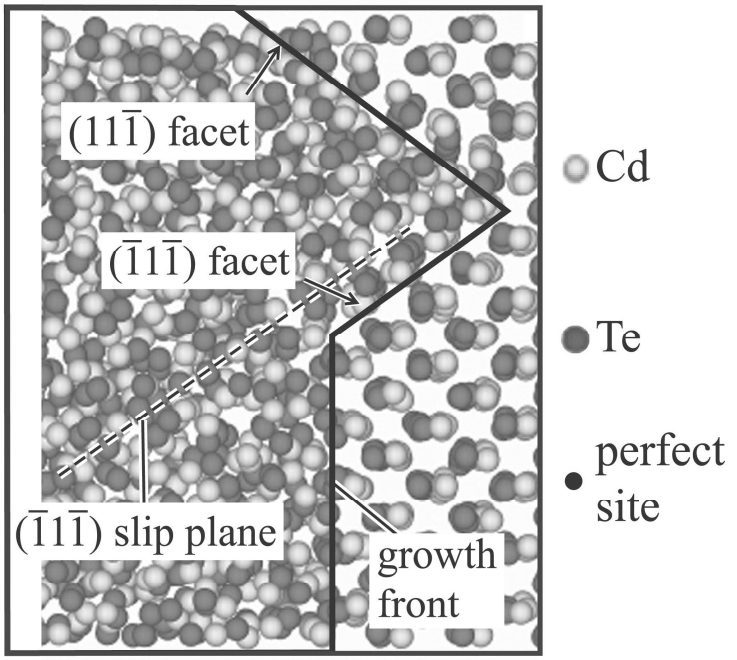


light balls: Cd; dark balls: Te

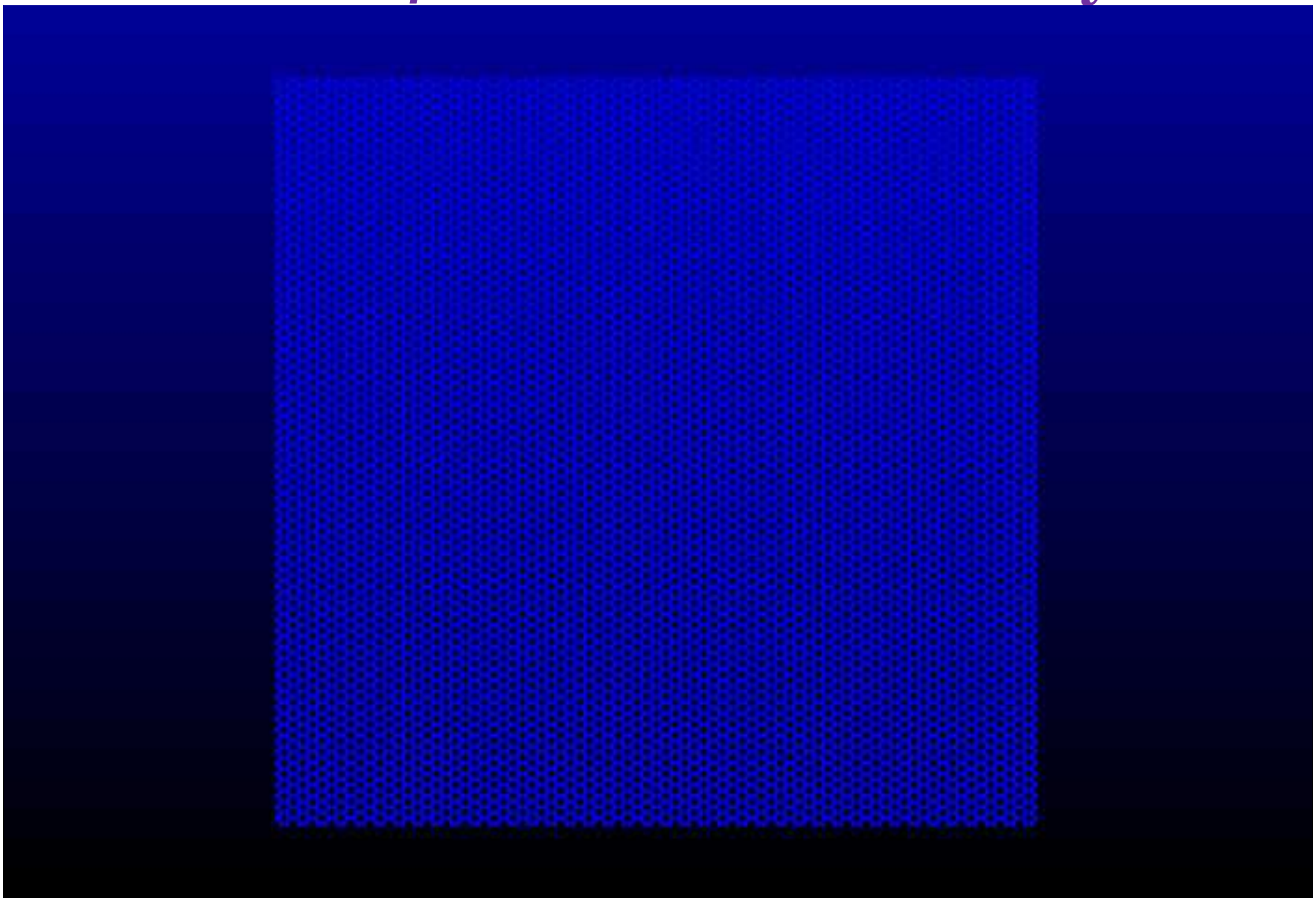
(a) framed region in Fig. 4(b)



(b) defect nucleation at local facets



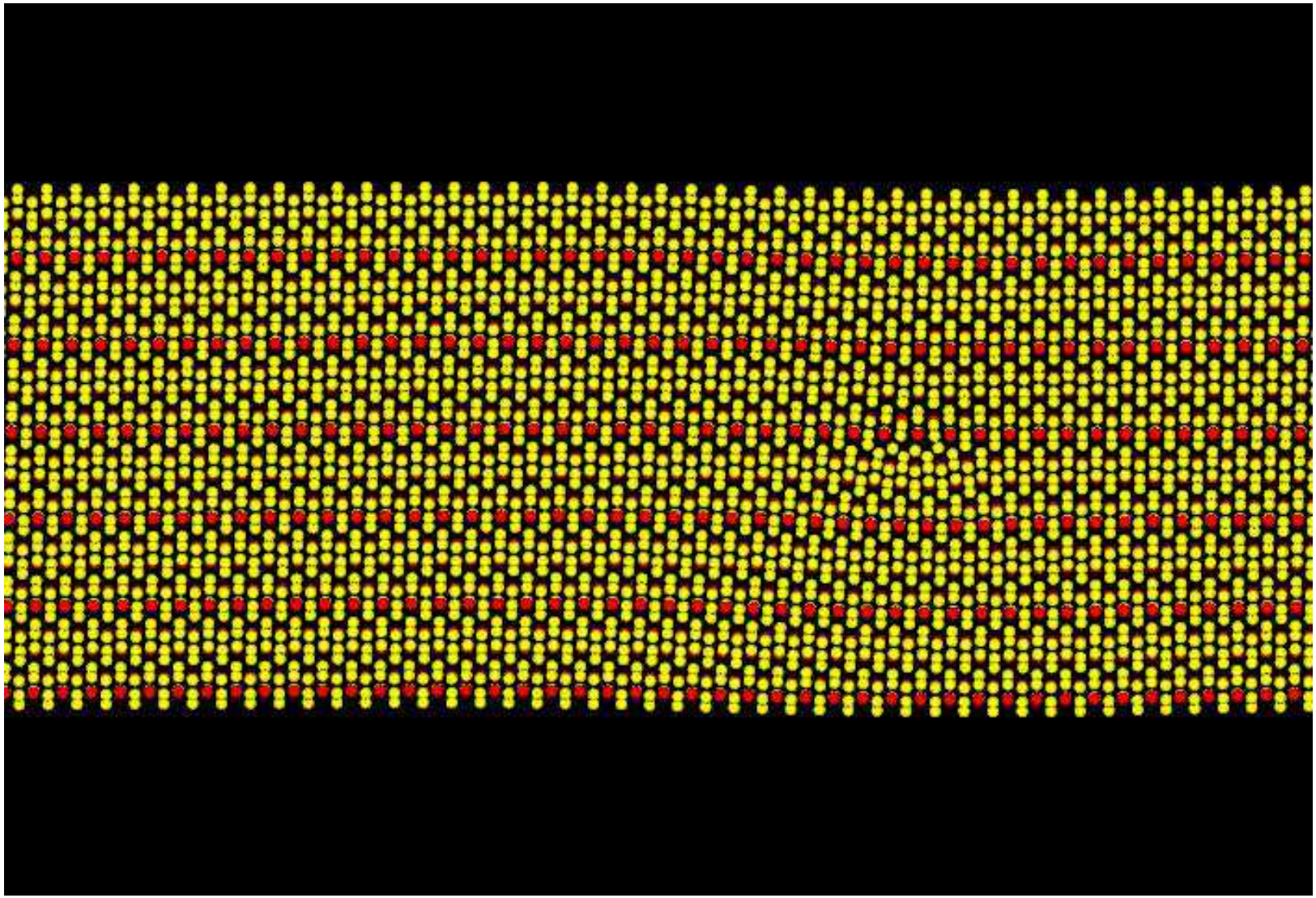
α and β Dislocation Mobility



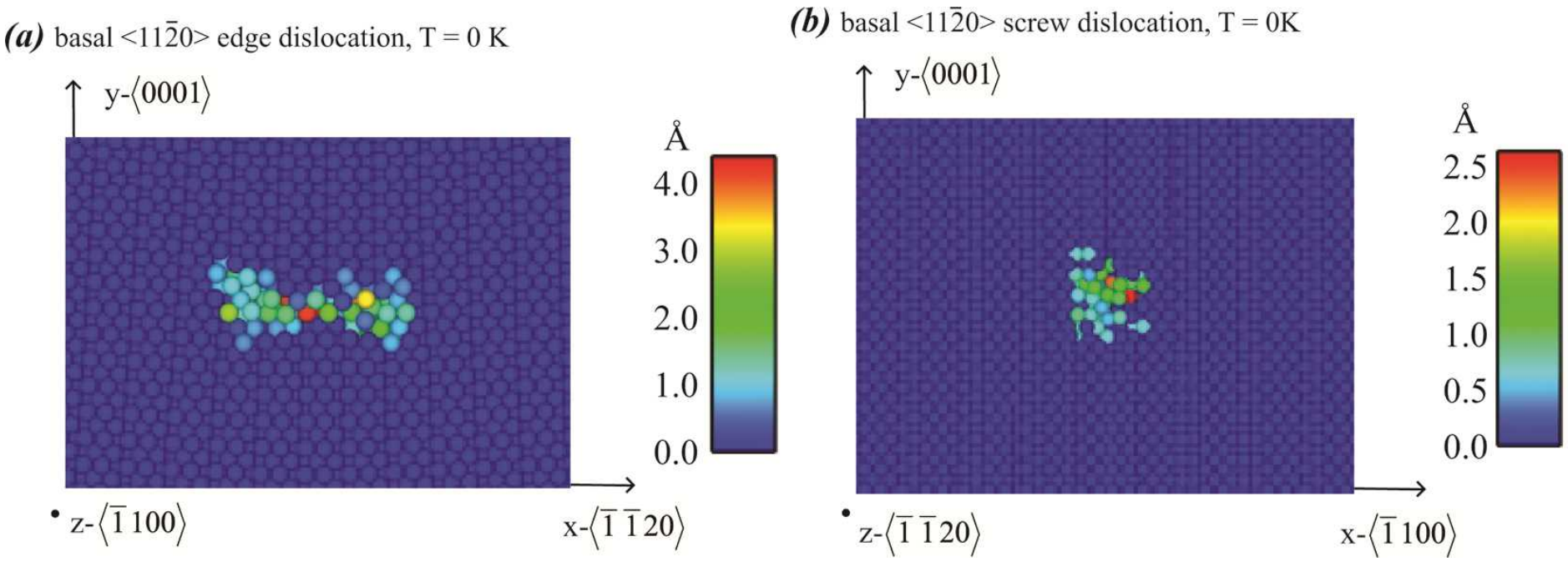
3. Some Examples

❑ Towards improved $LaBr_3$

Edge dislocation slip on <-1100>



Dislocation slip on <0001>



- **MD indicates that fracture cannot be prevented by promoting plastic deformation, but can be retarded by inhibiting dislocation motion**
- **This is proven in our experiments where aliovalent doping provides resistance to dislocation motion resulting in fracture free LaBr3 crystals**
- **A patent is issued and follow-on funding from NA22 is awarded**

Refresh, and think about 5-10 Years Later …

1. The Limiting Factor - **Potentials**
   - ❑ Growth simulation tests
   - ❑ 2D materials
   - ❑ Chemical reactions
   - ❑ Beyond EAM for metals
   - ❑ Database potentials
   - ❑ Temperature dependent potential
   - ❑ State-of-the-art potentials
2. MD Derived **Continuum Rules**
   - ❑ Misfit dislocation theory
3. Some **Examples**
   - ❑ CdTe/CdS solar cells
   - ❑ Dislocation behavior in compound semiconductors
   - ❑ Melt growth of CdTe
   - ❑ Towards improved $LaBr_3$

Thank You …