Overview of BES-GBS Project: The Role of Anisotropy on the Self-Organization of Gas Bubble Superlattice

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October 2017
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Prepared for the
U.S. Department of Energy
Under DOE Idaho Operations Office
Contract DE-AC07-05ID14517, DE-AC07-05ID14517
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September 4th, 2018

BES-GBS Project Review
Idaho Falls, Idaho
Acknowledgement

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The work was supported by the U.S. Department of Energy, Office of Science, Materials Sciences and Engineering Division under FWP C000-14-003.

The support from NSUF user project is acknowledged

The guidance and advice from the BES program managers Linda Horton, John Vetrano and Jim Horwitz are much appreciated
Outline of Project Review Presentations

- Project overview: Gan
- GBS formation windows: Sun
- Void/Bubble superlattice formation theory: Zhang
- DFT gas ↔ metal atom interaction: Jiang
- Synchrotron characterization of GBS: Ecker
**Motivation and Broad Impact**

- **Self-organization** has been widely observed in far-from-equilibrium systems in the fields of physics, chemistry, and biology.
- It can be driven by thermodynamics (e.g., crystallization), dynamics (e.g., instability), or both.
- Understanding the underlying self-organization mechanisms may help us control the pattern formation through a “materials-by-design” approach, e.g., nanopatterned structures with novel electronic, magnetic, photovoltaic, or radiation-resistant properties.
- Radiation, although often associated with damage, can be a powerful tool to create these tailored microstructures provided the physics is understood.
Key Science Questions & Hypothesis

• What drives the formation of superlattices instead of random voids or bubbles?

  Hypothesis:
  The anisotropy on elasticity and/or diffusion is responsible for the self-organization of void/bubble superlattice.

• How does the irradiation condition in terms of temperature, flux and fluence, affect void/bubble superlattice formation and their properties?

  Hypothesis:
  The self-organization involves the interplay of thermodynamics and defect dynamics, both dependent on the irradiation conditions. Therefore, the superlattice properties like superlattice parameter can be manipulated by varying irradiation conditions.

• What determines the thermal stability of void or bubble superlattice?

  Hypothesis:
  Voids and bubbles in superlattice are thermally more stable than random voids and bubbles. But they are not thermodynamically stable and they can coarsen upon annealing. The coarsening kinetics depends on the ordering and the type of gas atoms.
# Materials and Properties to address Hypothesis

<table>
<thead>
<tr>
<th>Property → Material</th>
<th>Elastically</th>
<th>SIA Diffusion</th>
<th>Note</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fe</td>
<td>anisotropic</td>
<td>isotropic</td>
<td>1-D diffusion for small int. clusters</td>
</tr>
<tr>
<td>W</td>
<td>isotropic</td>
<td>anisotropic</td>
<td>1-D along &lt;111&gt;</td>
</tr>
<tr>
<td>Mo</td>
<td>anisotropic</td>
<td>anisotropic</td>
<td>1-D</td>
</tr>
<tr>
<td>W(x) x=Re or Fe</td>
<td>isotropic</td>
<td>isotropic</td>
<td>To be acquired or fabricated</td>
</tr>
</tbody>
</table>

**Inert Gas Atom Size**

- **He**
- **Ne**
- **Ar**
- **Kr**
- **Xe**

Shown in atomic diameter computed using quantum mechanical calculation (0.98 Å for He, 2.48 Å for Xe)

$\left(\frac{2.48}{0.98}\right)^3 \approx 16$
Integrated Research Approach

Modeling

Anisotropic elastic interactions from MD and experiments

AKMC

Phase field

Anisotropic defect diffusion from MD and DFT

Moose

Gas-metal interaction from DFT

Experiment

Theory

Gas implantation

Superlattice

- Symmetry
- Parameter

Material

- Elasticity
- Defect kinetics
- Grain size
- Dislocation

Condition

- Temperature
- Dose rate
- Dose
- Gas type

TEM characterization

7-10 at % He, depth: 300-450 nm

Synchrotron measurement

3D GIAMS patterns
Beyond FY19

- $700k/yr* + $300k seed.
- This project is crucial to promote science and fundamental research at INL.
- This BES core project has a large impact.
The Synergies between INL and BNL

- Joint effort on proposal development.
- Complementary and unique strength, experience and capabilities.
- INL leads effort on irradiation, TEM characterization and modeling.
- BNL leads effort on synchrotron x-ray characterization.
- Bi-weekly BlueJean teleconf promotes new ideas, coordinated research, joint publication and in-depth discussion on the work in progress.
- Synergy on this project promotes joint research proposal between INL-BNL in other research areas.
BES & NE User Facilities are Critical to Facilitate Synergies
User Proposals Awarded (Led by INL, 6)


Pending:
**User Proposals Awarded (Led by BNL, 5)**


Published (5)


Submitted (2)

Presentations (2 meetings, 3 conf., 3 seminar)

- L. He, et al. (poster), “Advanced microstructural characterization of nuclear fuels”, NS&T Strategic Advisory Committee Meeting, Idaho Falls, May, 2018
Significant Outcomes

Much better correlation with shear modulus than charge density

MoHe

Ex-situ heating result agrees with in-situ heating result.
Coarsening leads to He GBS disorder at 1000°C/60min

Mo irradiated at 300 °C

This work is the first in this area not found in literature, a journal paper is published
What have we learned so far?

1. What define the lattice parameter and structure of void superlattice? A complete rate theory formulation was developed which can now predict both lattice parameter and structure. The predictions are consistent with experiments and simulations with regards to the effects of temperature, flux and fluence (Zhang’s talk).

2. How radiation conditions in terms of temperature, flux and fluence affect gas bubble superlattice characters: lattice parameter and structure? Radiation conditions for He GBS formation have been successfully established experimentally, with TEM analysis and non-destructive and high-resolution synchrotron X-ray techniques to determine lattice parameter and structure, and lattice strain and pressure in GBS (Ecker & Sun’s talk).

3. How gas atom interact with lattice atoms? A unified model with four input parameters trained by first principle calculations is developed to describe the incorporation energy of various inert gases in many bcc metals. (Jiang’s talk)

What we will answer in a short future

1. Are void and gas bubble superlattice (with various inert gases) fundamentally different? They share the same lattice structure; the theoretical predictions on void superlattice seem applicable to gas superlattice as well; their formation processes can be similar. These suggest void and gas superlattices may be explained using a unified theory, particularly when elastic strain energy is not important.

2. What is the effect of gas bubble pressure? Over-pressurized bubbles could cause different behavior, particularly elastic strain energy. This will be studied in future work.

3. Is effect of elasticity anisotropy important for large gas atoms? This will be studied in future work.
Other Important Questions

- Why is Kr and Xe GBS so difficult to form under ion irradiation?
- Why does Xe GBS in U-Mo have fcc structure on bcc host metal?
- Why Xe GBS is much better ordered than that of He GBS?
- Why void superlattice is in general much better ordered than that of He GBS?
- Why void thermal stability in void superlattice in Mo is much higher than that of isolated voids (1500 °C vs. 1100 °C)?
- Why Xe GBS thermal stability is much higher than He GBS (0.78T_m vs. 0.44T_m)?
SUMMARY

• A rate theory based instability analysis has been developed which can predict both lattice parameter and structure of void superlattice without using any fitting parameters. The theoretic predictions are consistent with both experiments and atomistic simulations.

• DFT calculation on inert gas atom interaction with bcc metal atoms generated important insight to help understanding the GBS formation mechanism and thermal stability.

• Many similarities have been observed between gas bubble and void superlattices, suggesting that they may form by the similar mechanisms, particularly in situations without substantial contribution from elastic strain energy.

• The synergies between TEM and Synchrotron analysis, and the experimental and modeling are critical to this BES project.

• We are facing the big challenges to close the knowledge gap on (1) creating Kr or Xe GBS through ion irradiation and (2) fully understanding its exceptional thermal stability (3) explaining the exceptional structural relationship (fcc on bcc). This is a critical step and it has never been done before. The predictions from modeling will be used to optimize the experimental conditions.