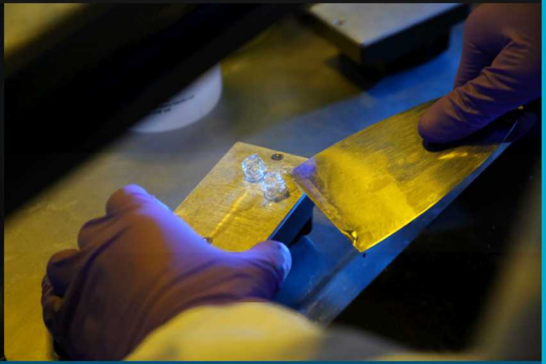
SAND2018-5113C

Nucleation and Growth of Helium Nanobubbles in Palladium Alloy Tritides



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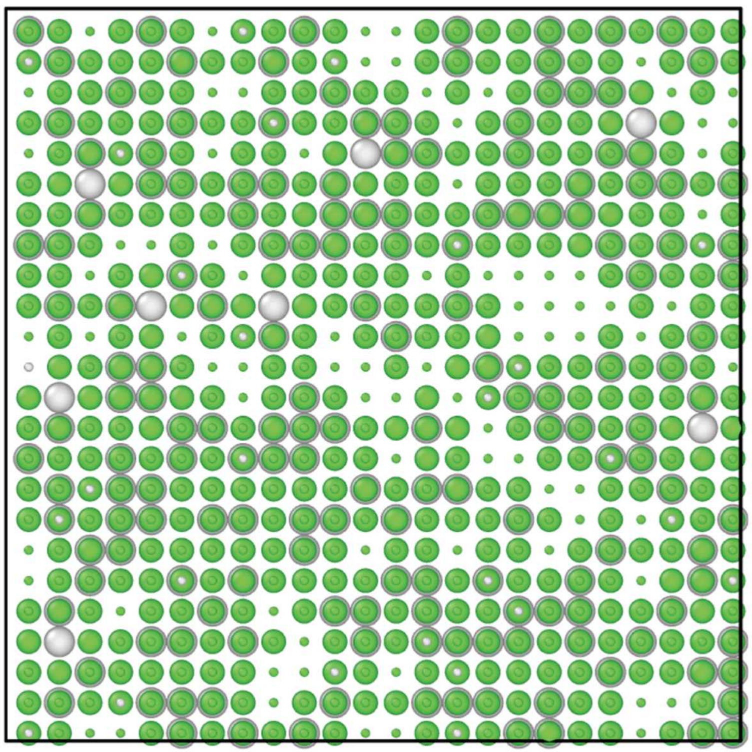
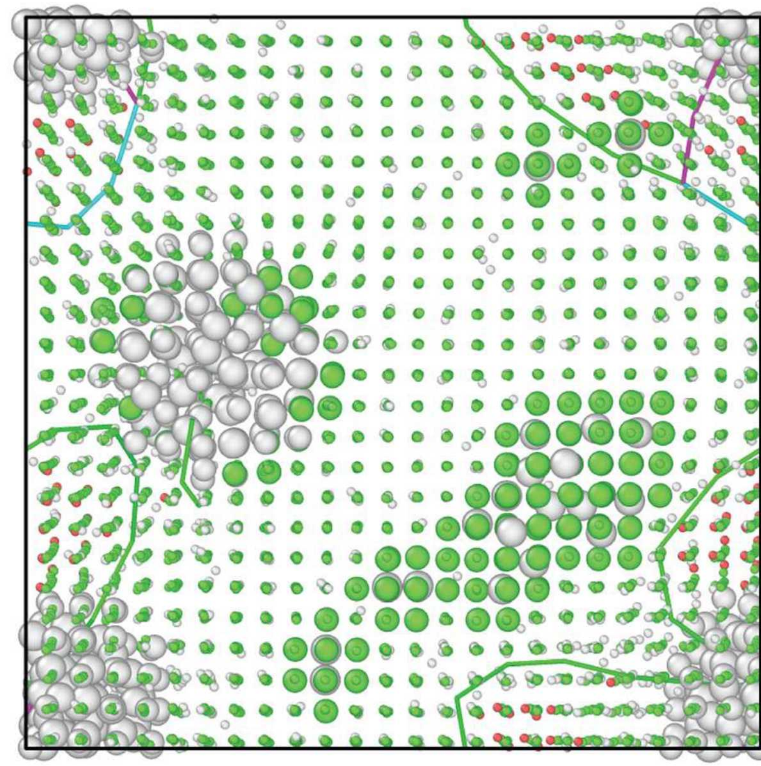
Tritium decays to insoluble helium-3 within metal lattices



^{3}H decays to ^{3}He without damage to lattice

^{3}He occupies interstices



Interstitial ^{3}He diffuses rapidly

^{3}He becomes trapped at point defects

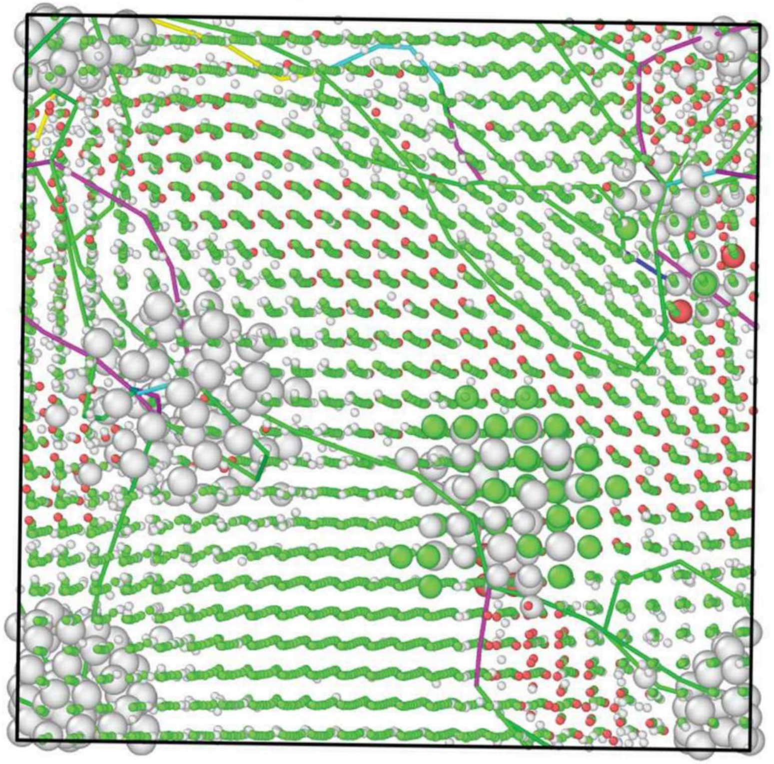
Pd

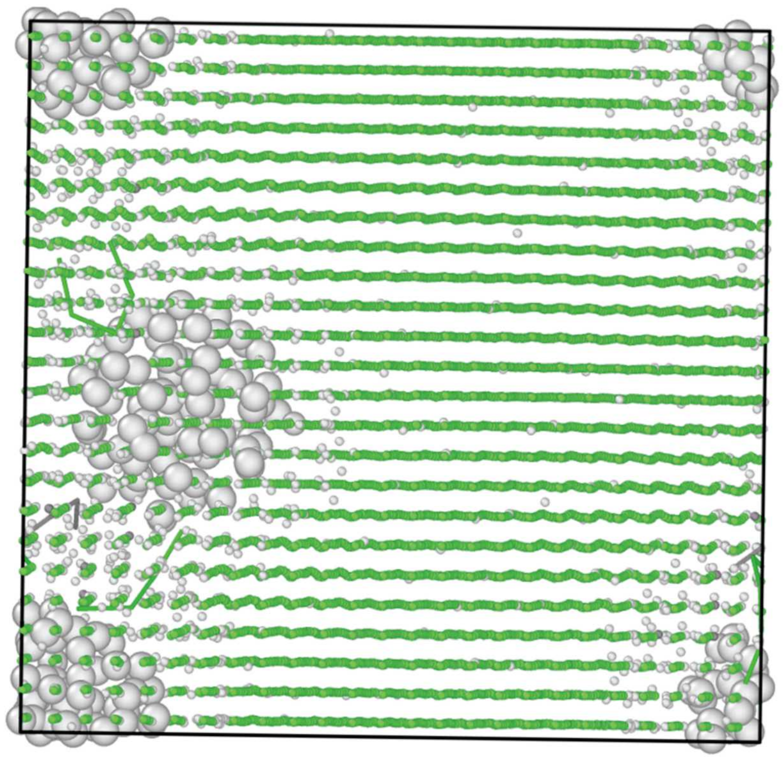
Pd with ^{3}He in O_h hole

^{3}He at lattice site

^{1}H

Helium-3 forms nanobubbles that cause defects in metal





^{3}He forms clusters

Clusters displace metal atoms and cause dislocations

Clusters diffuse within lattice

Clusters form bubbles that grow in volume

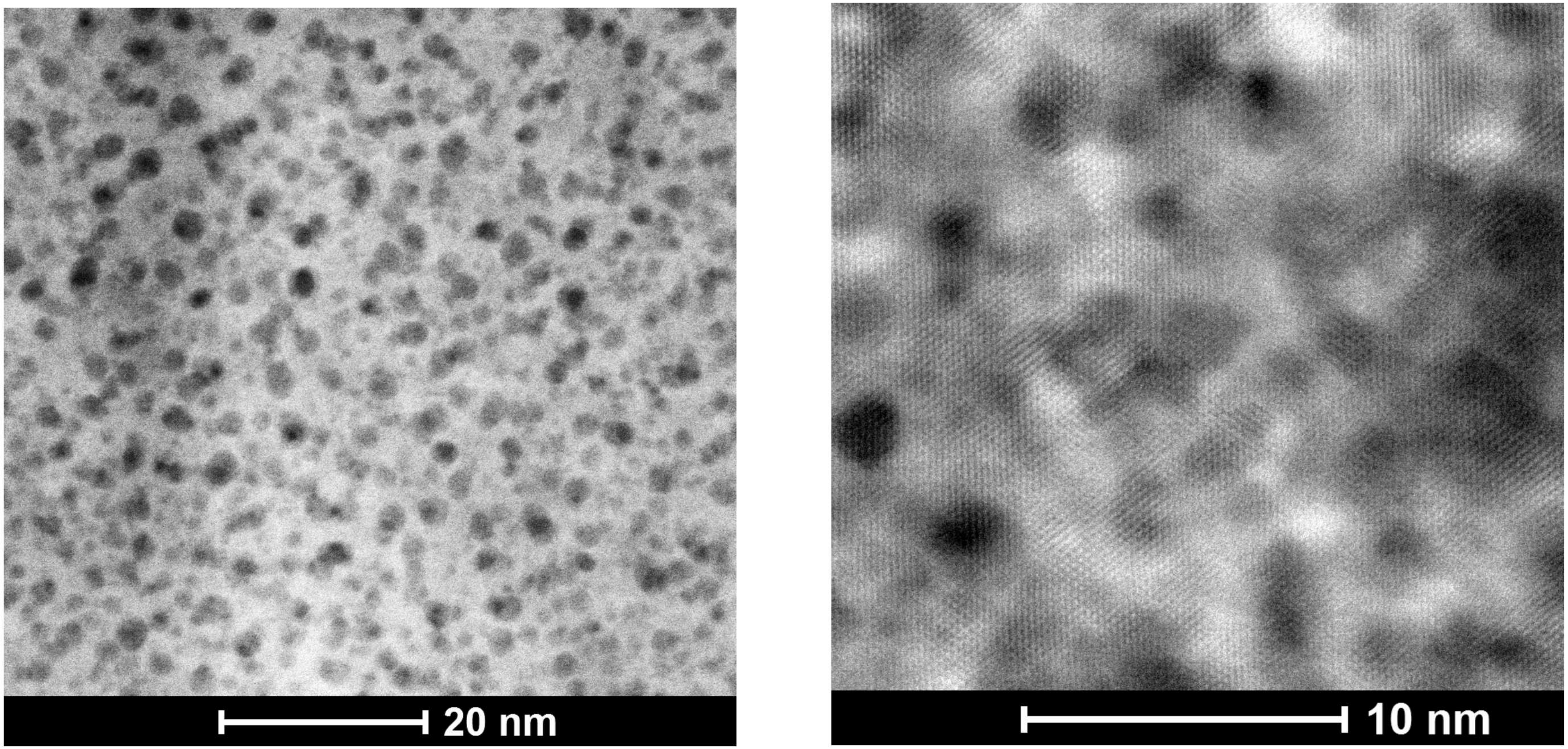
Pd

Pd with ^{3}He in O_h hole

^{3}He at lattice site

^{1}H

Helium nanobubbles were found in a PdNi tritide



High-angle annular dark field STEM images

Thinned section of a PdNi alloy previously exposed to tritium

Bubbles appear dark

Bubbles are a few nm in diameter and spaced a few nm apart

Open questions of helium bubble nucleation and growth

Questions

- What determines helium bubble size and spatial distribution?
- Do all helium bubbles nucleate within a narrow time range?
- What mediates bubble nucleation?

Goals

- To develop a model for helium bubble formation and evolution
- To validate model with experimental observations

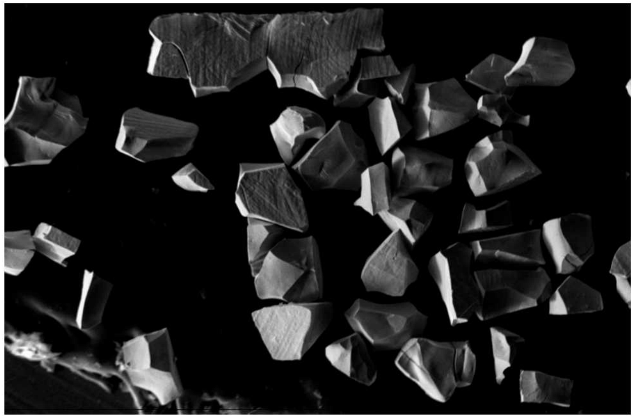
Approach

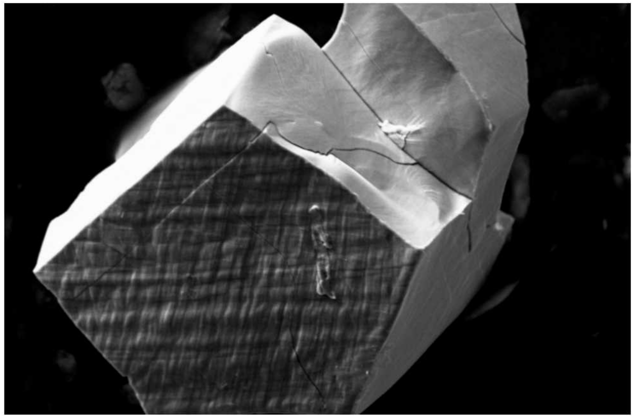
- Generate three-dimensional maps of helium bubbles trapped in metals by electron tomography
- Detect helium and measure pressure by electron energy loss spectroscopy (EELS)

Overarching aim is to develop a well-validated, comprehensive model for the nucleation, growth, and release of helium bubbles in metals

Recent prior work: (a) Cowgill, D. F. *Fusion Sci. Tech.* **2005**, *48,* 539. (b) Fabre, A.; Decamps. B.; Finot, E.; Penisson, J. M.; Demoment, J.; Thiébaut, S.; Contreras, S.; Percheron-Guegan, A. *J. Nuc. Mater.* **2005**, *342,* 101. (c) Montheillet, F.; Delaplanche, D.; Fabre, A.; Munier, E.; Thiébaut, S. *Mat. Sci. Eng. A* **2008**, *494*, 407.

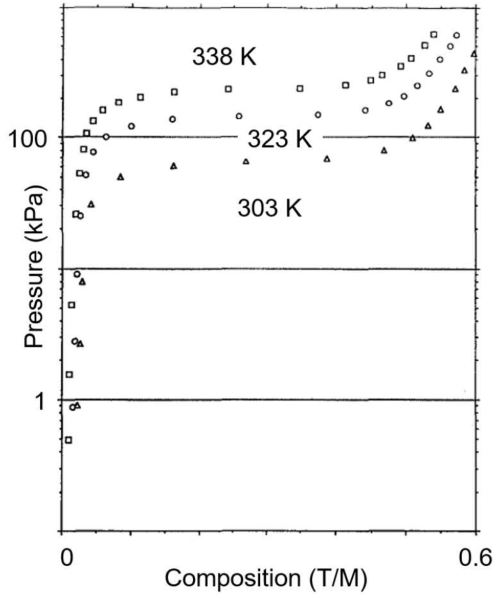
PdNi foil was exposed to tritium, aged, and decontaminated





SEM images

Tritium desorption isotherms



Sample characteristics

5 atom % Ni

Aged 3.8 years

Foil fragmented during aging

He/Pd ratio: 0.12

Tritium removed for analysis by cycling D_2 and vacuum near room temperature

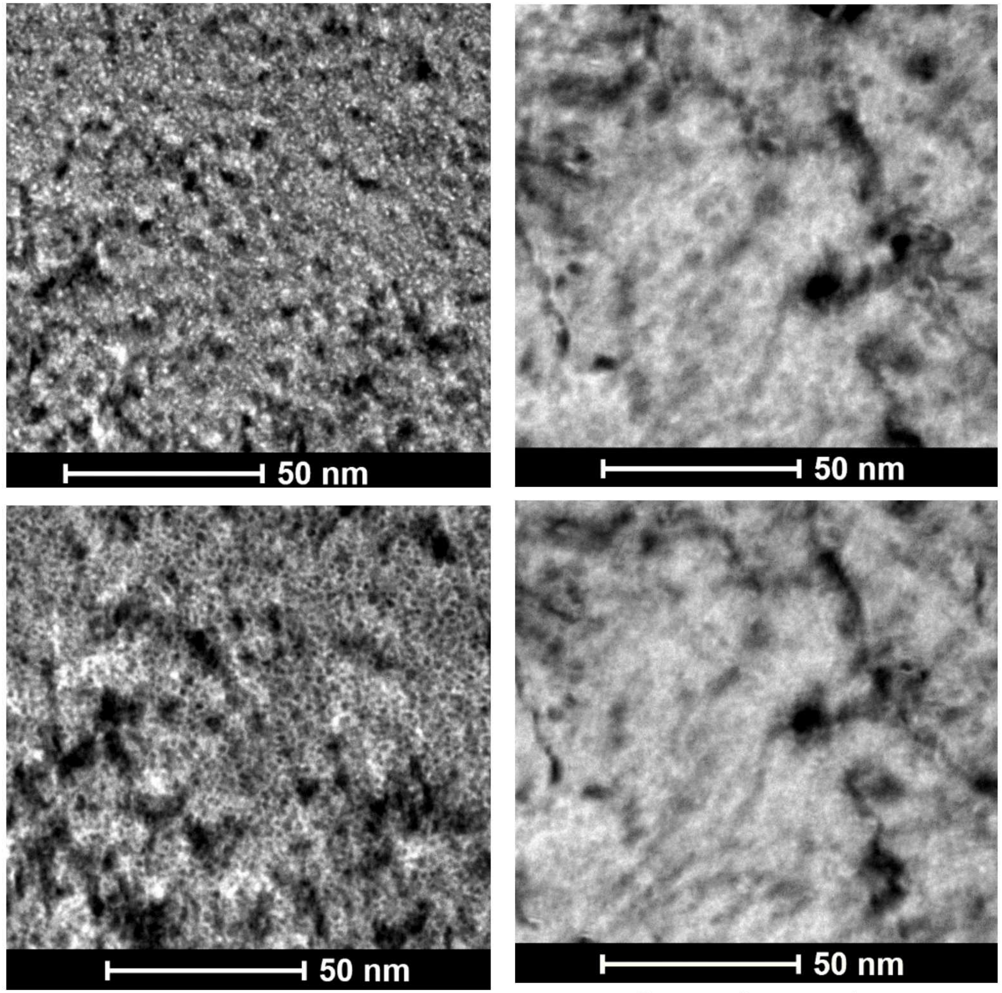
40 μCi / g by dissolution followed by liquid scintillation counting

(a) Hölder, J. S.; Wermer, J. R. *J. Alloys Compounds* **1995**, *231*, 773. (b) Shanahan, K. L. *Fusion Sci. Tech.* **2017**, *71*, 555.

Fresnel contrast seen in tritium-exposed sample but not in control

Under-focused images

Over-focused images



Tritium-exposed sample

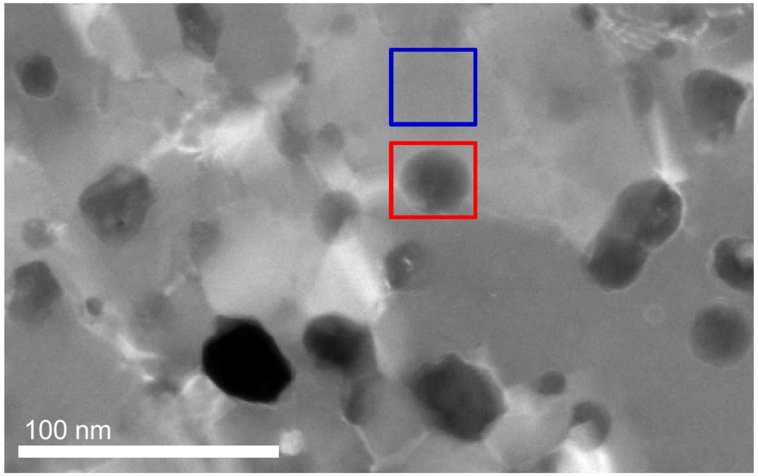
Control sample

Change in contrast with focus indicates round features (bubbles) are regions of lower density

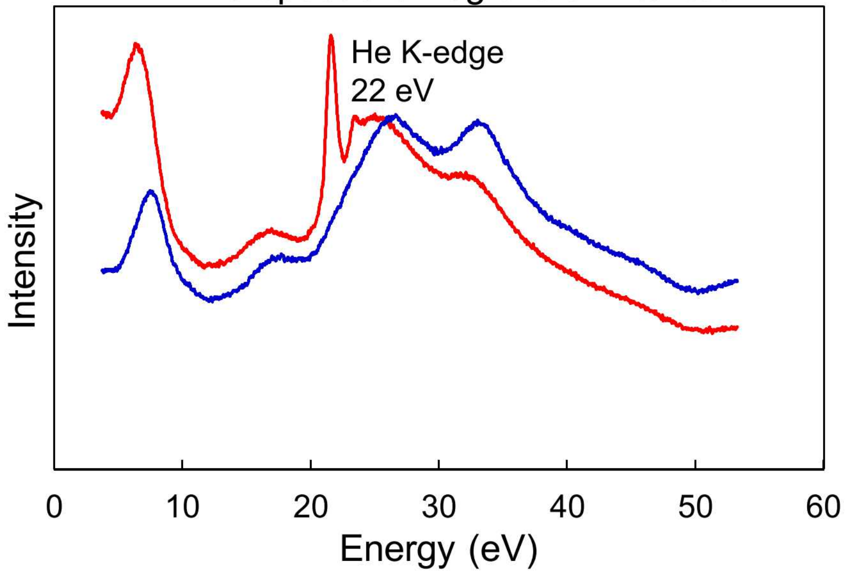
No contrast reversal occurs in the PdNi control sample that was never exposed to tritium

Electron energy loss spectroscopy detects He in He^+ implanted Pd

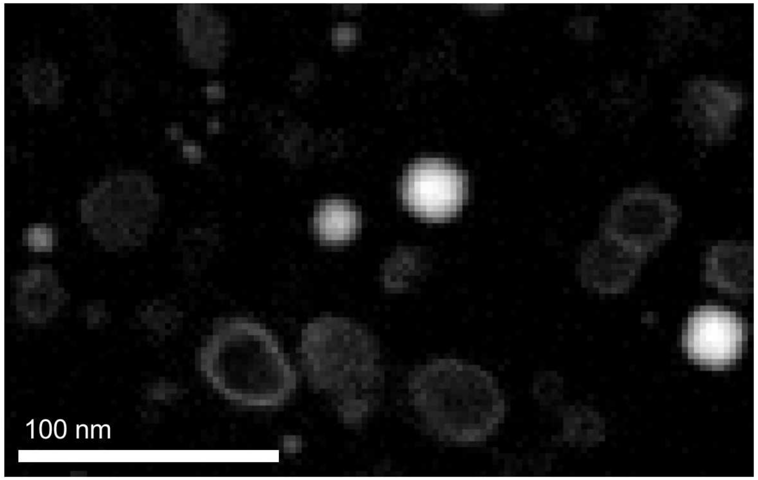
Annular dark-field STEM spectrum image



EELS spectra of regions of interest



Map of He K-edge



Pd implanted with 1 x 10^{17} helium ions / cm^2

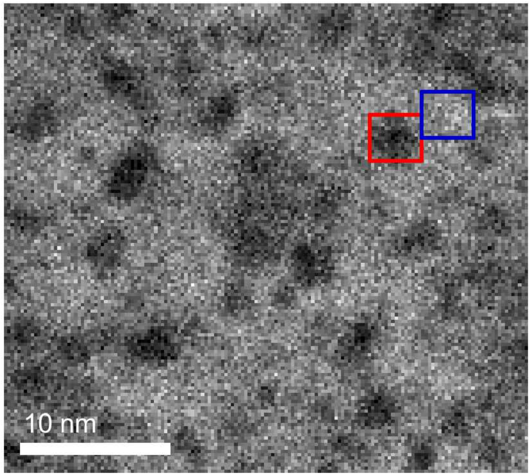
Annealed at 600°C for 2 hours

Large, sparse, low-pressure bubbles

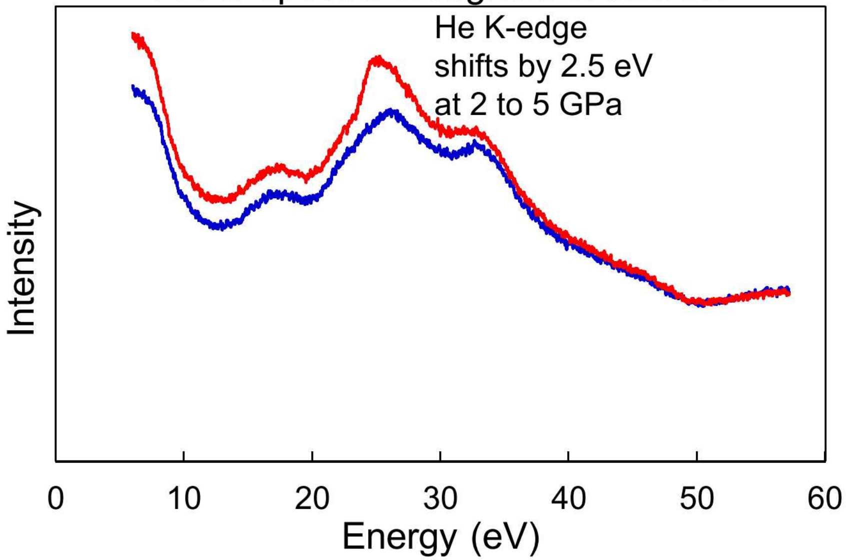
He signal maps on to bubble locations

Electron energy loss spectroscopy detects He in tritium-exposed PdNi

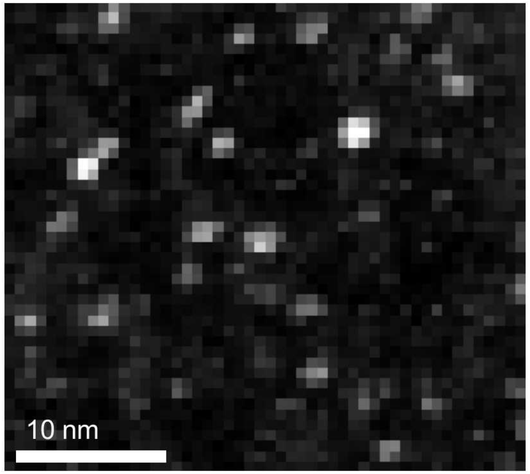
Annular dark-field STEM spectrum image



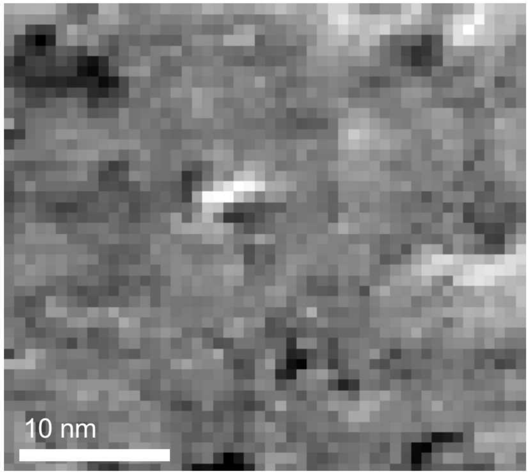
EELS spectra of regions of interest



Map of shifted He K-edge

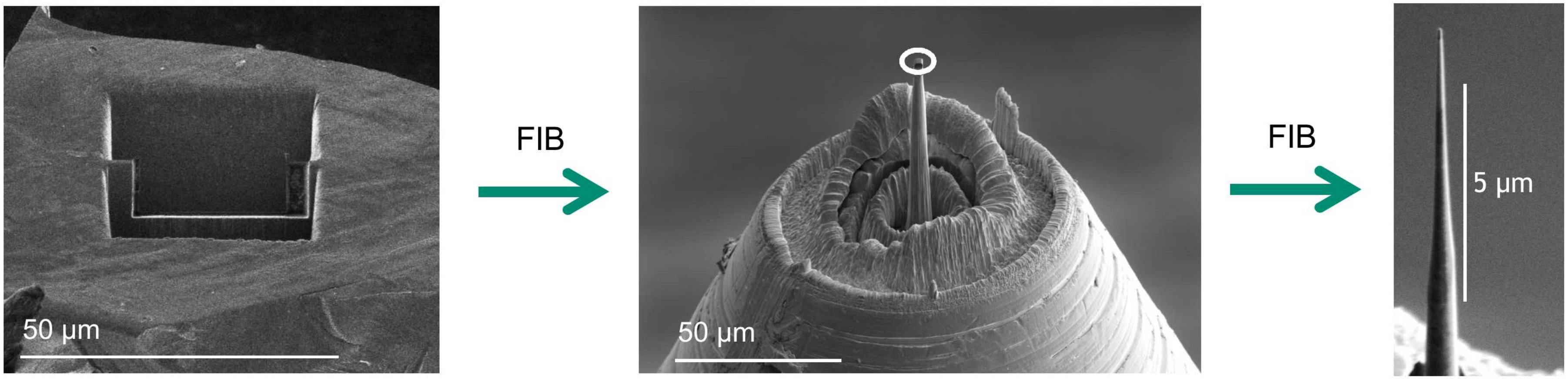


Map of Pd spectrum



Walsh et al., Phil. Mag. A 80 1507 (2000); Taverna et al., Phys. Rev. Lett. 100 035301 (2008).

PdNi was thinned to a tip for collection of tomography data set

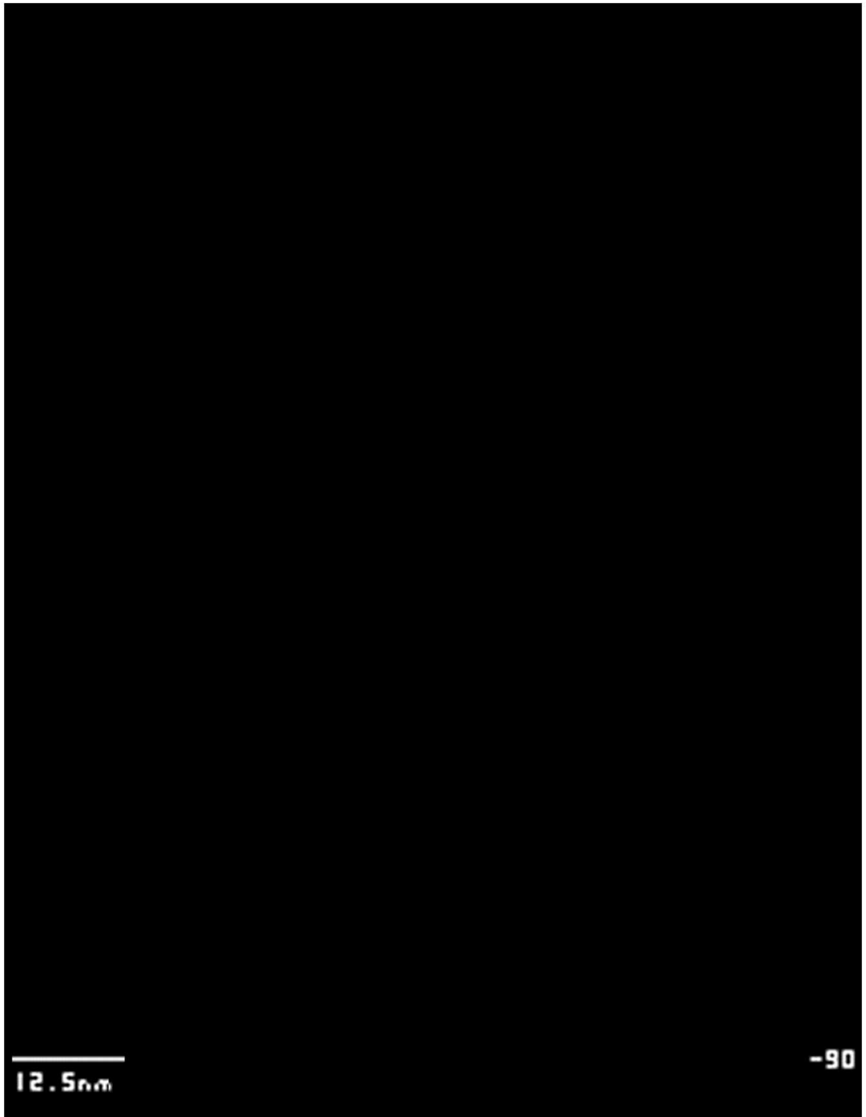


SEM images

Measurement of 3D bubble volumes and locations are needed for validation of He transport models

Specimen from PdNi particle was extracted and thinned with a focused ion beam to acquire a series of images at various angles by tilting sample

Set of images for tomographic reconstruction



Series of images from -70° to 70° in 1° increments

First set of images:

- 2D high-angle annular dark-field STEM images
- Raw images from microscope
- Helium bubbles appear dark

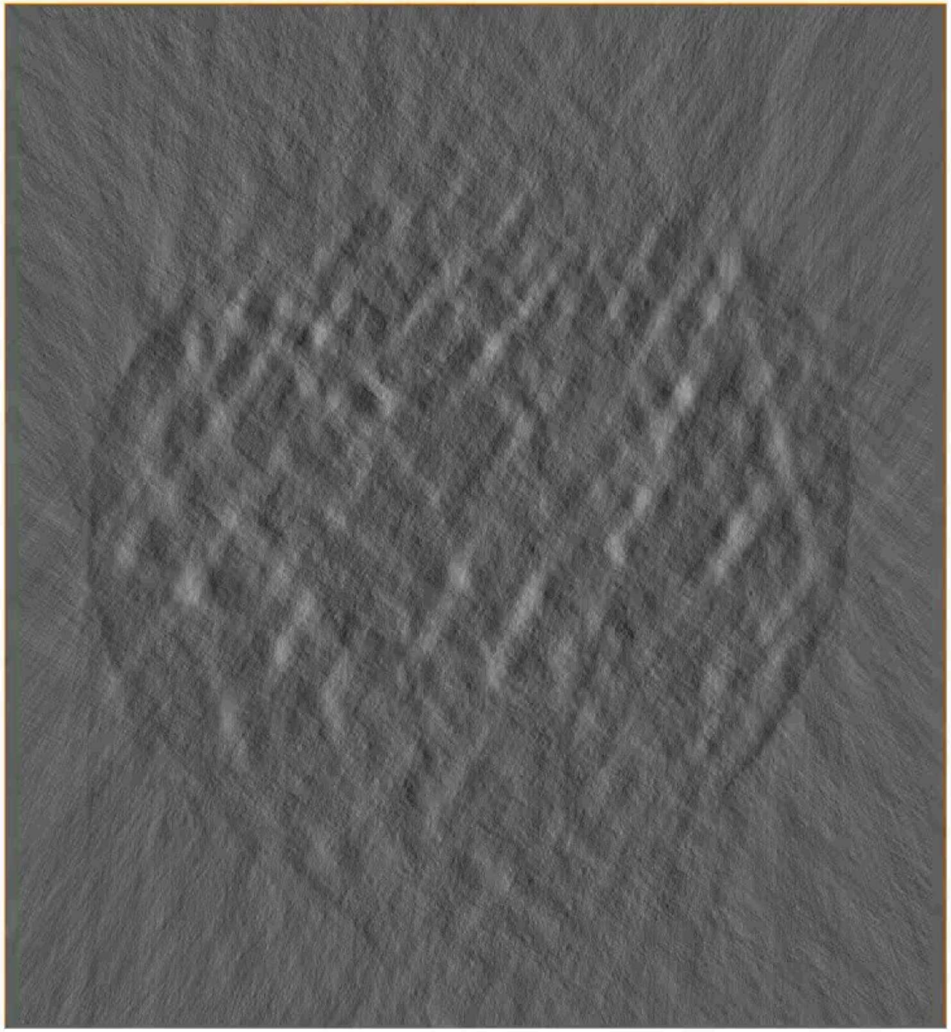
Second set of images:

- Filtered, contrast-inverted images
- Low-frequency background removed
- Input into reconstruction algorithm

3D reconstruction of TEM tomography images

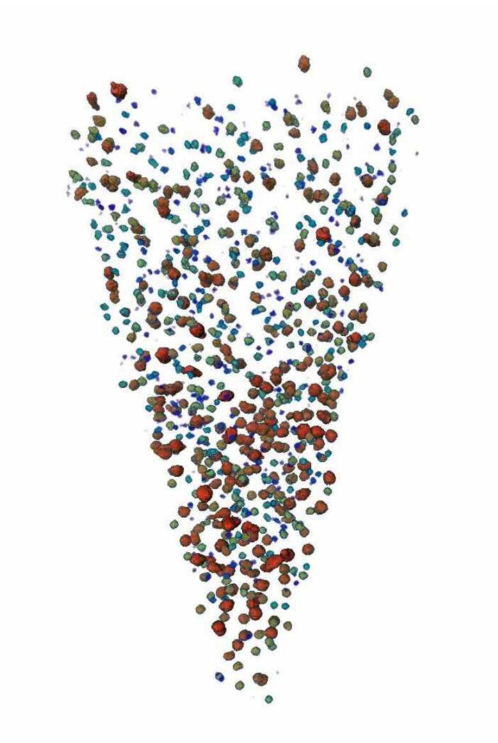
Visualization of helium bubble-impregnated PdNi tip parallel to tilt axis

3D reconstruction of TEM tomography images



Visualization of helium bubble-impregnated PdNi tip along tilt axis

Thresholding of reconstruction yields discrete bubbles of differing sizes



3D map of bubbles within PdNi tip

2D Voronoi diagram

1248 bubbles

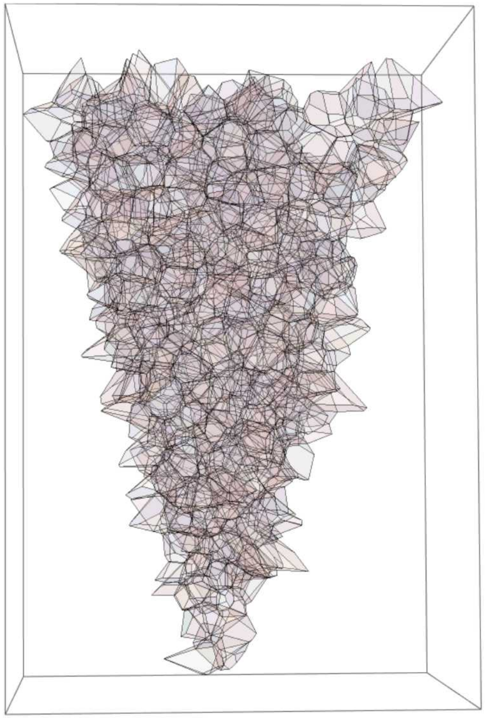
Colored from largest (red) to smallest (blue)

Capture volume is described by Voronoi tessellation

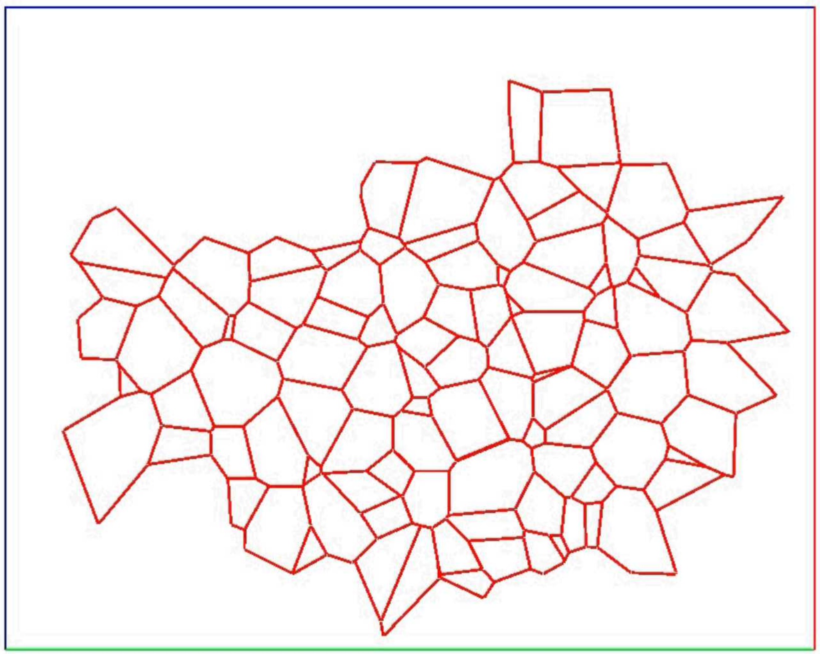
Larger capture volume → larger bubble?

Fortune, S. *Handbook of Discrete and Computational Geometry,* 2nd ed.; Goodman, J. E.; O'Rourke, J., Eds.; Chapman & Hall/CRC: Boca Raton, FL, 2004; pp 514.

Spacing of bubbles yields capture volumes of various sizes



3D Voronoi polyhedral formed by bubbles



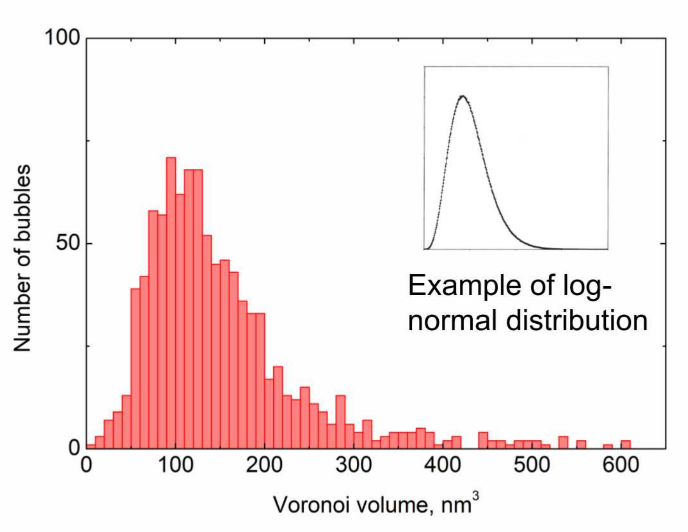
Voronoi cross sections along tip axis

PdNi tip was divided into polyhedral based on distribution of bubbles

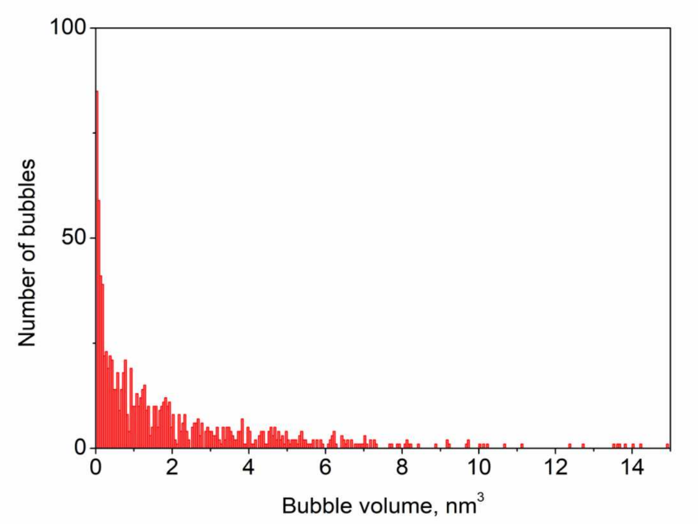
Each bubble has a corresponding polyhedron representing its capture volume

Experimental results differ from predictions of capture volume theory

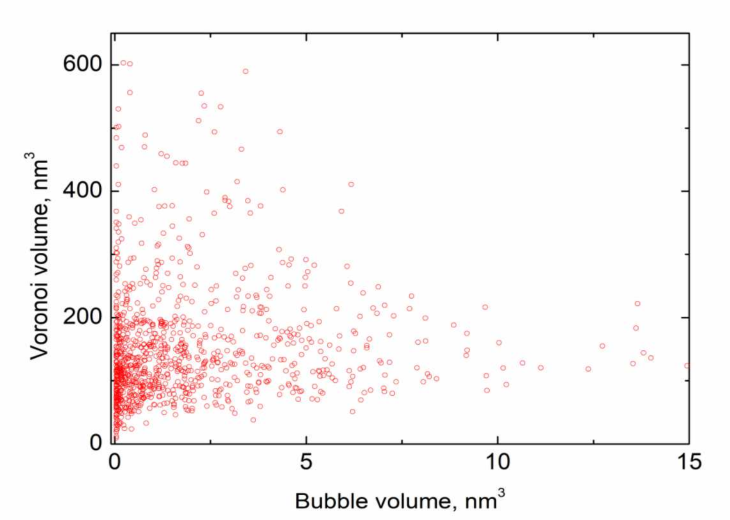
Voronoi volume distribution: log-normal



Bubble volume distribution: not log-normal



Capture volume and bubble volume: no correlation

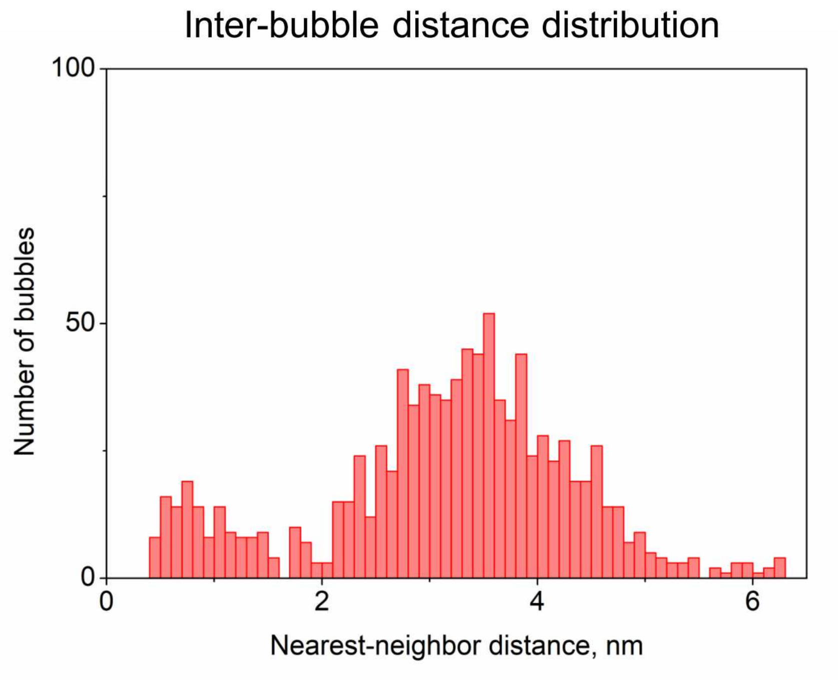


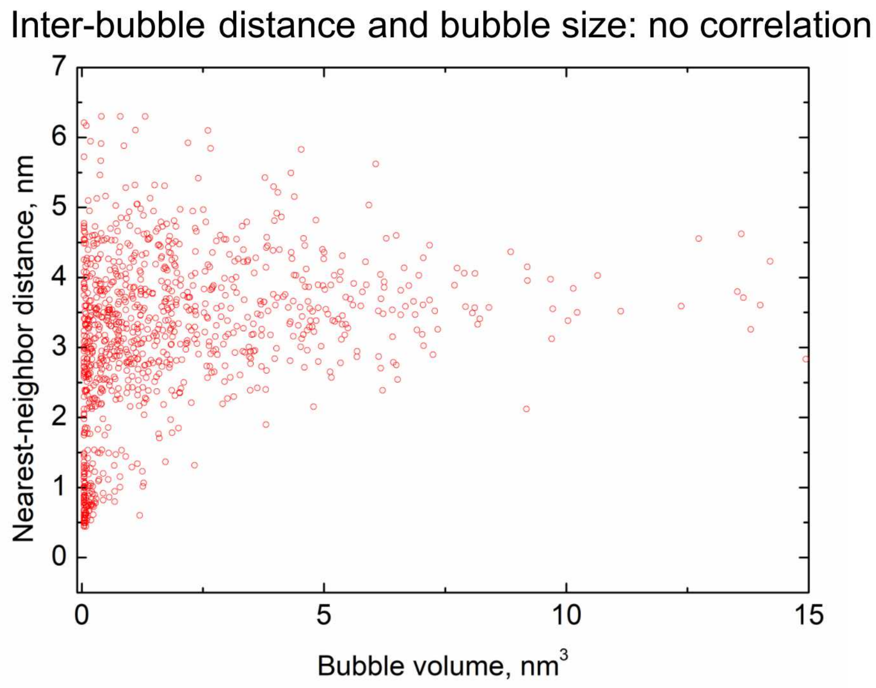
Theory suggests:

- Capture volume and bubble volume adhere to log-normal distributions
- Capture volume determines bubble volume

Tanemura, M. *Forma* **2003**, *18*, 221.

Experimental results differ from capture volume theory prediction





Nearest neighbors tend to be 3.7 nm away, regardless of bubble volume

Further evidence that bubble spatial and size distribution are not related

Conclusions

What determines helium bubble size distribution?

No correlation between capture volume and bubble volume → Bubble size is not primarily determined by proximity to other bubbles

What determines helium bubble spatial distribution?

Capture volume distribution is log-normal → Nucleation is random and homogeneous

Do all helium bubbles nucleate within a narrow time range?

Bubble size distribution is not log-normal → Raises possibility of late nucleation of bubbles

What additional factors explain spatial and size distribution?

Simple nucleation and growth prediction is incomplete → Bubbles may become activated toward motion, leading to bubble migration and coalescence

Acknowledgements

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