SAND2011-9172 C

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Size Dependent Fracture of Si Nanoparticles during Lithiation: Implication to Li-Ion Batteries

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DEPARTMENT OF ENERGY
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NANOSTRUCTURES for ELECTRICAL ENERGY STORAGE

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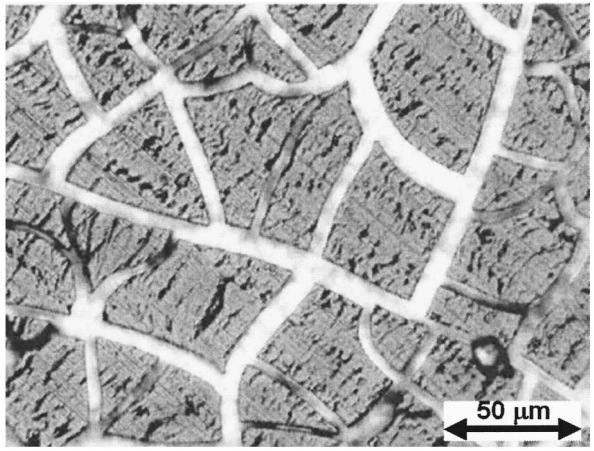
Challenging issues in Si electrode

- **Si has the highest energy density among all the anode materials, but it experiences large volume changes during cycling, which causes mechanical failure of the electrode.**

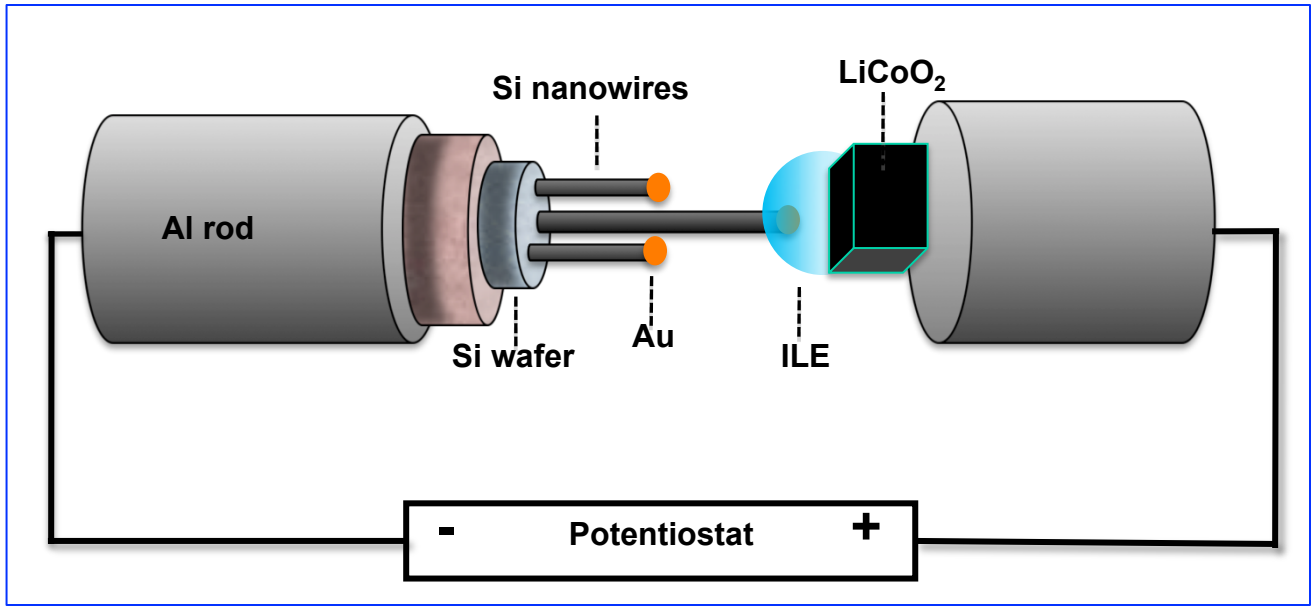
- **How does the mechanical failure occur? How can structural stability during cycling be achieved?**

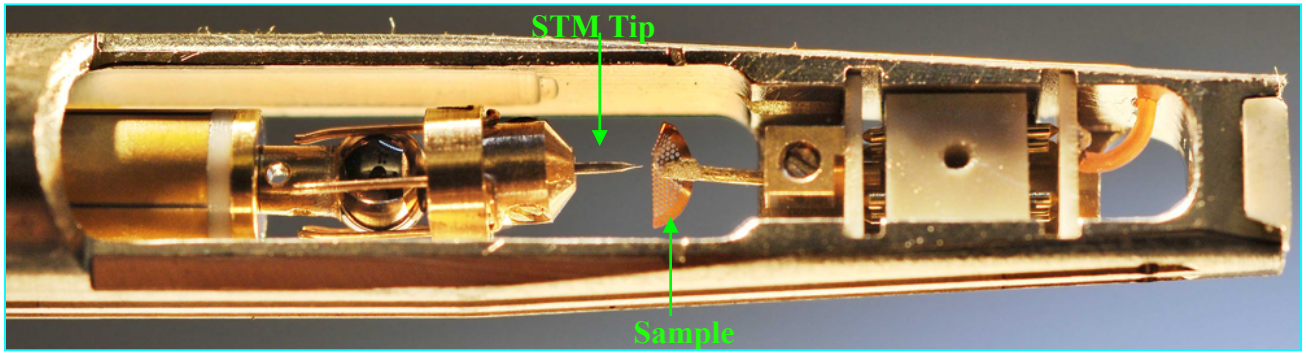
Created a working nano Li-ion cell inside a TEM! Understanding the fundamental science of mechanical degradation!



J. R. Dahn et al., ***Electrochemical and Solid-State Letter 4, A137 (2001)***

Our approach: A single nanowire Li-ion cell (open cell)

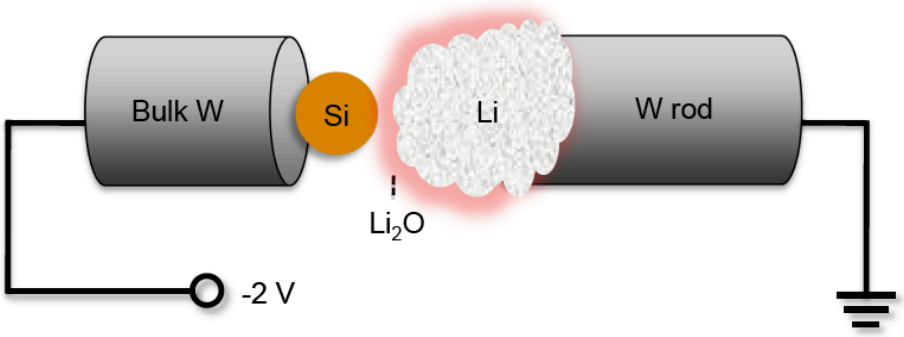


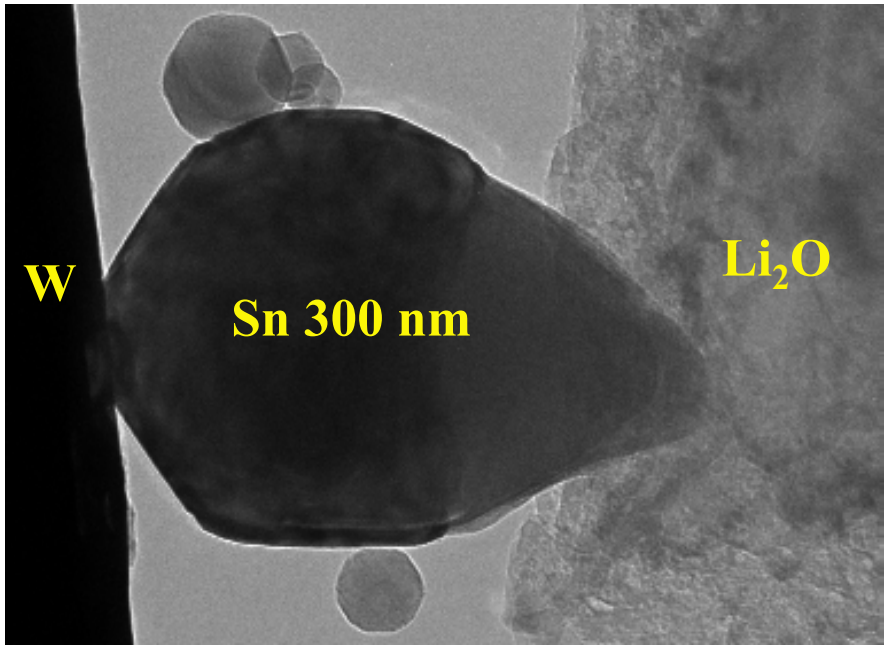


Very clean system, no binder

Huang *et al.*, Science 330, 1515 (2010)

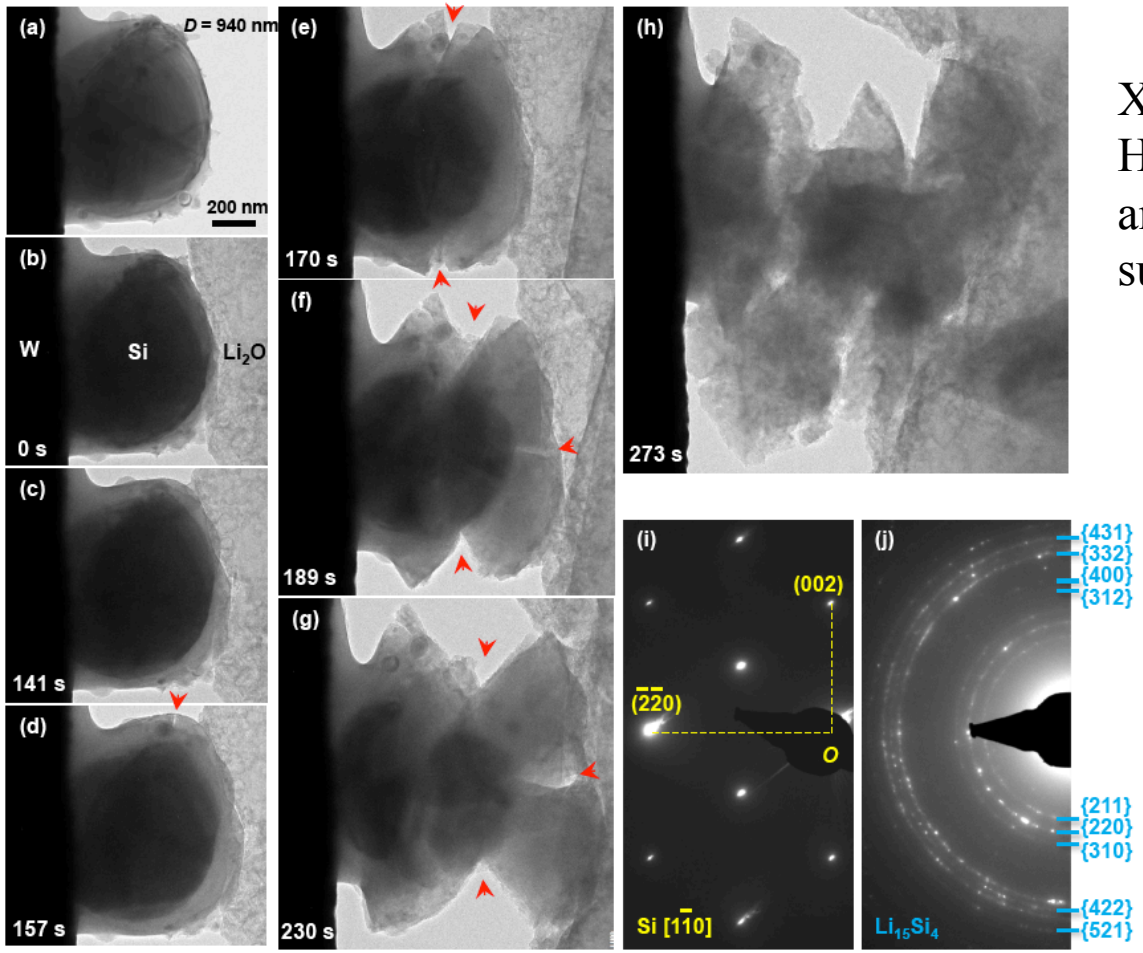
An all solid electrochemical cell

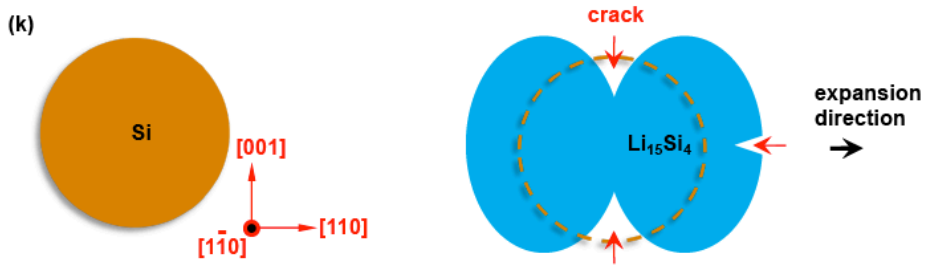




Liu, Huang, Zhu, Li et al., Nano Lett. 11, 3312 (2011)

Size dependent fracture of Si nanoparticles





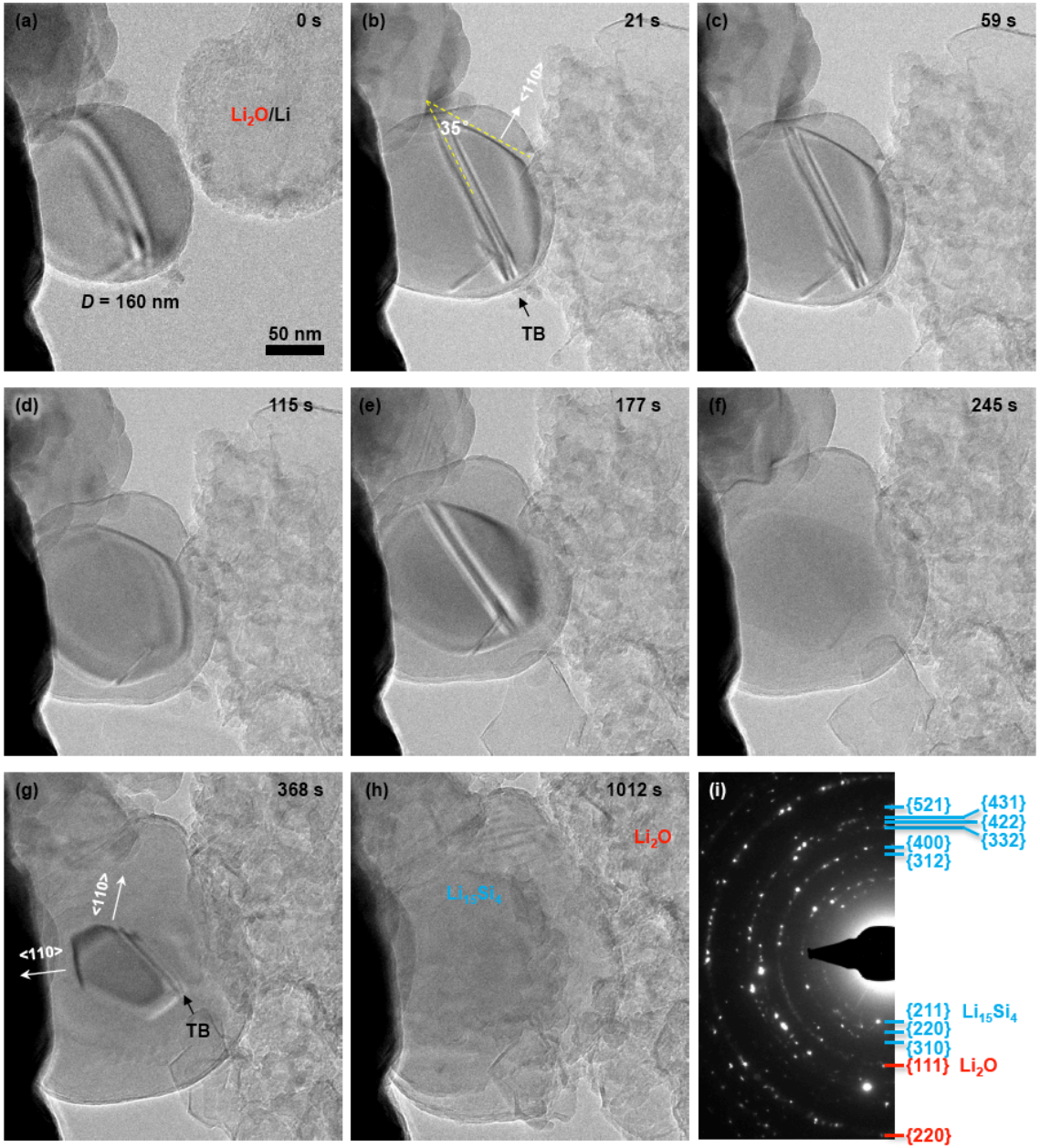
Xiao Hua Liu, Li Zhong, Shan Huang, Scott X. Mao, Ting Zhu, and Jian Yu Huang
submitted

Hatchard, T. D.; Dahn, J. R. J. Electrochem. Soc. 2004, 151 (6), A838–A842.

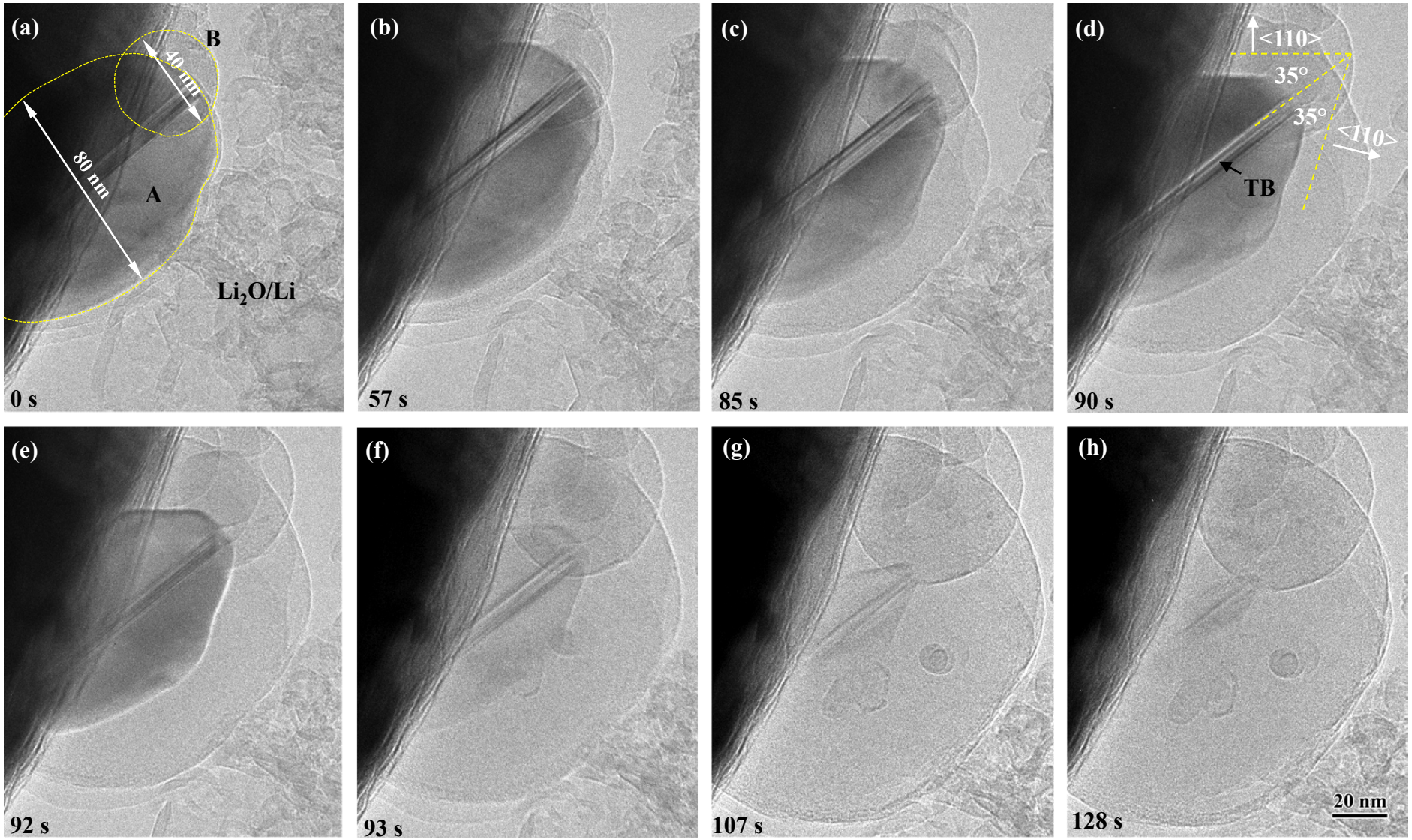
Larcher, D.; Beattie, S.; Morcrette, M.; Edstroem, K.; Jumas, J.; Tarascon, J. J. Mater. Chem. 2007, 3759–3772

Obrovac, M. N.; Krause, L. J. J. Electrochem. Soc. 2007, 154 (2), A103–A108.

Size dependent fracture of Si nanoparticles



Size dependent fracture of Si nanoparticles



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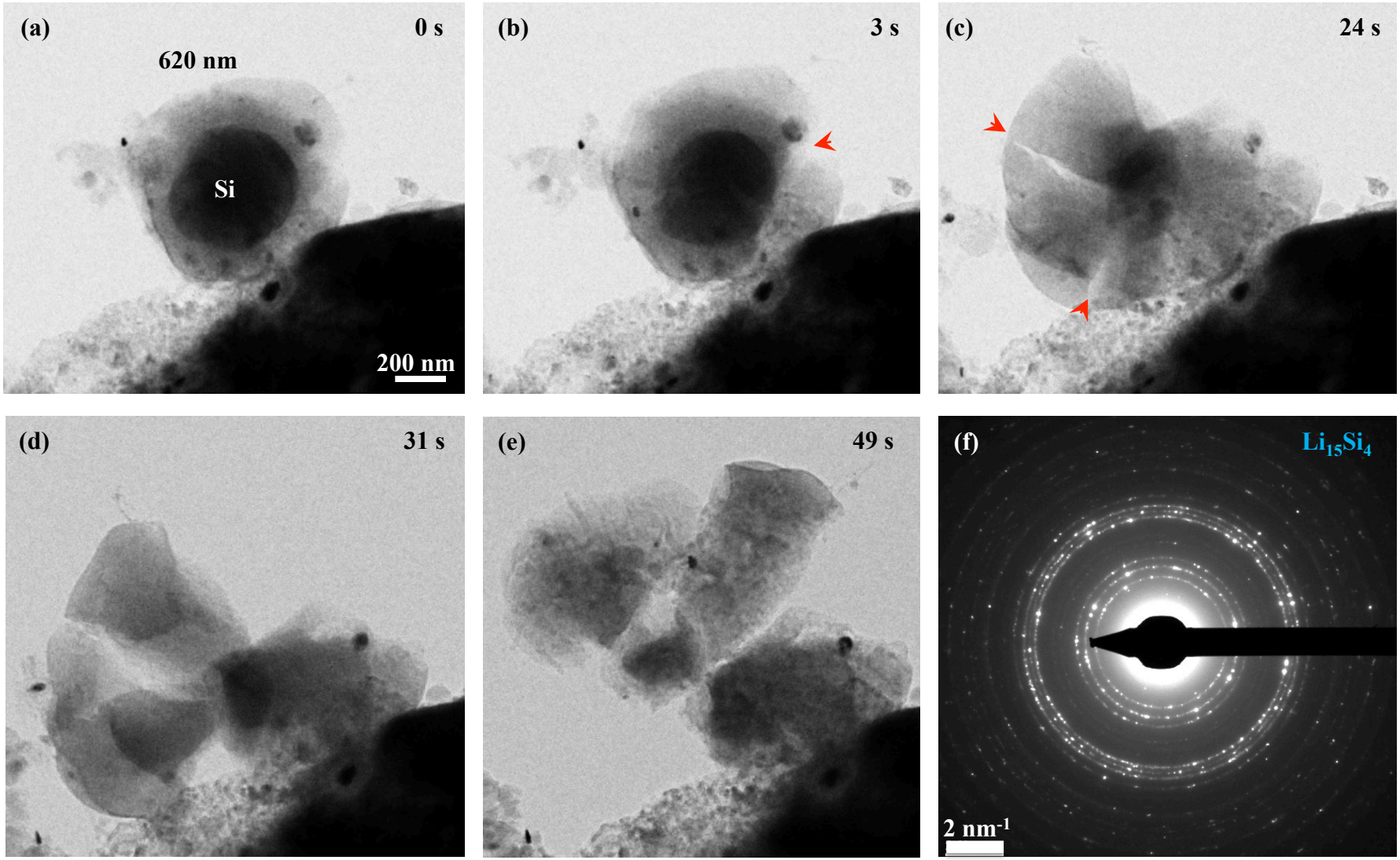

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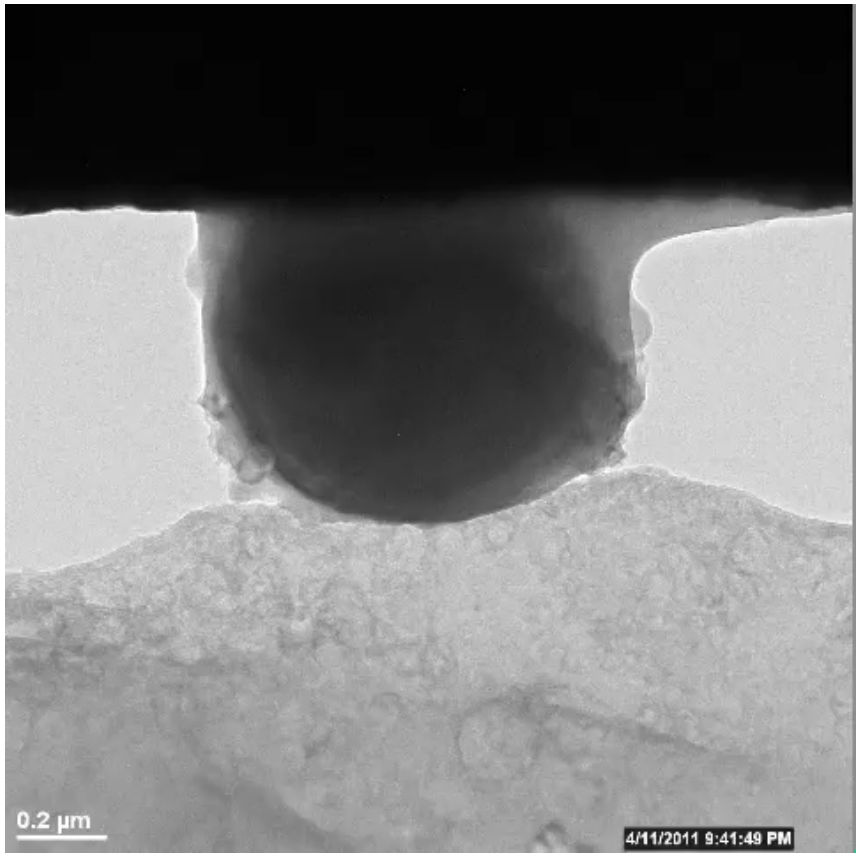
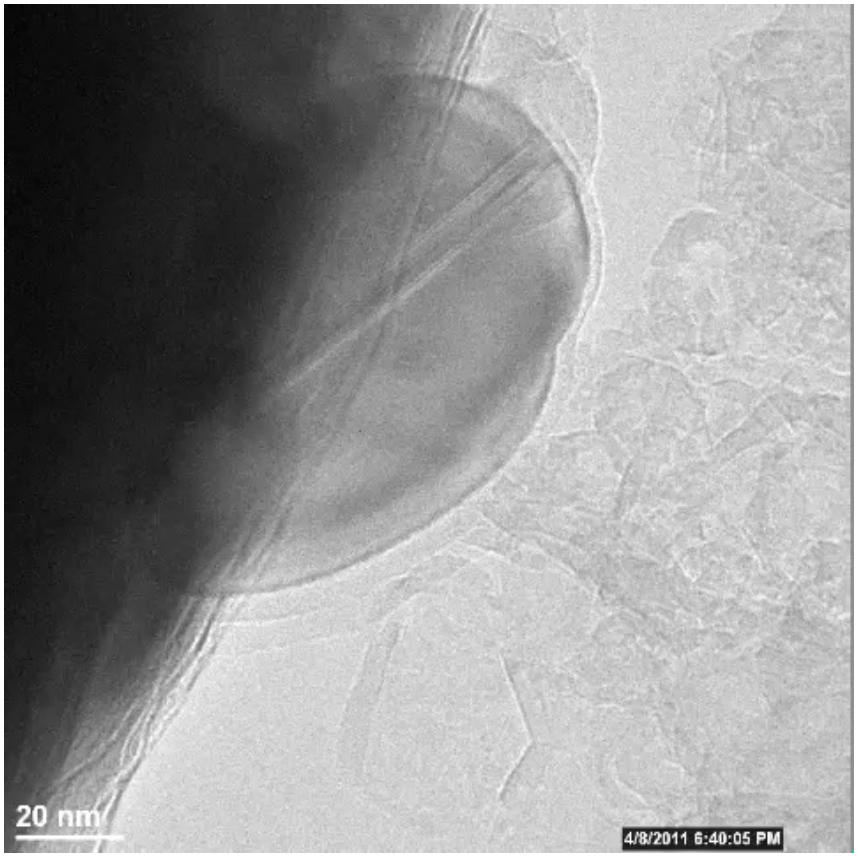
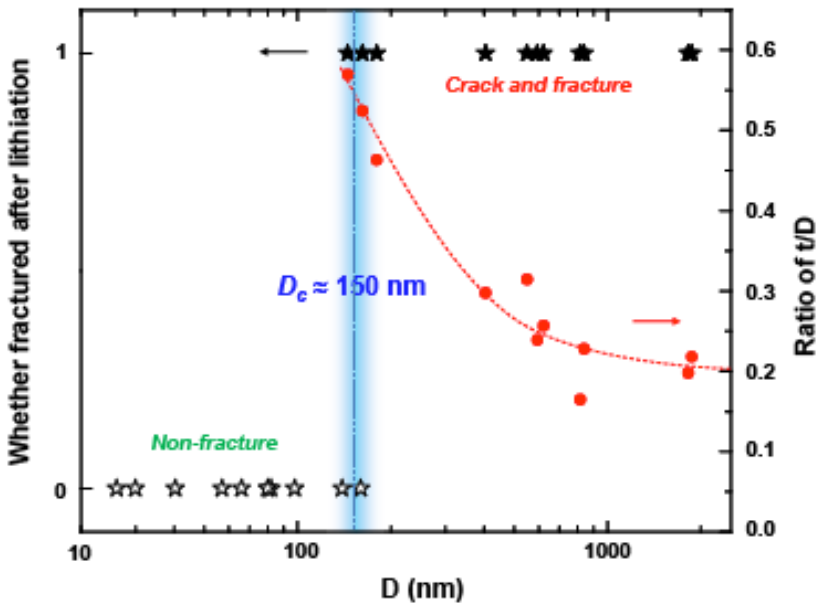
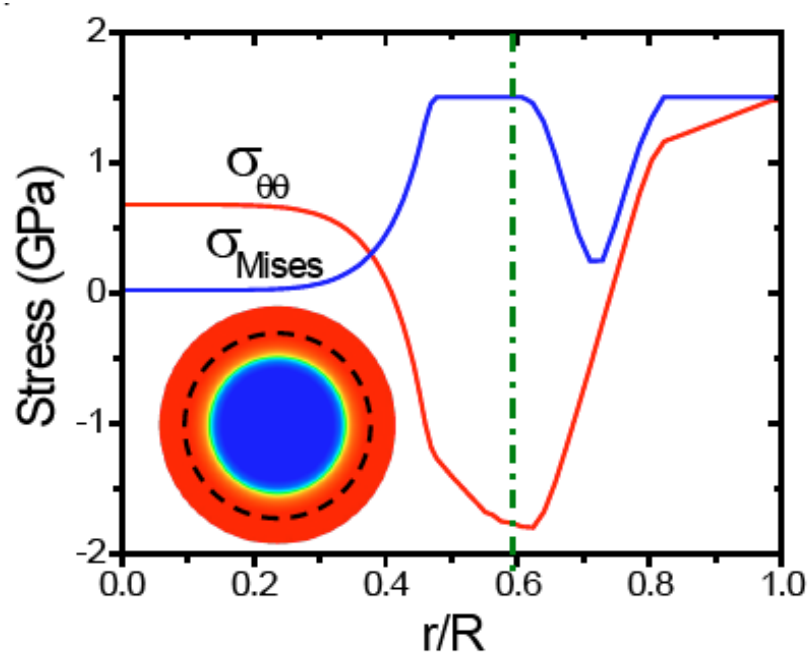
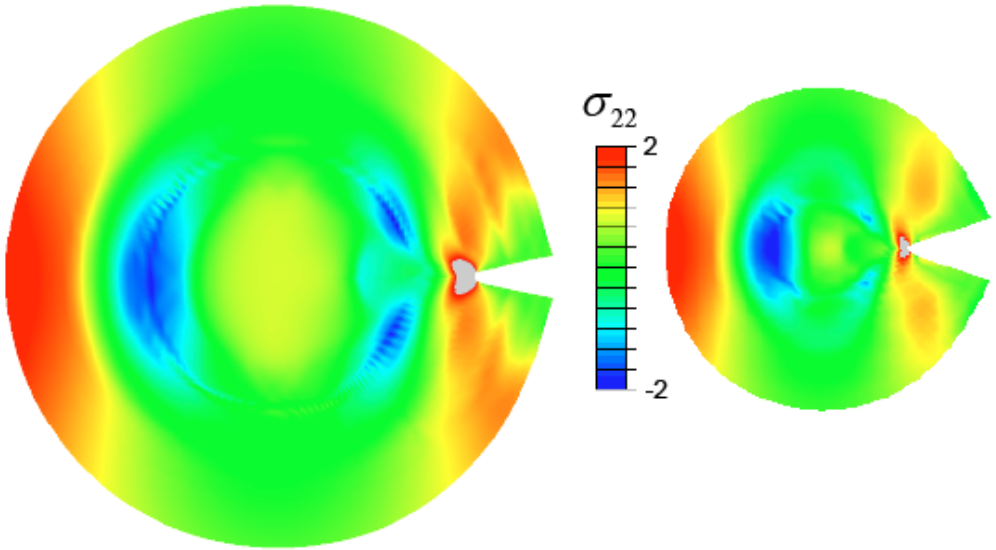




Size dependent fracture of Si nanoparticles





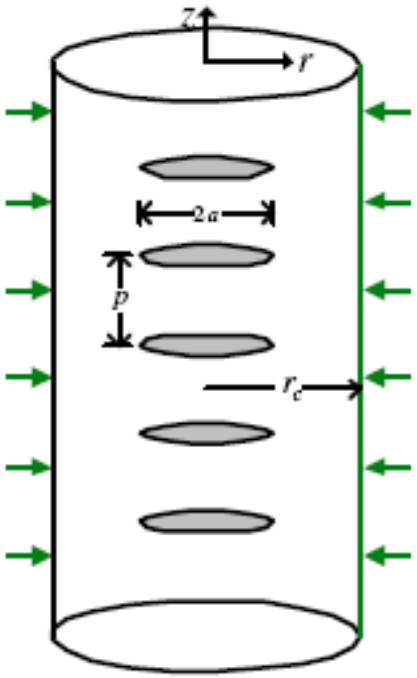


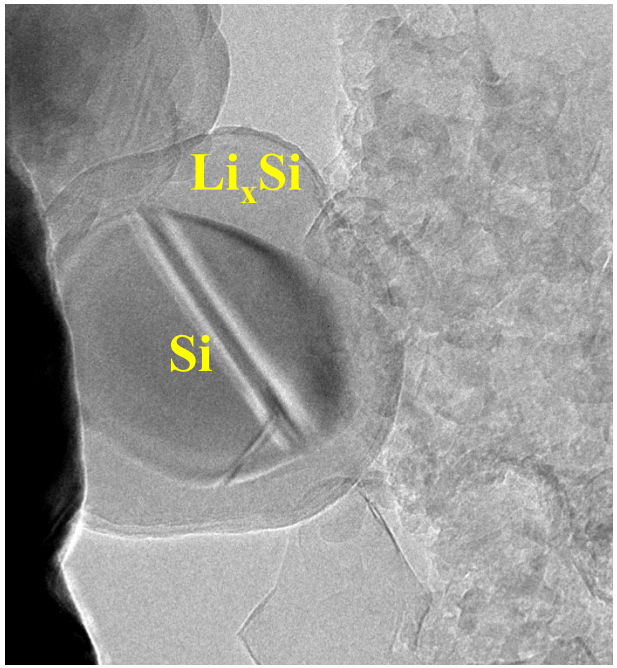
T. Zhu , X.H. Liu, J.Y. Huang et *al.*, submitted

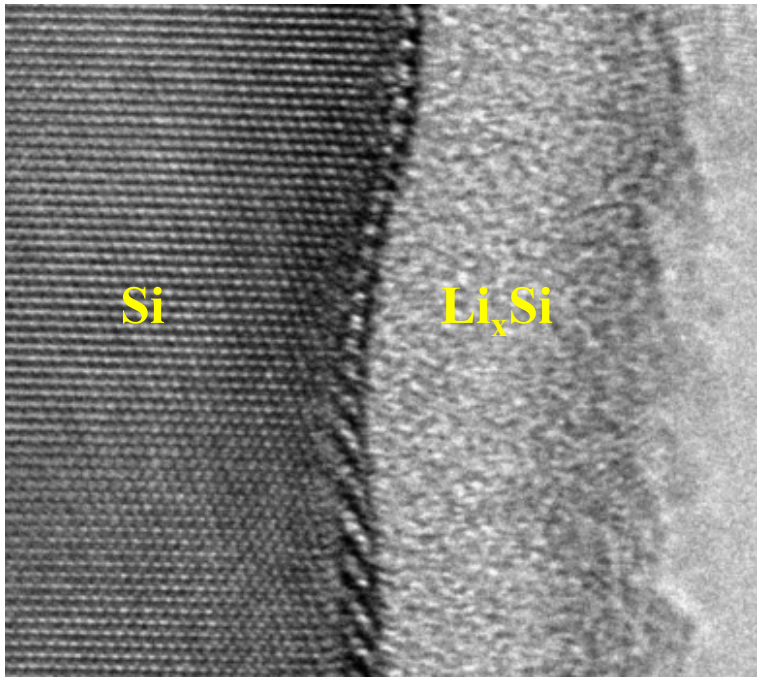
K.J. Zhao, Z.G. Suo *et al.*, Journal of Power Sources 2011

S.L. Zhang, T. Zhu, J. Li, J.Y. Huang *et al.*, in preparation

Size dependent fracture of Si nanoparticles







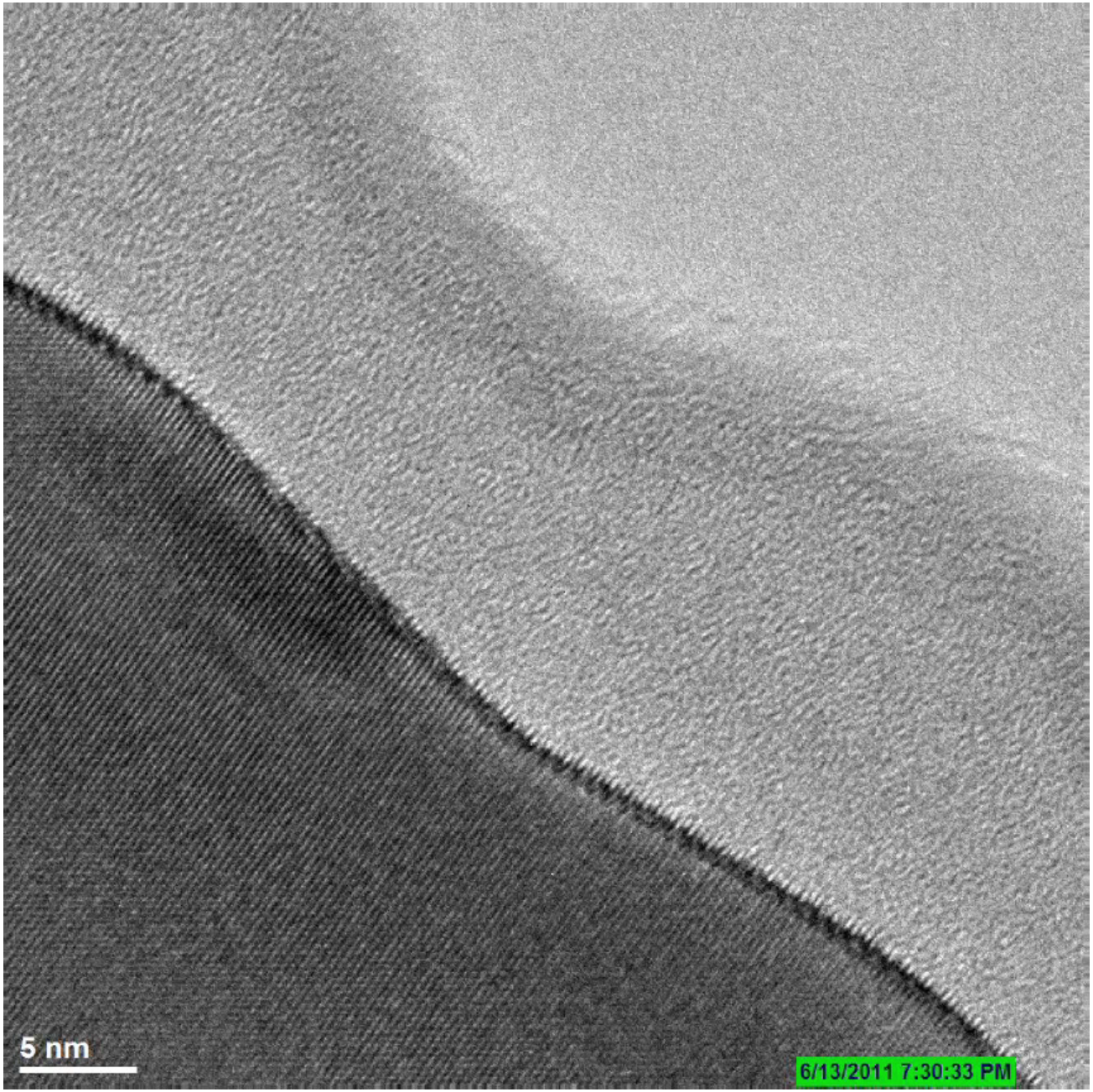
- **Single-phase model**
- **Surface stress always compression**
- **Cracks nucleate from center**

Yang-Tse Chenga, MarkW. Verbruggeb, Journal of Power Sources 190 (2009) 453–460
Tanmay K. Bhandakkar 1, Huajian Gao, International Journal of Solids and Structures 48 (2011) 2304–2309
Kejie Zhao, Matt Pharr, Joost J. Vlassak, and Zhigang Suo, Journal of Applied Physics **109**, 016110 2011

- **Two-phase model**
- **Surface stress is tensile**
- **Cracks nucleate from surface**

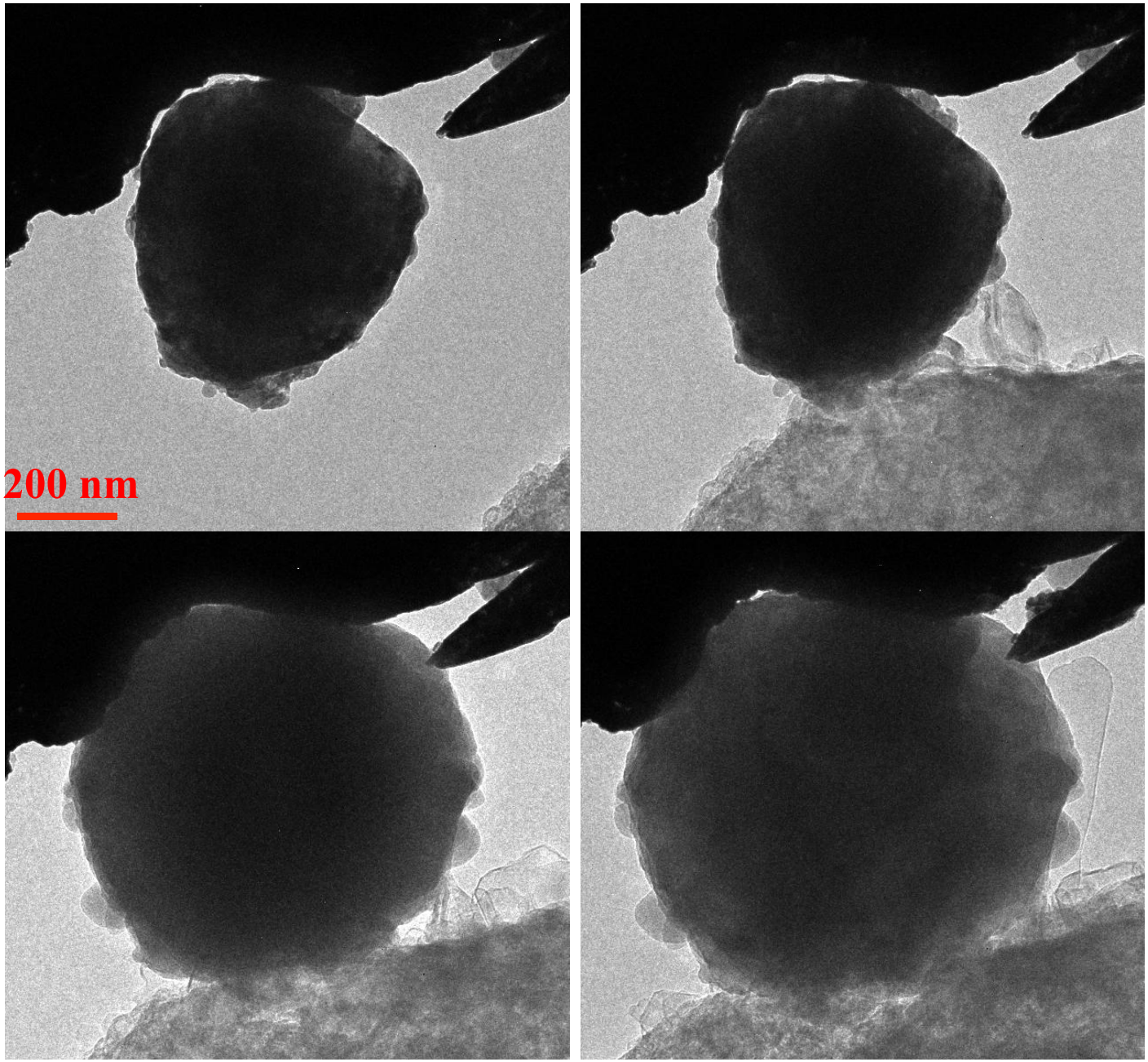
Chon et al., PRL 2011
Zhao et al., J. Power Source, 2011
Zhu, Liu, Huang *et al.*, submitted

Atomically sharp interface during lithiation Si



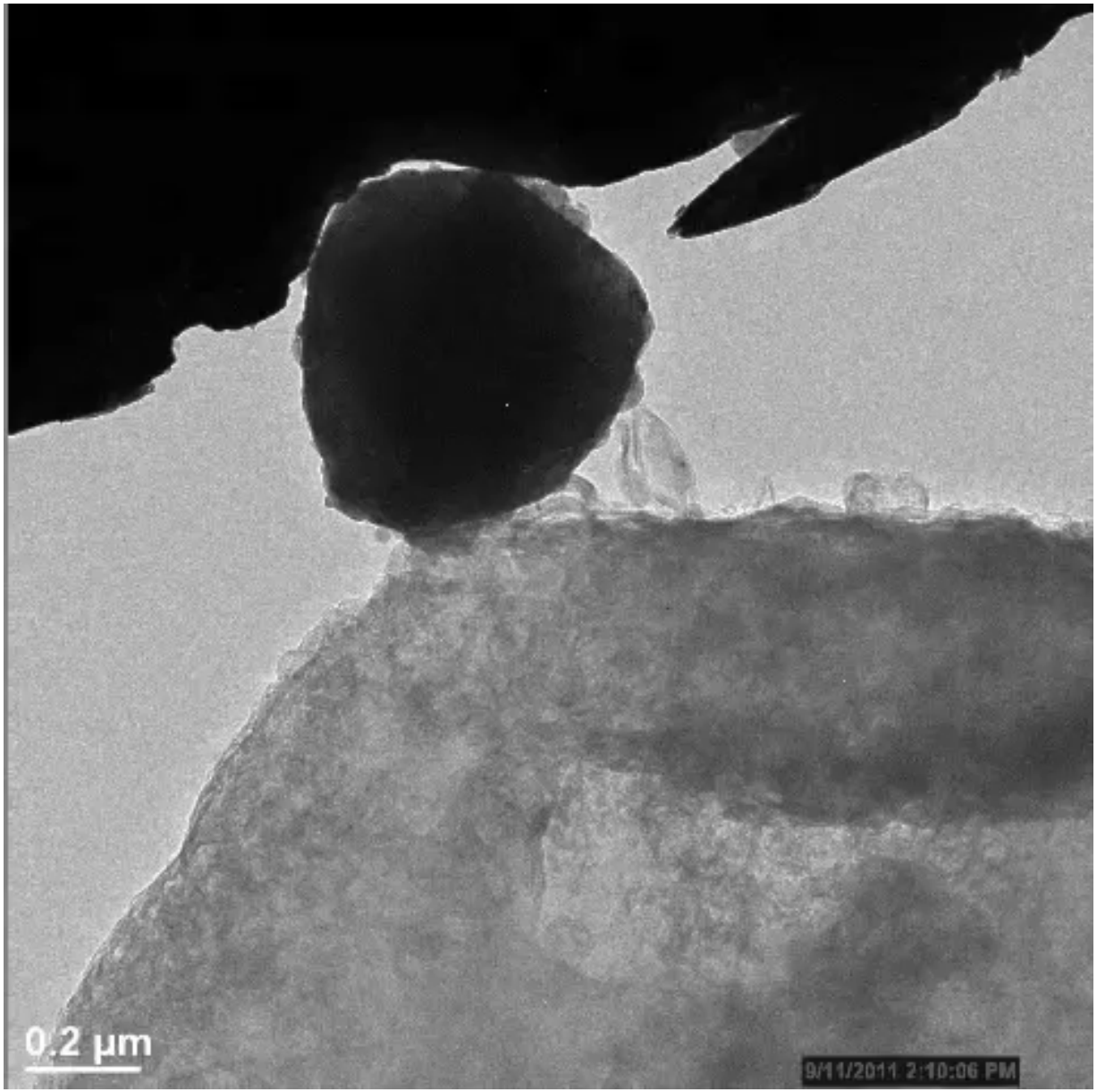
X.H. Liu, T. Zhu, J.Y. Huang *et al.*, unpublished

Lithiation of Ge nanoparticles



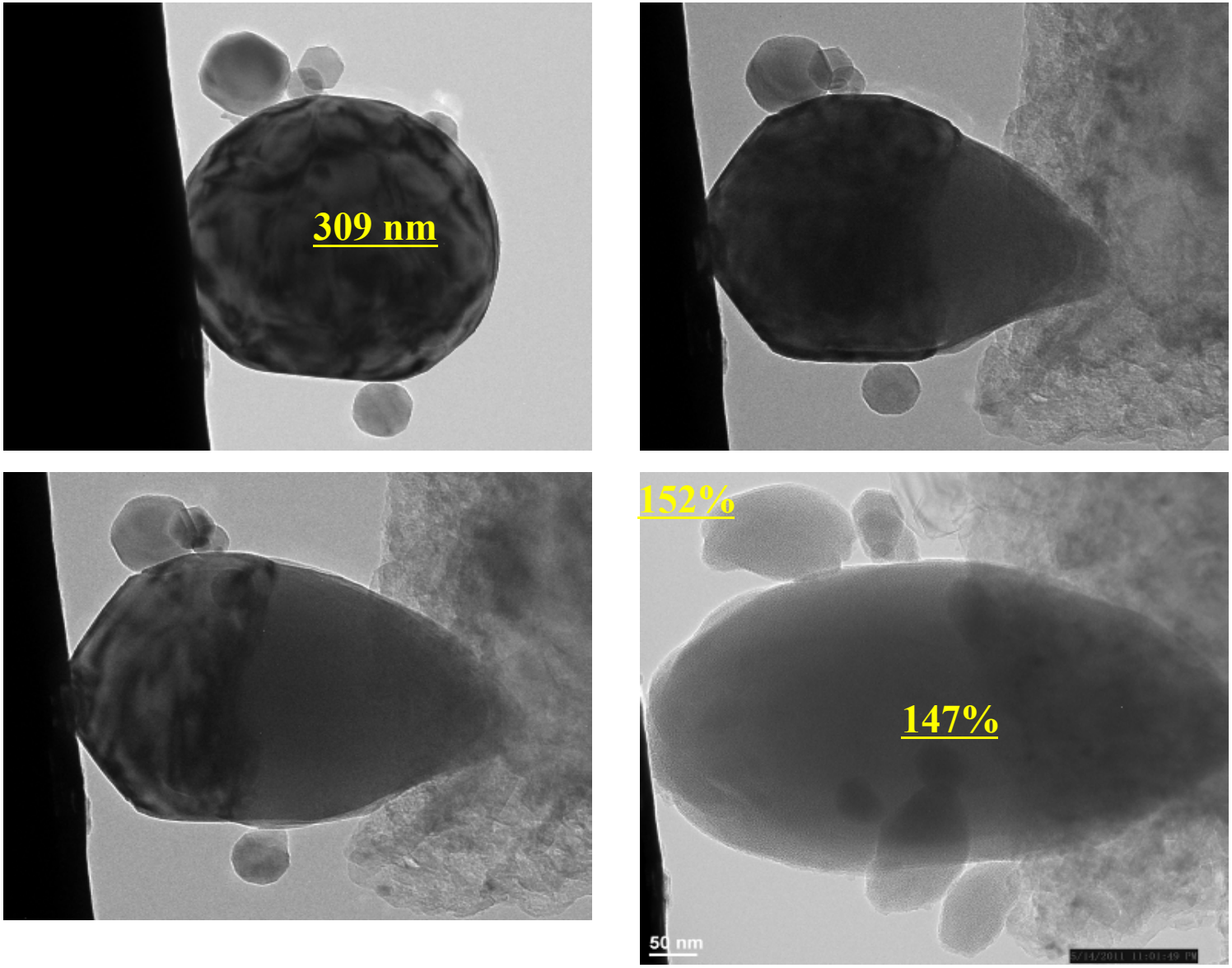
W.T. Liang,
S.L. Zhang,
J.Y. Huang
unpublished

Lithiation of Ge nanoparticles



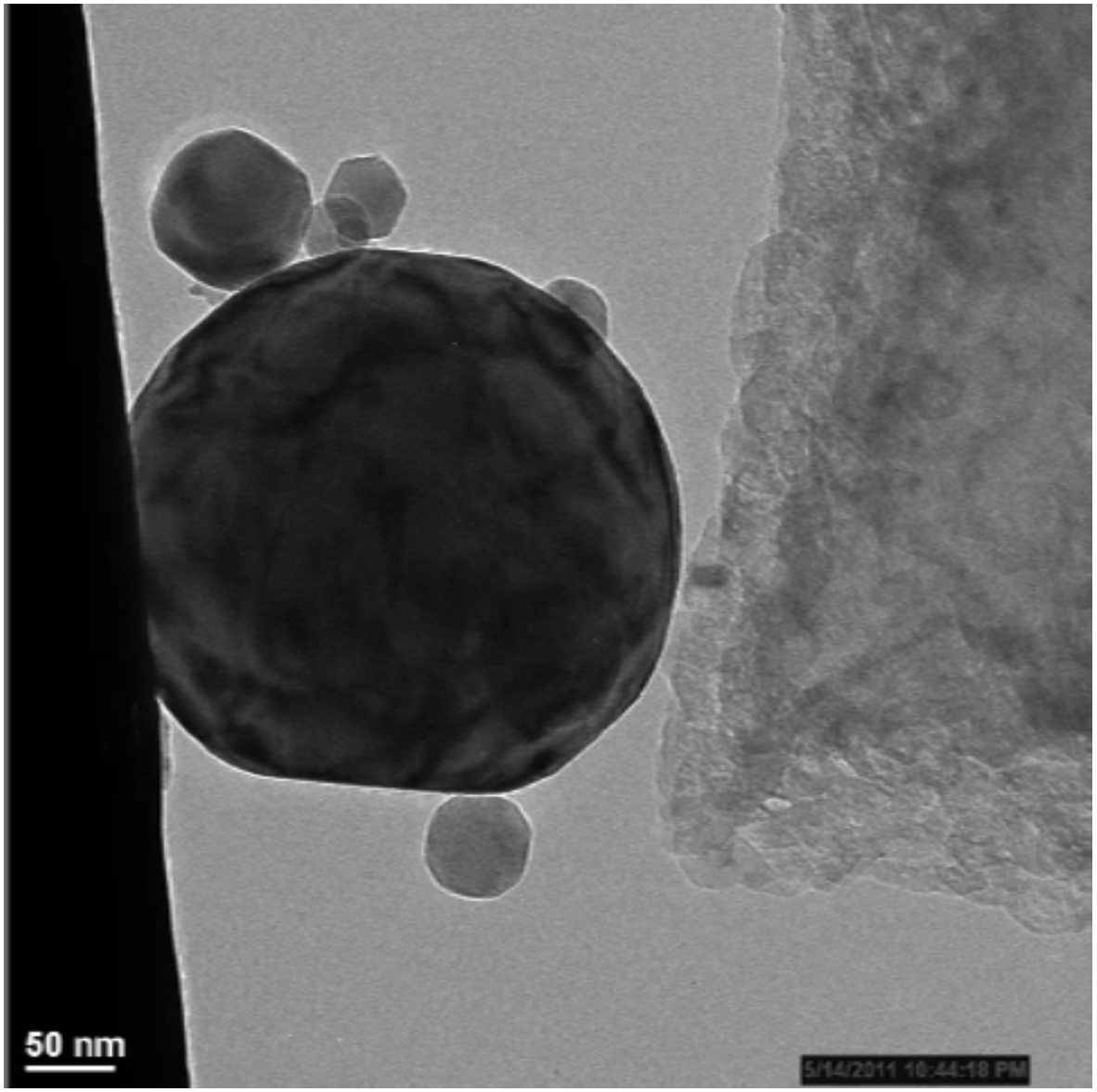
W.T. Liang,
S.L. Zhang,
J.Y. Huang
unpublished

Lithiation of Sn nanoparticles



J.W. Wang, S. Mao, J.Y. Huang, unpublished

Lithiation of Sn nanoparticles



J.W. Wang, S. Mao, J.Y. Huang, unpublished

Summary

- **Lithiation induced fracture of Si NPs is strongly size dependent, critical particle diameter of ~ 150 nm, below which the particles neither cracked nor fractured upon first lithiation, above which the particles initially formed surface cracks and then fractured due to lithiation induced swelling.**

- **The surface fracture arose owing to the buildup of large tensile hoop stress in the surface layer that reversed the initial compression.**

- **The small-sized nanoparticles nevertheless averted fracture because of insufficient mechanical energy to drive crack propagation during the electrochemical energy storage.**

- **D < 150 nm for LIB**

Acknowledgement

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