Resolving the surface structure of iron-disilicide alloy nanocrystals

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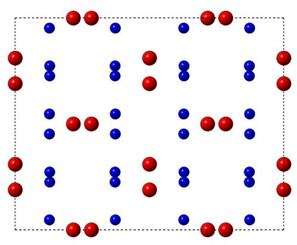
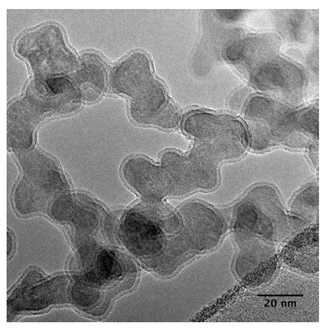
Growth of β-$FeSi_2$ nanocrystals

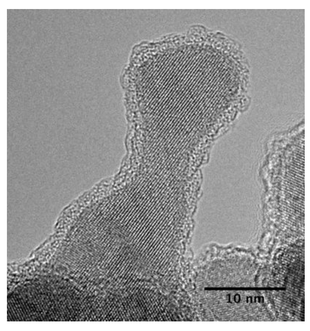
Beta-iron-disilicide (β-$FeSi_2$) is a promising thermoelectric material (Seebeck coef. ~0.2 mV/K), because of its chemical stability and low cost. Fabrication of β-$FeSi_2$ nanostructures, however, is challenging due to its complex structure and narrow compositional range of single phase formation.



The β-$FeSi_2$ phase - space group 64, symbol *Cmce*

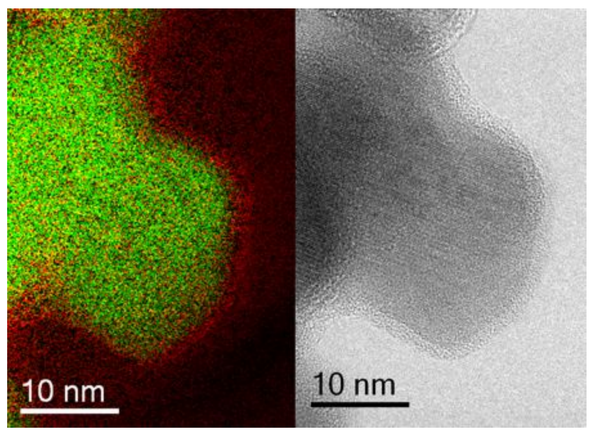
β-$FeSi_2$ nanocrystals were fabricated by thermal decomposition of silane (SiH_4) and iron pentacarbonyl ($Fe(CO)_5$) in a gas phase reactor.



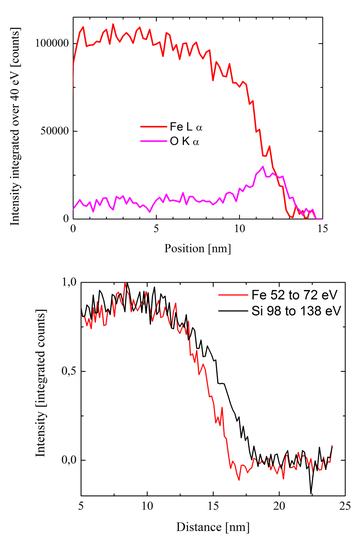


The nanoparticles form agglomerates and surrounded by an amorphous shell as shown in BF images.

Shell composition – EELS



High-resolution EFTEM maps were recorded in the Cc-Cs-corrected PICO instrument at 80 kV. Energy range from 450 to 780 eV, window size 20 eV, step size 20 eV. Colored map shows the Fe and O distribution - green=Fe, red=O.

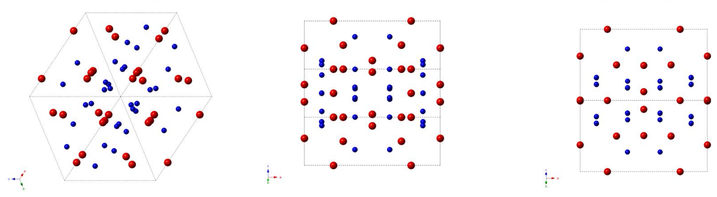


Extracted distribution of Fe and O from the core-loss and Fe and Si from the low-loss EELS regions suggest that the the amorphous shell is SiO_x.

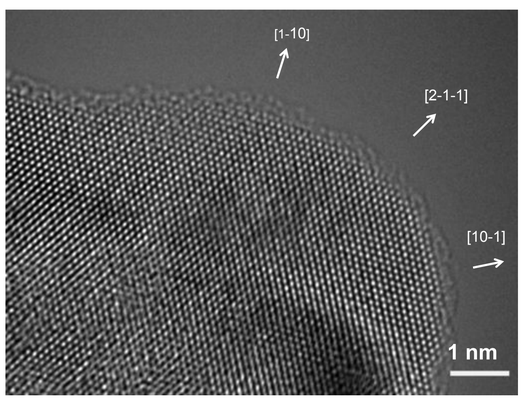
Resolving the atomic positions

The surface structure was verified by comparing experimental aberration-corrected images to simulated images of the structure.
Location of Fe and Si atoms in high-resolution TEM images can be identified from certain crystallographic direction.

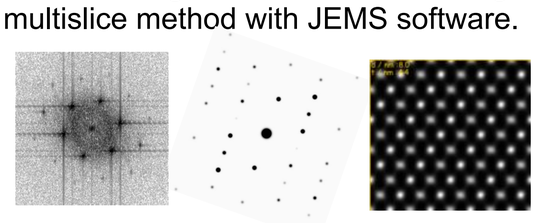
[111] [021] [010]



β-$FeSi_2$ nanocrystal viewed from [111] direction.

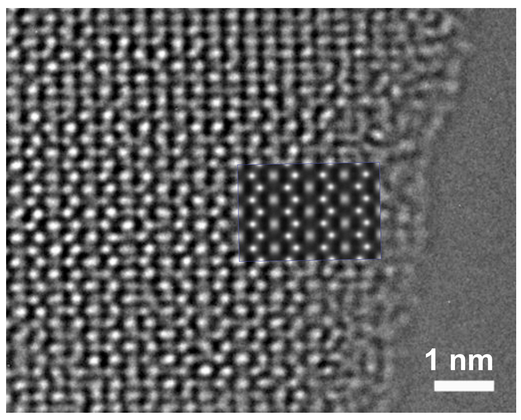


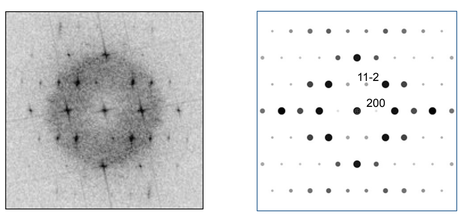
HRTEM images were simulated using multislice method with JEMS software.



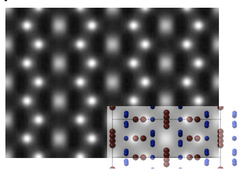
FFT of HRTEM image, simulated diffraction pattern and image of β-$FeSi_2$.
Fe and Si columns could not be distinguished from [111] direction.

β-$FeSi_2$ nanocrystal viewed from [021] direction.



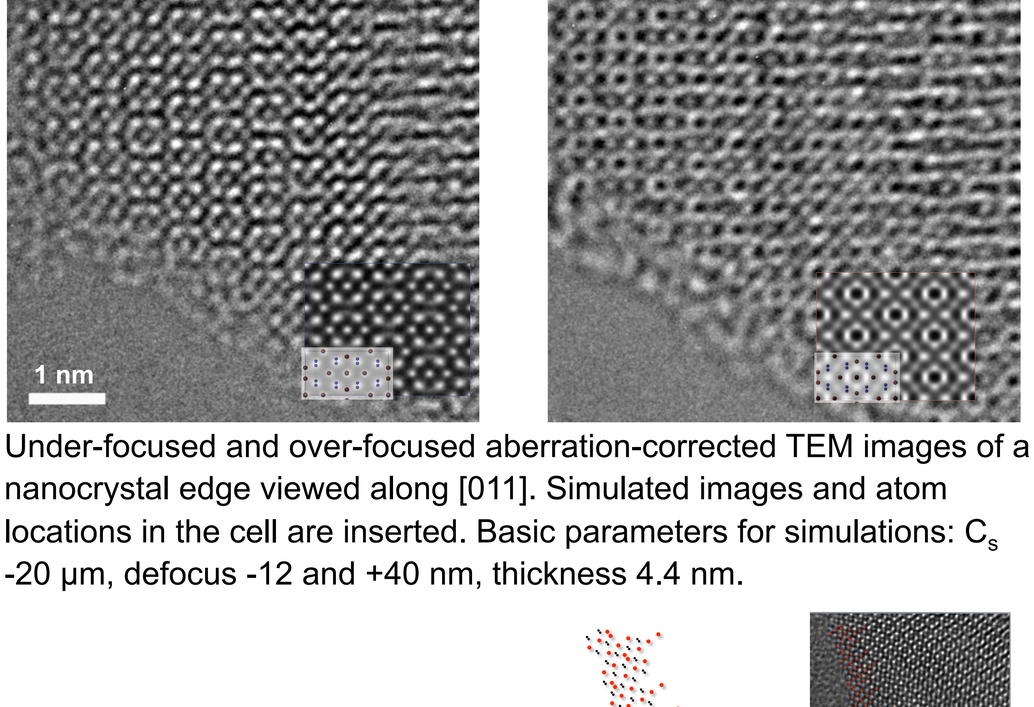


Digital diffractogram and simulated diffraction pattern .



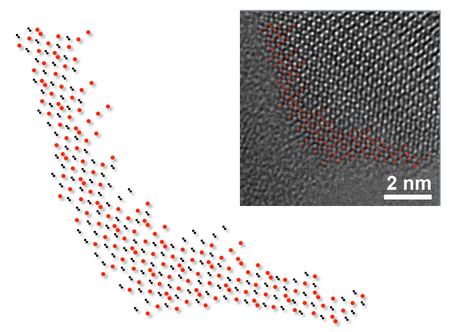
Fe and Si columns are separated.

β-$FeSi_2$ nanocrystal viewed from [011] direction.



Under-focused and over-focused aberration-corrected TEM images of a nanocrystal edge viewed along [011]. Simulated images and atom locations in the cell are inserted. Basic parameters for simulations: C_s -20 µm, defocus -12 and +40 nm, thickness 4.4 nm.

Results revealed no distinct facets of the β-$FeSi_2$ nanocrystals and the surface contains both Fe and Si columns.



Reconstructed Fe and Si locations.

Summary

- β-$FeSi_2$ nanocrystals were successfully produced from gas phase;
- an amorphous shell was observed;
- the shell composition was determined as SiO_x ;
- no segregation or distinct facets were observed;
- surfaces of crystalline cores contain both Fe and Si;
- this work was supported by BMF, grant 03SF0402A.