Probing Atomic and Electronic Structure of Catalysts by Combination of *In Situ* and *Ex Situ* Chemical Imaging

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Purpose

Develop chemical imaging capabilities that drive the atomic-level imaging and spectroscopic analysis of catalytic materials under *in situ/operando* conditions.

Directly relate structural information with the catalytic performance, and establish the guiding principles for accelerated discovery of next-generation catalytic materials.

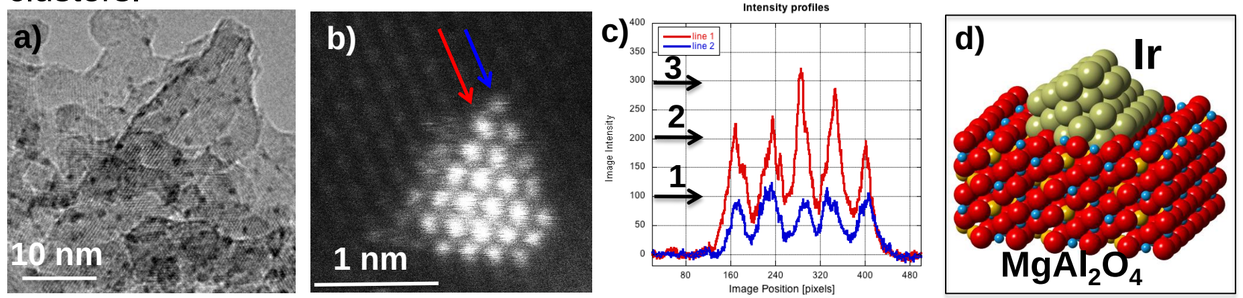
Expected Outcomes

New capabilities that enable integrated imaging and spectroscopy studies of catalytic materials under *in situ* and *ex situ* conditions.

New insights on atomic-level structure and electronic nature of oxide-supported transition-metal (TM) catalytic clusters under *ex situ* and *in situ/operando* conditions.

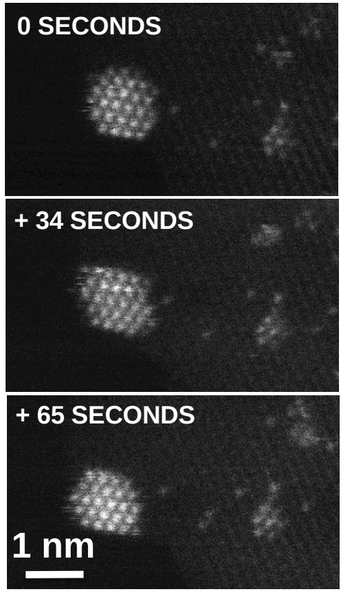
Accomplishment to Date

- Derived morphological models of Ir nanoclusters and the way they are epitaxially related to the spinel oxide substrate.
- Based on STEM image simulation, determined the limits of "single-shot" STEM HAADF imaging for direct determination of 3D atomic nature of crystalline clusters.



(a) Conventional TEM of iridium clusters supported by $MgAl_2O_4$. (b) Detail STEM HAADF of a sub-nanometer Ir cluster. (c) For small clusters, the intensity varies proportionally with the number of atoms in the atomic columns. (d) Analysis of the image intensities and the lattice correspondence enables us to propose a full 3D model.

Cluster stability during TEM observations



Why It Matters

Derived 3D model of Ir clusters enables us to quantify the nature of atomic sites on metal cluster surfaces, which is important for modeling effort in rationalizing the catalytic properties

Publications and Presentations

- 3 conference presentations (NAM 2011, M&M 2011, Fall MRS 2011).
- Manuscripts in preparation (fully and partially supported by CII).

Path Forward

- Establish capability for aberration-corrected S/TEM for *in situ* heating experiments.
- Establish capability that enables *in situ* gas flow experiment in aberration-corrected ETEM.
- Integrate imaging and spectroscopy methods for a physical-based understanding of catalytic phenomena.

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