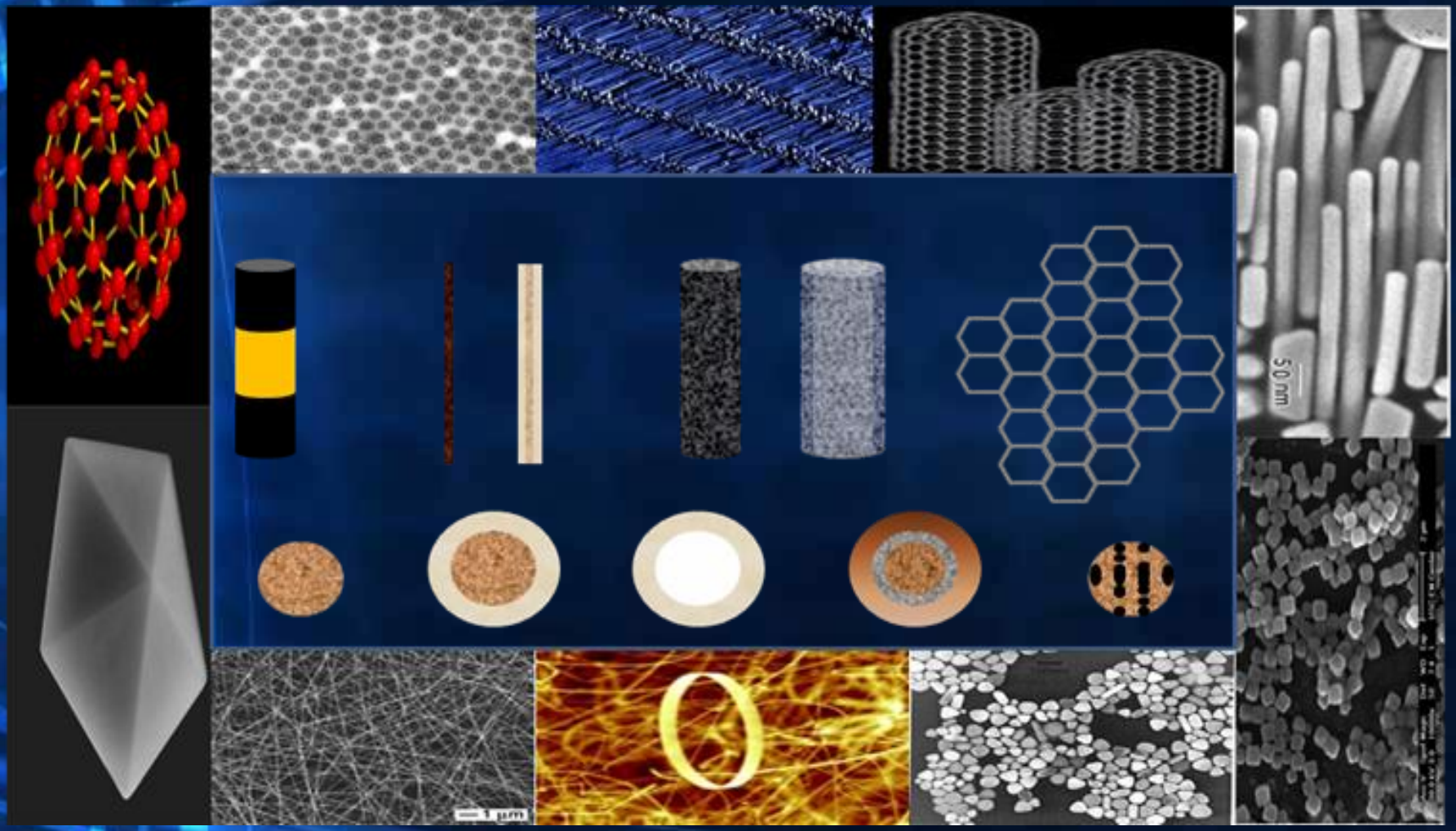
Center for Atomic-Level Catalyst Design

Design and Engineering of Core-shell Magnetic Nanomaterials
Current Challenges

Challa Kumar
Director, Nanofabrication & Nanomaterials
LSU-CAMD

ALS Workshop 10/14/10

Different architectures of Nanomaterials



Why Core-shell Nanomaterials ?

Core and Shell have different material compositions in a single particle

- Composition and microstructure varies through radial direction
- The dimensions and composition can be modulated
- Opportunity for tailor-made properties

A shell can be utilized as a protective shield to sensitive core material

Inexpensive nanomaterials can be formed as shells around expensive cores

Why Core-shell Nanomaterials ?

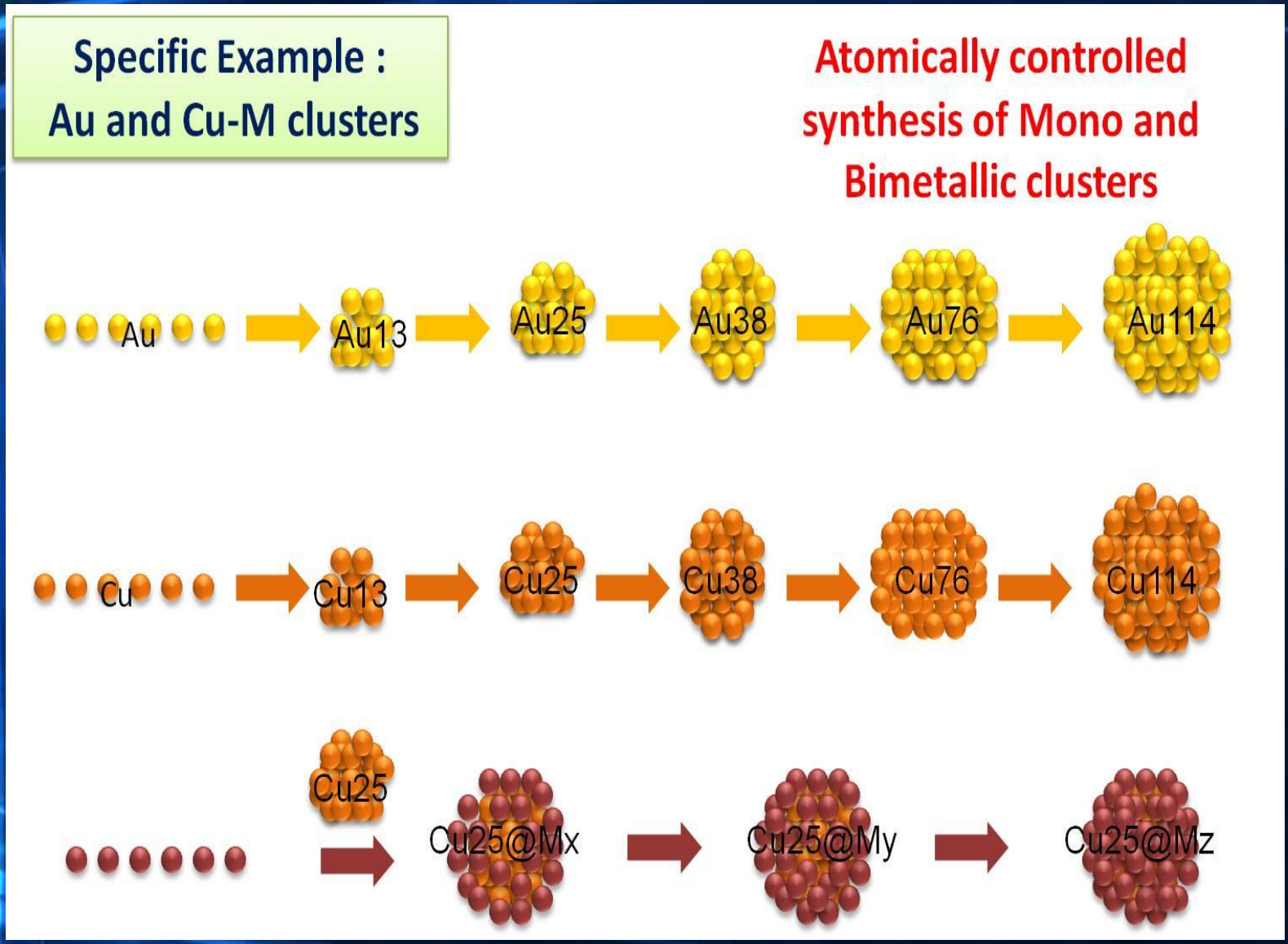
Biocompatible shell around toxic core material enables reduction in their toxicity

The shell surface can be utilized for bio-functionalization

Multiple functionalities can be incorporated

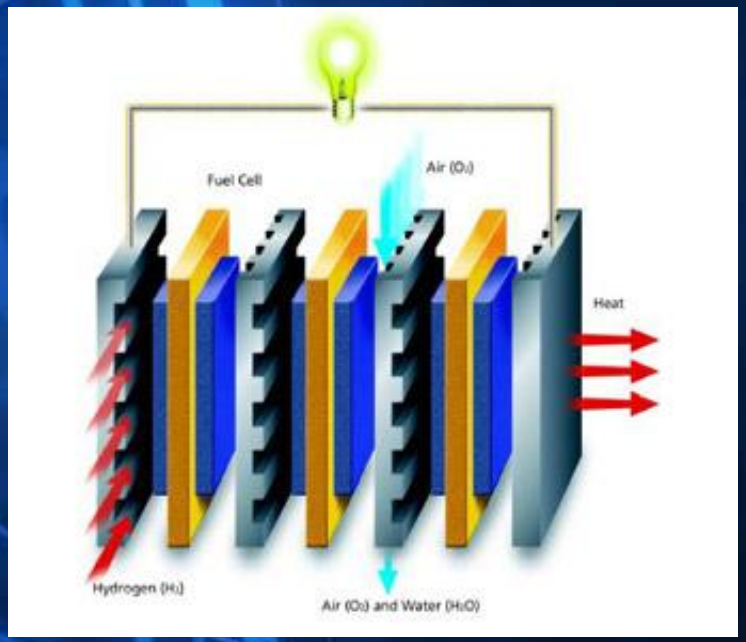
CATALYSIS

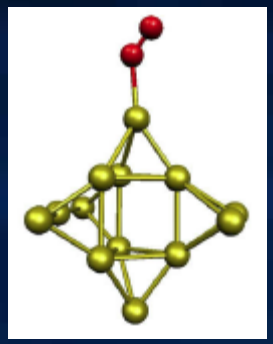
Atomically controlled metal clusters



Specific Example :
"CO Activation"

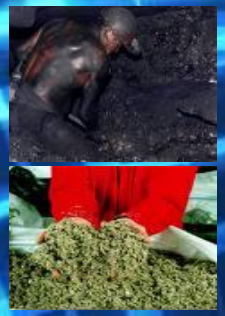
$$2CO + O_2 \rightarrow 2CO_2$$





$$CO + H_2O \rightarrow CO_2 + H_2$$

Specific Example
"CO Activation"



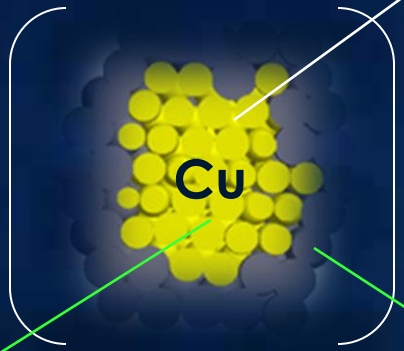
Coal / Biomass

Gasification

$CO + H_2$

Syngas

Co/Mn/Fe/Zn



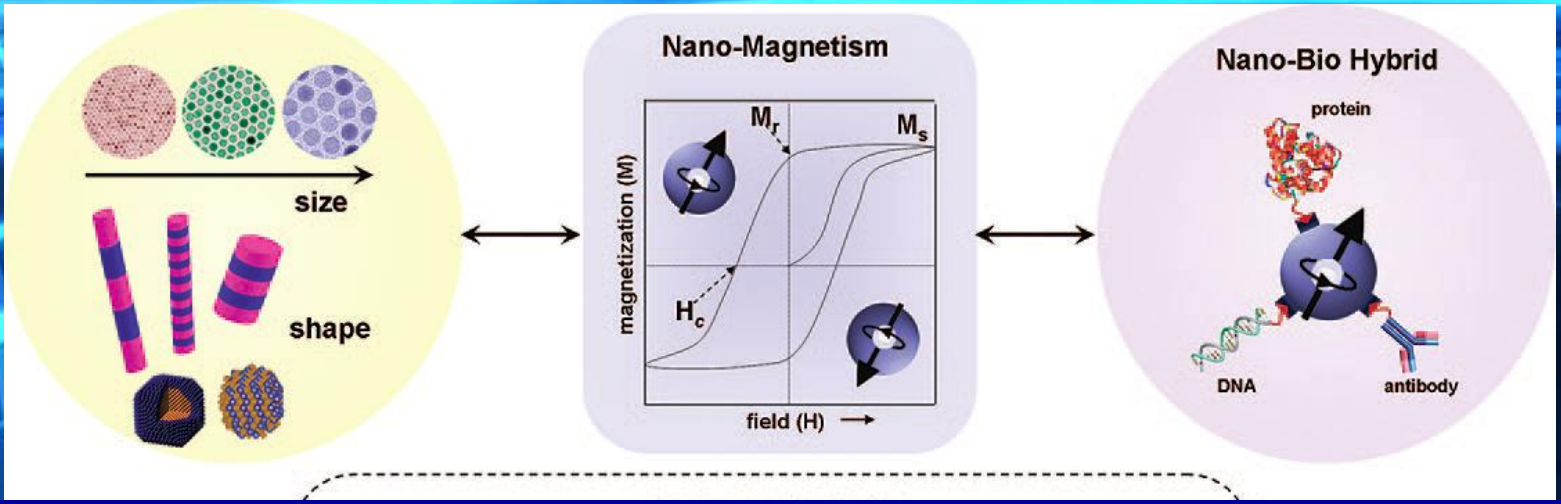
Core-shell
Atomic clusters

Composition, size, structure of the cluster will be determined from computation & Surface Science studies

EtOH

- Methanol Synthesis Catalyst
- Non-dissociative Activation of CO
- Ability to dissociate CO
- Helps in carbon chain growth

BIOMEDICINE



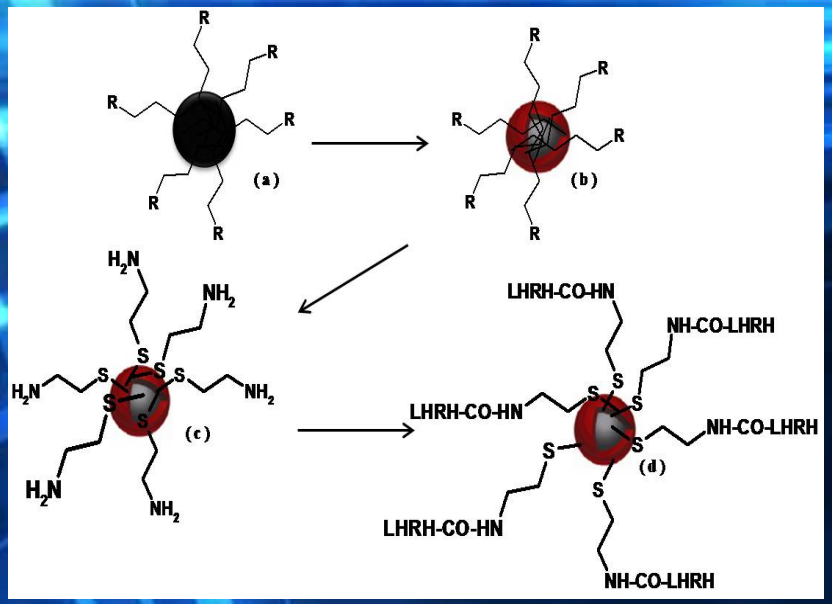
- **Their magnetic properties can be altered**
  - **Size, shape**, **composition**, crystal structure, surface function, vacancies, defects
- **They can be conjugated to bio-molecules**
  - **DNA, Peptides, Antibodies** etc
- **They offer multi-functionality- Remote Control**
  - **Controlled release, imaging, drug delivery** etc

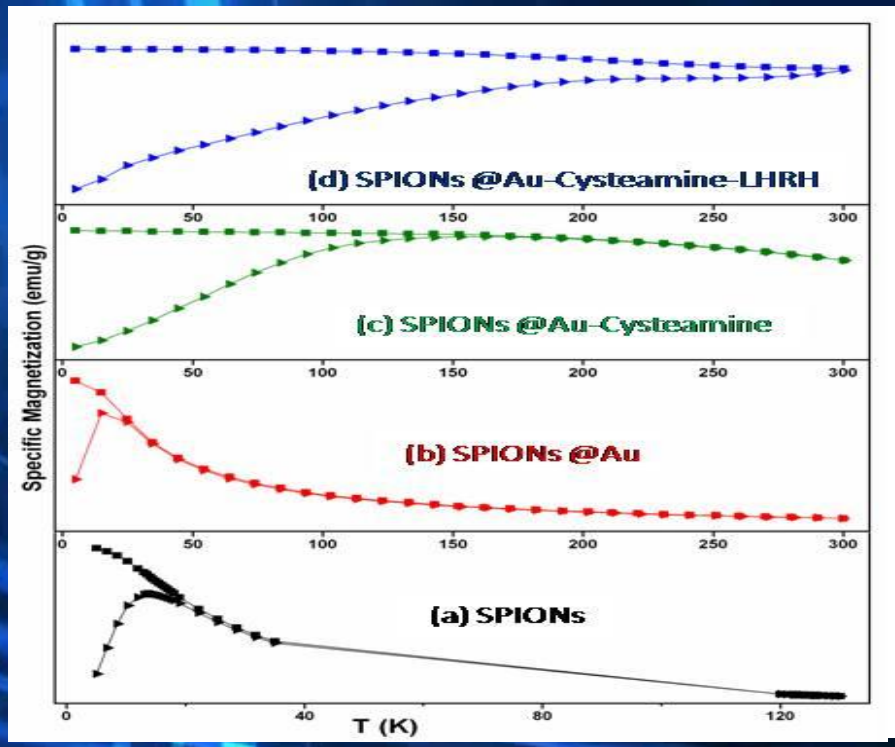
Acc. Chem. Res., 41(2), 179-189(2008)

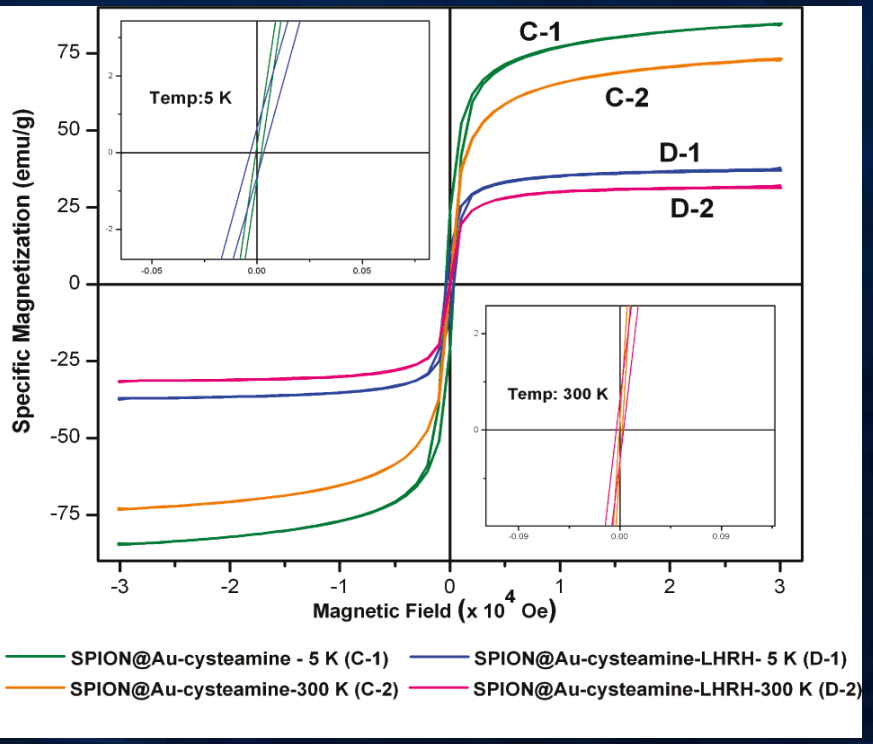
Magnetic Gold Nanoshells

Step-wise Changing of Magnetism through Step-wise Bio-functionalization

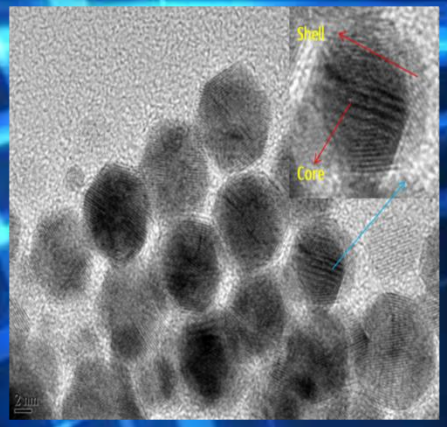
J. Phy. Chem. Lett., 2010

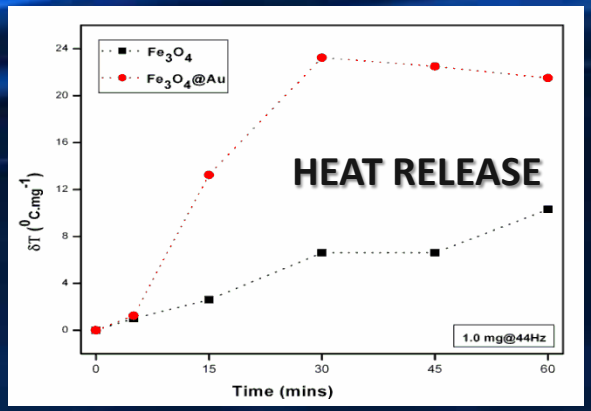


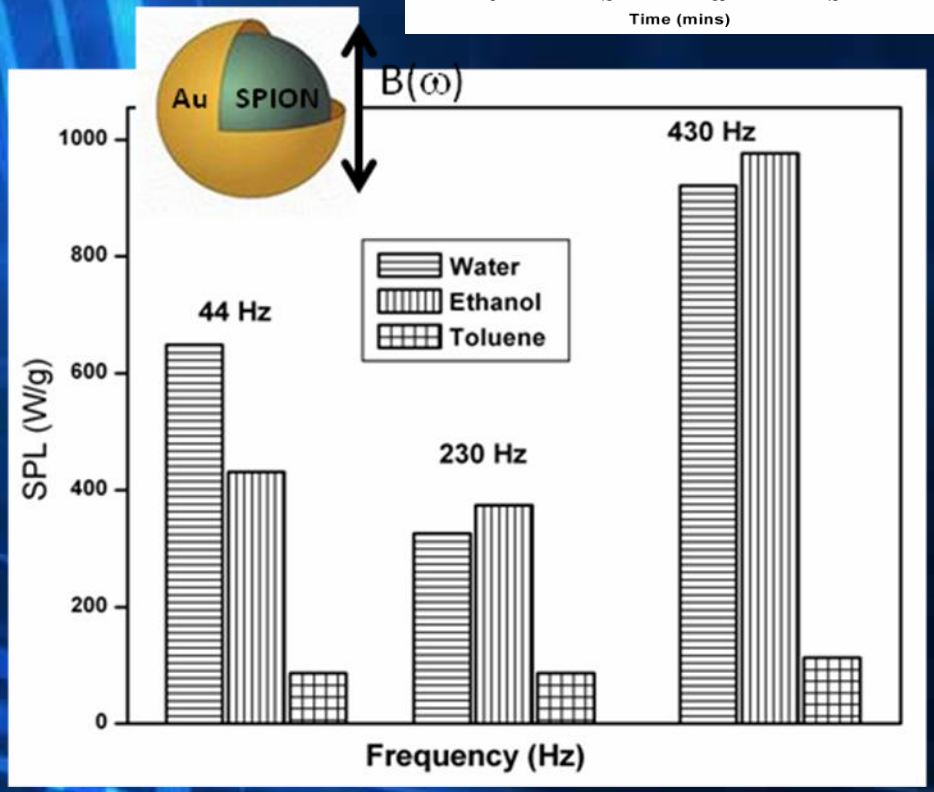




Influence of Gold Nanoshell on Hyperthermia



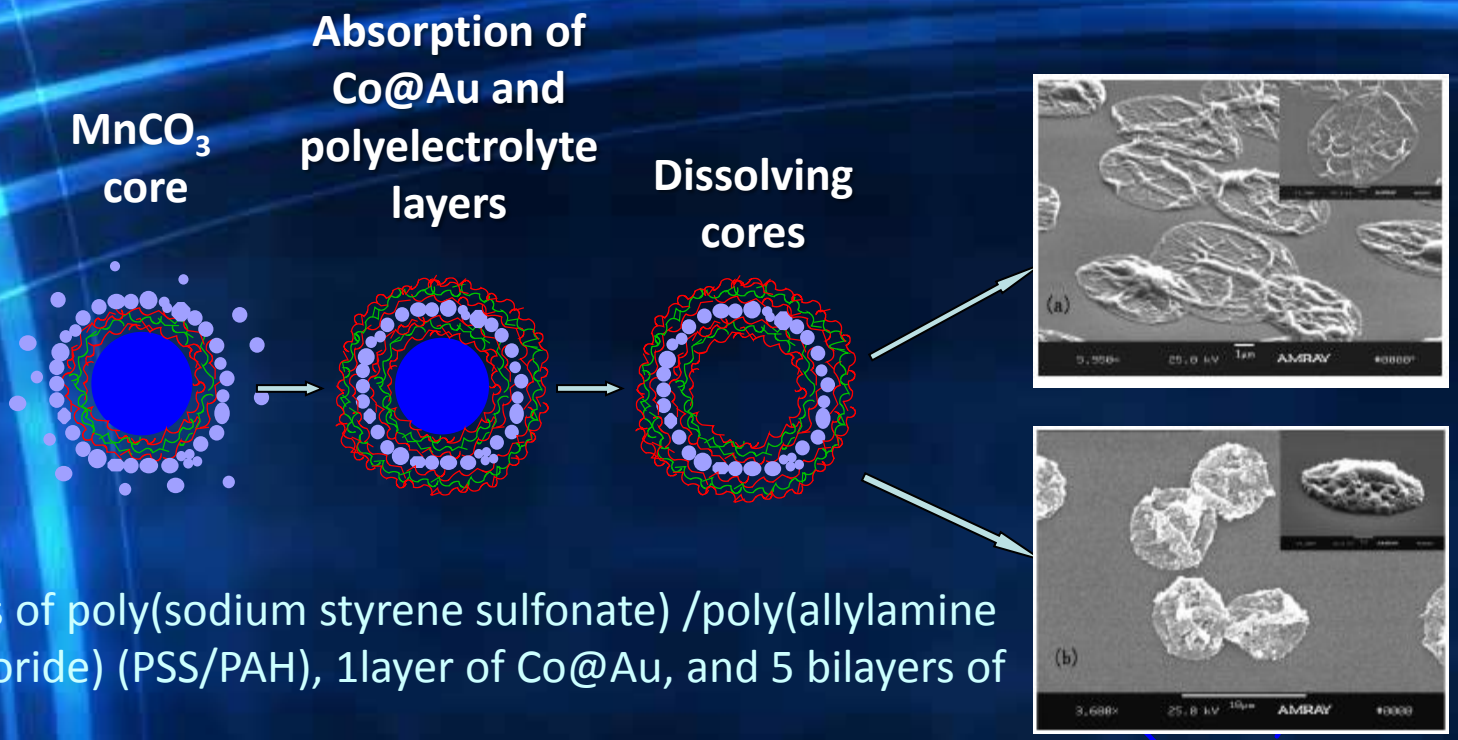




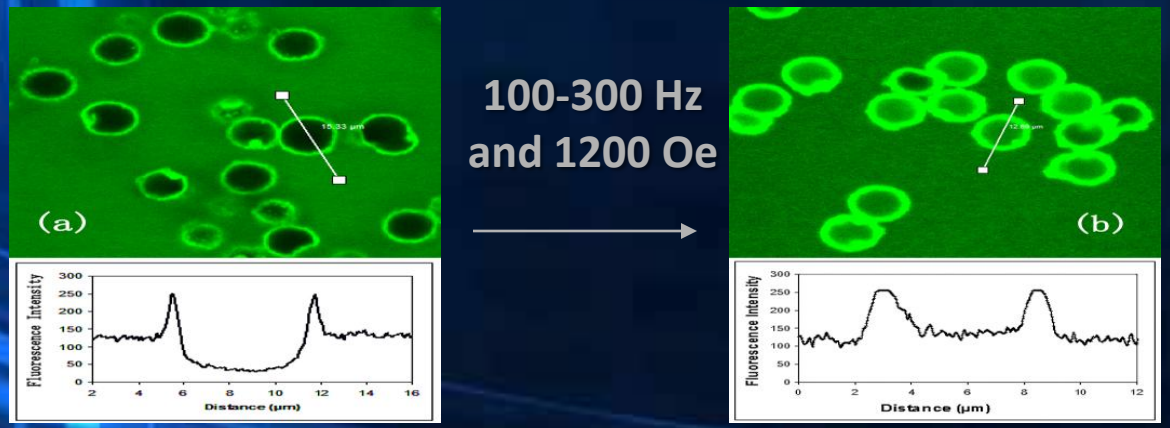
- A gold coating of ~ 0.4 to 0.5 nm thickness
- Application of low frequency oscillating magnetic fields (44 – 430 Hz)
- 4-5 fold increase in the amount of heat released
- Highest value of 976W/g in ethanol at 430Hz frequency

J.Phy.Chem. C., 2010

REMOTE CONTROLLED RELEASE



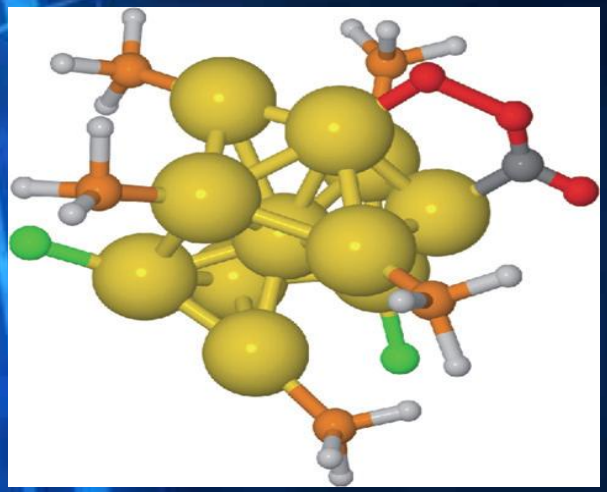
4 bilayers of poly(sodium styrene sulfonate) /poly(allylamine hydrochloride) (PSS/PAH), 1layer of Co@Au, and 5 bilayers of PSS/PAH.

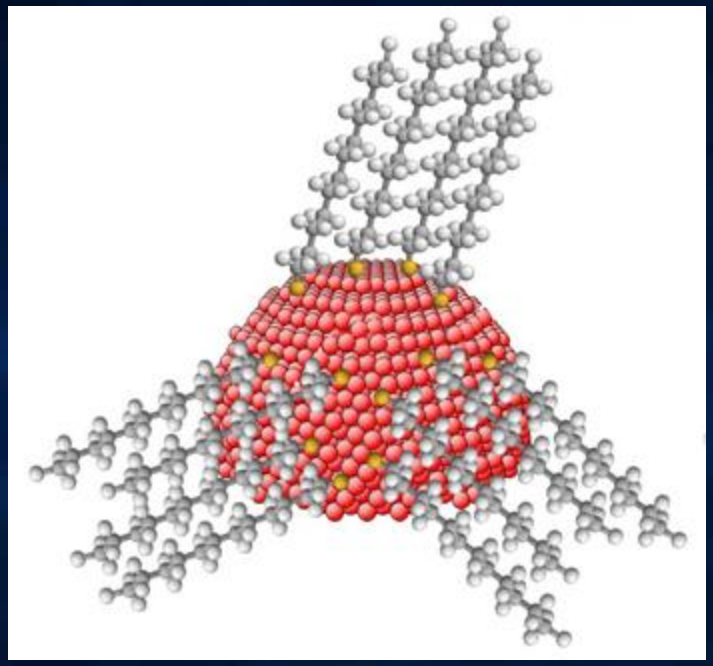


Z. Lu et al., Langmuir, 2005

Magnetism in small metal clusters

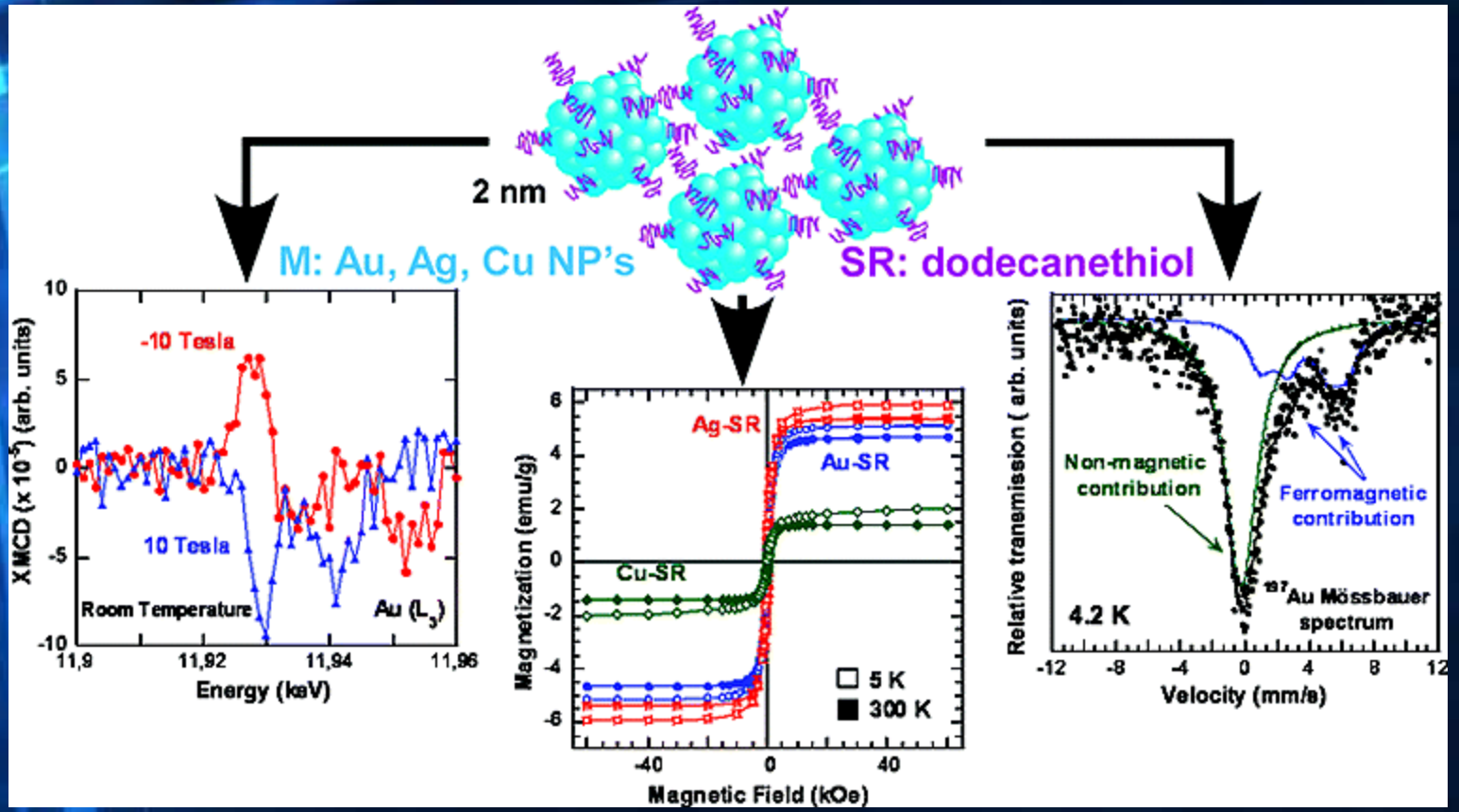
Magnetism in small Au clusters





Nanotechnology 19 (2008) 175701

Chemically induced permanent magnetism in small Au, Ag, and Cu Clusters



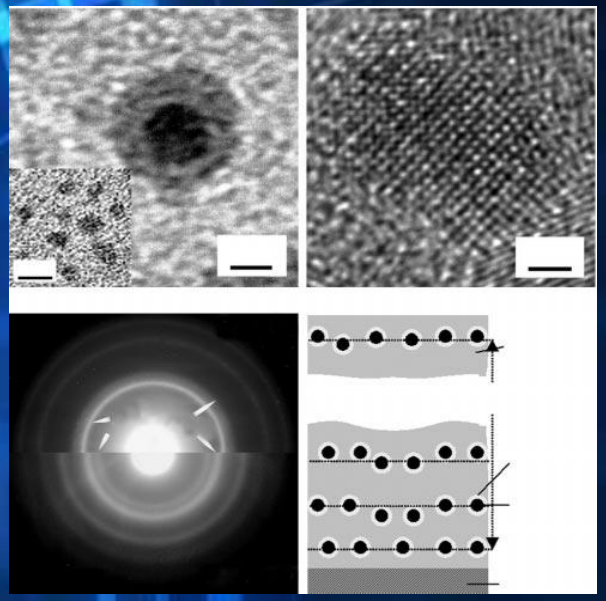
Nano Letters 2008, 8(2), 661-667.

Challenges

Core-shell interface

Understanding microscopic mechanism underlying the Exchange Bias effect is important

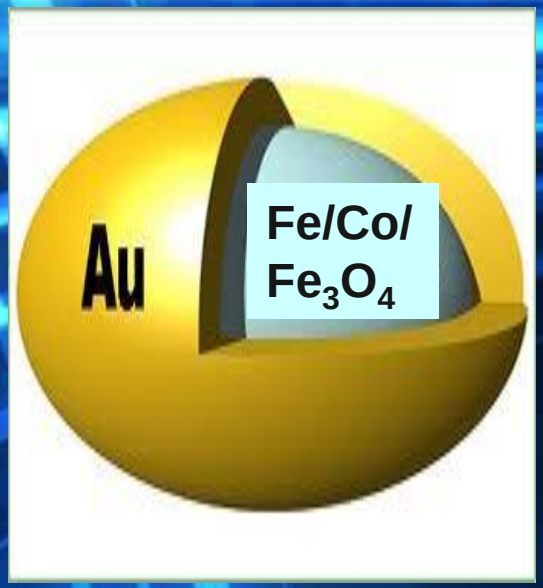
- Studied extensively in planar geometries
- Complicated in Core-shell NPs due to intrinsic disorder and finite-size effects



NATURE, 423, 2003

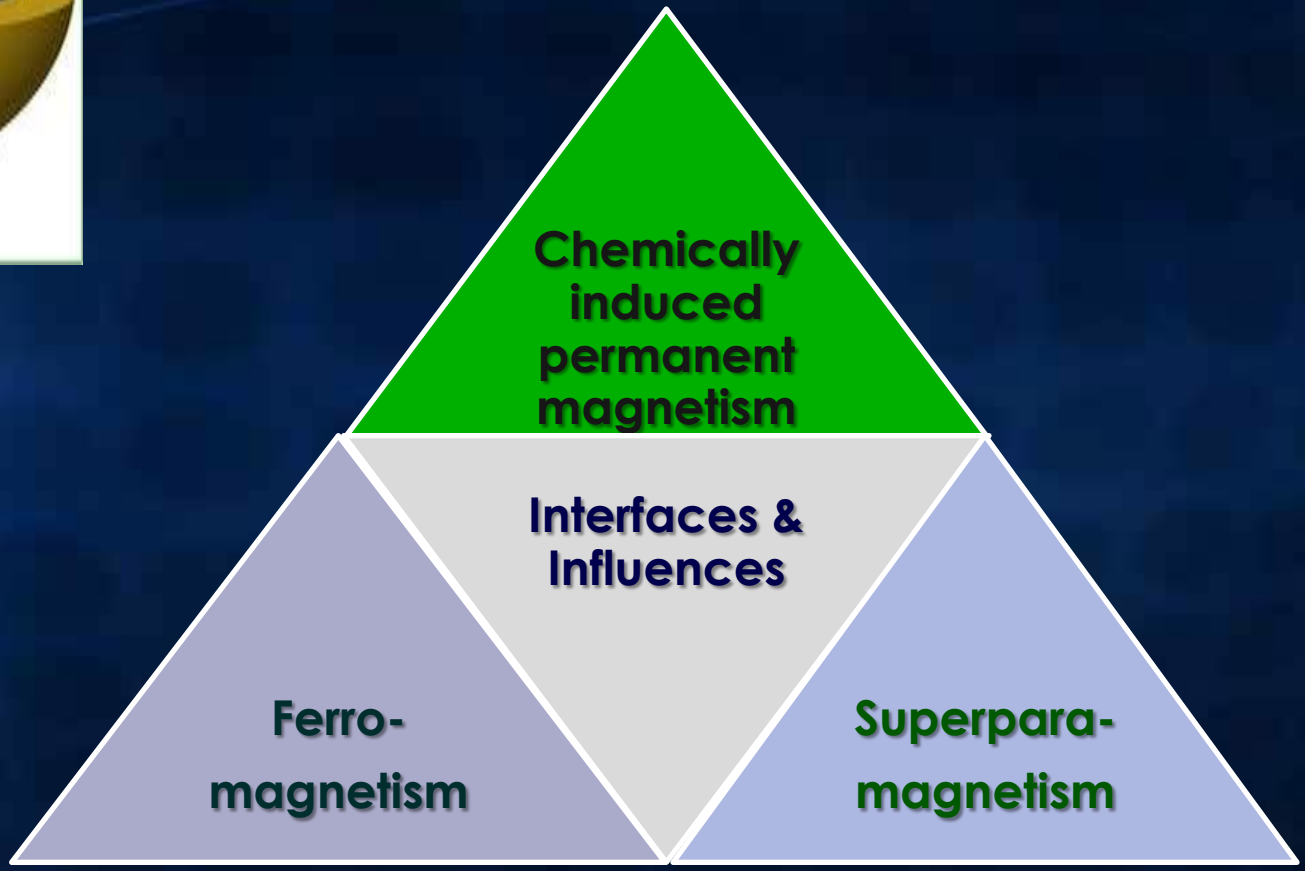
The intimate contact between the core and shell leads to effective exchange coupling

- Cooperative magnetic switching leads to tunable properties
- How to investigate these?

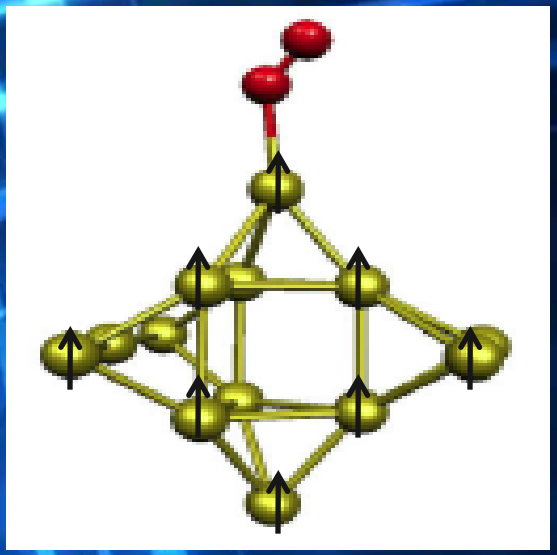


Magnetic Gold Shells

Microscopic examination of magnetism and magnetic influences is challenging



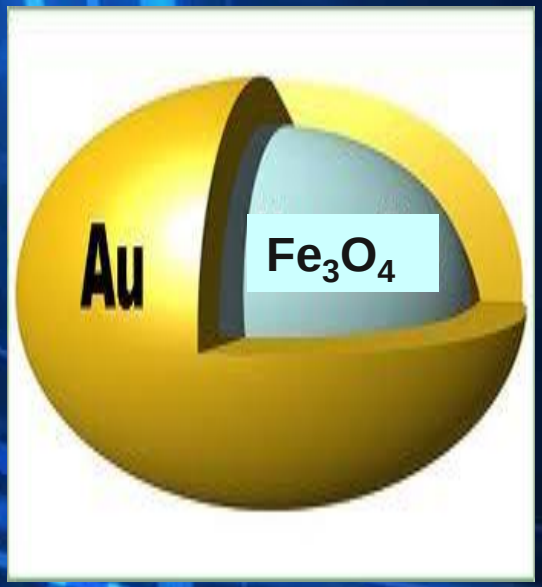
Implications for Catalysis & Medicine



Magnetic modulation of spin dynamics

Influence on HOMO-LUMO gap?

Magnetically modulated Au Catalysis?



Fundamental understanding of the origin of hyperthermia

Ultrafast spin dynamics at the interface?

What we need?

Sub-10nm spatial resolution with elemental specificity with a fsec time resolution with the capability to take instantaneous snapshot images of ultrafast spin dynamics .


Thank you for your attention !
LSU

ACKNOWLEDGEMENTS

LSU-CAMD
Josef Hormes, John Scott, Jost Goettert, Vadim Palshin

LSU-Chem. Eng
James Spivey

Princeton University
Wole Soboyejo

Virginia Tech
Beverly Rzigalinski

LSU-Bio & Agr. Eng
Christina Sabliov, Todd Monroe, Chandra Theegala

LSU-PBRC
Carola Leuschner, William Hansel

La Tech University
Yuri Lvov, Tabbetha Dobbins

Duke University
Warren Warren, Baastian Driehuys, Tamara Blanca