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# Laser-matter interactions and modern nanotechnology

Tatiana Itina

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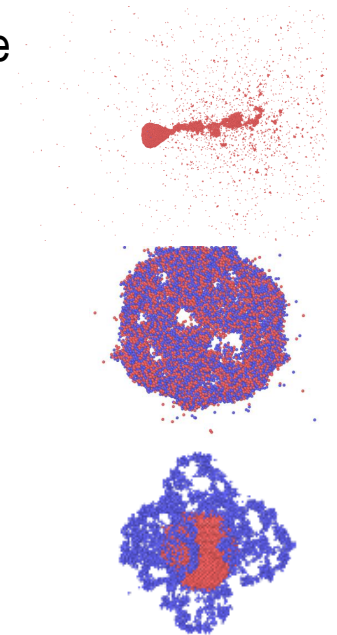
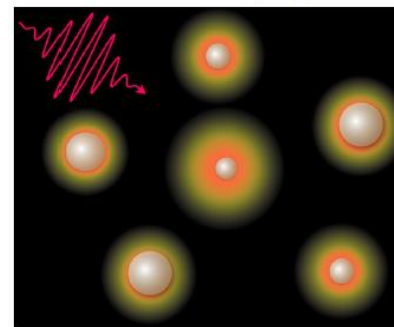
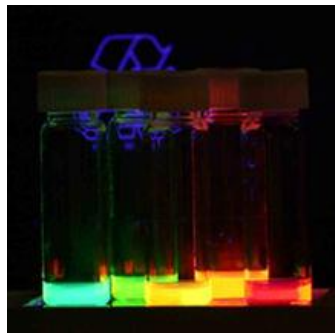
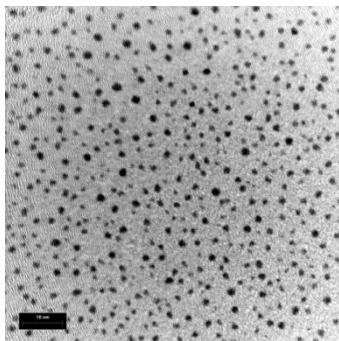
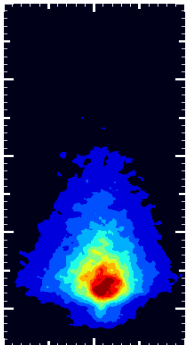
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# Laser-Matter Interactions and Modern Nanotechnology

**Tatiana E. ITINA**

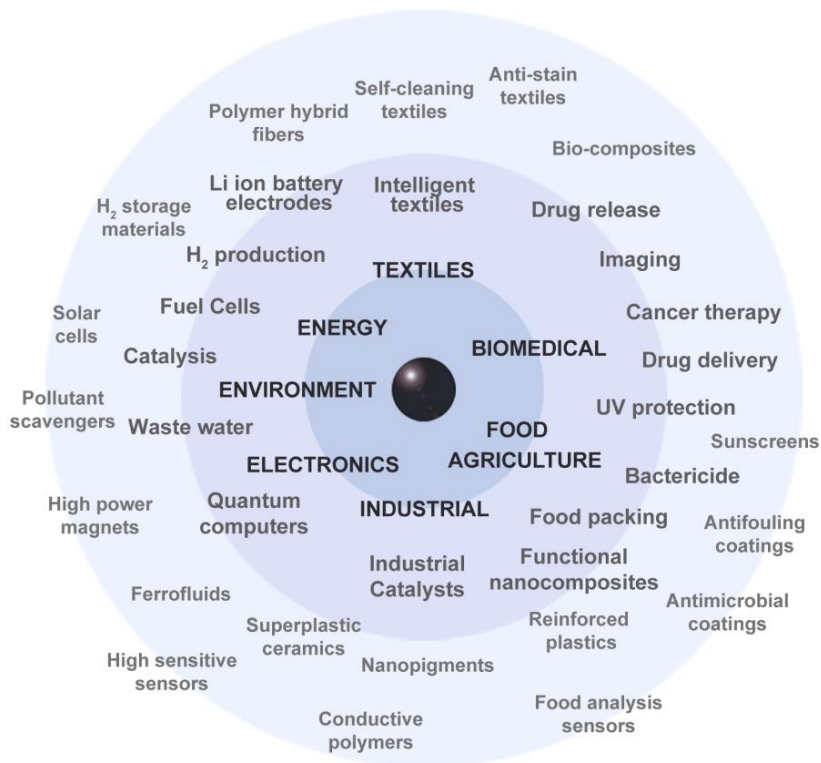
Laboratoire Hubert Curien, UMR CNRS 5516, Saint-Etienne, France

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**From experiments to simulations,  
from fundamental research to applications**

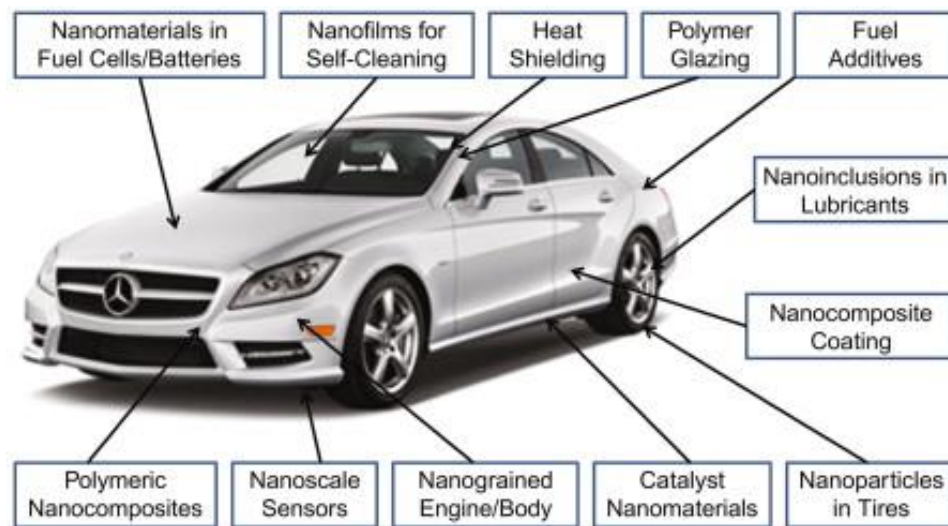
# Nanoparticles and Nanocomposites



## Aerospace and Defense



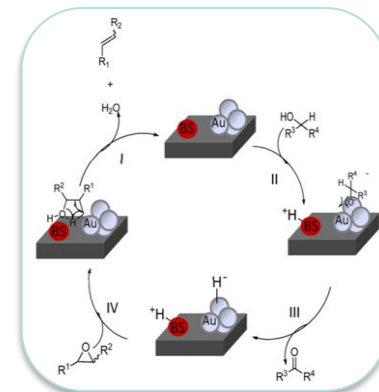
## Car industry



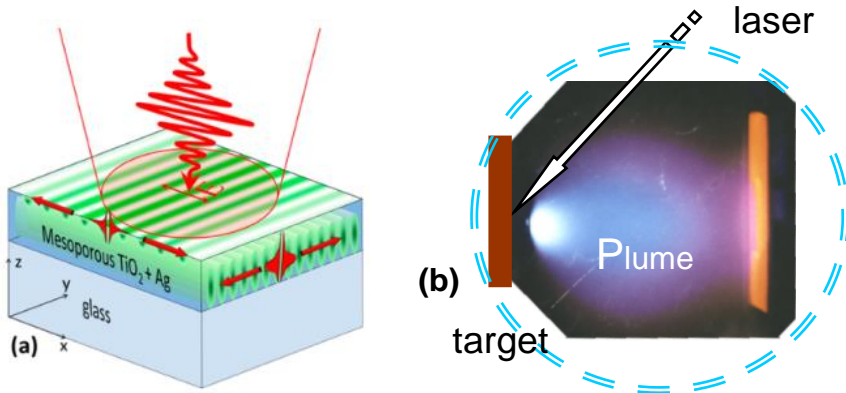
## Optics and security



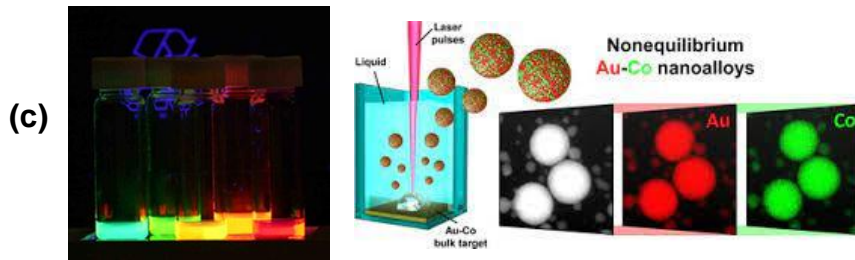
## Sensors and Catalysis



# Laser Interactions



In gas or in vacuum with different geometry



In liquids or in the presence of a colloids

## Laser interaction commonly involve:

- Energy propagation, electronic excitation, light scattering, reflection and absorption
- Electron-phonon relaxation, heat diffusion, pressure wave propagation, rarefaction and tensile stress, phase transitions, mechanical fragmentation
- Material removal (ablation), plasma plume expansion, reactions, clustering, etc...
- Often multi-pulse irradiation is used with various scanning and focusing conditions

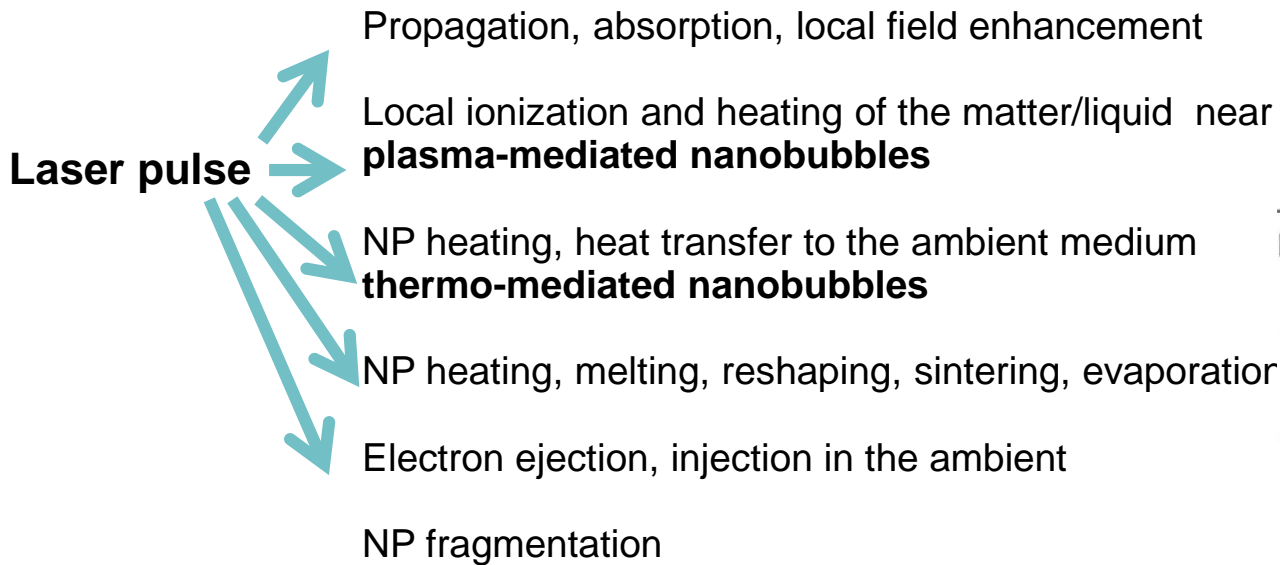
## Laser-induced nanoparticle formation

- Green synthesis methods, bio-compatibility
- Control over NP properties
- Scaling-up possibilities
- Possibility to create stable ultra-structures, nanoalloys, hybrids, etc.
- **Additional laser-particle interactions involve many physical and chemical processes**

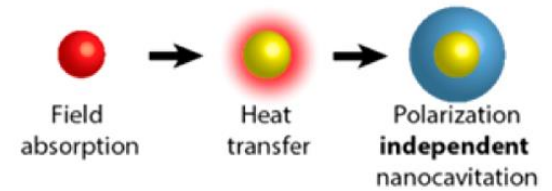




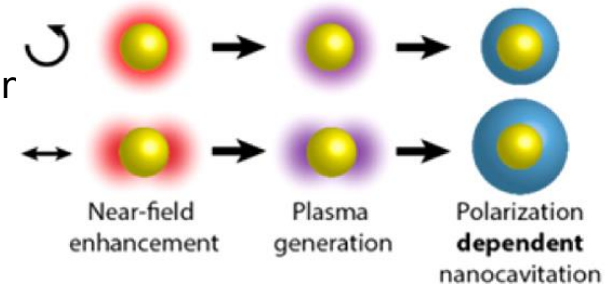
# Ultra-short Laser Interactions with NPs



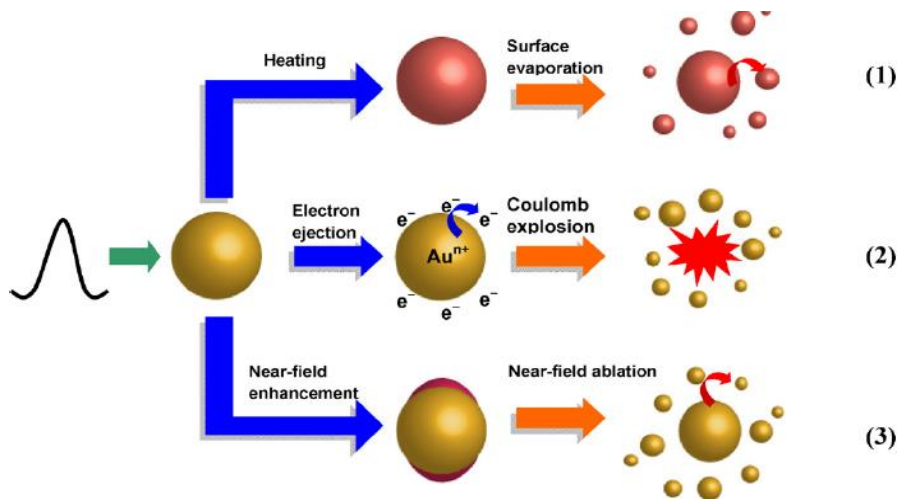
## Thermo-mediated nanobubbles



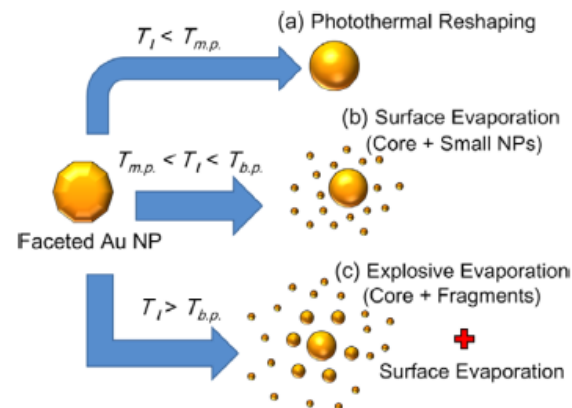
## Plasma-mediated nanobubbles



*Lachaine et al. Photonics (2013)*



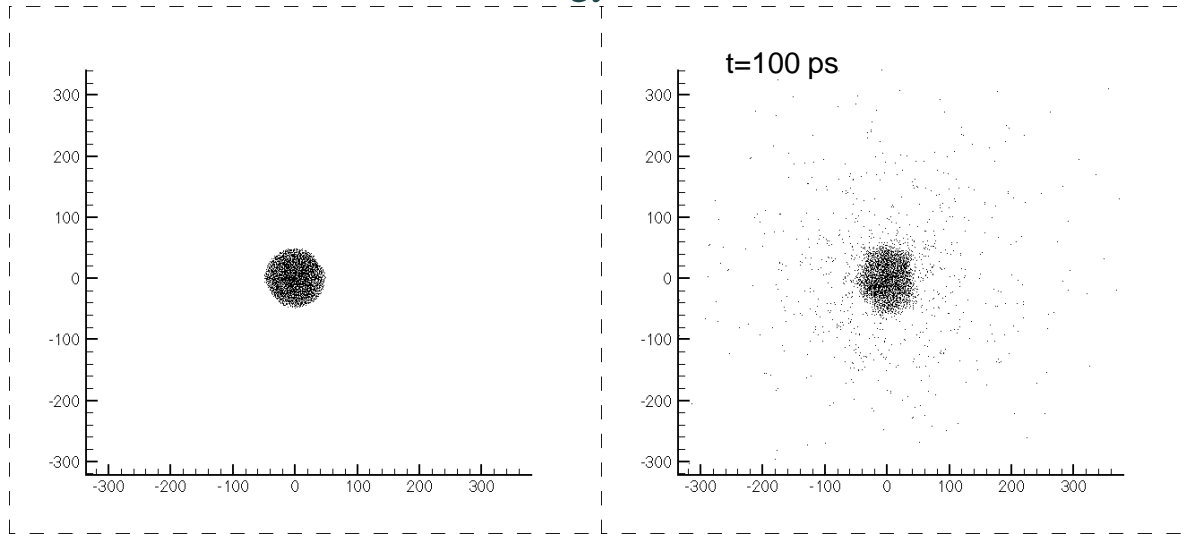
*Hashimoto et al. J. PhotChem&PhotBiology (2012)*



*Strasser et al., J. Phys.Chem. C 118, 25748 (2014)*

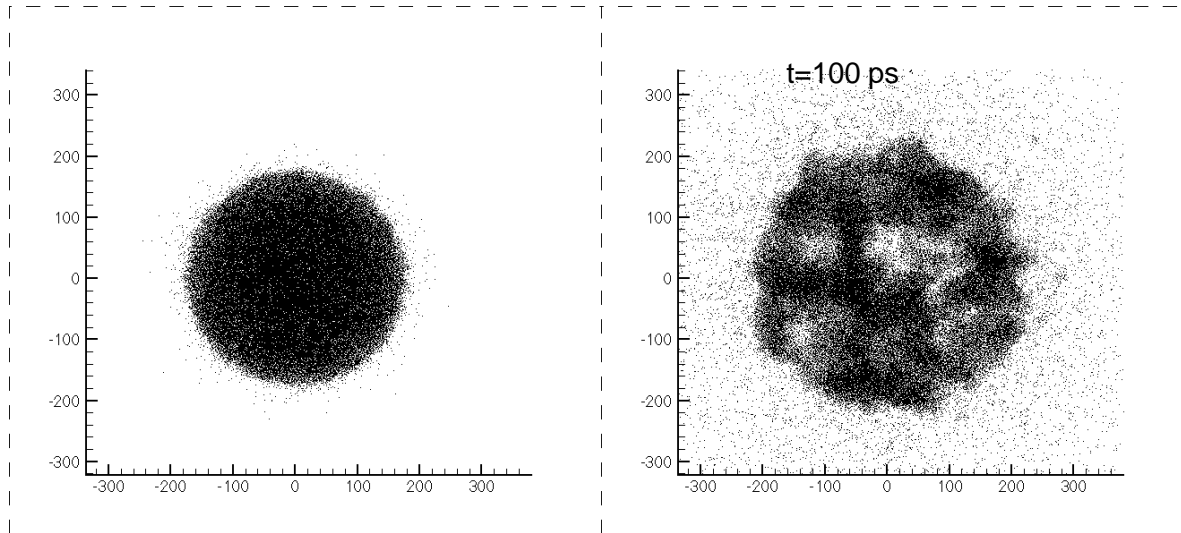
# Laser Fragmentation of NPs: MD simulations

$R=50 \text{ \AA}$ , absorbed energy  $e=0.6 \text{ eV/mol}$



Smaller NPs  
**Evaporation**  
(can be also due to Coulomb Explosion for small clusters and fs laser)

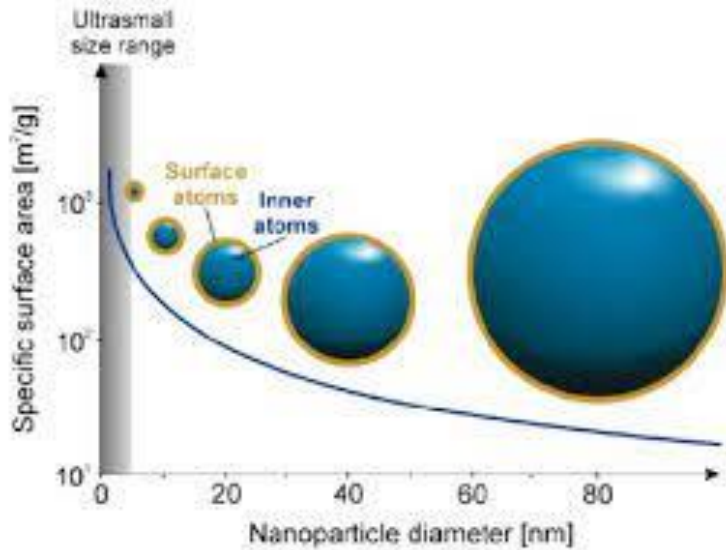
$R=150 \text{ \AA}$ , absorbed energy  $e=0.6 \text{ eV/mol}$ .



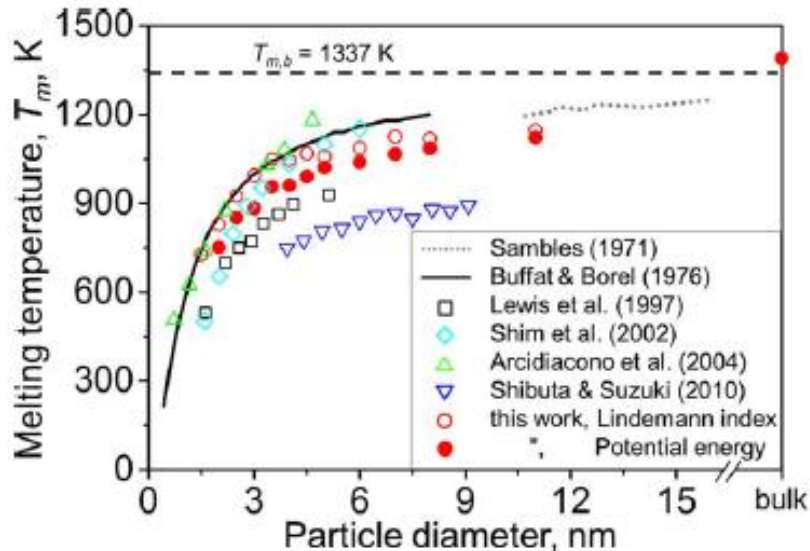
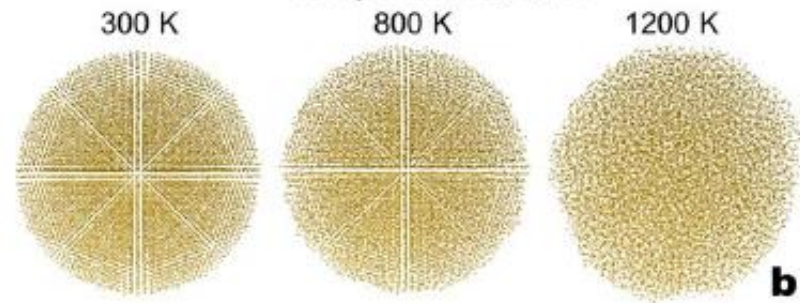
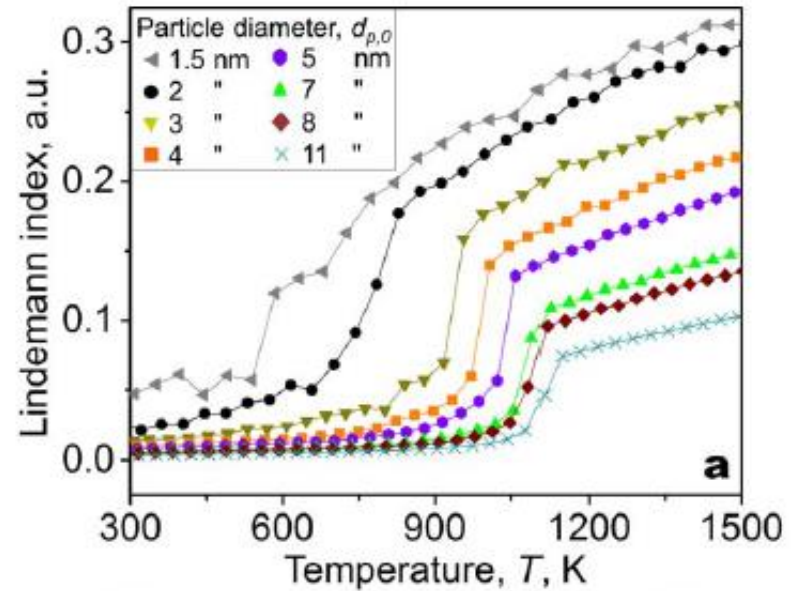
Larger NPs  
**Fragmentation due to thermo-mechanical effects**

# NP's Heating and Melting : Size Effects

## Melting and structural transformations: role of NP's sizes



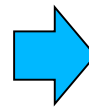
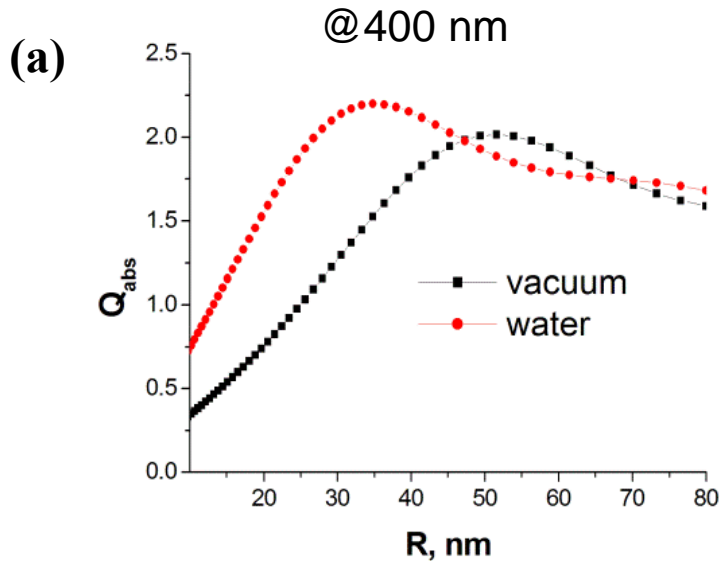
K. Zarschler et al. *Nanomed-Nanotechnol* 12, 1663-1701 (2016).



E. Goudeli et al., *AIChE* 2015

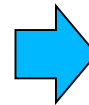
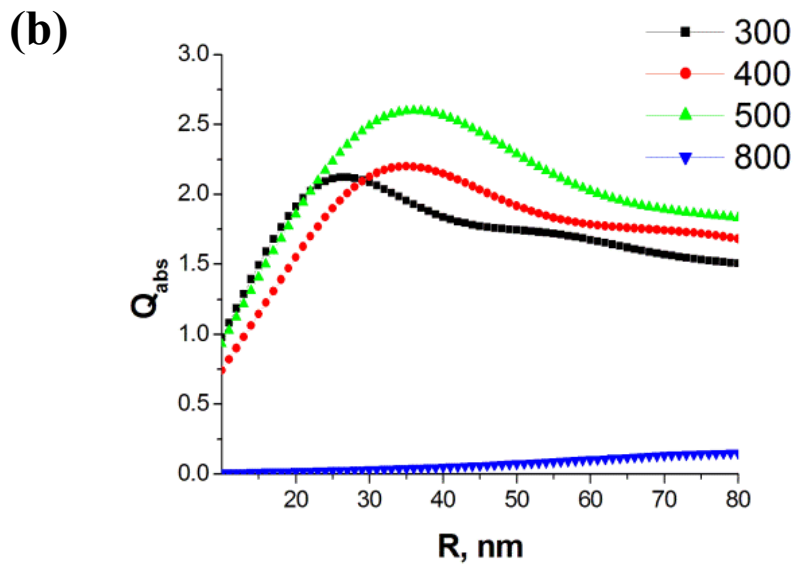
# Laser Energy Absorption vs NP's size

Absorption coefficient / efficiency  
(beyond SPR for gold)



**Generalized Mie theory** =>  
Red – Gold NPs in for water  
Black – Gold NPs in vacuum

**peak in absorption at**  
 $R=R^* \sim 30$  nm for Au NP in  
water



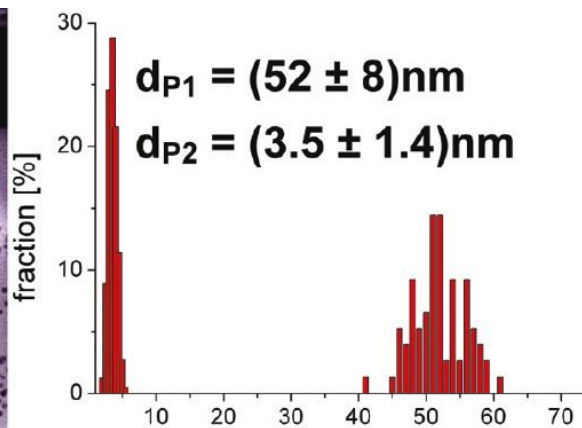
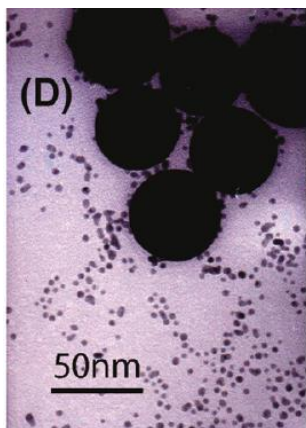
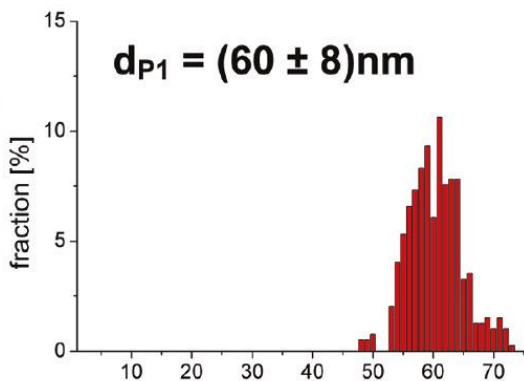
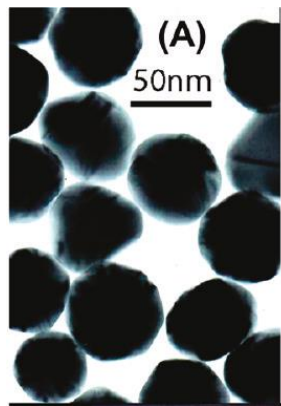
Gold NPs in water for **different**  
**wavelength @300-800nm**

**peak position is shifted to**  
**larger  $R=R^*$  if wavelength rises**



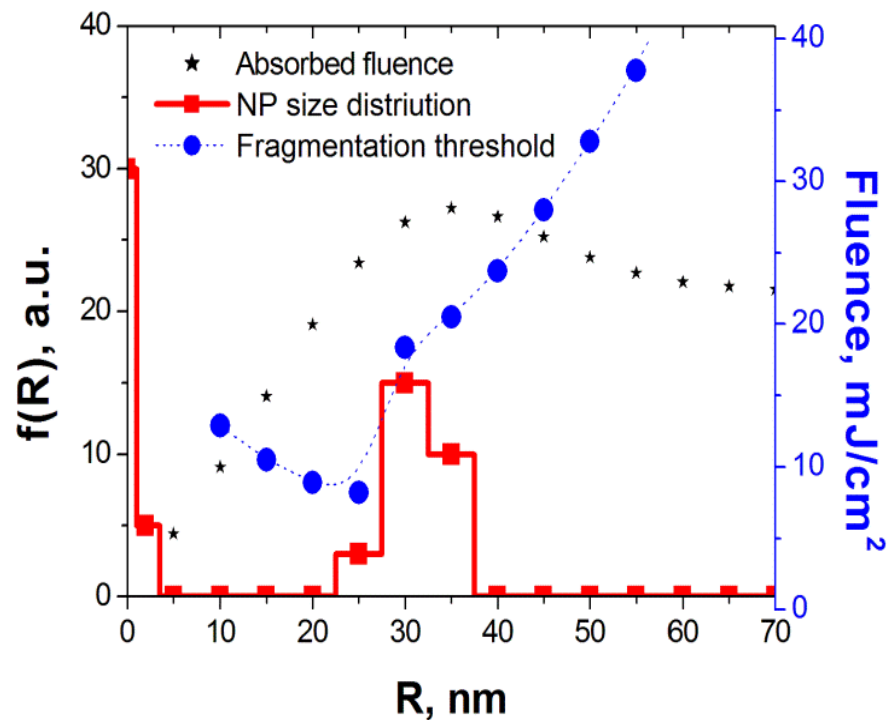
# Ultra-short Laser Interaction with Au NPs in Water

## Experiments



Werner et al. *J. Phys. Chem. C*, 2011, 115, 8503

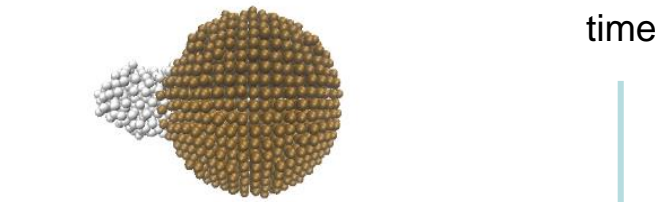
## Modeling



Good correlation with  
Experimental results

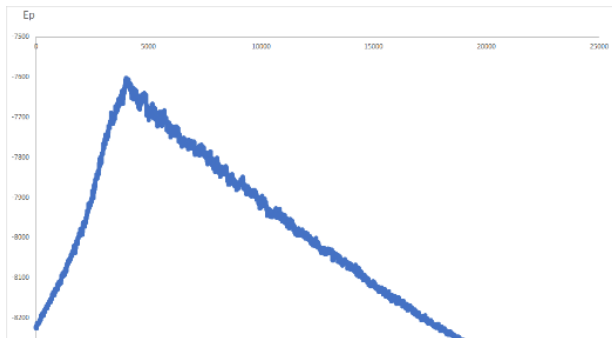
# NP Sintering or Alloying

FeNP R=10 uc   AuNP R = 20 uc   FeNP 1700+AuNP 1700



Different sizes and species, but the same temperatures

Time-evolution of energy



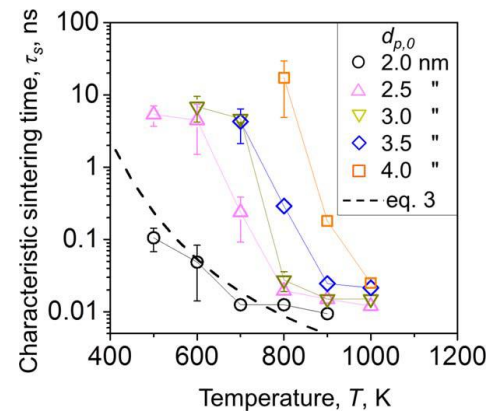
time

100ps



- Simulation starts from a Janus configuration
- Then both particles are heated to the same temperature
- Then, the system is cooled down back to room temperature

Core-shells can be formed when one particle is much larger and is composed of a material with different cohesive energy



E. Goudeli et al.,  
AIChE 2015

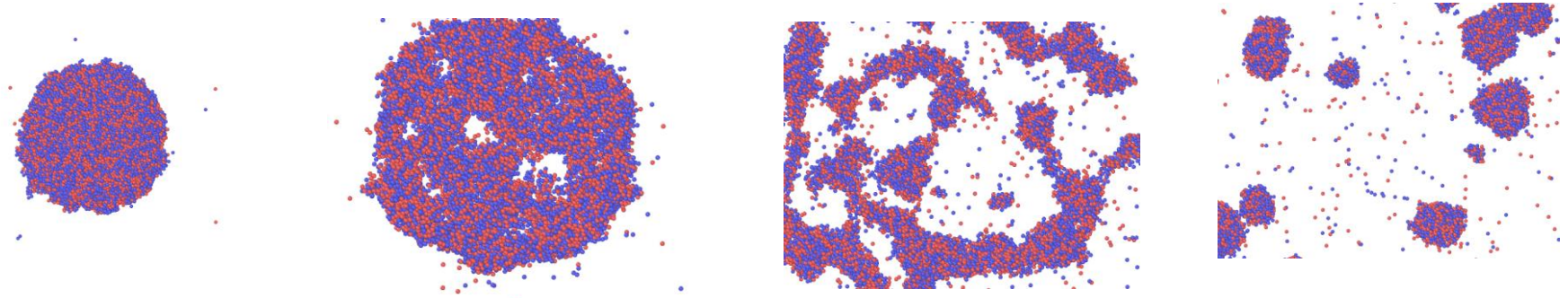
# Fragmentation vs Sintering, Additional Fragmentation

Simulations starts from a larger janus AuCo NP, heating to 1700 K

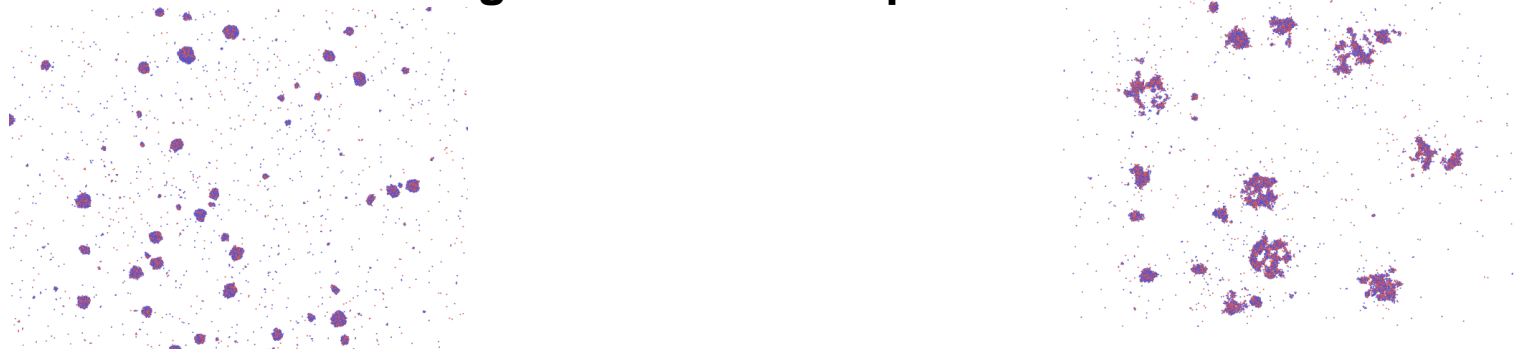
AuCo, Au NP R = 40 un, rapidly heated up to 1700 K



Simulations starts from Alloy AuCo NP, R=40 uc rapid heating above  $T_c$



Simulations starts from the fragmentation results, additional fragmentation takes place



# Ion-impregnated Porous Thin Films Treated by Laser

- sol-gel mesoporous films are prepared and impregnated by salts
- these samples are then irradiated by laser

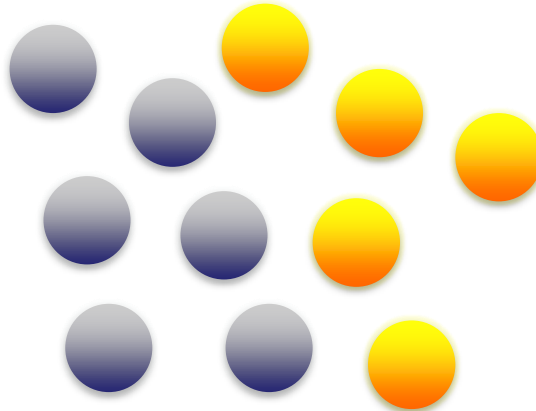
Porous films:  
 $\text{SiO}_2$  ,  $\text{TiO}_2$

Sol-gel synthesized  
Porosity ~ 10%  
Porous size 5-7 nm

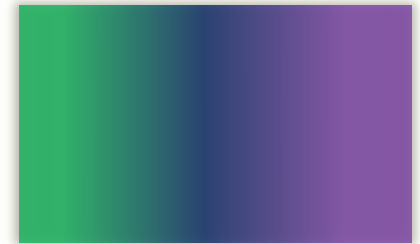


Metallic NPs:  
Ag, Au

Produced by soaking in metallic  
salts solutions with consequent  
reduction  
Initial size < 3-5 nm

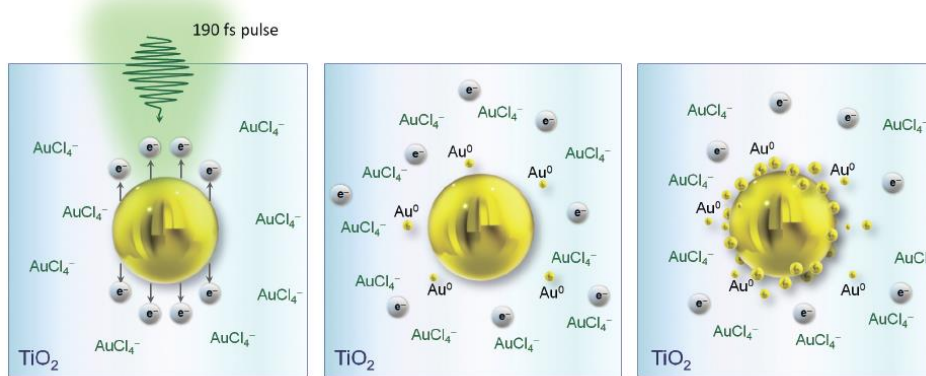


Laser irradiation:  
CW  
Nanosecond  
Picosecond  
Femtosecond



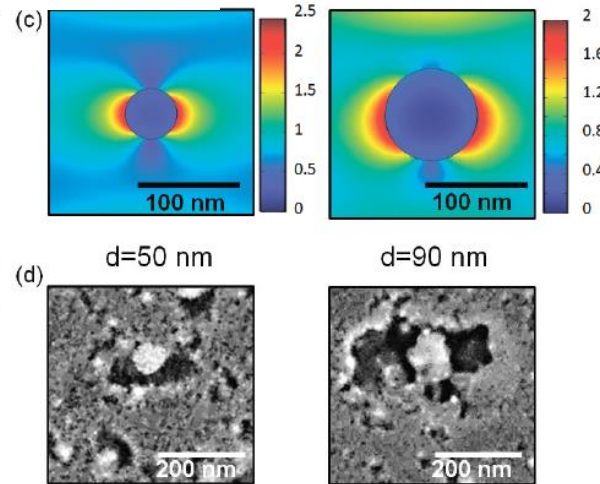
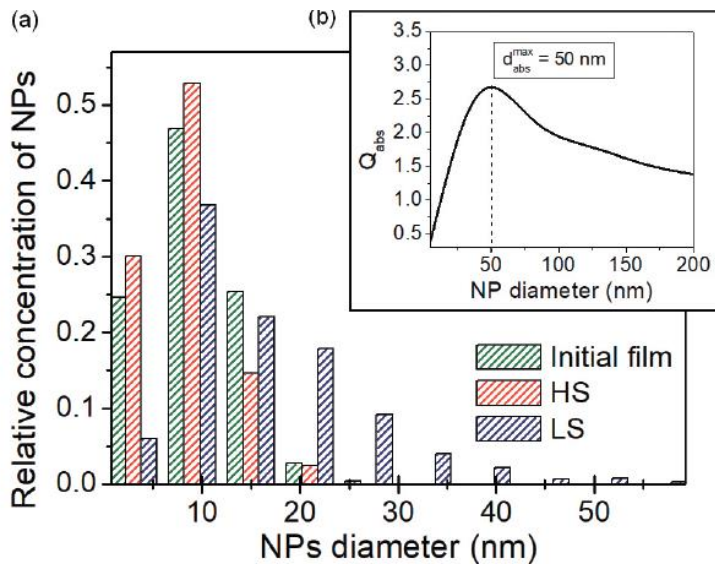
**Ion-impregnated porous glasses and semiconductors**  
can be used as samples for following laser-induced nano-structuring

# Au NPs in TiO<sub>2</sub> under Ultra-Short Laser Irradiation



**At the beginning, the matrix is solid**

*Y Andreeva et al.  
The Journal of Physical Chemistry C  
124 (18), 10209-10219, 2020*



fs laser  
 $\lambda = 515 \text{ nm}$

(a) Size distribution of gold NPs in before and after laser treatment.

(b) Calculated absorption efficiency of the nanocomposite based on TiO<sub>2</sub> as a function of the diameter of gold NPs

(c) Calculated field distribution of gold nanoparticles in TiO<sub>2</sub> ( $n=1.6$ ) for spheres with the diameters of 50 and 90 nm.

(d) SEM images of the NPs produced in LS mode ( $F = 57 \text{ mJ/cm}^2$ ,  $V_{sc} = 0.16 \text{ mm/s}$ )



## Summary and Conclusions

- Laser is a very powerful and versatile tool suitable not only for laser ablation but also for nanoparticle synthesis.
- Particle size, composition, geometry, atomic masses, binding energies, excess energy, environment, heating/cooling protocol affect the results of laser interactions.
  - NPs of smaller sizes easily melt and evaporate, while larger NPs behave more like bulk material, so that thermo-mechanical fragmentation of over-heated material plays a role in their fragmentation. Phase diagrams of NPs smaller than ~30-100 nm should account for NP's size.
  - Fragmentation threshold of alloy nanoparticle strongly depends on their composition (excess energy).
  - When different nanoparticles interact they may **either sinter, mix or even be fragmented** depending on their sizes and heating/cooling rate. To reach alloying without fragmentation NP should not be over-heated.
- Optical properties are strongly related to size and distribution of NPs after laser treatment, are controlled by laser processing parameters.