Computational Materials Design: DFT Application in Nuclear Medicine

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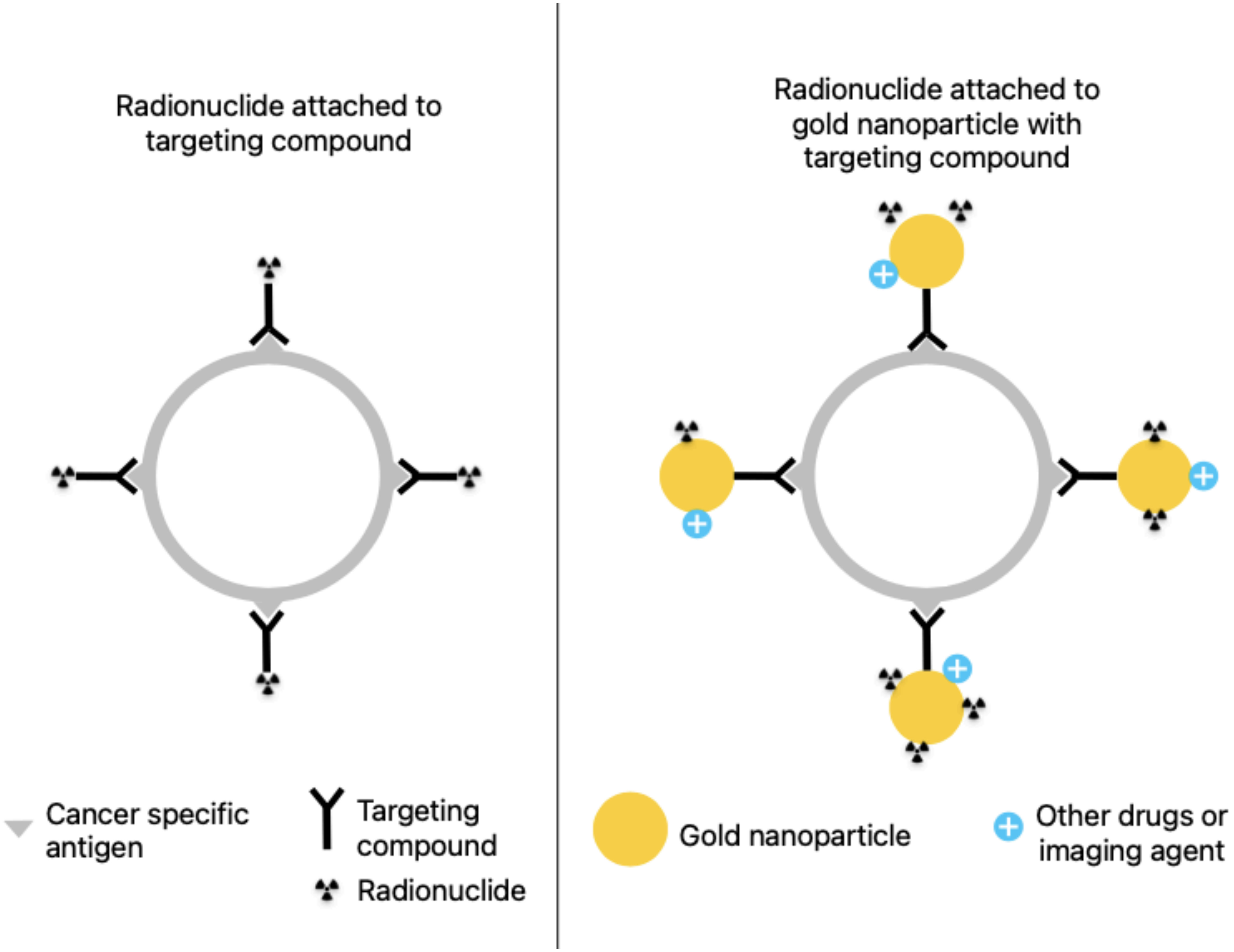
2025/03/25

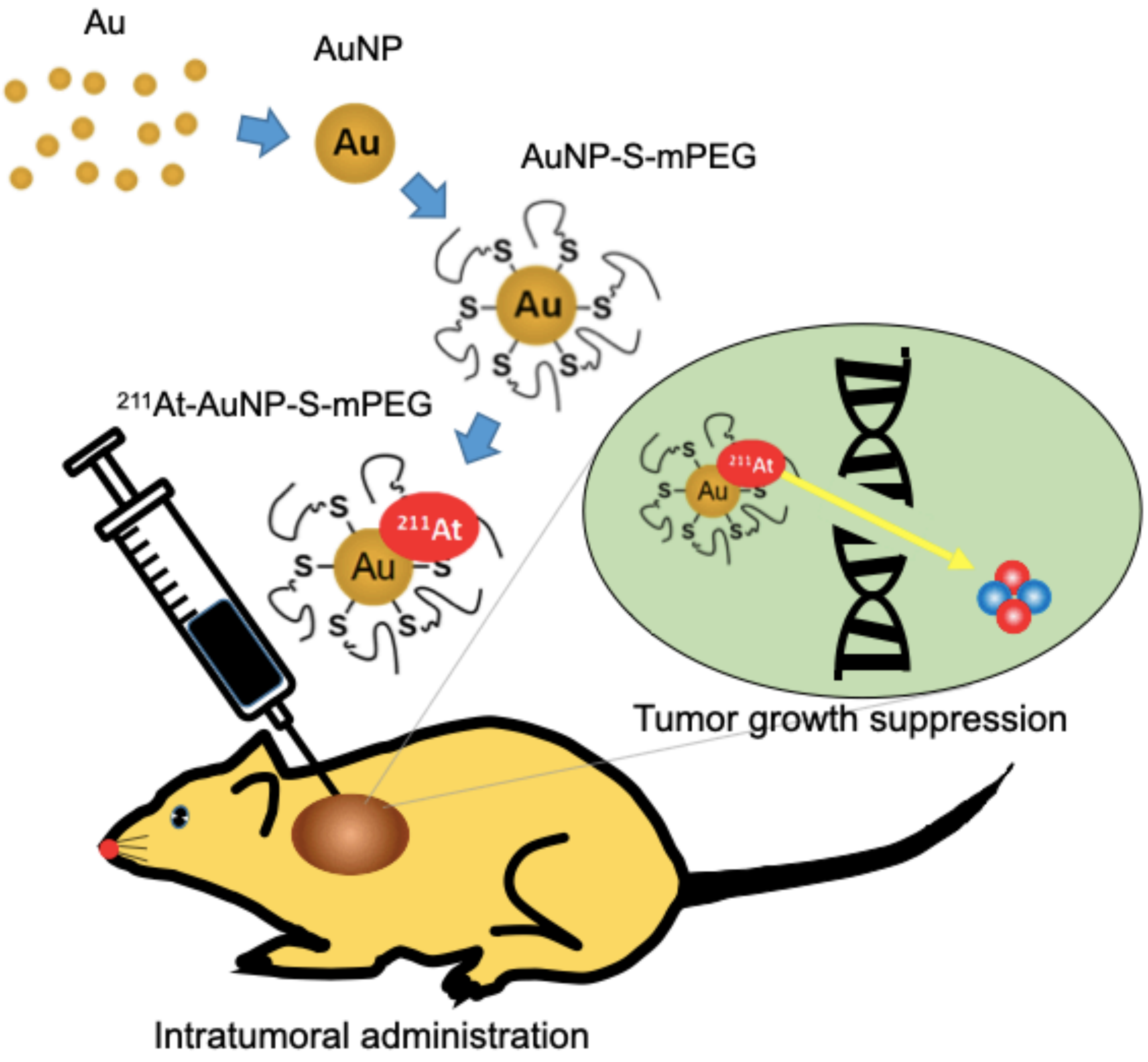
Introduction

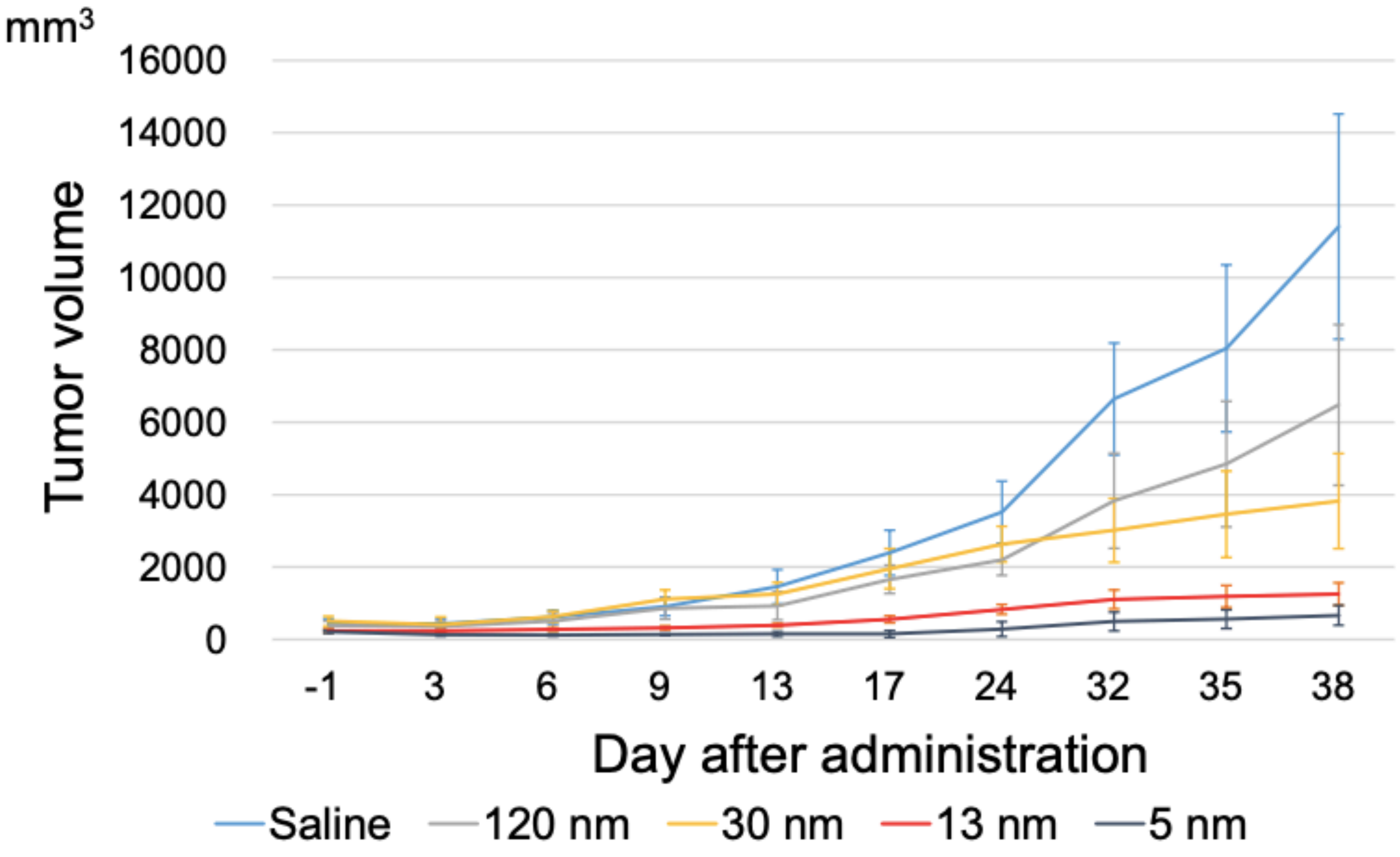
- Targeted alpha therapy (e.g. with ^{211}At) is being developed as a new method for cancer therapy
- Compared to traditional methods, using gold nanoparticle as carrier could potentially bring more radionuclides to the target area, along with other drugs or imaging agents



[1] J. Tanudji et al., Phys. Chem. Chem. Phys 26, 12915 (2024)

Experimental findings of ^{211}At/Au use



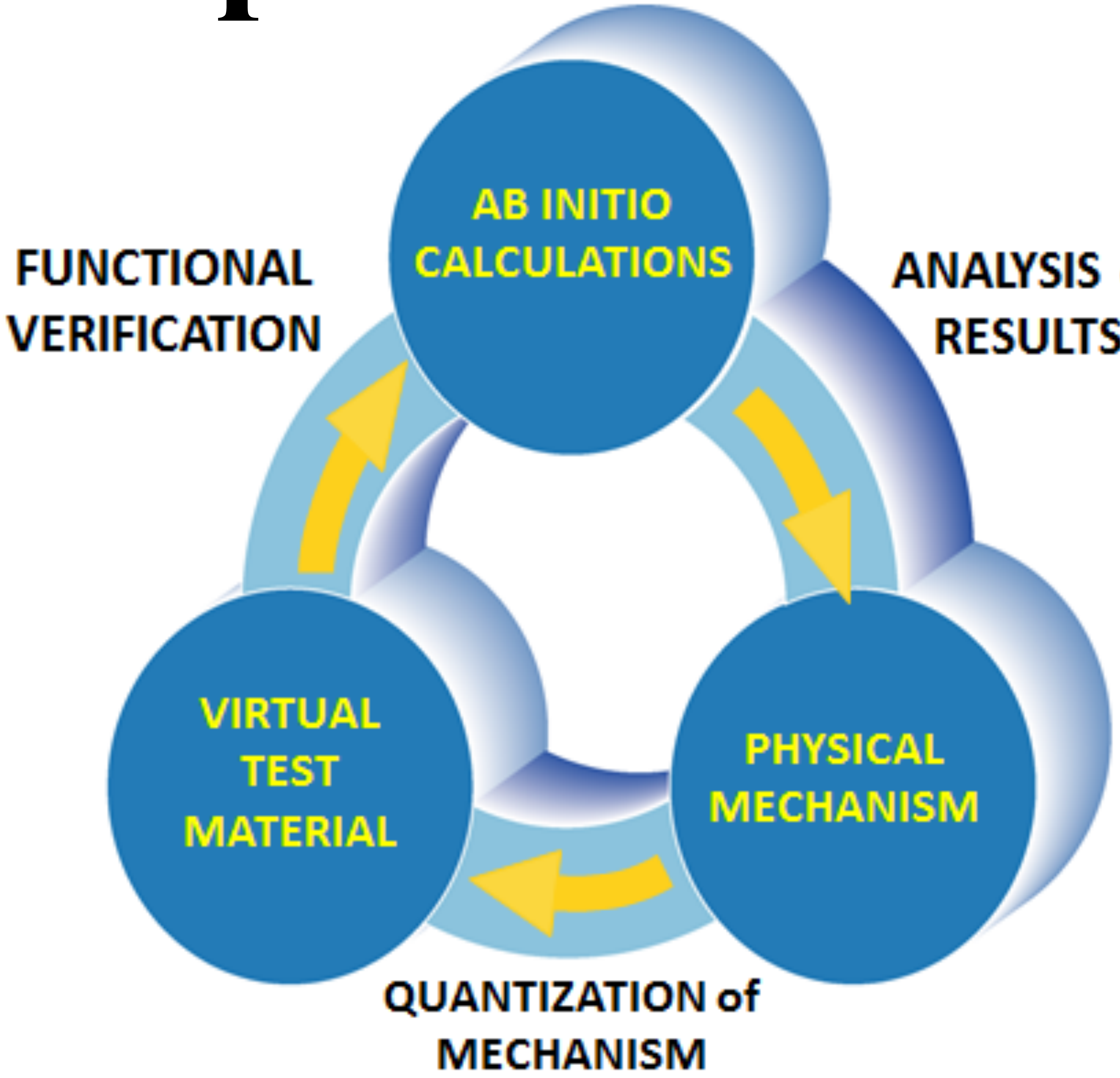


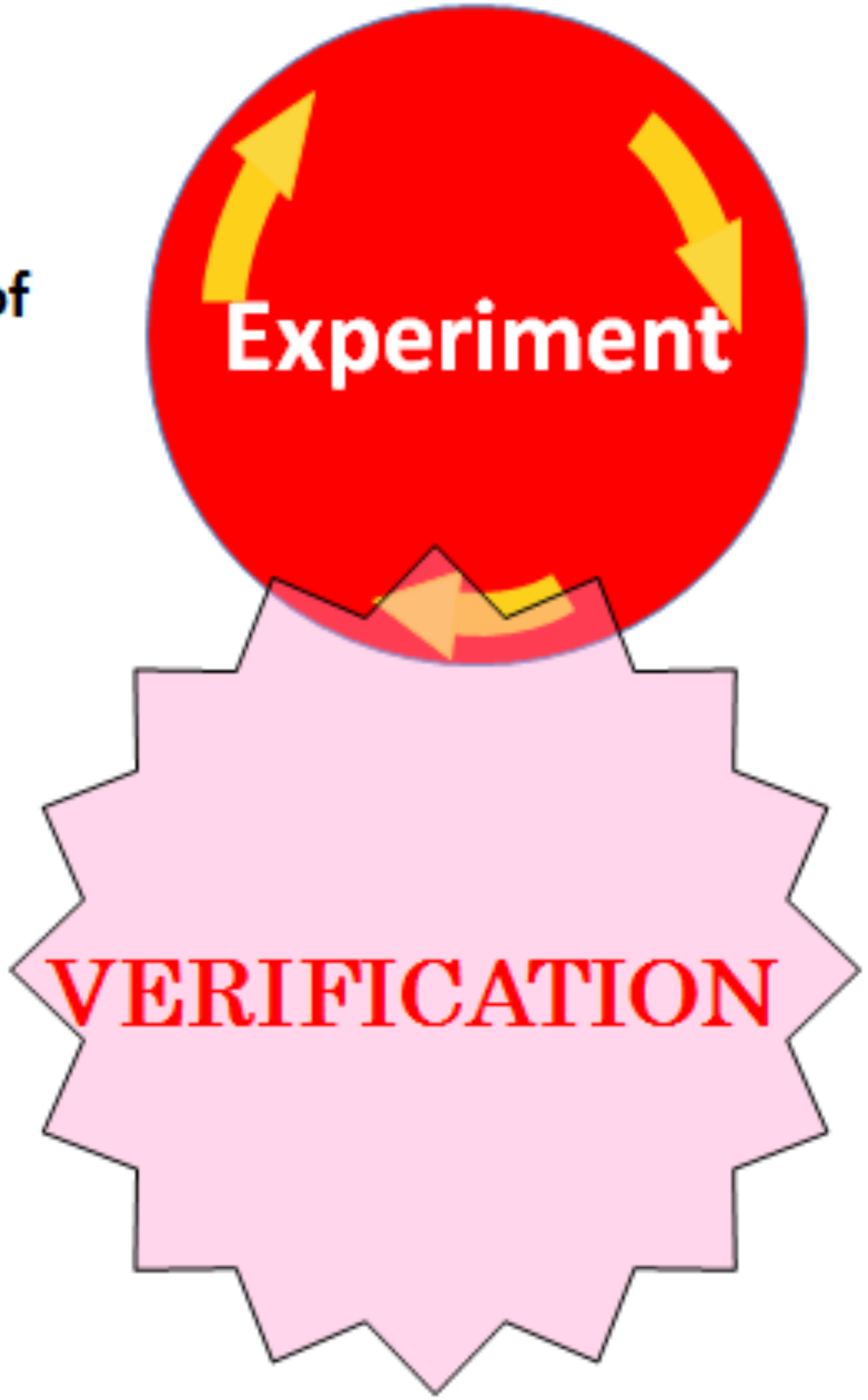
[2] H. Kato et al., J. Nanobiotechnol. 19, 233 (2021)

Motivation

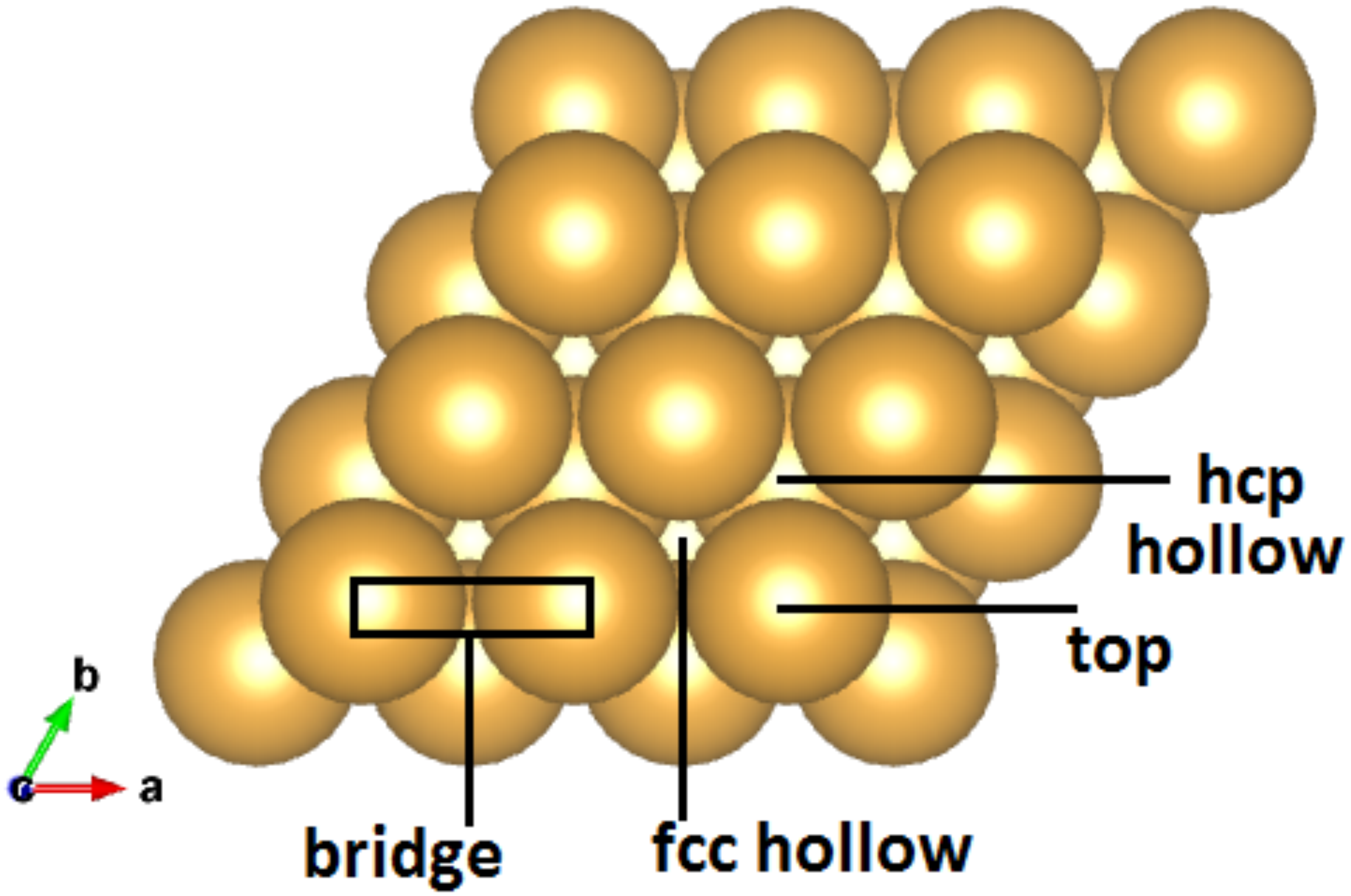
- Experiments have shown gold nanoparticles to be able to hold astatine
- Lack of fundamental understanding to allow us to design better or more efficient carrier
- The goal of this series of studies is to design nanoparticles that can be used to carry astatine more effectively

Computational Materials Design





Adsorption of astatine on Au(111)



Adsorption energy calculation

$$E_{ads} = \mathrm{At/Au(111)} - (0.5\mathrm{At_2} + \mathrm{Au(111)})$$

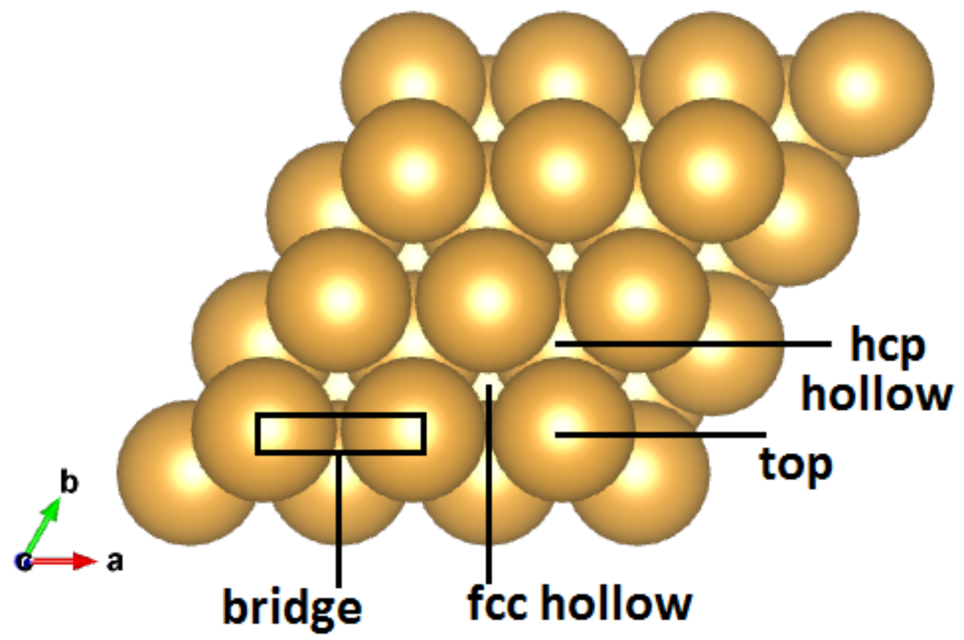
One astatine on Au surface

Isolated astatine

Clean Au surface

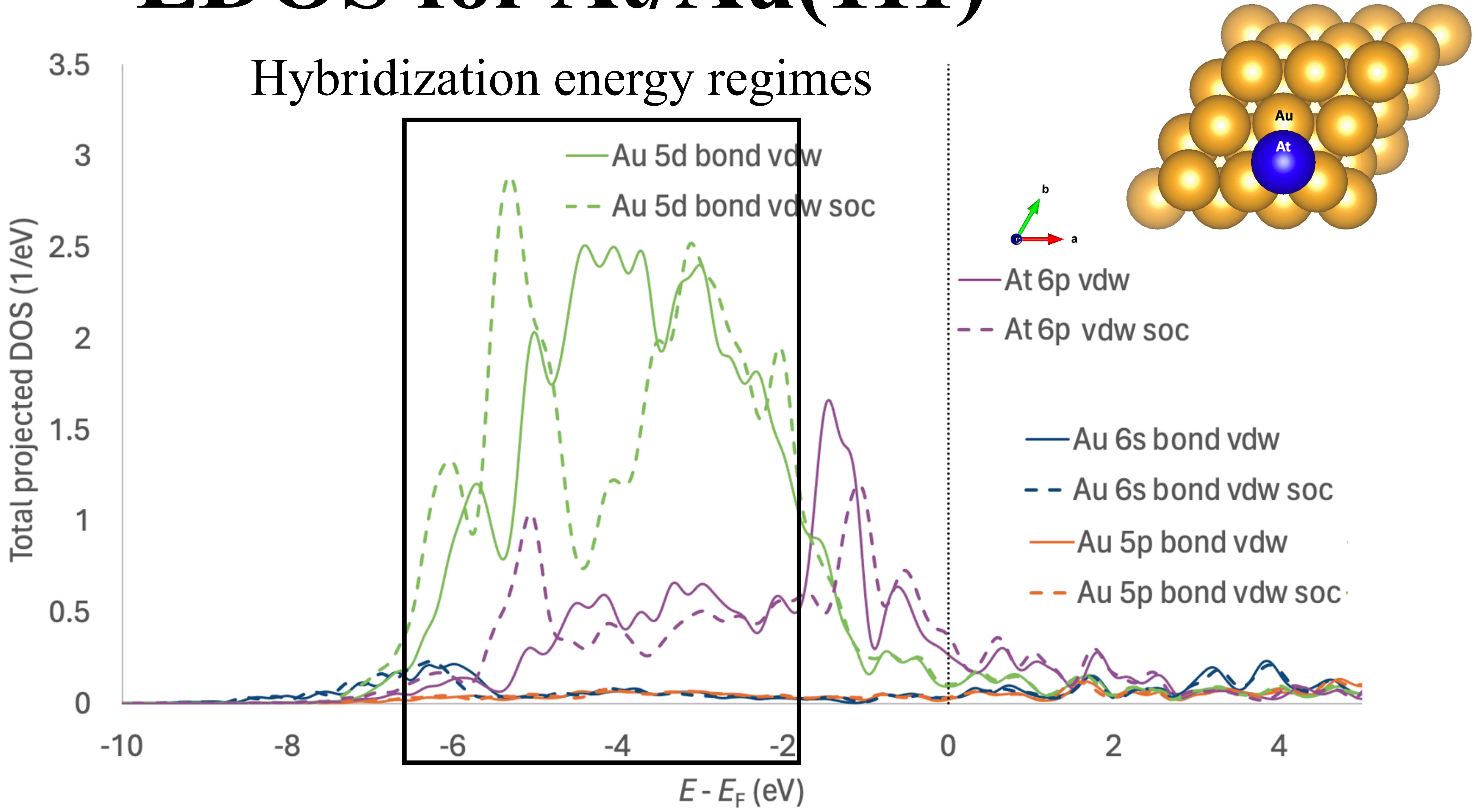
	Adsorption energy of At/Au(111)		
	w/o vdW [eV]	**w/ vdW [eV]**	**w/ vdW + SOC [eV]**
fcc hollow	-0.91	-1.43	-1.43
hcp hollow	-0.90	-1.42	-1.43
bridge site	-0.85	-1.37	-1.40
top site	-0.60	-1.20	-1.26

Adsorption energies on Au(111)

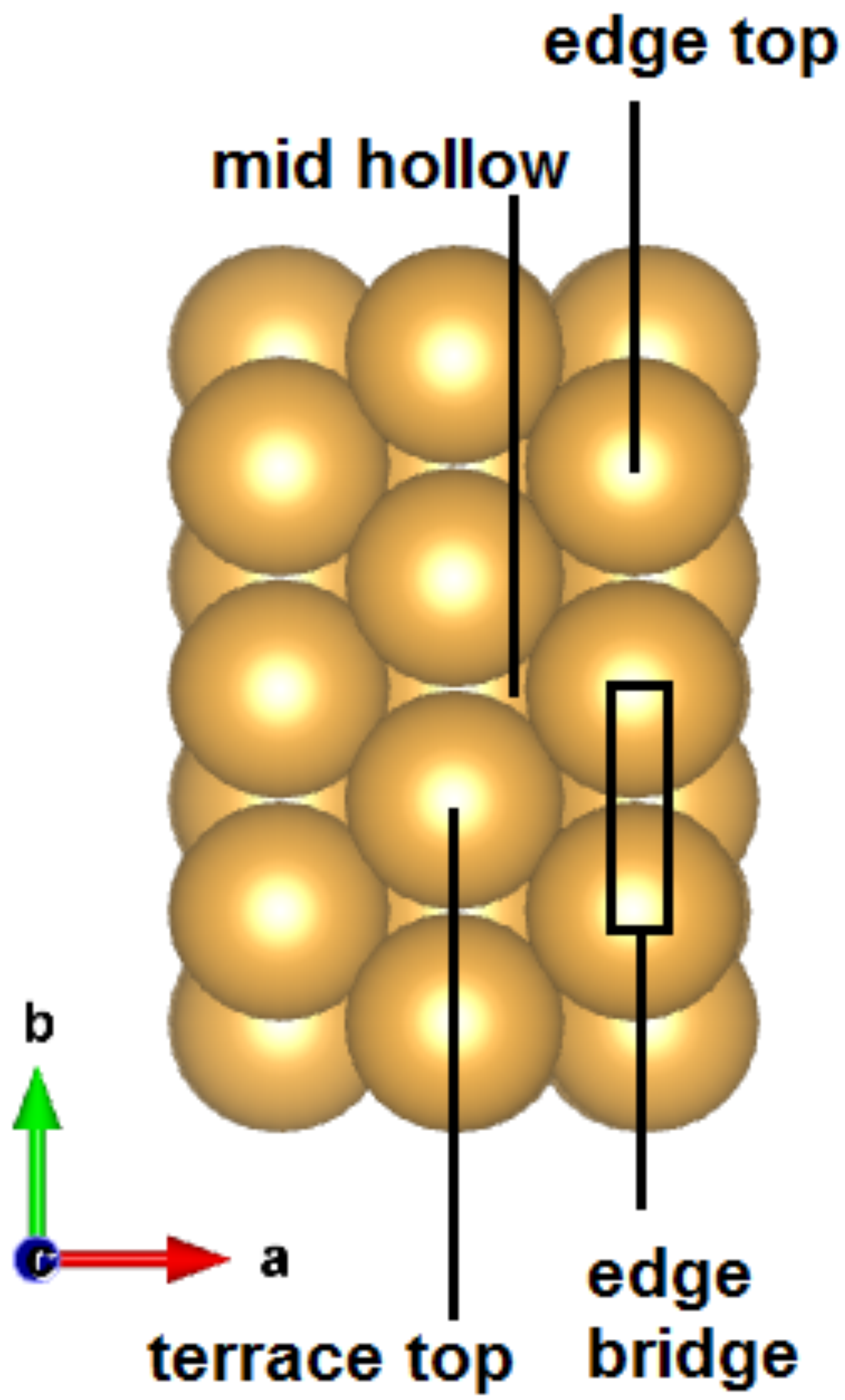


	At/Au(111) adsorption energy			I/Au(111) adsorption energy		
	w/o vdW(eV)	w/ vdW(eV)	w/ vdW+ SOC (eV)	w/o vdW (eV)	w/ vdW (eV)	w/ vdW + SOC (eV)
fcc hollow	-0.91	-1.43	-1.43	-0.92	-1.33	-1.34
hcp hollow	-0.90	-1.42	-1.43	-0.90	-1.33	-1.34
bridge site	-0.85	-1.37	-1.40	-0.85	-1.29	-1.30
top site	-0.60	-1.20	-1.26	-0.62	-1.11	-1.14

LDOS for At/Au(111)



Adsorption on Au(211)



One M adsorbed on Au surface

$$E_{ad} = \mathrm{M/Au(211)} - (0.5\mathrm{M}_2 + \mathrm{Au(211)})$$

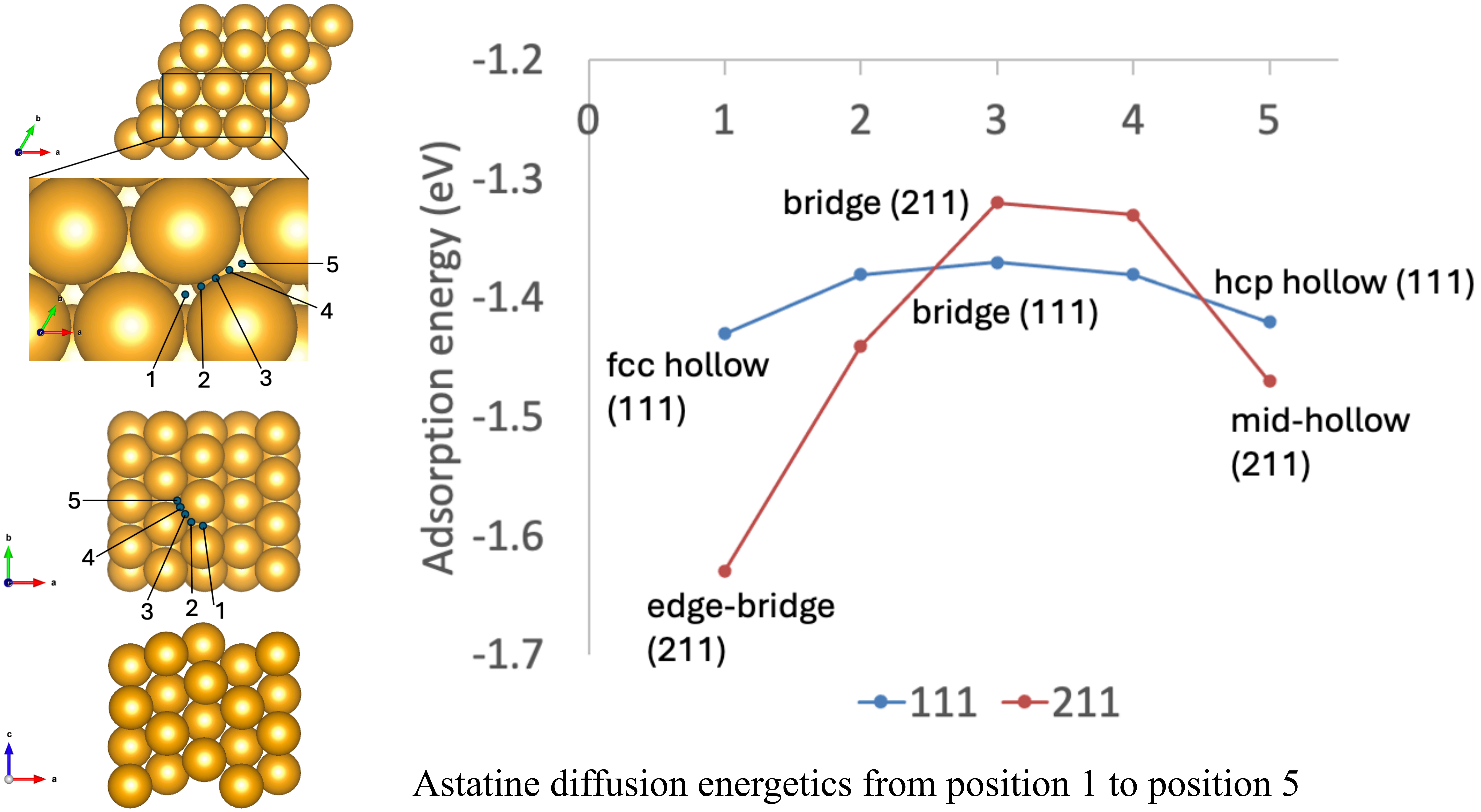
Isolated M

Clean Au surface

where M is the adsorbate (iodine or astatine)

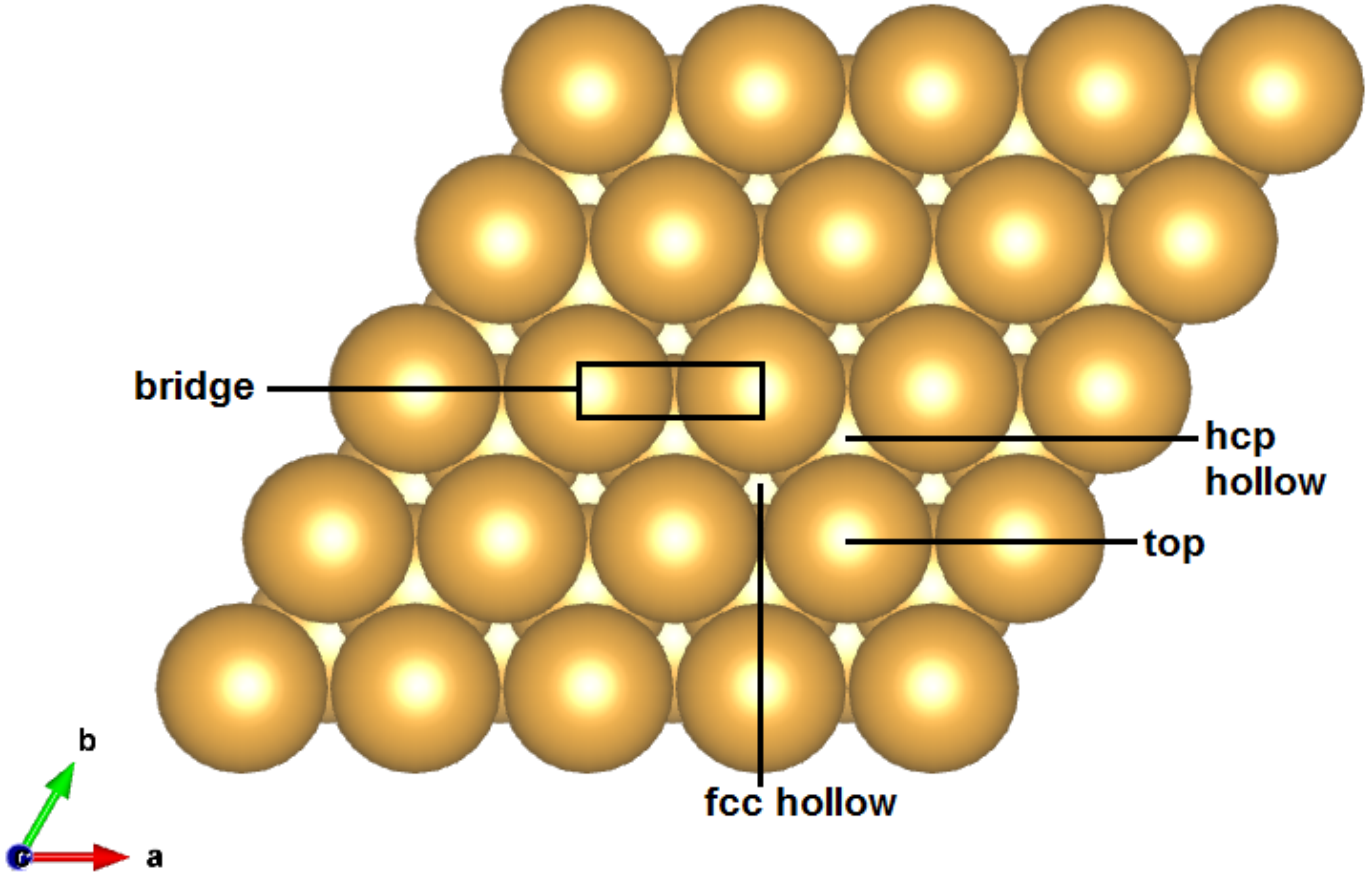
	I adsorption energy (eV)	At adsorption energy (eV)
Edge top	-1.18	-1.19
Edge bridge	-1.63	-1.63
Mid hollow	-1.38	-1.47
Terrace top	-1.12	-1.22

Diffusion between sites



Astatine diffusion energetics from position 1 to position 5

Adsorption of At atom on different metals



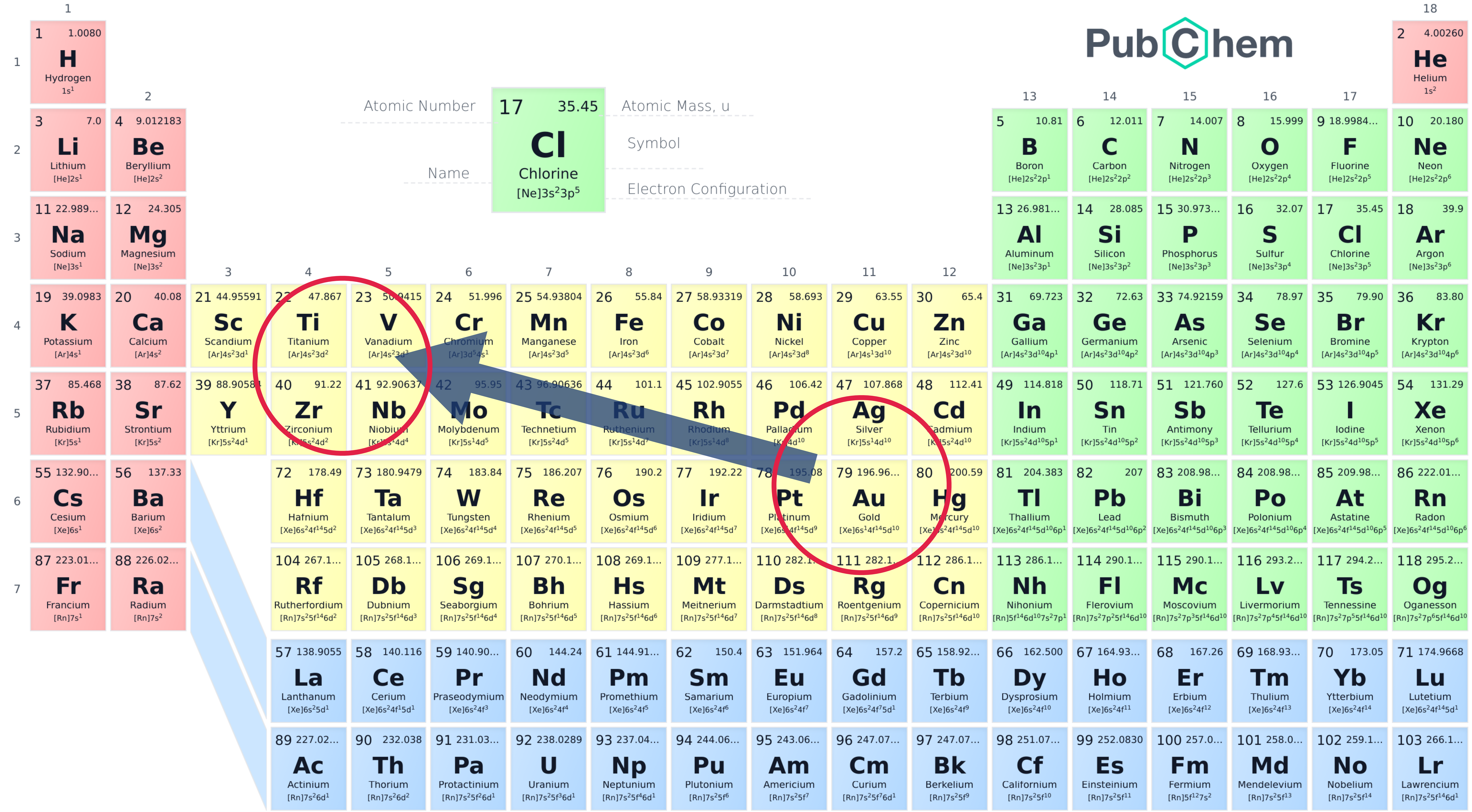
Top view of (111) surface

$$E_{ads} = \underbrace{\mathrm{At/M(111)}}_{\text{One At adsorbed on metal surface}} - (0.5\underbrace{\mathrm{At_2}}_{\text{Isolated At}} + \underbrace{\mathrm{M(111)}}_{\text{Clean metal surface}})$$

	hollow sites [eV]
Au(111)	-1.57
Ag(111)	-1.63
Cu(111)	-1.73
Pt(111)	-1.95

PERIODIC TABLE OF ELEMENTS

Electron Configuration



National Center for Biotechnology Information (2024). Periodic Table of Elements.

Current results

- Hybridization of astatine 6p and gold 5d and 6s upon adsorption
- A more uneven surface allows for stronger adsorption of astatine
- Other biocompatible/noble metals can be potentially useful as astatine carriers

Calculations were performed in the computer clusters of NIT Akashi College and YITP Kyoto University

Challenges and future works

- Other materials such as oxides (e.g. Ti and TiO_2) that are biocompatible may be used as alternatives
- Things to be considered
  - Actual operating conditions (temperature, pressure, etc.)
  - Effect of astatine decay
- Corroboration with experiments will be useful in understanding the accuracy of the simulation

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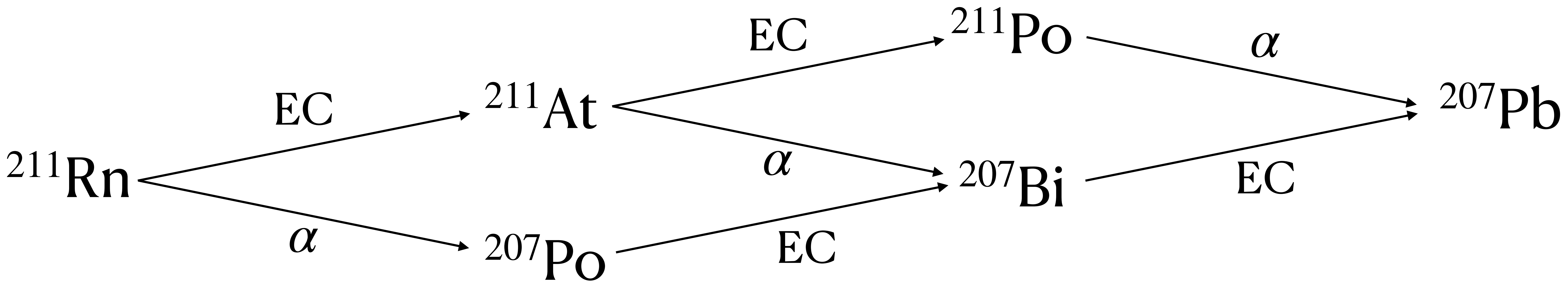
Production of ^{211}At

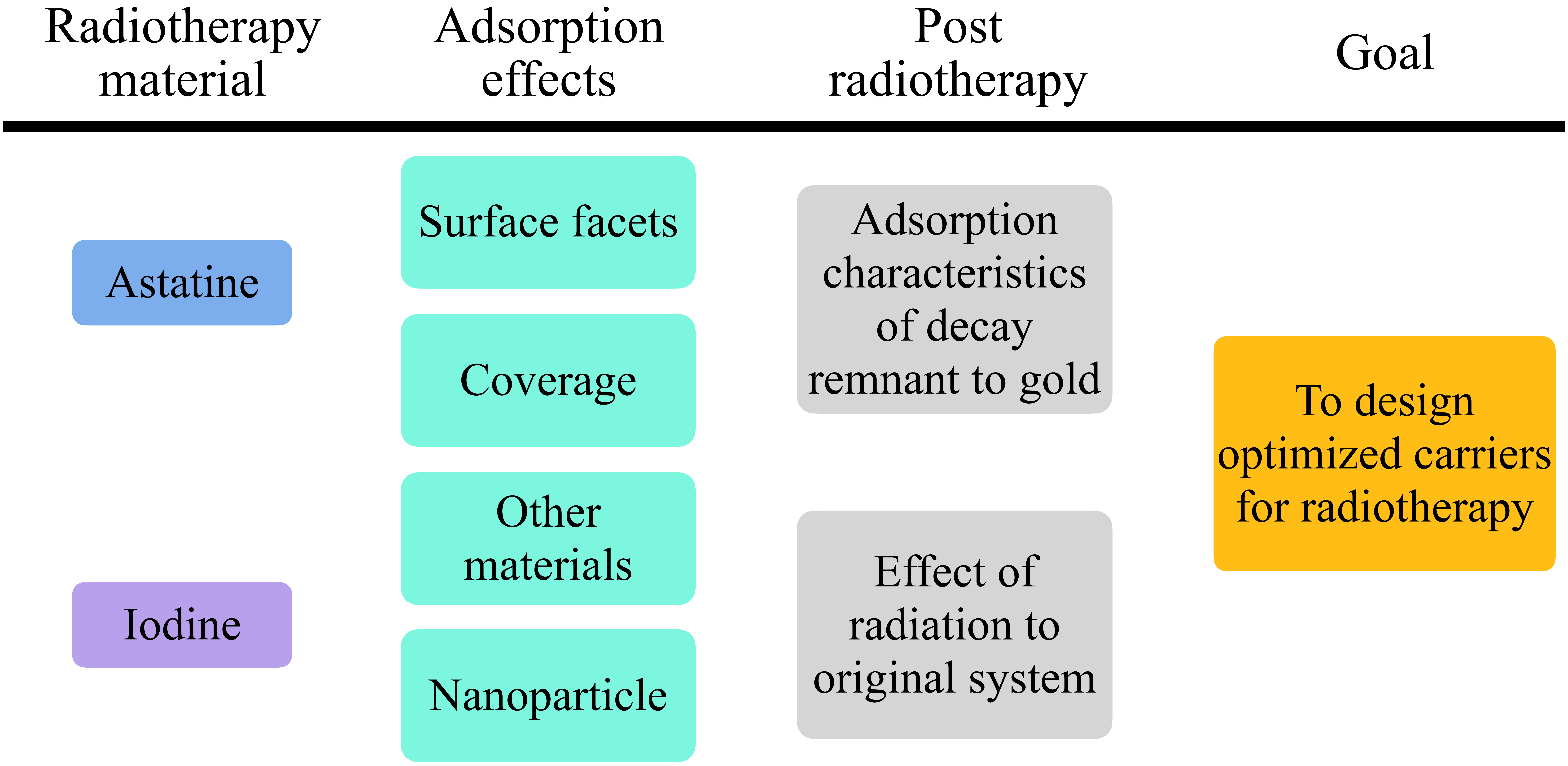
Equation	Energy
$^{209}Bi\ (^{4}He,2n)\ ^{211}At$	28 MeV
$^{209}Bi\ (^{7}Li,5n)\ ^{211}Rn$	50-60 MeV
$^{209}Bi\ (^{6}Li,4n)\ ^{211}Rn$	42 MeV

- Production energy cannot exceed 28 MeV due to possibility of creating ^{210}At, which decays into toxic ^{210}Po

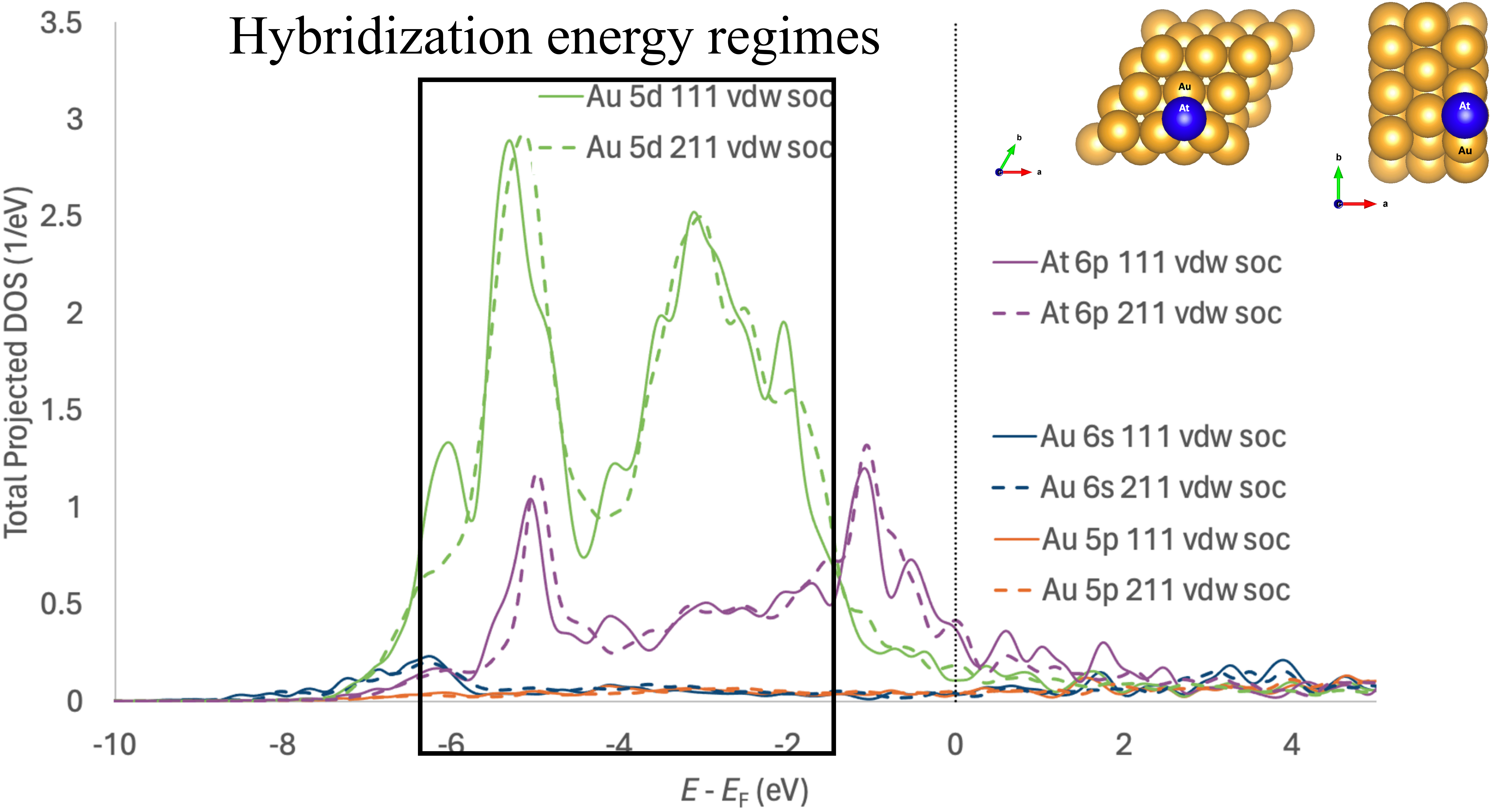


Use of gold nanoparticle

- Many shapes and sizes of AuNP can be created by
  - Reducing $HClAu_4$ with reducing agents such as sodium citrate or sodium borohydride
  - Add anti-coagulant to stop AuNPs from aggregating
- Attaching AuNP with targeting compounds
- Label AuNP with ^{211}At

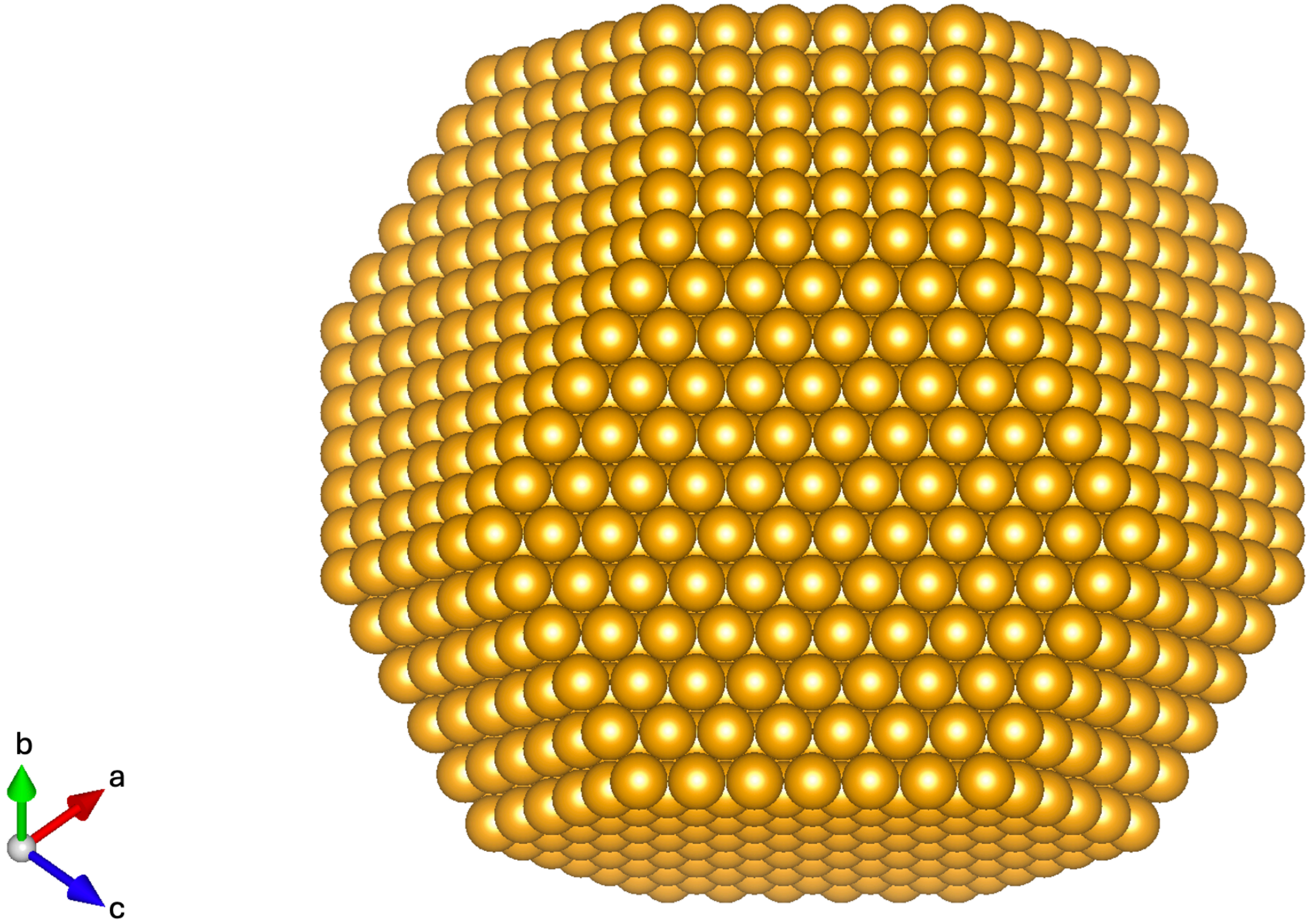

Radiotherapy material
Adsorption effects
Post radiotherapy
Goal
Astatine
Iodine
Surface facets
Coverage
Other materials
Nanoparticle
Adsorption characteristics of decay remnant to gold
Effect of radiation to original system
To design optimized carriers for radiotherapy

LDOS for At/Au(111) and At/Au(211)

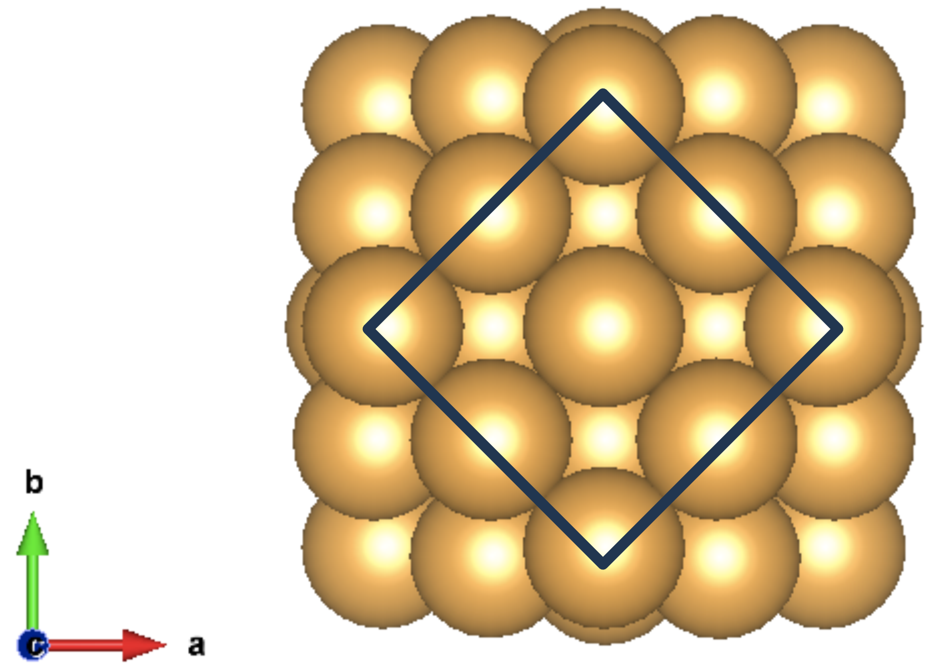


Adsorption on nanoparticle

Model diagram of a 5 nm AuNP



Model diagram of a 1 nm AuNP



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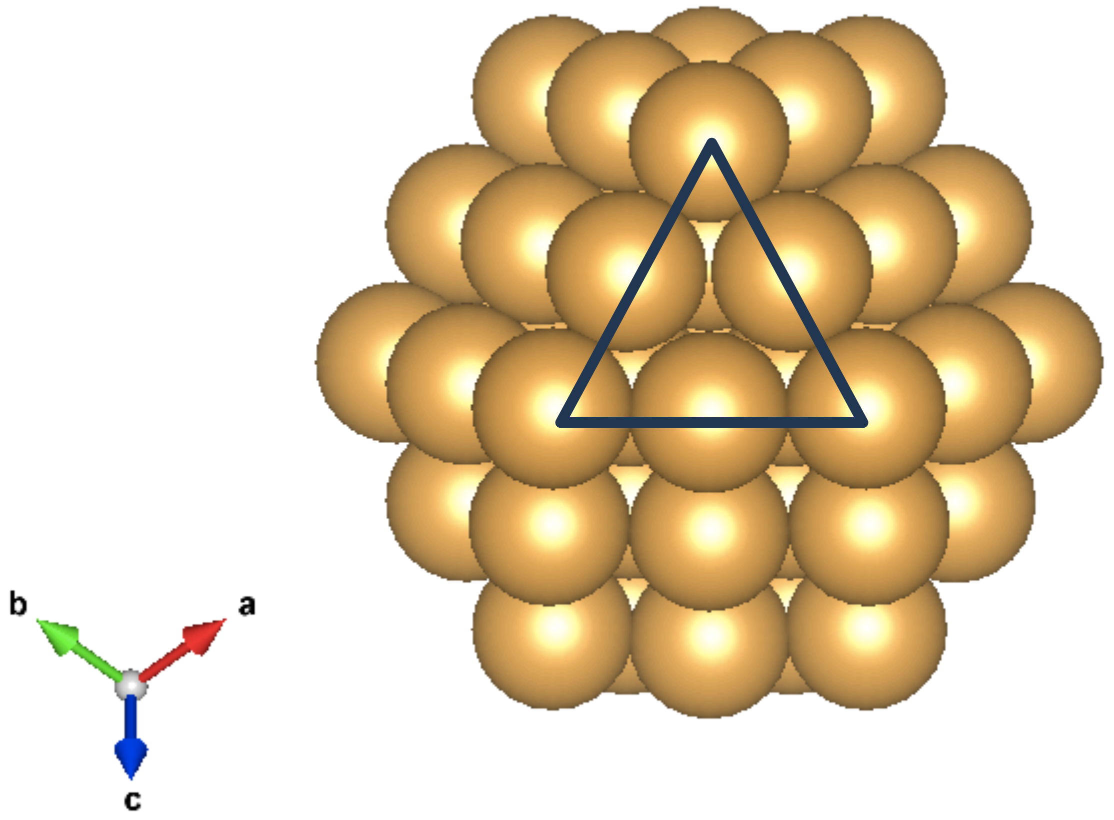
facet



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