

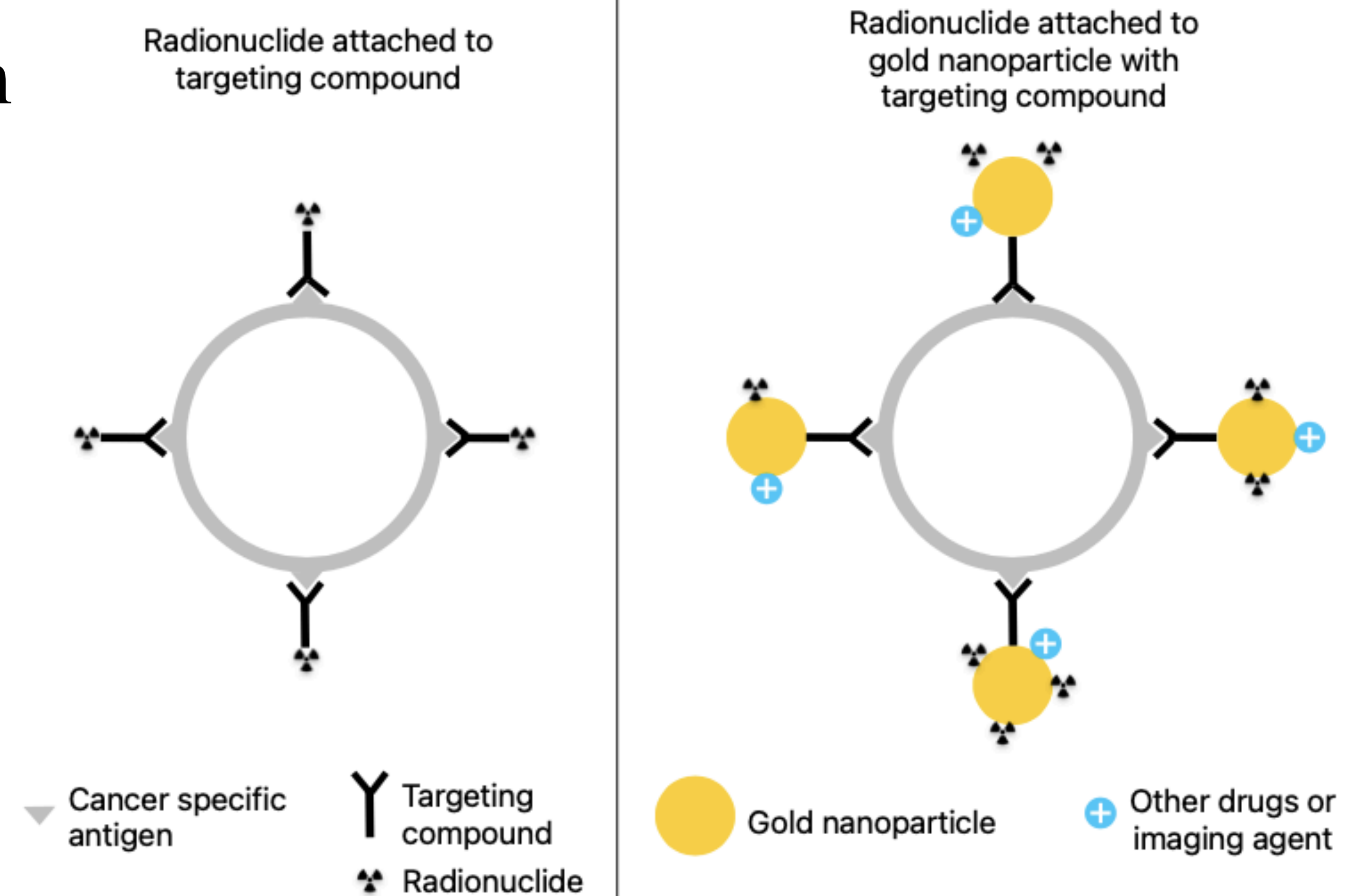
Computational Materials Design: DFT Application in Nuclear Medicine

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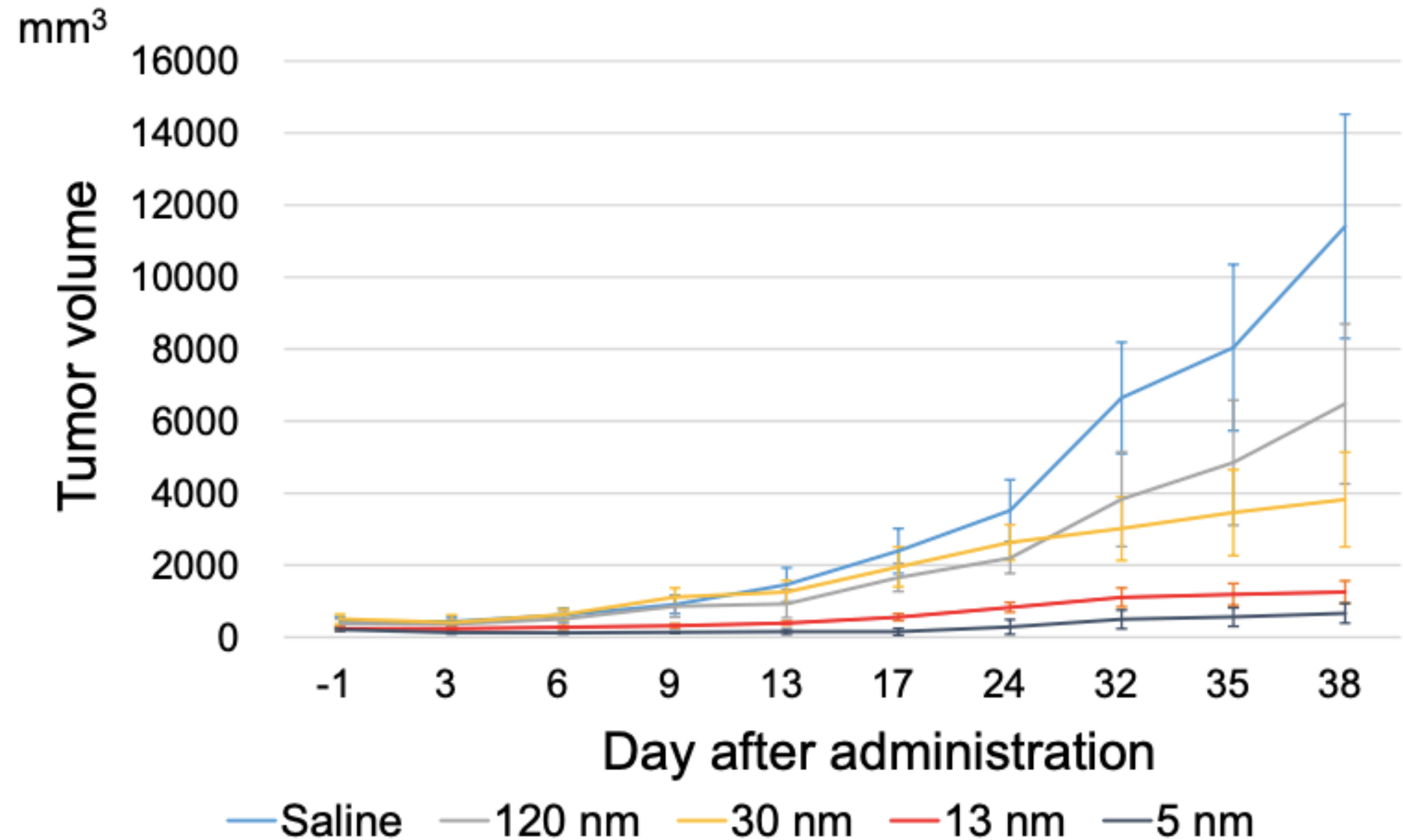
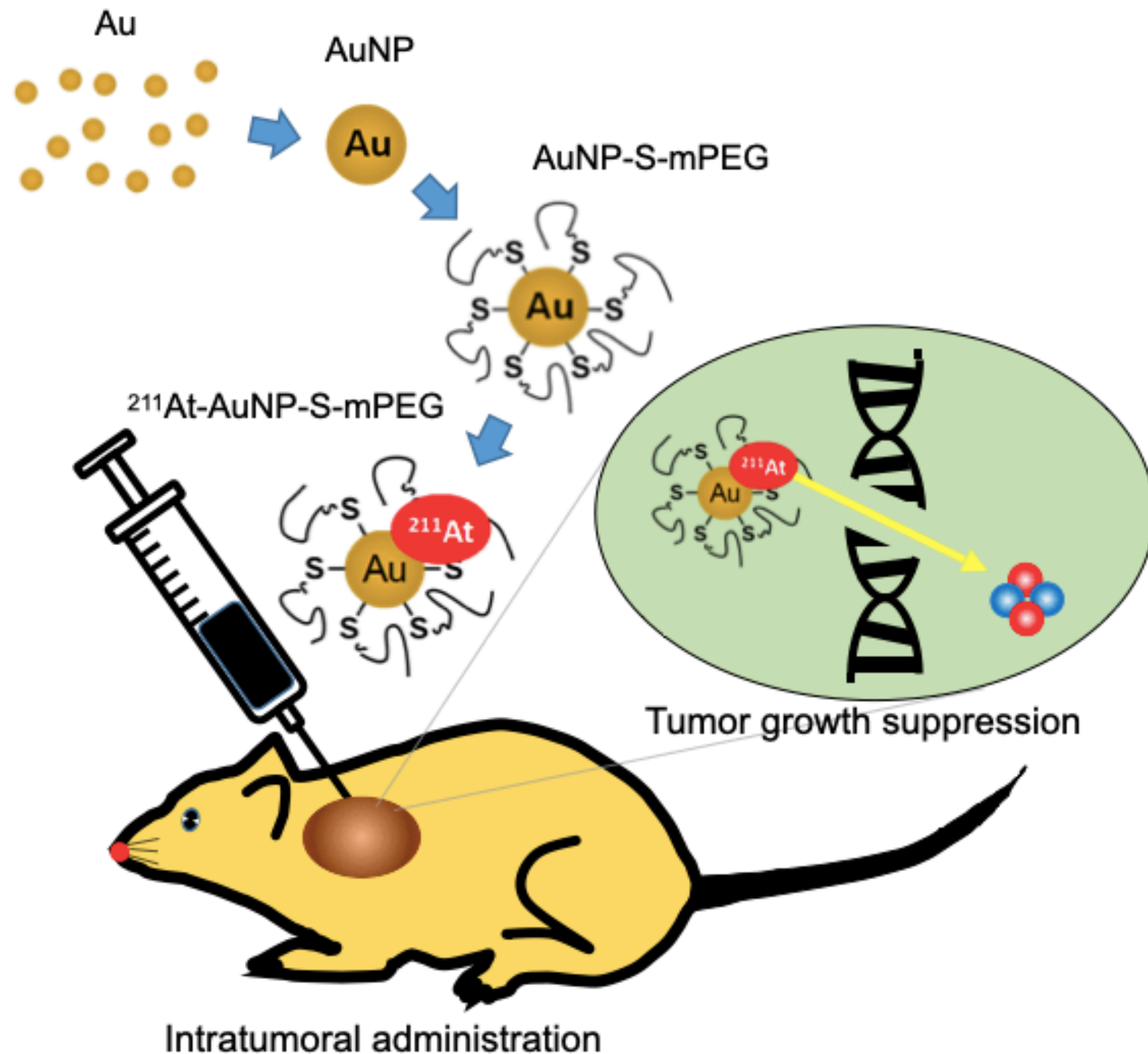
2025/03/25

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



Experimental findings of $^{211}\text{At}/\text{Au}$ use

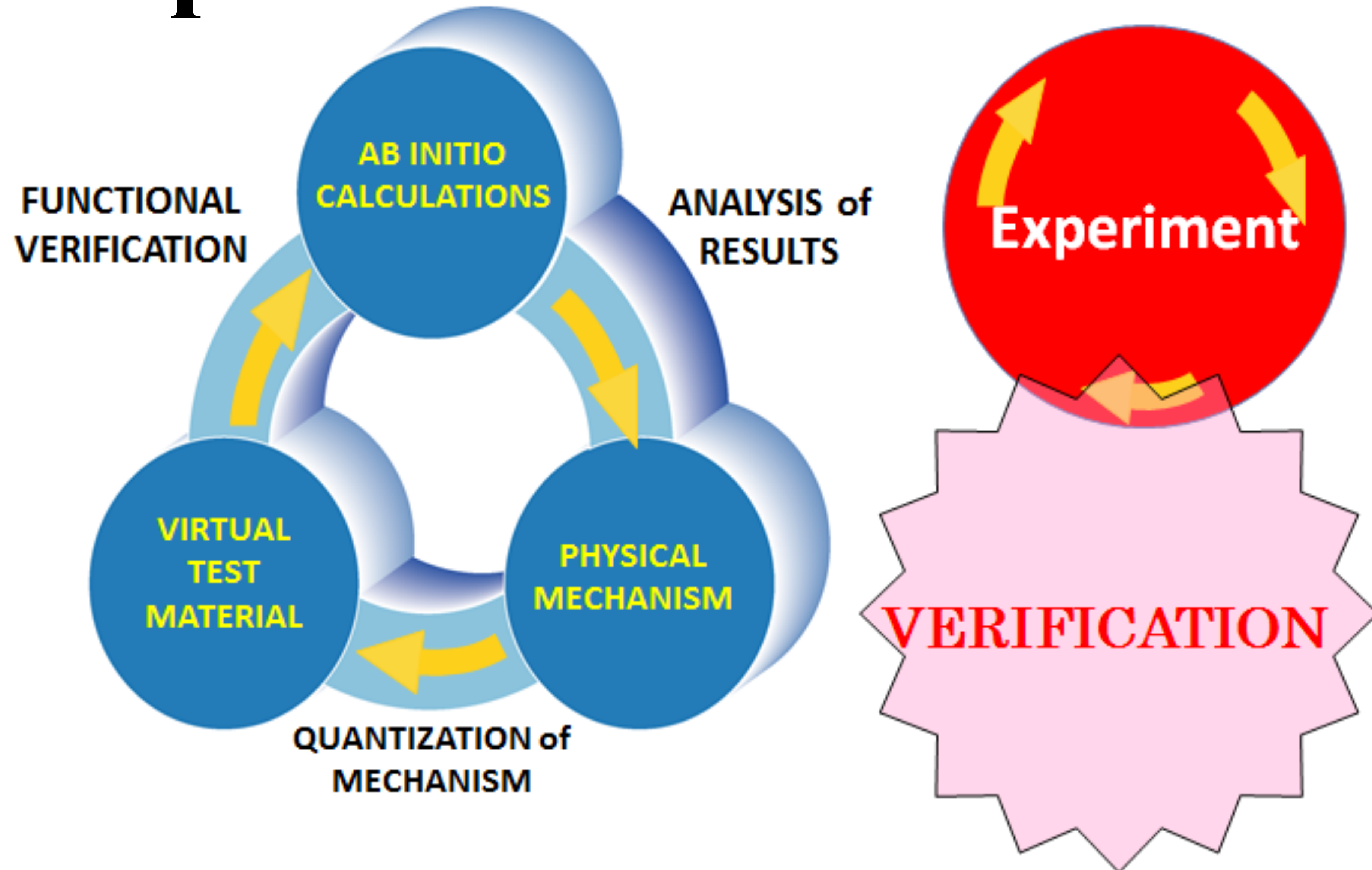


[2] H. Kato et al., J. Nanobiotechnol. 19, 233 (2021)

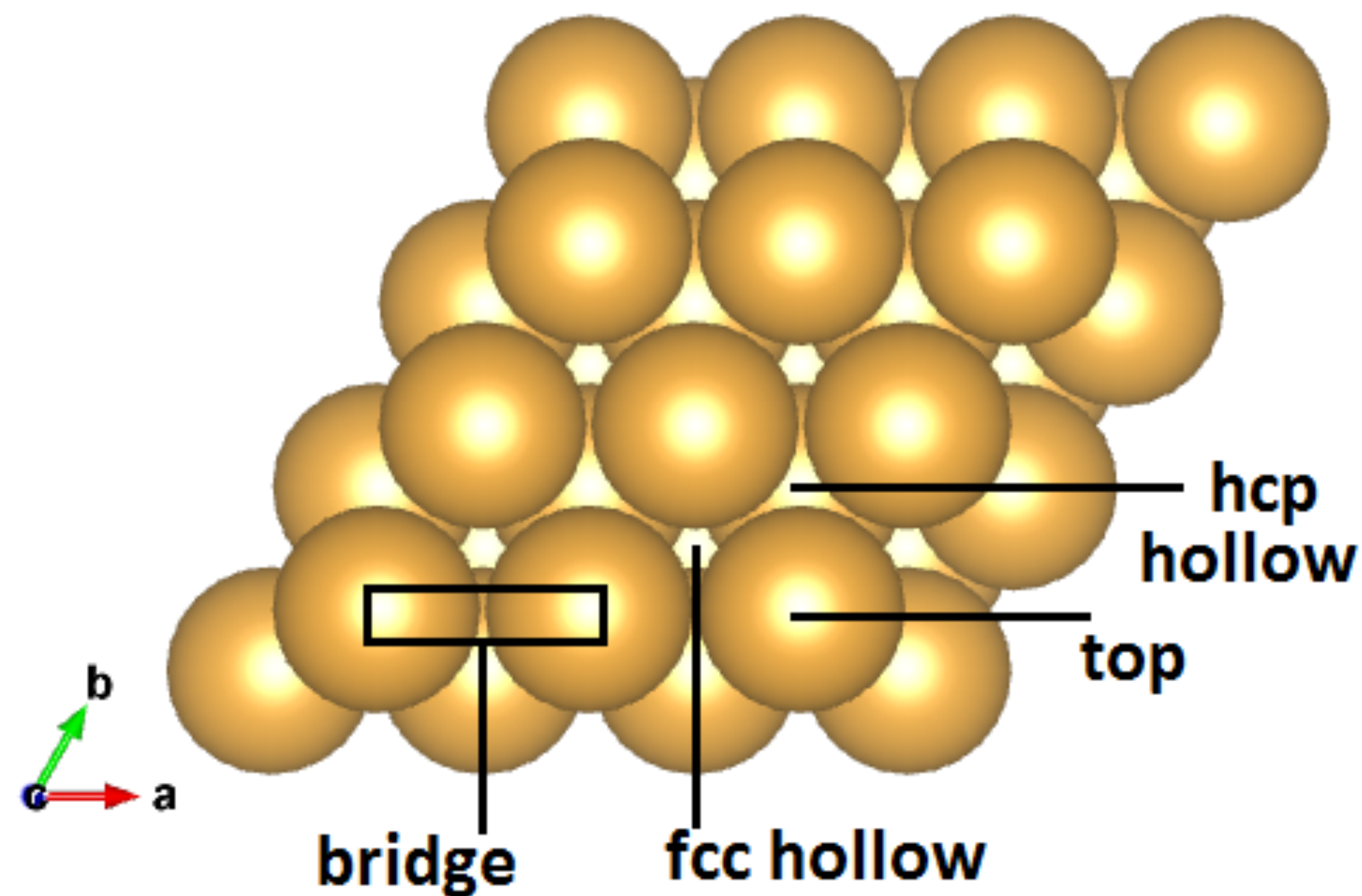
Motivation

- 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)



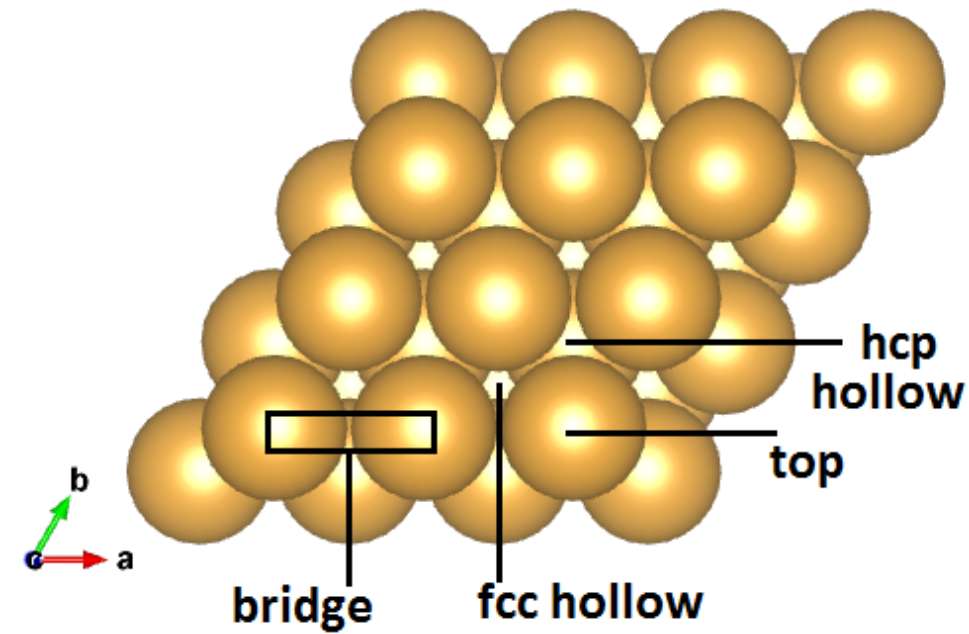
Adsorption energy calculation

$$E_{ads} = \text{At/Au(111)} - (0.5\text{At}_2 + \text{Au(111)})$$

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

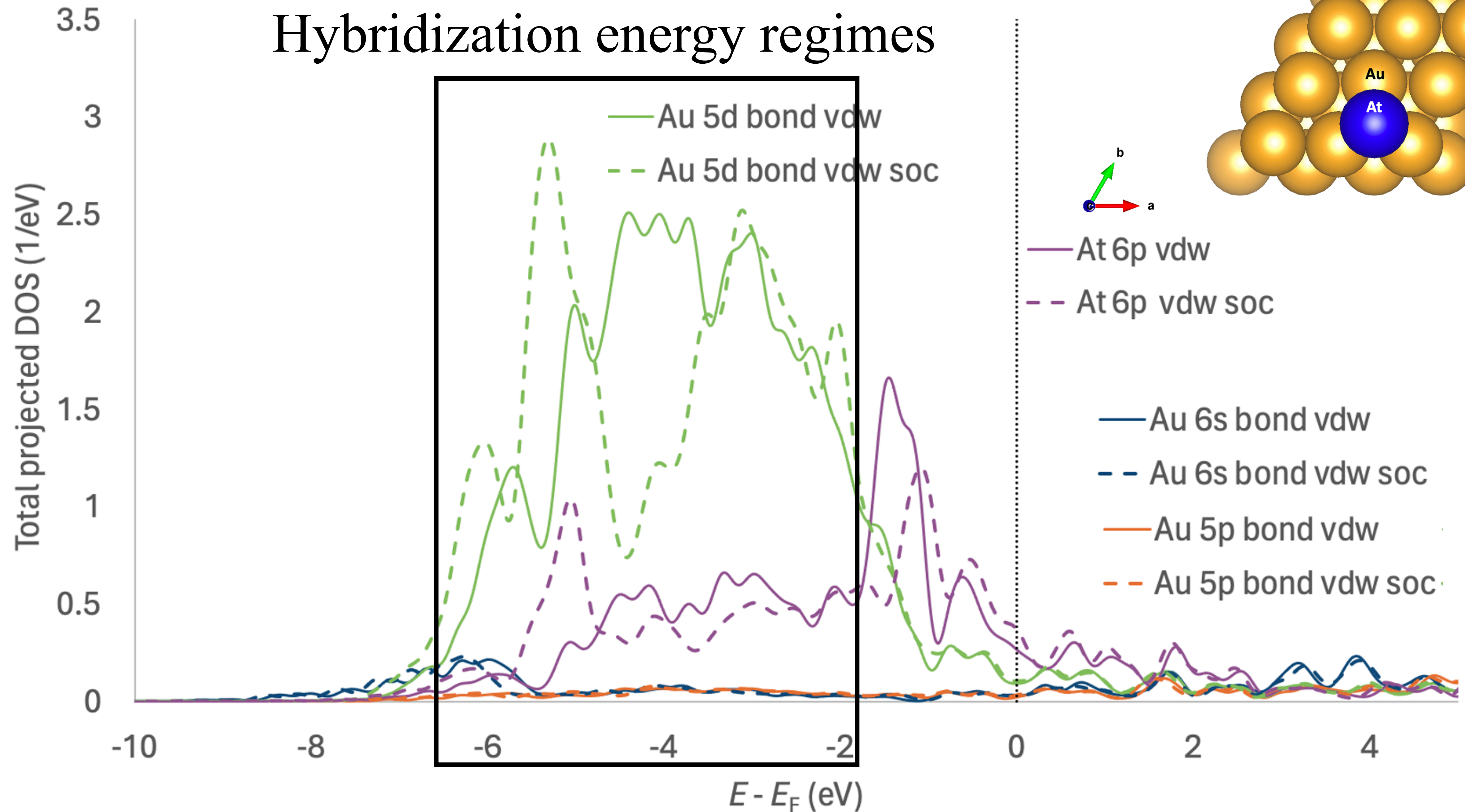
Adsorption energies on Au(111)



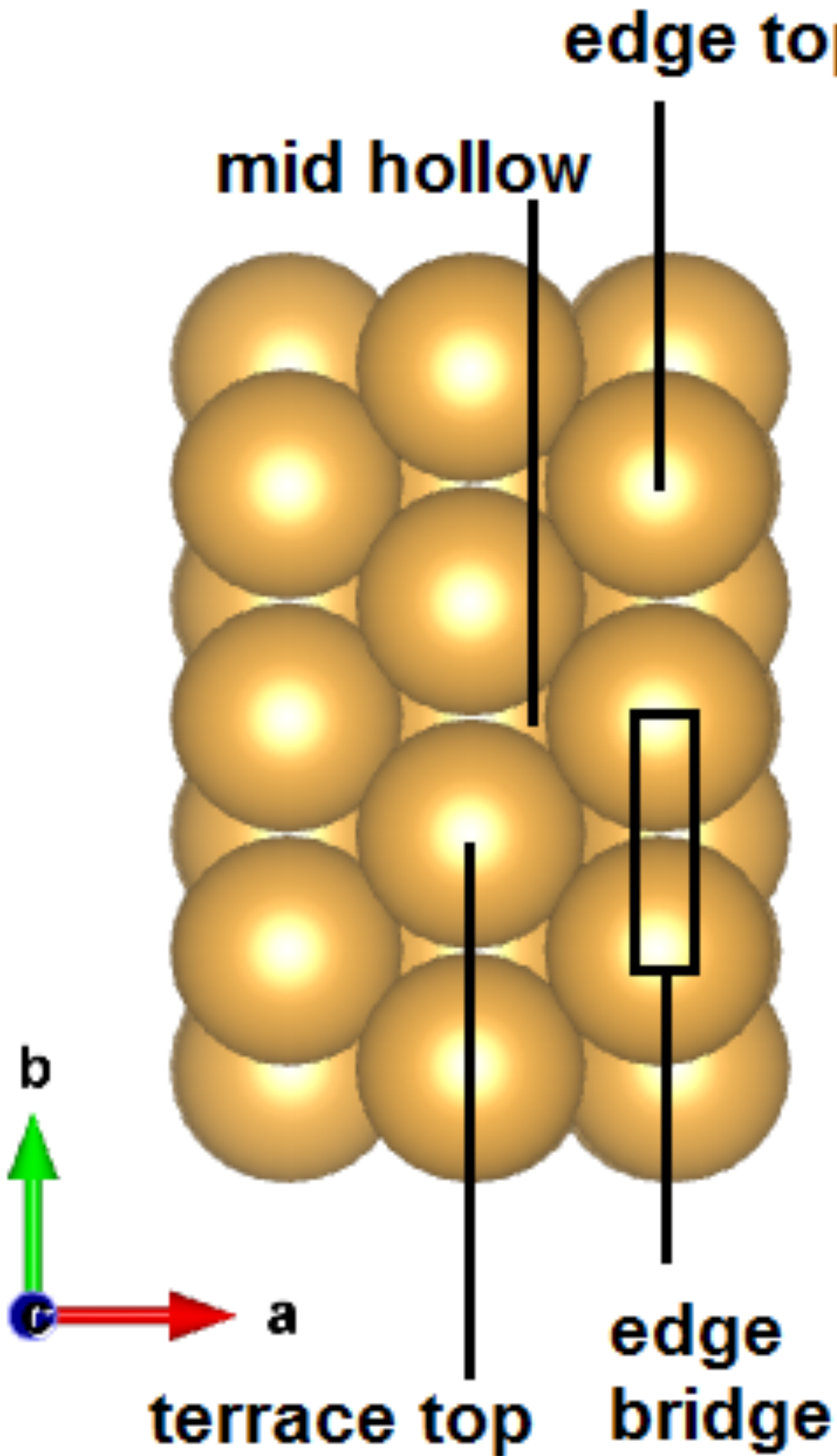
	At/Au(111) adsorption energy			I/Au(111) adsorption energy		
	w/o vdW(eV)	w/ vdW(eV)	w/ vdW+ SOC (eV)	w/o vdW (eV)	w/ vdW (eV)	w/ vdW + SOC (eV)
fcc hollow	-0.91	-1.43	-1.43	-0.92	-1.33	-1.34
hcp hollow	-0.90	-1.42	-1.43	-0.90	-1.33	-1.34
bridge site	-0.85	-1.37	-1.40	-0.85	-1.29	-1.30
top site	-0.60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

LDOS for At/Au(111)

Hybridization energy regimes



Adsorption on Au(211)



One M adsorbed on Au surface

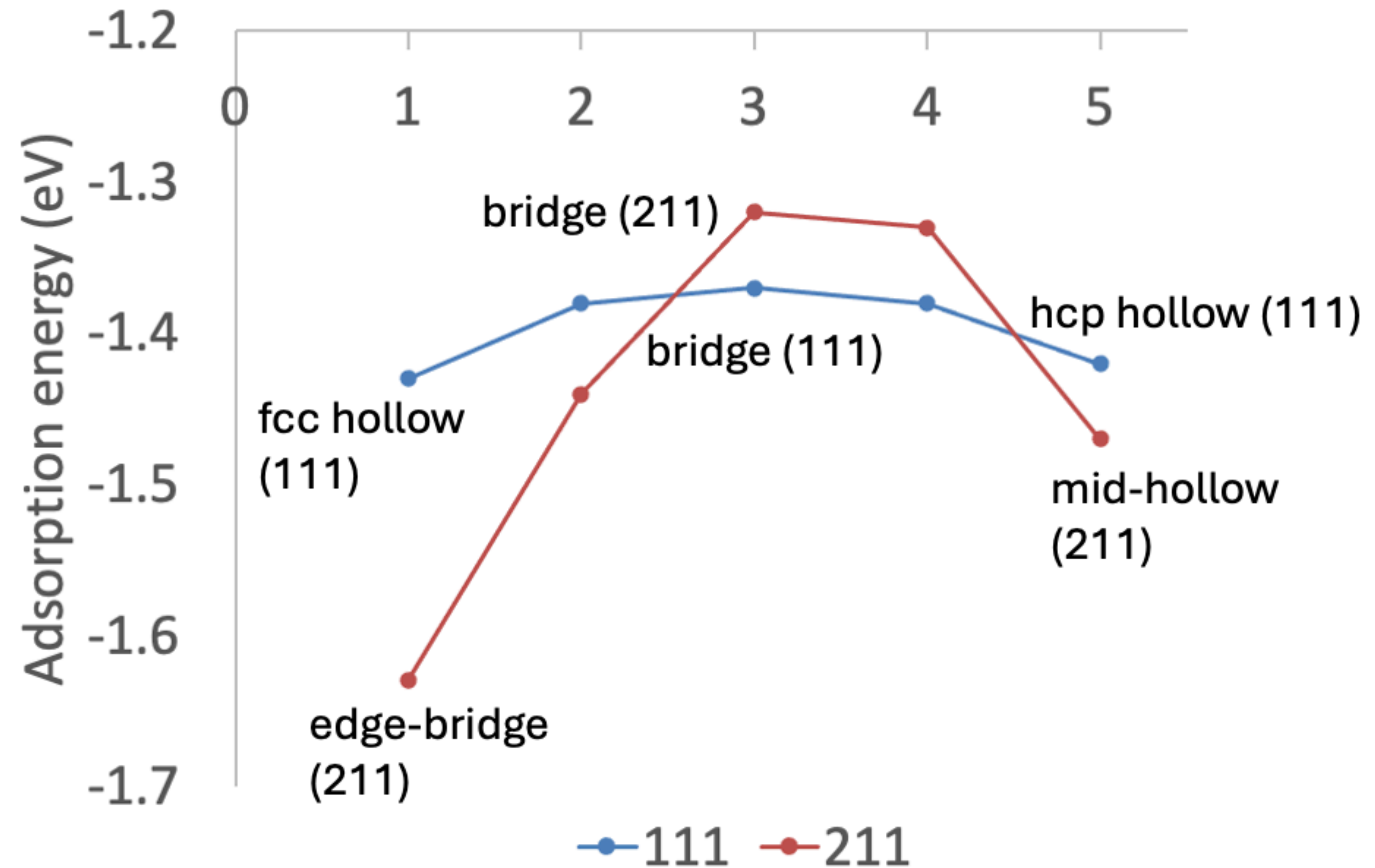
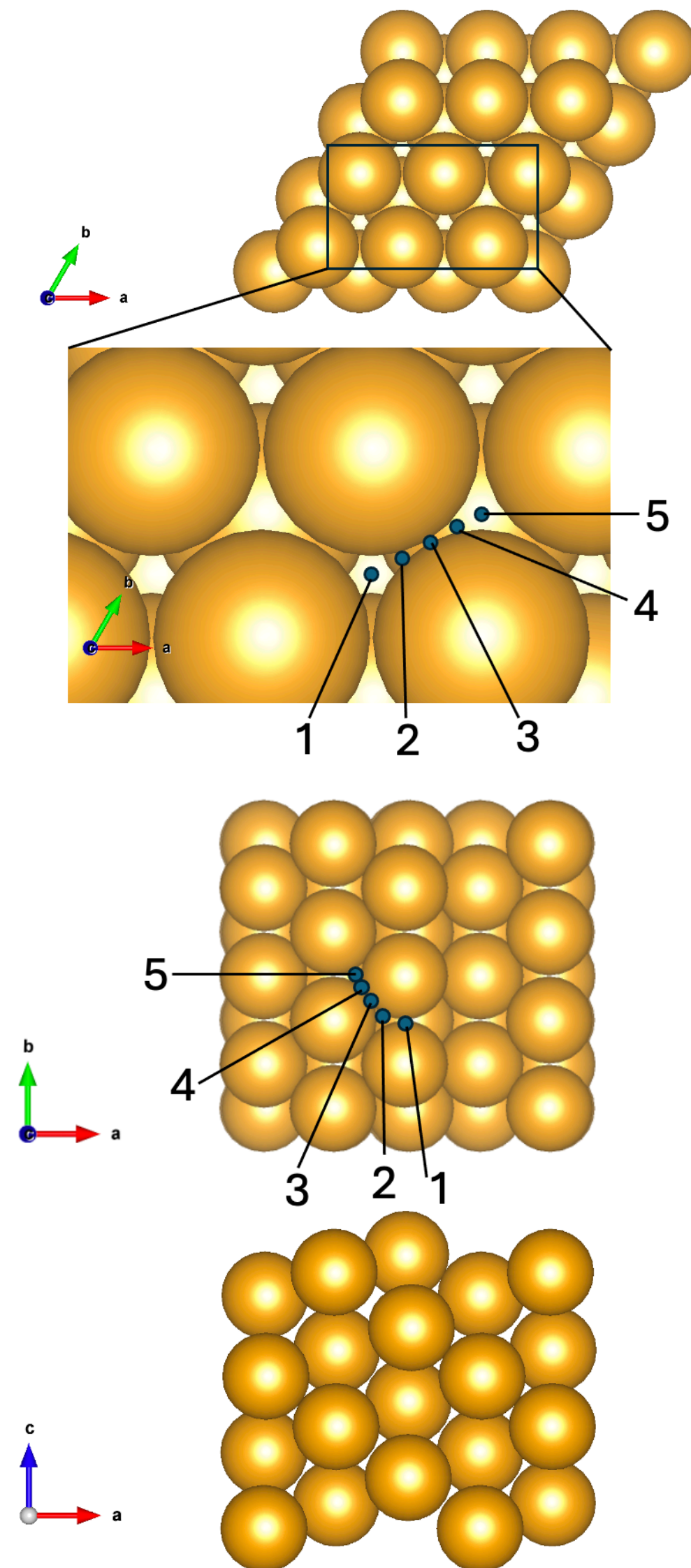
$$E_{ad} = M/\text{Au}(211) - (0.5M_2 + \text{Au}(211))$$

↑ ↑
Isolated M Clean Au surface

where M is the adsorbate (iodine or astatine)

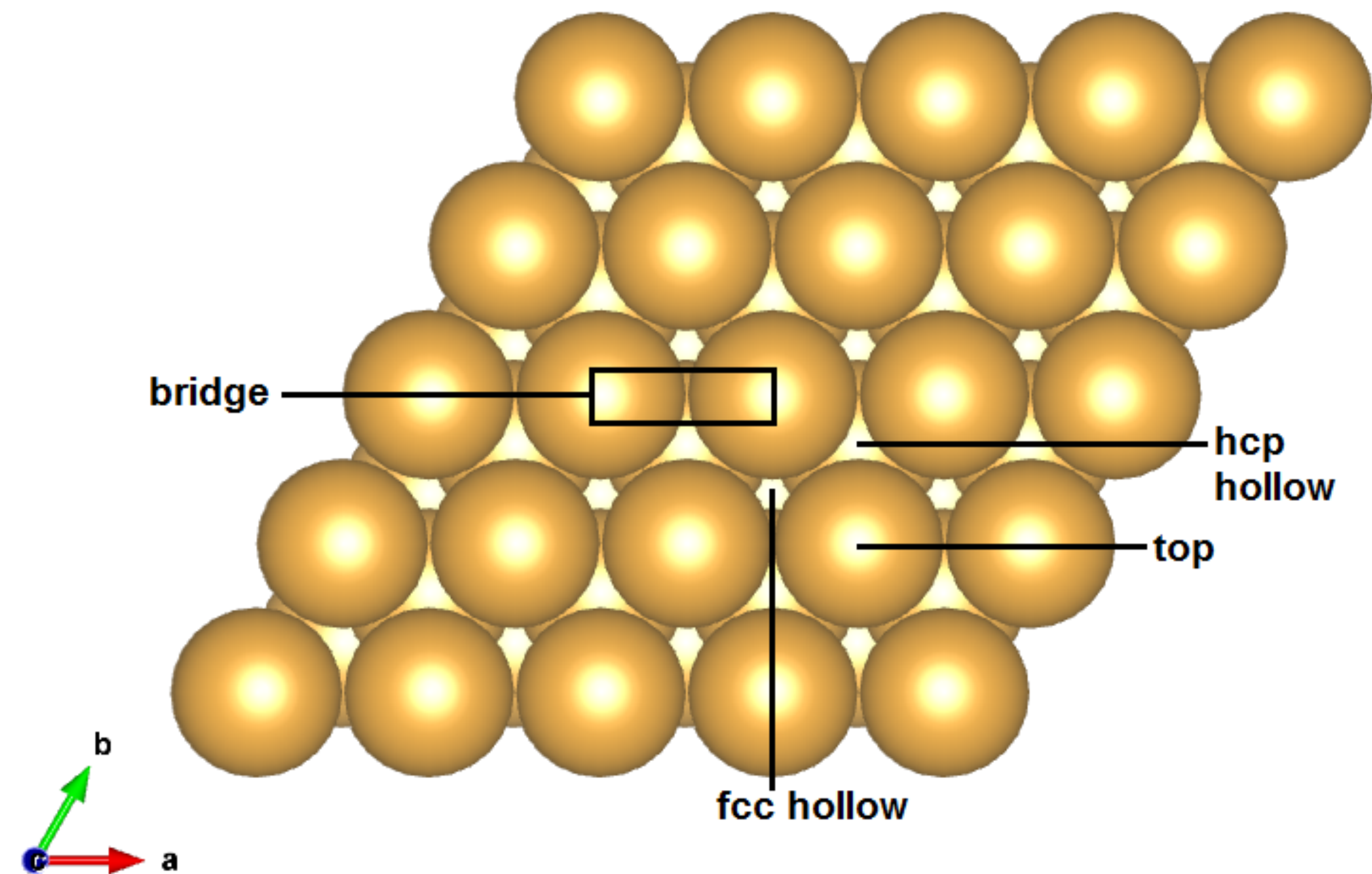
	I adsorption energy (eV)	At adsorption energy (eV)
Edge top	-1.18	-1.19
Edge bridge	-1.63	-1.63
Mid hollow	-1.38	-1.47
Terrace top	-1.12	-1.22

Diffusion between sites



Astatine diffusion energetics from position 1 to position 5

Adsorption of At atom on different metals



Top view of (111) surface

$$E_{\text{ads}} = \underset{\substack{\uparrow \\ \text{One At adsorbed} \\ \text{on metal surface}}}{\text{At/M(111)}} - \left(\underset{\substack{\uparrow \\ \text{Isolated At}}}{0.5\text{At}_2} + \underset{\substack{\uparrow \\ \text{Clean metal} \\ \text{surface}}}{\text{M(111)}} \right)$$

	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

1

1.0080

H

Hydrogen

1s¹

3

7.0

Li

Lithium

[He]2s¹

11

22.989...

Na

Sodium

[Ne]3s¹

19

39.0983

K

Potassium

[Ar]4s¹

37

85.468

Rb

Rubidium

[Kr]5s¹

55

132.90...

Cs

Cesium

[Xe]6s¹

87

223.01...

Fr

Francium

[Rn]7s¹

2

9.012183

Be

Beryllium

[He]2s²

12

24.305

Mg

Magnesium

[Ne]3s²

20

40.08

Ca

Calcium

[Ar]4s²

38

87.62

Sr

Strontium

[Kr]5s²

56

137.33

Ba

Barium

[Xe]6s²

88

226.02...

Ra

Radium

[Rn]7s²

21

44.95591

Sc

Scandium

[Ar]4s²3d¹

22

47.867

Ti

Titanium

[Ar]4s²3d²

23

50.9415

V

Vanadium

[Ar]4s²3d³

24

51.996

Cr

Chromium

[Ar]3d⁵4s¹

25

54.93804

Mn

Manganese

[Ar]4s²3d⁵

26

55.84

Fe

Iron

[Ar]4s²3d⁶

27

58.93319

Co

Cobalt

[Ar]4s²3d⁷

28

58.693

Ni

Nickel

[Ar]4s²3d⁸

29

63.55

Cu

Copper

[Ar]4s¹3d¹⁰

30

65.4

Zn

Zinc

[Ar]4s²3d¹⁰

39

88.90584

Y

Yttrium

[Kr]5s²4d¹

40

91.22

Zr

Zirconium

[Kr]5s²4d²

41

92.90637

Nb

Niobium

[Kr]5s²4d⁴

42

95.95

Mo

Molybdenum

[Kr]5s¹4d⁵

43

96.90636

Tc

Technetium

[Kr]5s²4d⁵

44

101.1

Ru

Ruthenium

[Kr]5s¹4d⁷

45

102.9055

Rh

Rhodium

[Kr]5s¹4d⁸

46

106.42

Pd

Palladium

[Kr]4d¹⁰

47

107.868

Ag

Silver

[Kr]5s¹4d¹⁰

48

112.41

Cd

Cadmium

[Kr]5s²4d¹⁰

72

178.49

Hf

Hafnium

[Xe]6s²4f¹⁴5d²

73

180.9479

Ta

Tantalum

[Xe]6s²4f¹⁴5d³

74

183.84

W

Tungsten

[Xe]6s²4f¹⁴5d⁴

75

186.207

Re

Rhenium

[Xe]6s²4f¹⁴5d⁵

76

190.2

Os

Osmium

[Xe]6s²4f¹⁴5d⁶

77

192.22

Ir

Iridium

[Xe]6s²4f¹⁴5d⁷

78

195.08

Pt

Platinum

[Xe]6s²4f¹⁴5d⁹

79

196.96...

Au

Gold

[Xe]6s¹4f¹⁴5d¹⁰

80

200.59

Hg

Mercury

[Xe]6s²4f¹⁴5d¹⁰

104

267.1...

Rf

Rutherfordium

[Rn]7s²5f¹⁴6d²

105

268.1...

Db

Dubnium

[Rn]7s²5f¹⁴6d³

106

269.1...

Sg

Seaborgium

[Rn]7s²5f¹⁴6d⁴

107

270.1...

Bh

Bohrium

[Rn]7s²5f¹⁴6d⁵

108

269.1...

Hs

Hassium

[Rn]7s²5f¹⁴6d⁶

109

277.1...

Mt

Meitnerium

[Rn]7s²5f¹⁴6d⁷

110

282.1...

Ds

Darmstadtium

[Rn]7s²5f¹⁴6d⁸

111

282.1...

Rg

Roentgenium

[Rn]7s²5f¹⁴6d⁹

112

286.1...

Cn

Copernicium

[Rn]7s²5f¹⁴6d¹⁰

57

138.9055

La

Lanthanum

[Xe]6s²5d¹

58

140.116

Ce

Cerium

[Xe]6s²4f¹5d¹

59

140.90...

Pr

Praseodymium

[Xe]6s²4f³

60

144.24

Nd

Neodymium

[Xe]6s²4f⁴

61

144.91...

Pm

Promethium

[Xe]6s²4f⁵

62

150.4

Sm

Samarium

[Xe]6s²4f⁶

63

151.964

Eu

Europium

[Xe]6s²4f⁷

64

157.2

Gd

Gadolinium

[Xe]6s²4f⁷5d¹

65

158.92...

Tb

Terbium

[Xe]6s²4f⁹

66

162.500

Dy

Dysprosium

[Xe]6s²4f¹⁰

67

164.93...

Ho

Holmium

[Xe]6s²4f¹¹

68

167.26

Er

Erbium

[Xe]6s²4f¹²

69

168.93...

Tm

Thulium

[Xe]6s²4f¹³

70

173.05

Yb

Ytterbium

[Xe]6s²4f¹⁴

71

174.9668

Lu

Lutetium

[Xe]6s²4f¹⁴5d¹

89

227.02...

Ac

Actinium

[Rn]7s²6d¹

90

232.038

Th

Thorium

[Rn]7s²6d²

91

231.03...

Pa

Protactinium

[Rn]7s²5f²6d¹

92

238.0289

U

Uranium

[Rn]7s²5f³6d¹

93

237.04...

Np

Neptunium

[Rn]7s²5f⁴6d¹

94

244.06...

Pu

Plutonium

[Rn]7s²5f⁶

95

243.06...

Am

Americium

[Rn]7s²5f⁷

96

247.07...

Cm

Curium

[Rn]7s²5f⁷6d¹

97

247.07...

Bk

Berkelium

[Rn]7s²5f⁹

98

251.07...

Cf

Californium

[Rn]7s²5f¹⁰

99

252.0830

Es

Einsteinium

[Rn]7s²5f¹¹

100

257.0...

Fm

Fermium

[Rn]5f¹²7s²

101

258.0...

Md

Mendelevium

[Rn]7s²5f¹³

102

259.1...

No

Nobelium

[Rn]7s²5f¹⁴

103

266.1...

Lr

Lawrencium

[Rn]7s²5f¹⁴6d¹

17

35.45

Cl

Chlorine

[Ne]3s²3p⁵

13

10.81

B

Boron

[He]2s²2p¹

14

12.011

C

Carbon

[He]2s²2p²

15

14.007

N

Nitrogen

[He]2s²2p³

16

15.999

O

Oxygen

[He]2s²2p⁴

17

18.9984...

F

Fluorine

[He]2s²2p⁵

31

69.723

Ga

Gallium

[Ar]4s²3d¹⁰4p¹

32

72.63

Ge

Germanium

[Ar]4s²3d¹⁰4p²

33

74.92159

As

Arsenic

[Ar]4s²3d¹⁰4p³

34

78.97

Se

Selenium

[Ar]4s²3d¹⁰4p⁴

35

79.90

Br

Bromine

[Ar]4s²3d¹⁰4p⁵

49

114.818

In

Indium

[Kr]5s²4d¹⁰5p¹

50

118.71

Sn

Tin

[Kr]5s²4d¹⁰5p²

51

121.760

Sb

Antimony

[Kr]5s²4d¹⁰5p³

52

127.6

Te

Tellurium

[Kr]5s²4d¹⁰5p⁴

53

126.9045

I

Iodine

[Kr]5s²4d¹⁰5p⁵

67

164.93...

Ho

Holmium

[Xe]6s²4f¹¹

68

167.26

Er

Erbium

[Xe]6s²4f¹²

69

168.93...

Tm

Thulium

[Xe]6s²4f¹³

70

173.05

Yb

Ytterbium

[Xe]6s²4f¹⁴

71

174.9668

Lu

Lutetium

[Xe]6s²4f¹⁴5d¹

1

1.0080

H

Hydrogen

1s¹

2

4.00260

He

Helium

1s²

10

20.180

Ne

Neon

[He]2s²2p⁶

18

39.9

Ar

Argon

[Ne]3s²3p⁶

36

83.80

Kr

Krypton

[Ar]4s²3d¹⁰4p⁶

54

131.29

Xe

Xenon

[Kr]5s²4d¹⁰5p⁶

86

222.01...

Rn

Radon

[Xe]6s²4f¹⁴5d¹⁰6p⁶

118

295.2...

Og

Oganesson

[Rn]7s²7p⁶5f¹⁴6d¹⁰

Atomic Number

Symbol

Name

Electron Configuration

Atomic Mass, u

PubChem

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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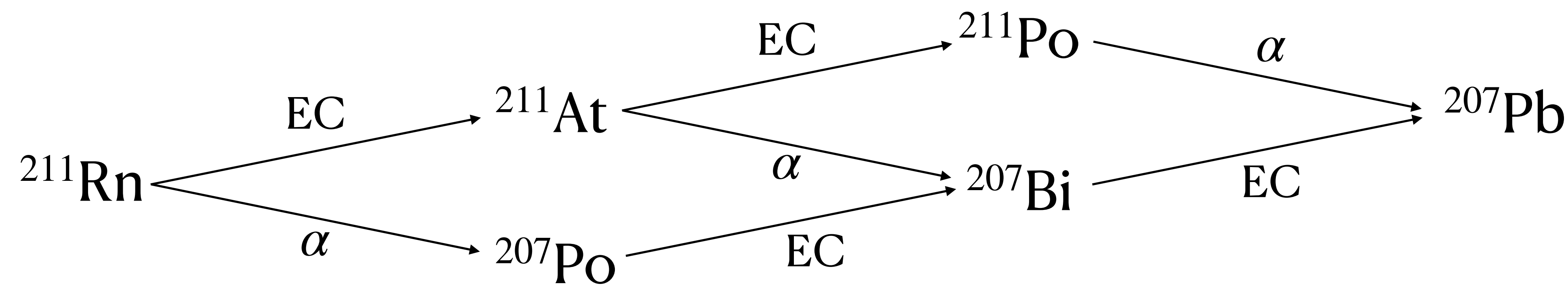
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Production of ^{211}At

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

Surface facets

Coverage

Other
materials

Iodine

Nanoparticle

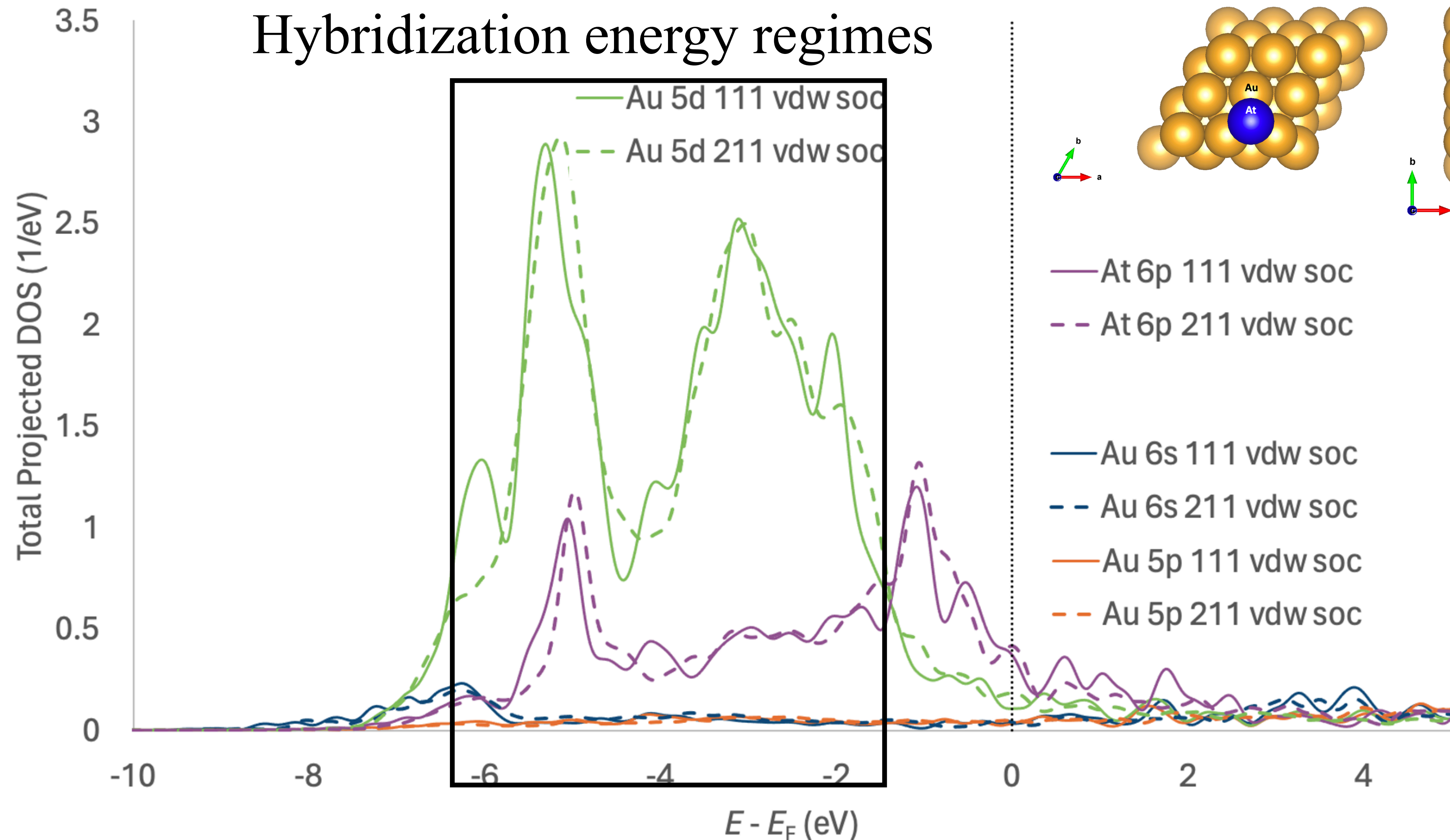
Adsorption
characteristics
of decay
remnant to gold

Effect of
radiation to
original system

To design
optimized carriers
for radiotherapy

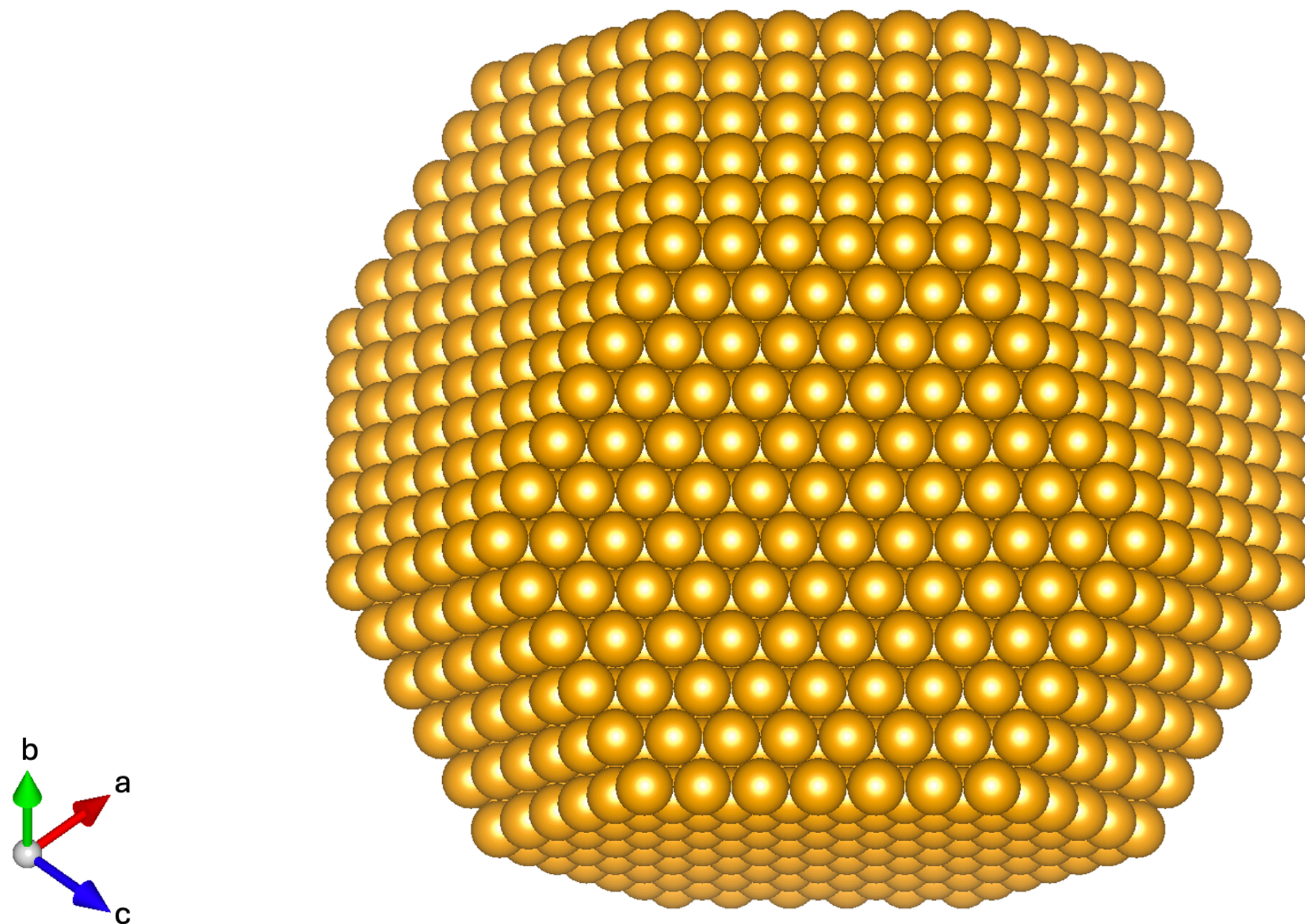
LDOS for At/Au(111) and At/Au(211)

Hybridization energy regimes



Adsorption on nanoparticle

Model diagram of a 5 nm AuNP



Model diagram of a 1 nm AuNP

