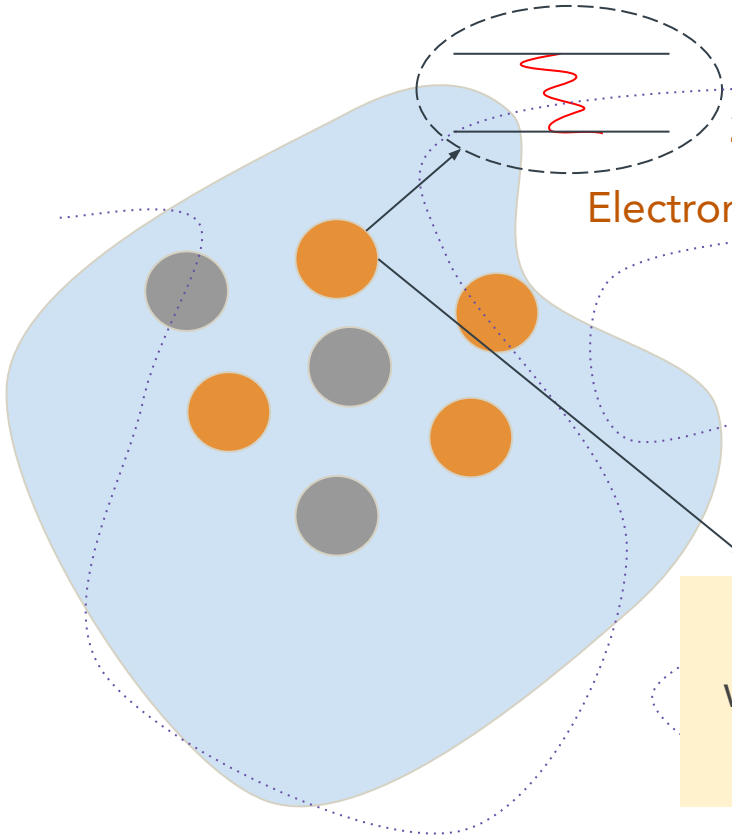
Enhancement Due to Photonic Coupling in Nanocavity Structures

Bumbia, Amal;
Abudayyeh, Hamza;
Liu, Zhida;
Li, Xiaoqin

Department of Physics UT Austin

Background

How do quantum emitters work?



2-level Quantum emitter

Electromagnetic environment

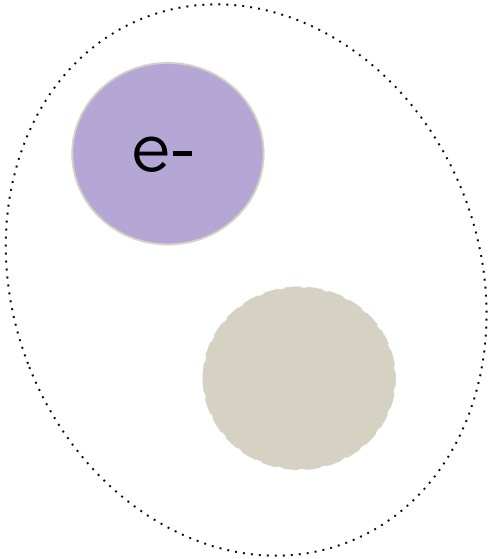
Optical properties!

Spontaneous photon emission!

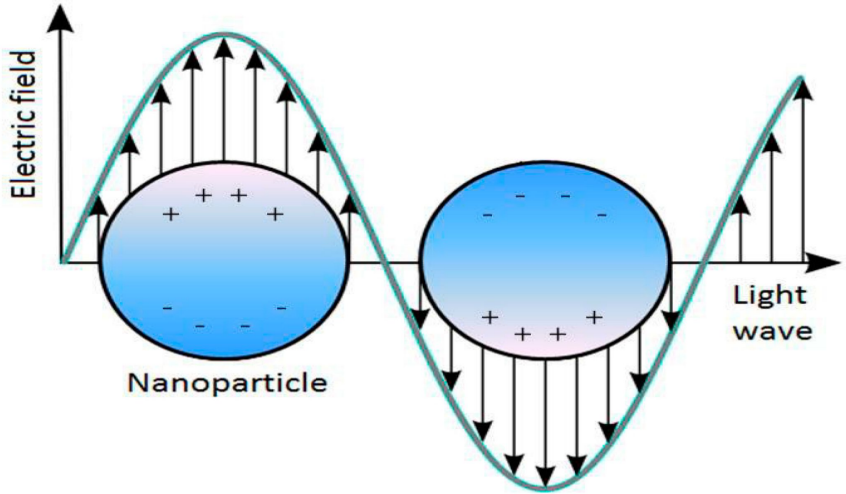
How can we alter this environment to favorably influence the decay rate of the system?

Multiple 2-level systems with differing polarization or wavelength

Why can a nanoscale emitter can have emitting behavior?



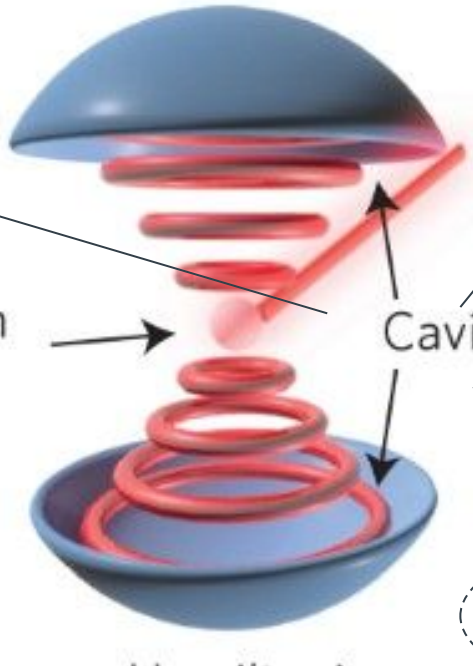
Exciton - electron-hole pair



Plasmon - EM field via the collective oscillations of material's conduction electrons

interaction between the excitons of the emitter and the plasmons of the nanoparticles — emitter is one of the excitonic transitions

What is the interplay between quantum emitters and cavities?



coupled resonantly exchange photons

enhanced photon emissions into free space

contains plasmonic resonances

emits into the plasmonic cavity

focuses the emission fields into a mode volume

Cavities can impact emitters in a few ways:

- decay rate enhancement --- weak coupling of a 2-level emitter to a cavity -> modification of the spontaneous emission rate so emitter can have multiple radiative modes and some nonradiative processes

- emission redirection --- intrinsic radiation pattern of a quantum emitter is omnidirectional, making it hard to collect emitted photons. Cavities can redirect the emission to the desired optical mode with an efficiency ξ. Different modes can have different efficiencies.

- absorption enhancement --- cavities alter the absorption cross-section of the emitter causing absorption enhancement and lowering saturation power needed from the pump laser

Coupling parameters:

- emitter's decay in free space Γ_0
- cavity's loss rate κ
- emitter-cavity coupling coefficient g

- Parameters depend on the overlap of the photonic cavity and the resonances of the 2-level system.
- Due to the interplay between the photonic cavity and the system, we are able to enhance some processes at the expense of others.
- For instance, if the resonances of the plasmonic cavity are vertical, they selectively enhance dipole emitters with a vertical dipole.
- Not only are we increasing the lifetime, but we are also altering our characterization and probability of the emission.
- What matters is coherence and its impact on the photonic field. We aim to enhance all photons in the emission.

Weak Coupling Regime

We are concerned with the weak coupling regime $g < \kappa, \Gamma_0$.

Here, the photon is released by an excited emitter due to spontaneous emission and lost to far-field radiation modes leaving the cavity in a vacuum state.

In this case, the cavity becomes the system's environment and results in a modified decay rate $\Gamma = F \Gamma_0$ where $F = 3/(4\pi^2)\, Q\, (\lambda^3)/V$ (Purcell factor - represents the shortening of lifetimes; losses ensure that it is not directly related to the number of emitted photons), $Q \sim 1/\kappa$ (cavity quality factor), and the cavity mode volume V/λ^3.

The advantage of the weak coupling regime is that for emitted photons, the probability that they will be reabsorbed by the emitter is low compared to the likelihood of the photon escaping the cavity.

What are the effects of weak coupling between plasmonic nanocavities and excitons?

How does plasmon-exciton coupling effect emission enhancement?

Background

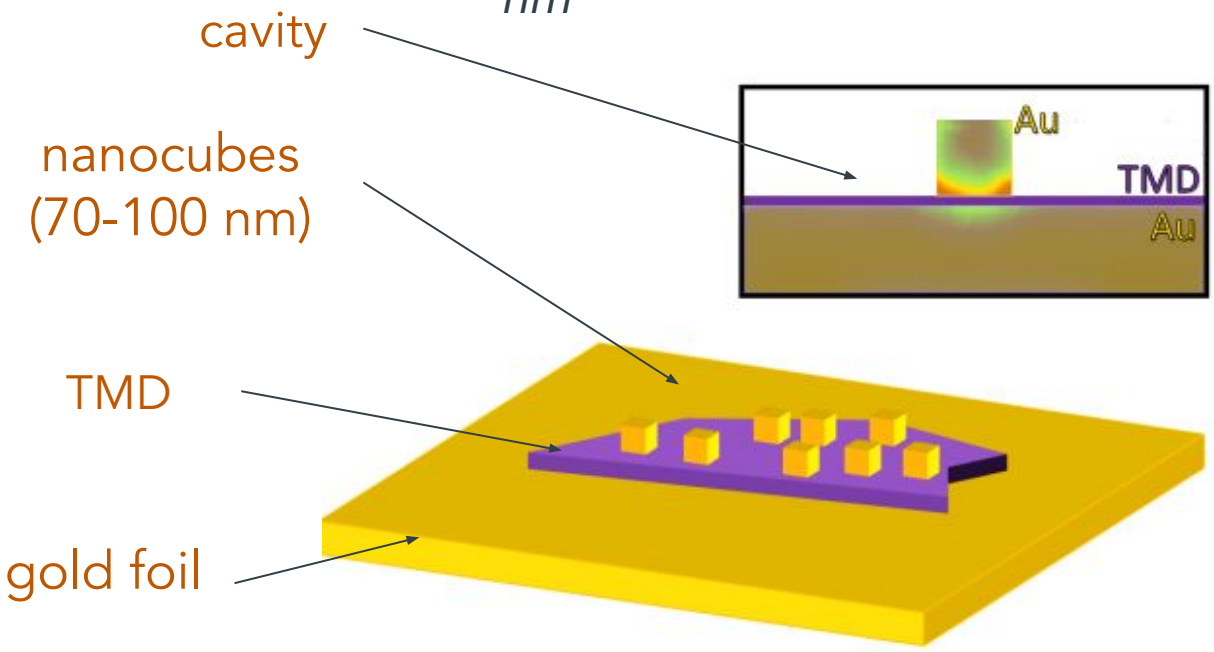
Sample 1 : Gold Nanocube

Sample 1: Gold Nanocube : Optical Data

The lightning-rod effect (EM fields concentrating) causes the corners of the cubes to be the regions of greatest enhancement

Material: *Monolayer TMD*

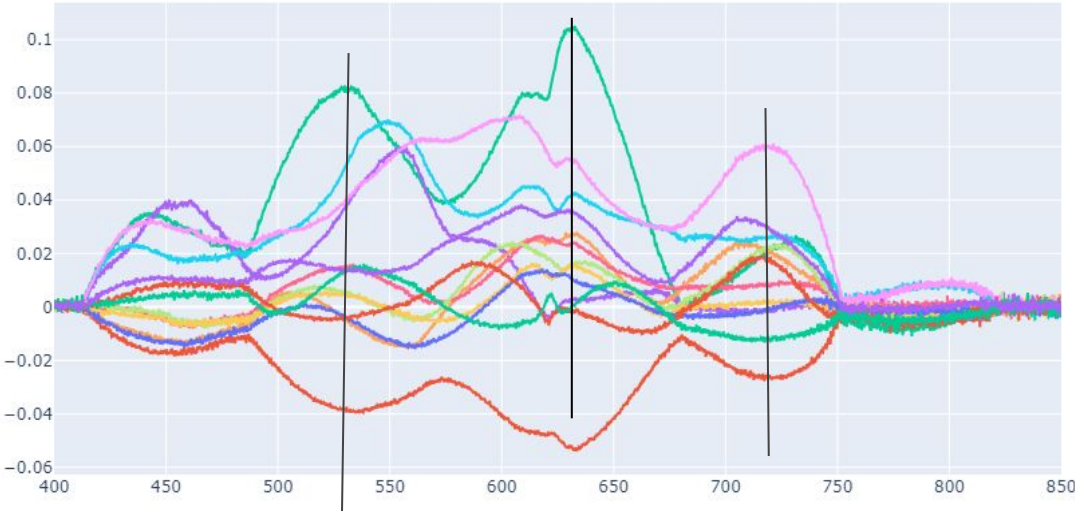
Film-Cube *Separation: 8-9 nm*



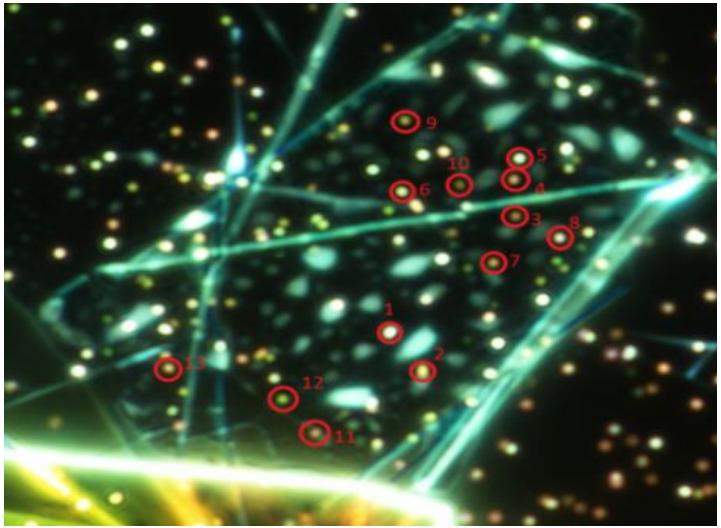
Dark Field Spectroscopy

Where do the plasmonic resonances occur?
This is a point where the data gets murky…

- 540 nm
- *625 nm !!!*
- 725 nm

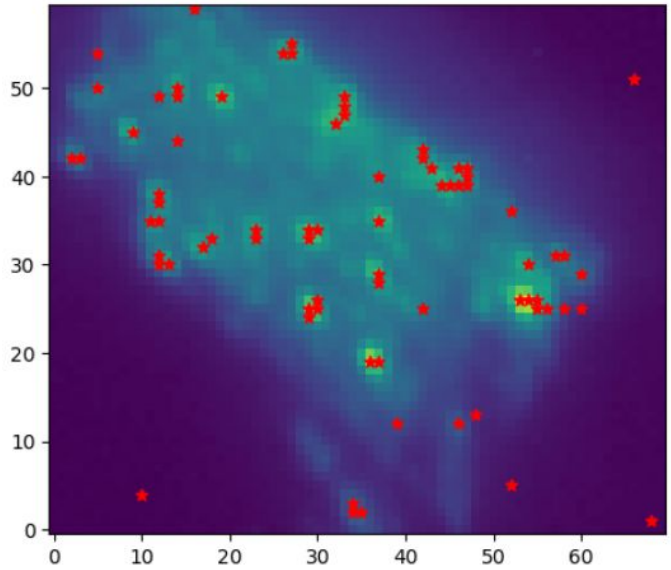


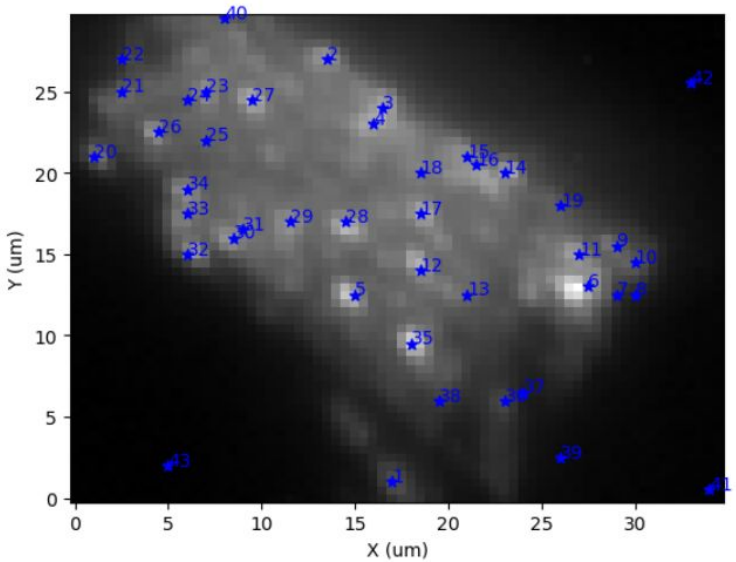
- white light scattering by nanocavities
- measure plasmonic resonances



Hyperspectral Spatial Image

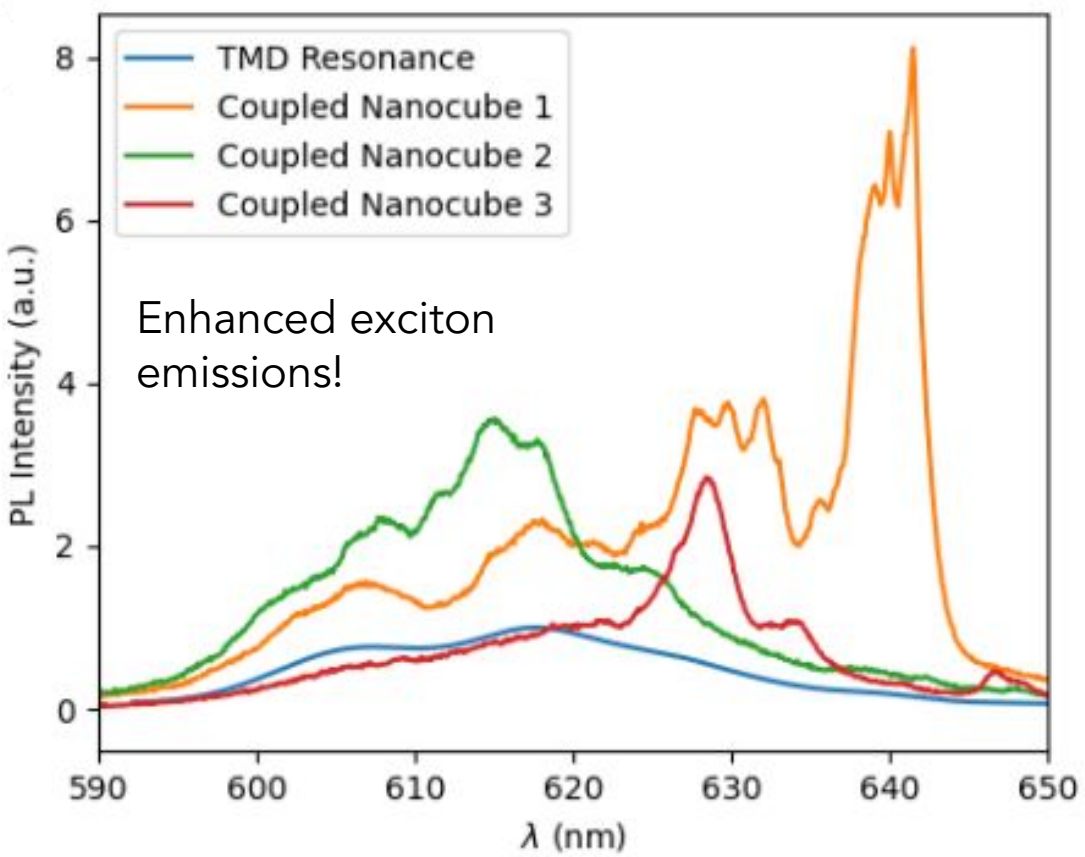
- well localized points with nanocubes
- intensity coming out of these points is enhanced

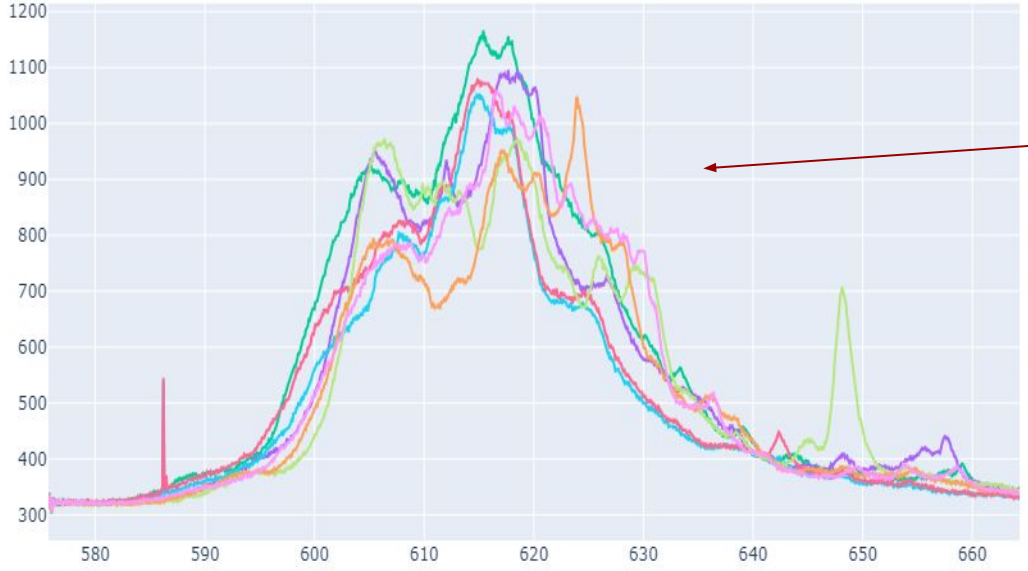




Photoluminescence (PL)

- excite sample at different wavelengths
- measure exciton resonances
- longer wavelengths enhanced
- greater, non-uniform intensity enhancement along nanocubes
- find overlap between the wavelengths where plasmonic resonances and enhancement occur

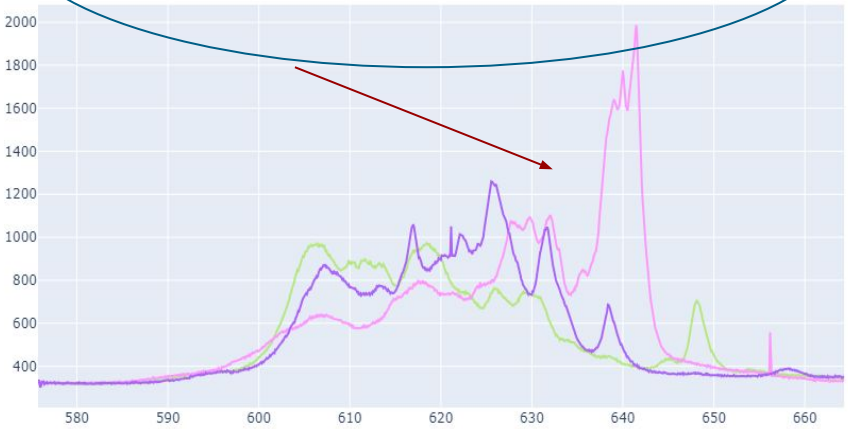




Notice the peaks between 603-609 nm and 612-620 nm and around 625 nm

Sight trough at 608-611 nm and again at 620-625 nm

These were the brightest spots - less uniformity between them but we still have peaks at around 605, 630 , 640 and 615 nm



Recall the plasmonic spectrum via dark field spectroscopy and the peaks around 625 nm — possible overlap between plasmonic and exciton resonances?

Background

Sample 1 : Gold Nanocube

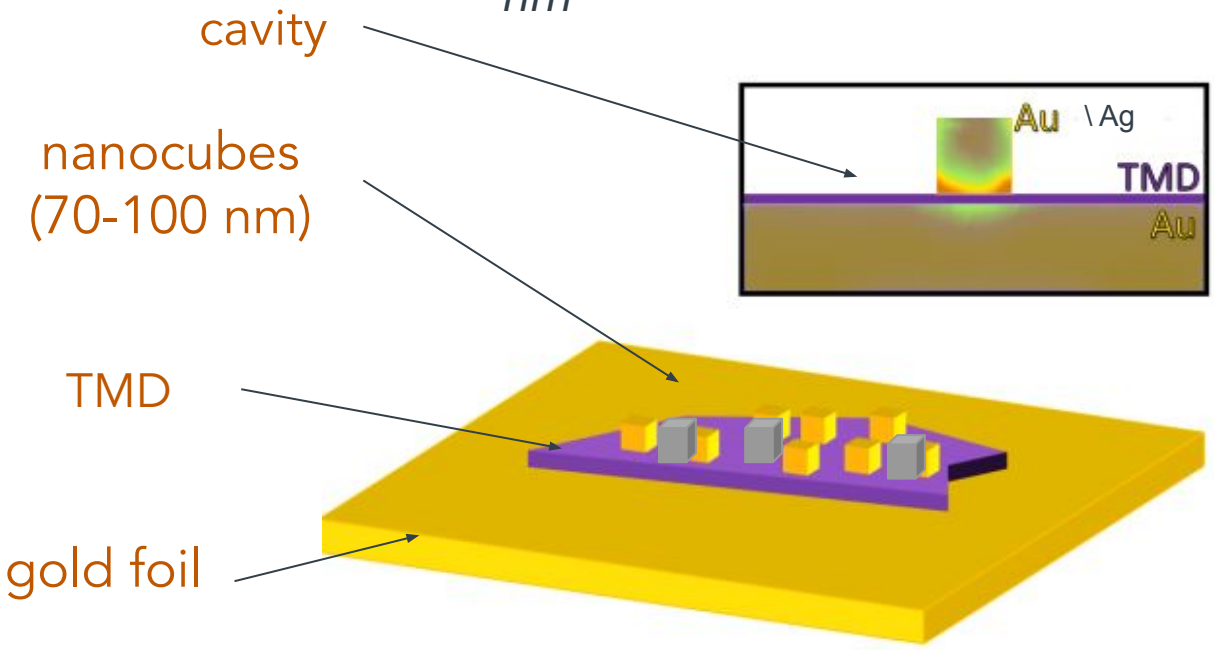
Sample 1 : Silver Nanocube

Sample 1: Silver Nanocube : Optical Data

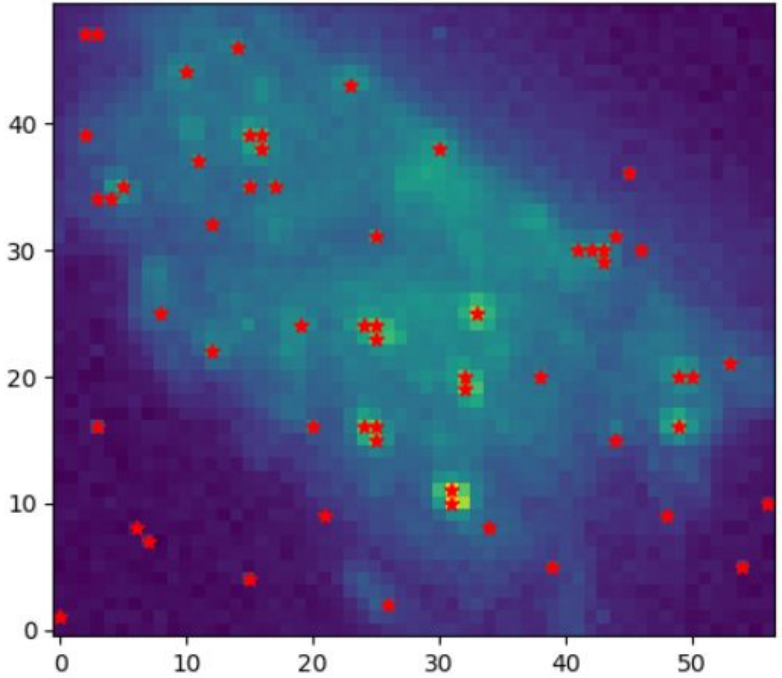
Initially, the silver nanocubes were placed onto the original gold nanocube sample. There was an issue with the trilayer sample so it needs to be remade.

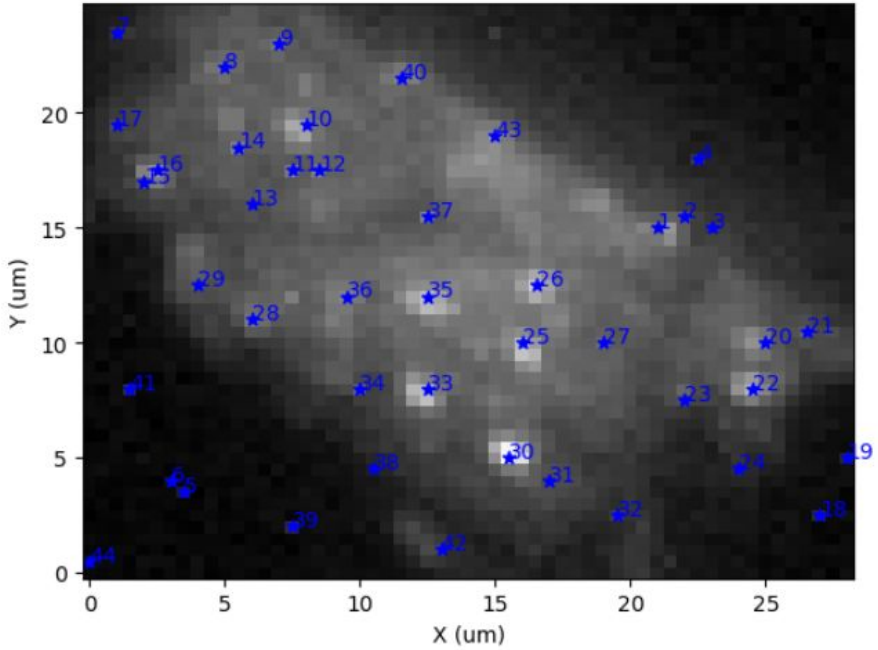
Material: *Monolayer TMD*

Film-Cube *Separation: 8-9 nm*

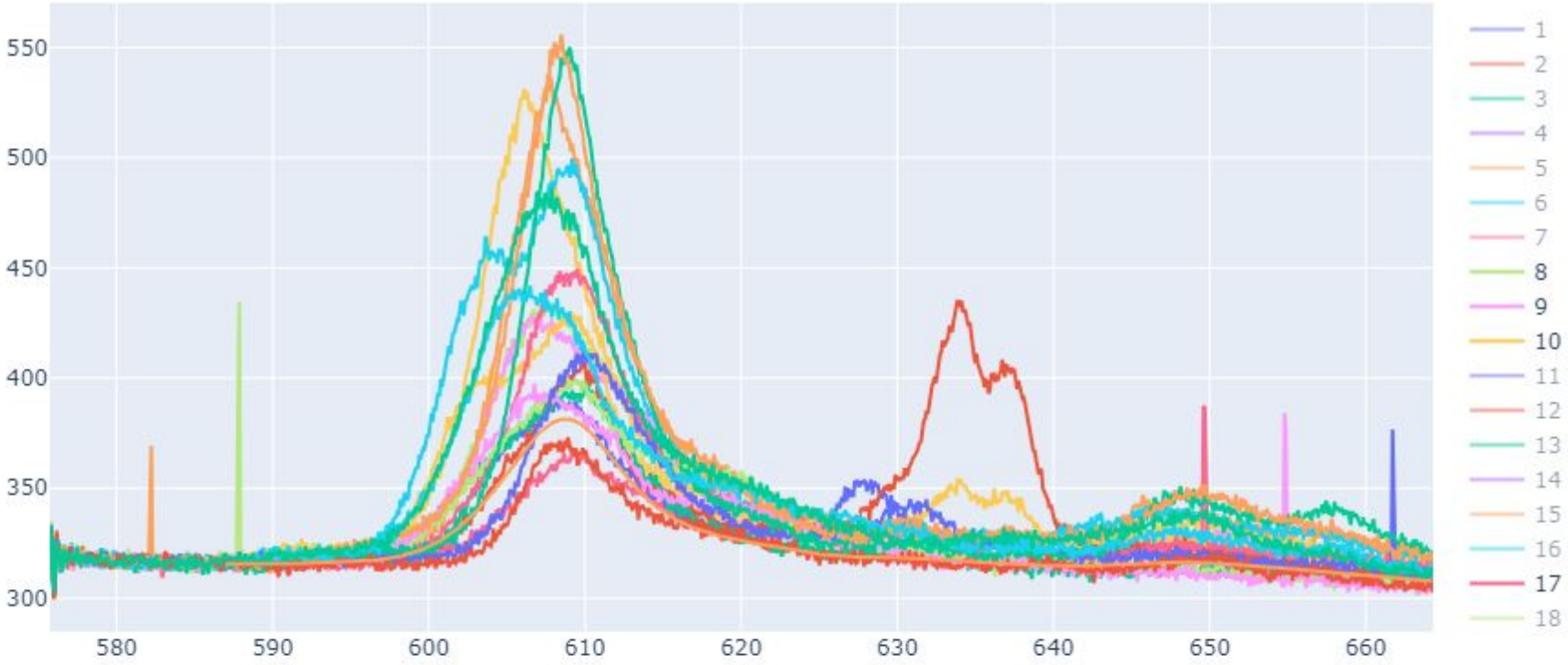


Hyperspectral Spatial Image - Cryogenic





Cryogenic Spectra - cleaned



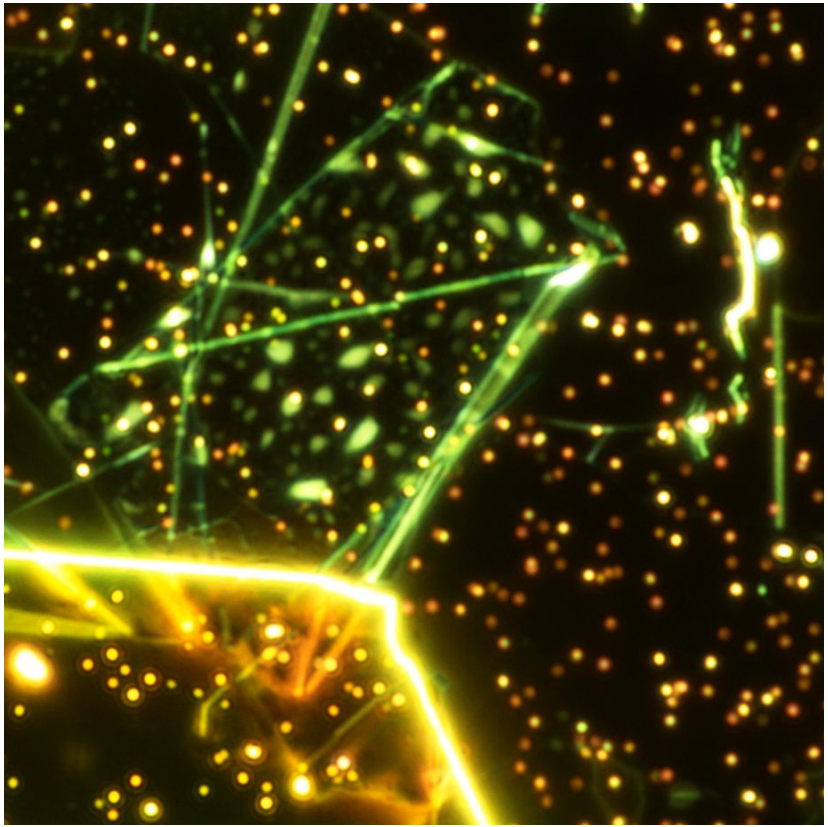
Comparing Cryogenic Results

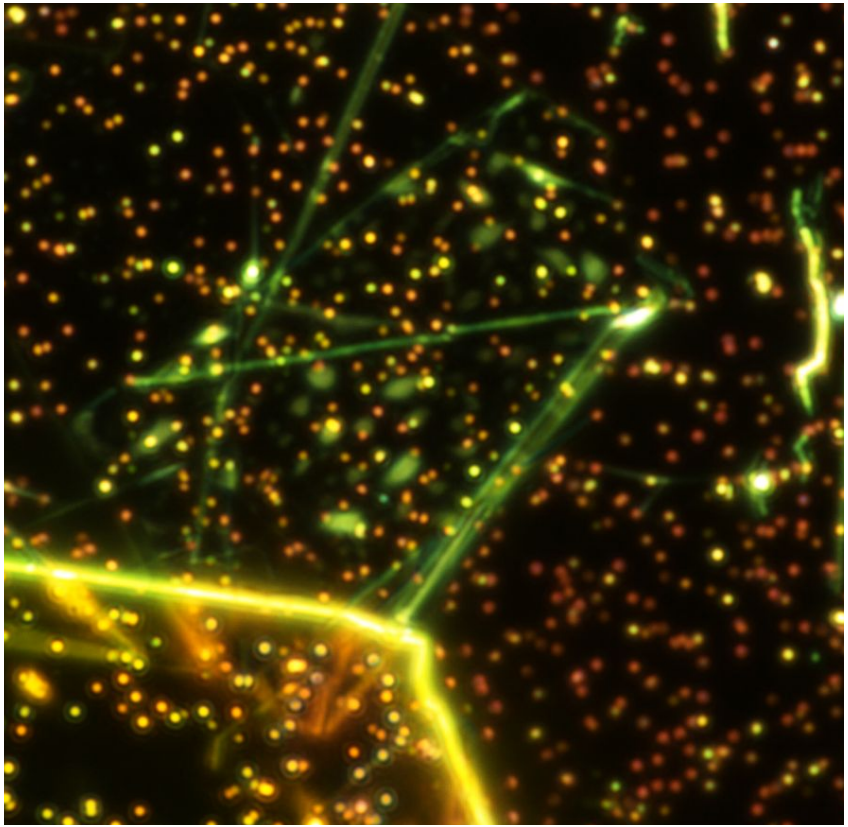
Do the spectra of the same points peak at the same wavelengths between the samples? How does the presence of silver nanocubes impact the measurements?

Can we identify which points are likely silver nanocubes?

In general, how do the peaks differ between the gold and silver nanocubes? Does either provide more obvious enhancement?

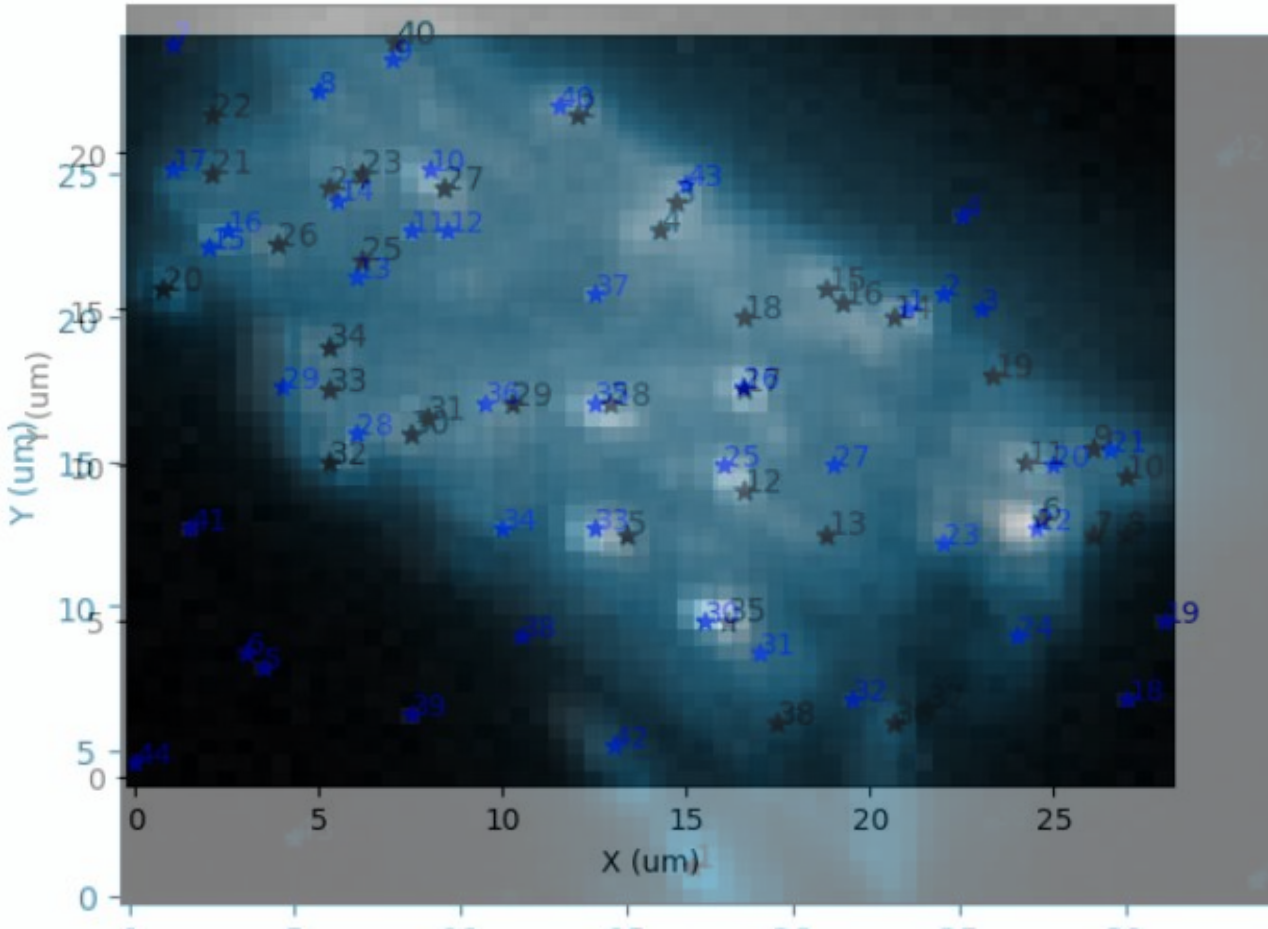
Sample 1 Before and After Silver Nanocube Placement





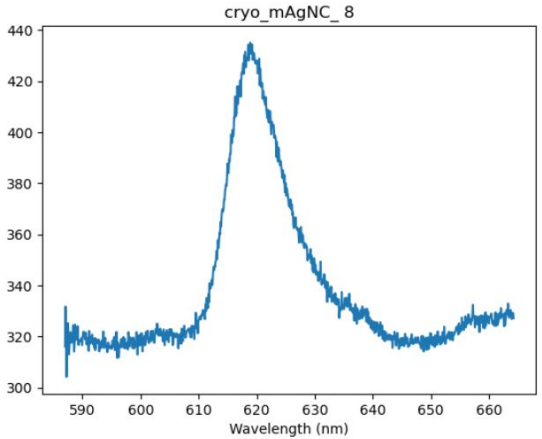
Grey points - Cryo Au
Blue points - Cryo Ag

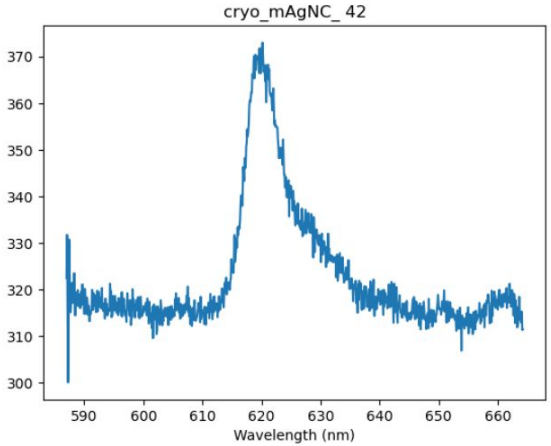
Ag — Au
8 - new silver
9 - 40
10 - 27
13 - 25
17 - 21
20 - 11
21 - 9
22 - 6
23 - new silver
25 - 12
26 - 17
27 - new silver
29 - 33
31 - new silver
33 - 5
35 - 28
36 - 29
42 - new silver
43 - 3

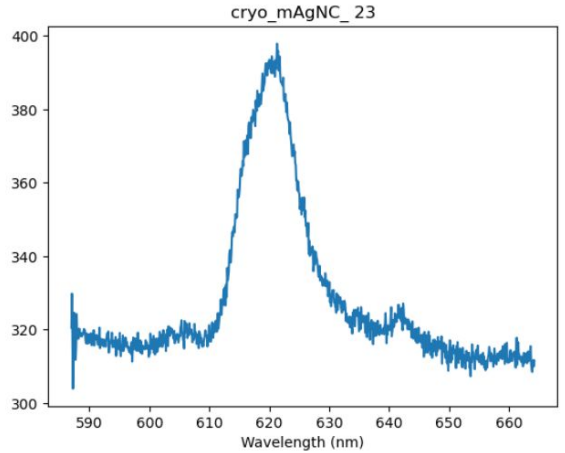


Can we identify which points are likely silver nanocubes?

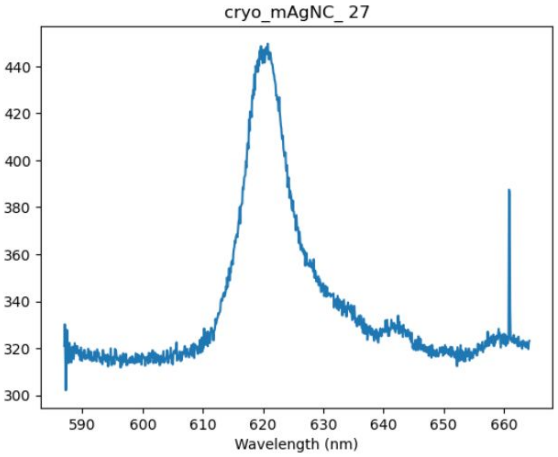
Yes!

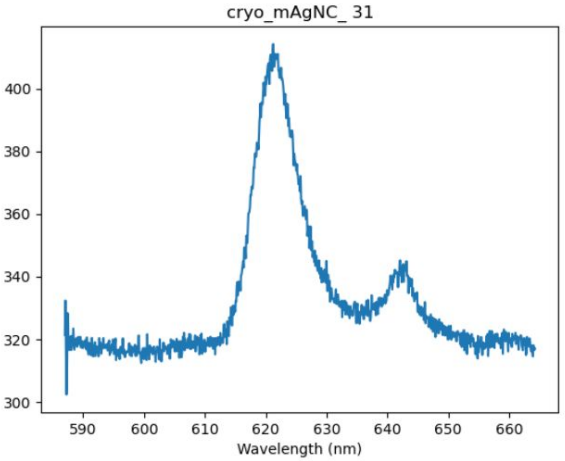






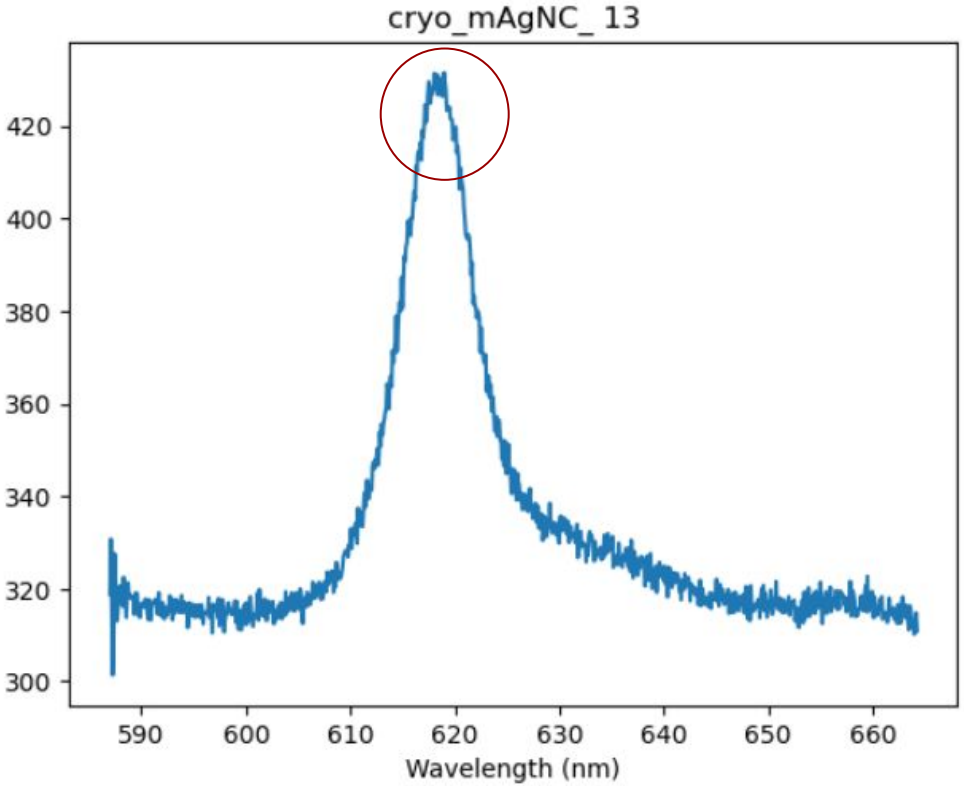
Likely silver nanocubes — notice peaks around 620 nm and similar spectra

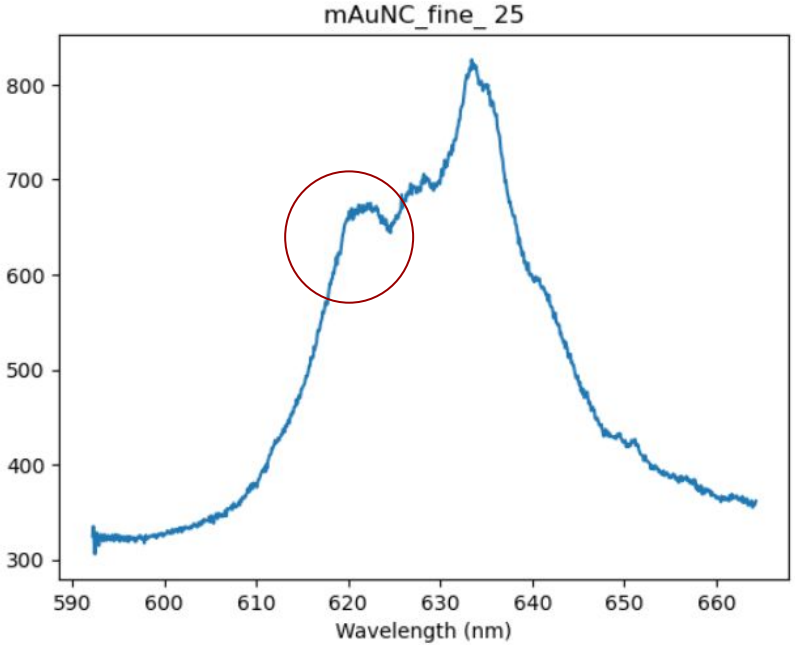


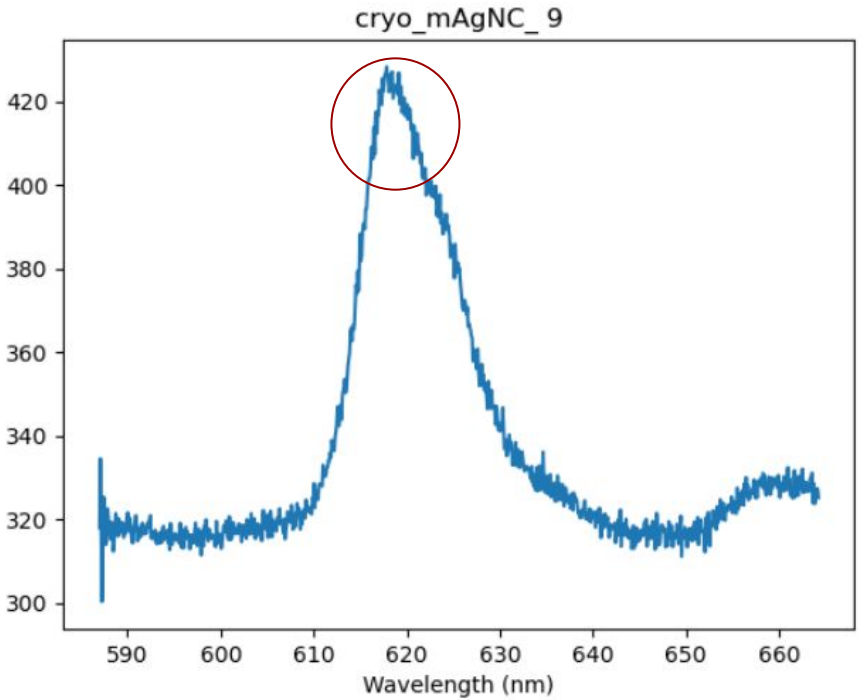


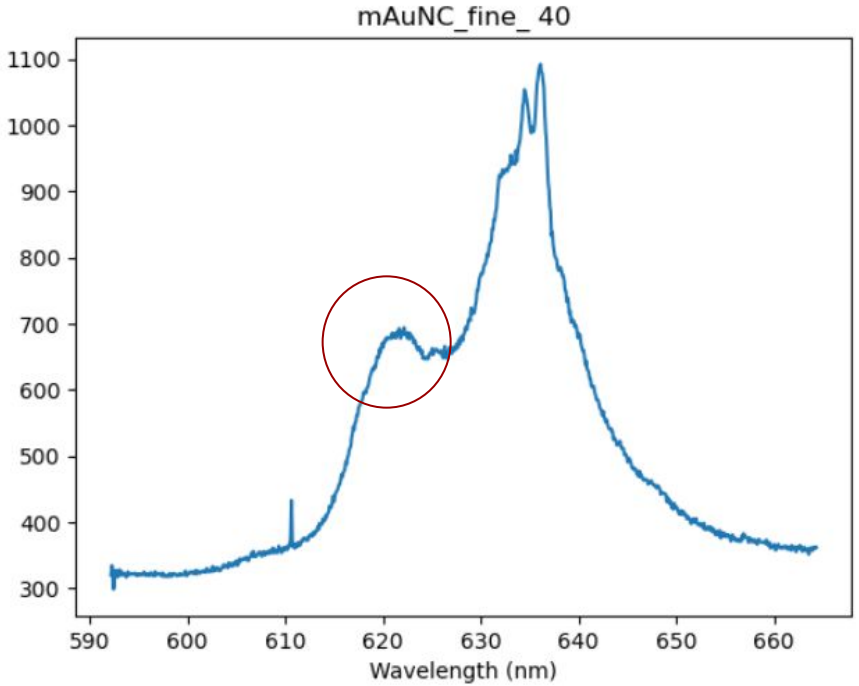
Group 1

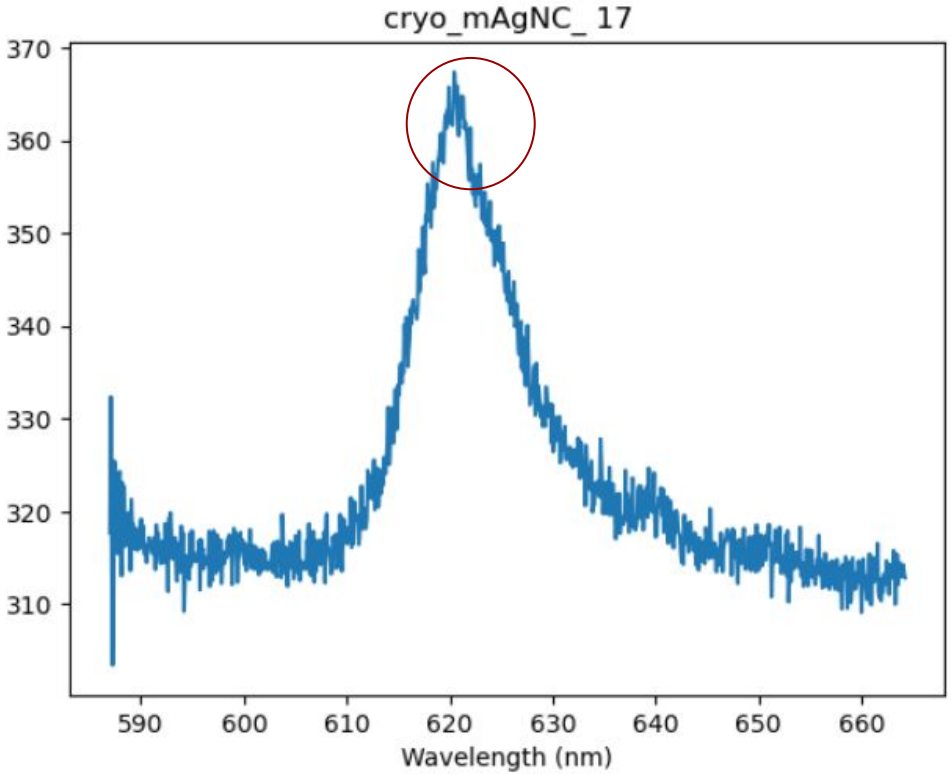
The following points are interesting because they resemble the silver nanocube spectra but are correlated with gold nanocubes

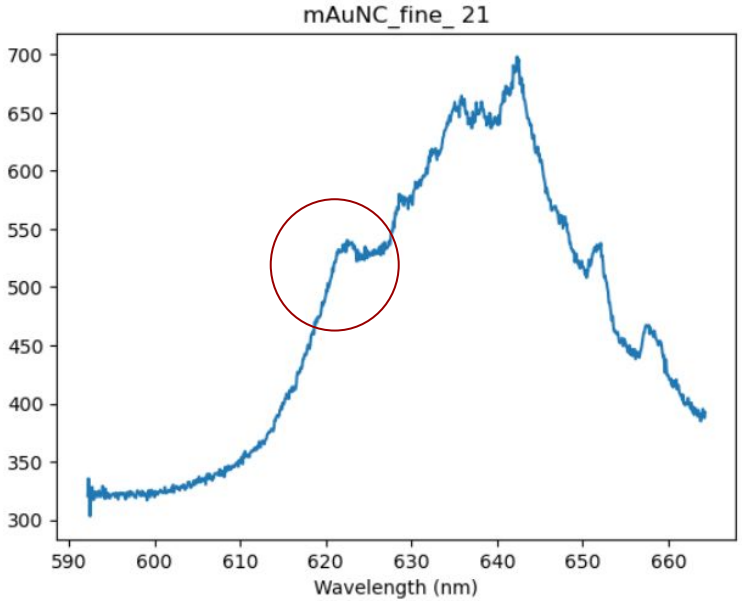

cryo_mAgNC_ 13
420
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

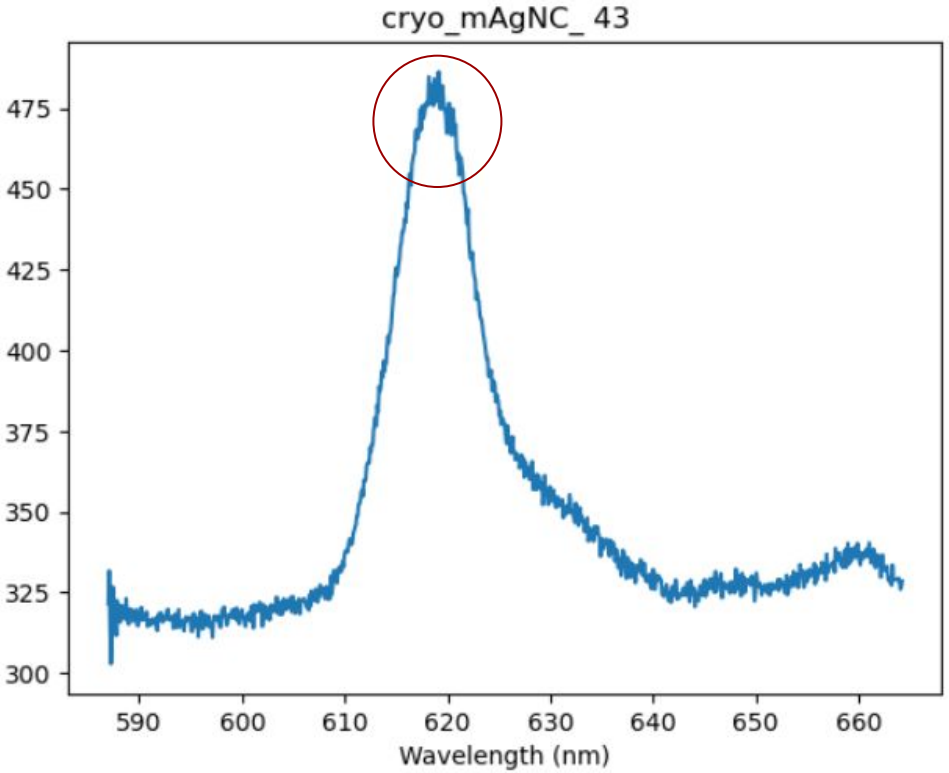

mAuNC_fine_ 25
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

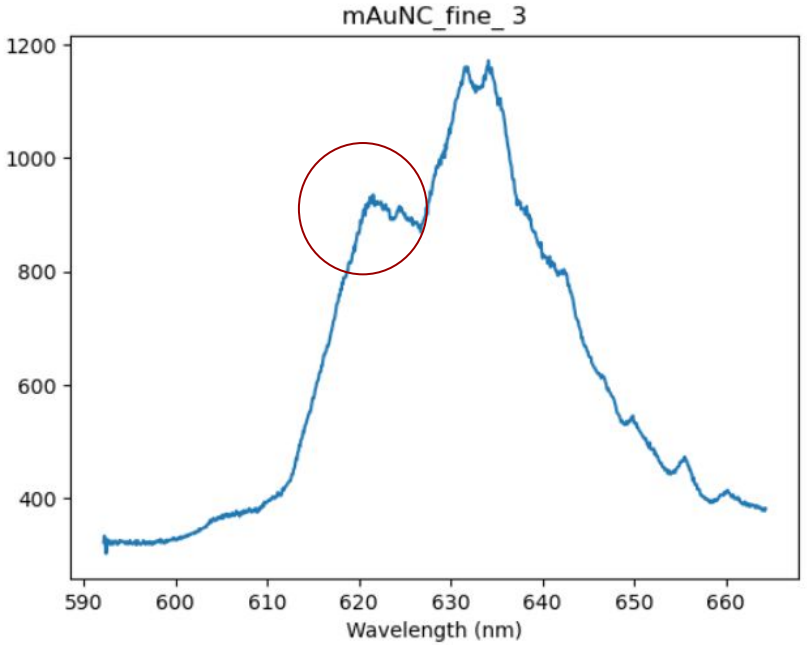

cryo_mAgNC_ 9
420
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

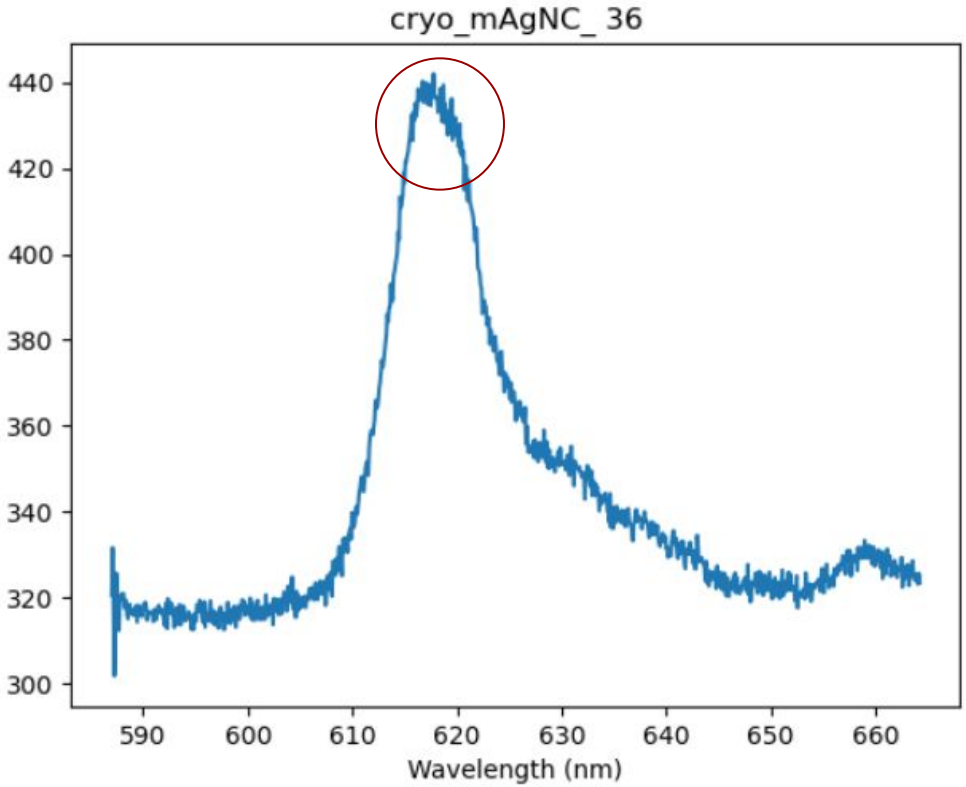

mAuNC_fine_ 40
1100
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

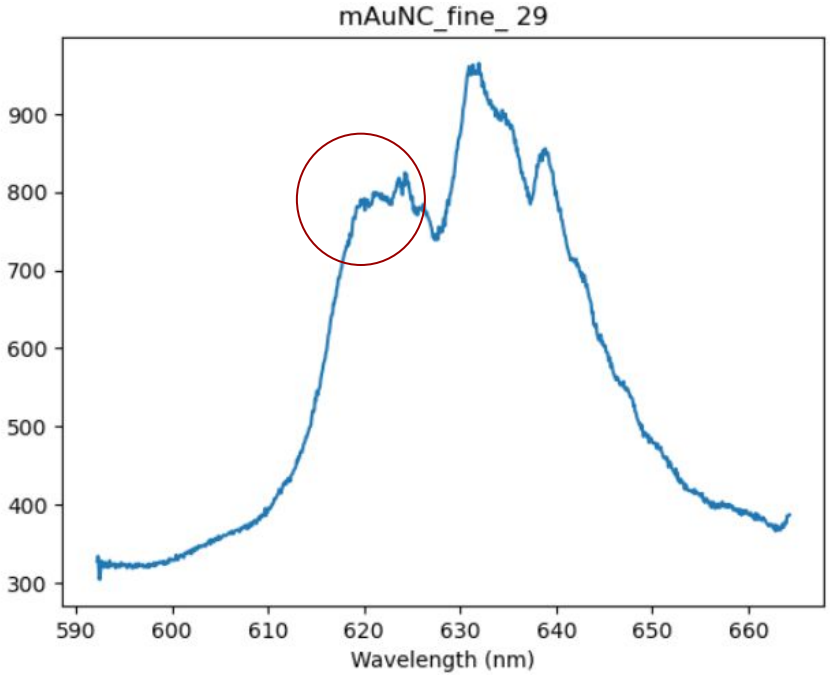

cryo_mAgNC_ 17
370
360
350
340
330
320
310
590
600
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 21
700
650
600
550
500
450
400
350
300
590
600
610
620
630
640
650
660
Wavelength (nm)

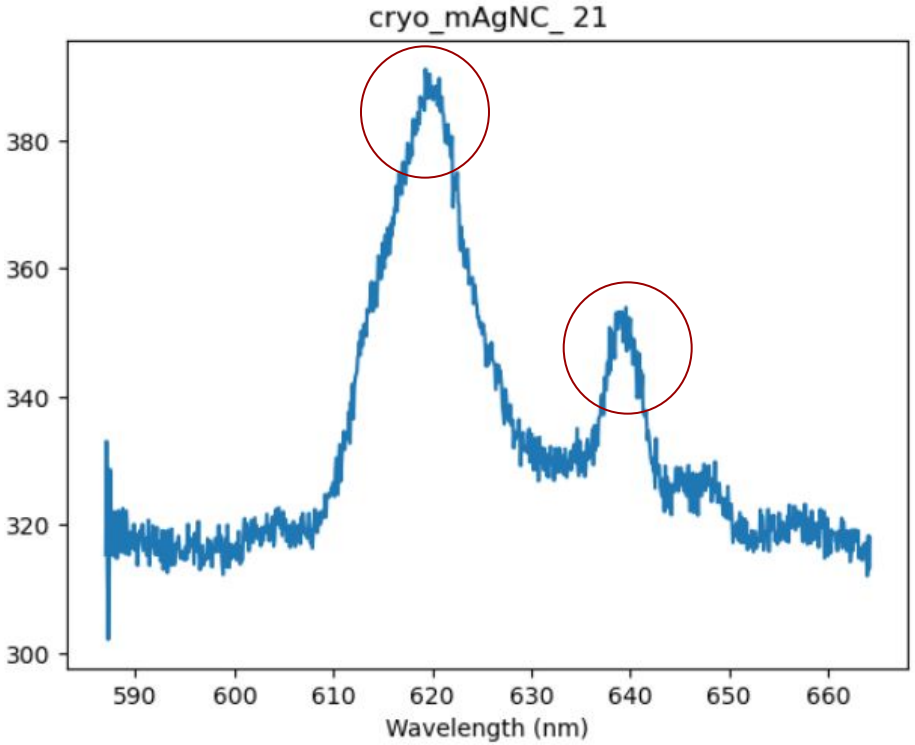

cryo_mAgNC_ 43
475
450
425
400
375
350
325
300
590
600
610
620
630
640
650
660
Wavelength (nm)

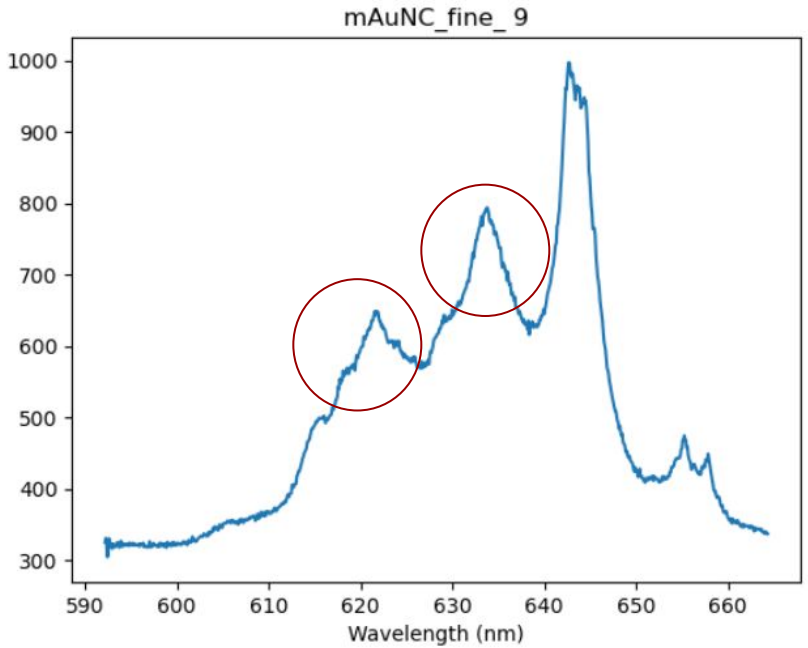

mAuNC_fine_ 3
1200
1000
800
600
400
590
600
610
620
630
640
650
660
Wavelength (nm)

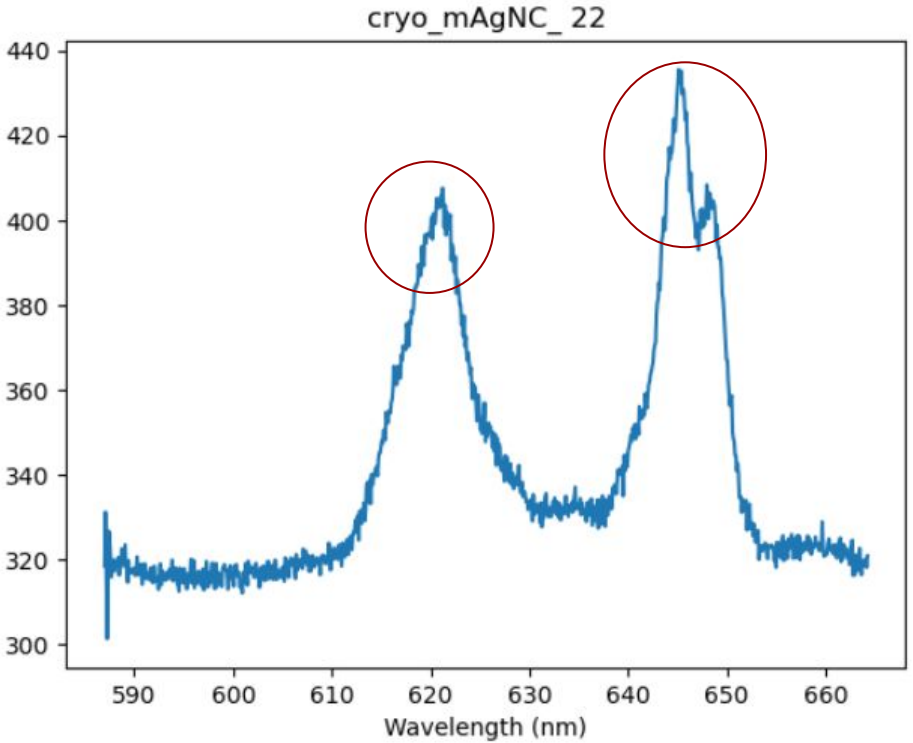

cryo_mAgNC_ 36
440
420
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

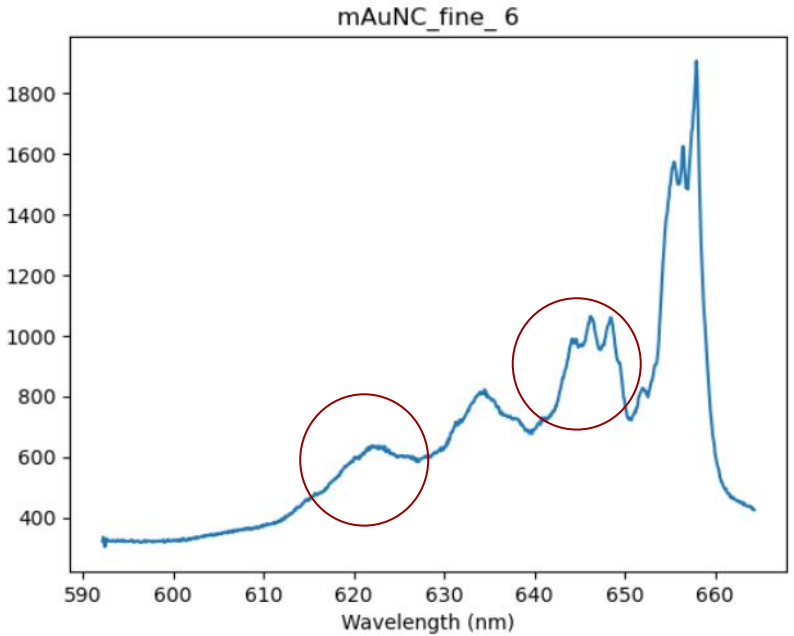

mAuNC_fine_ 29
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

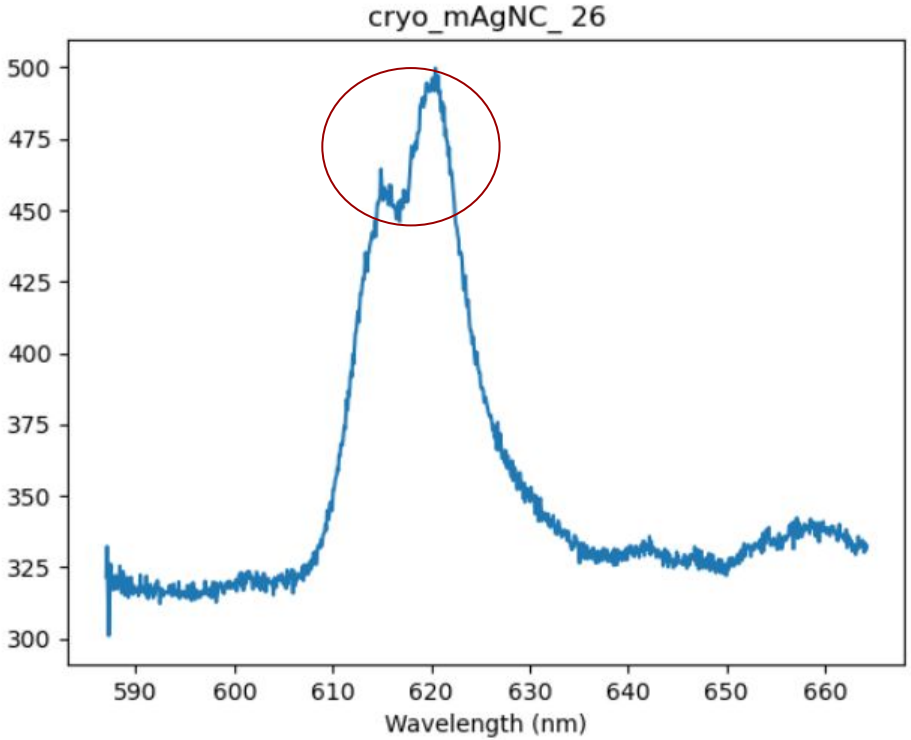
Group 2

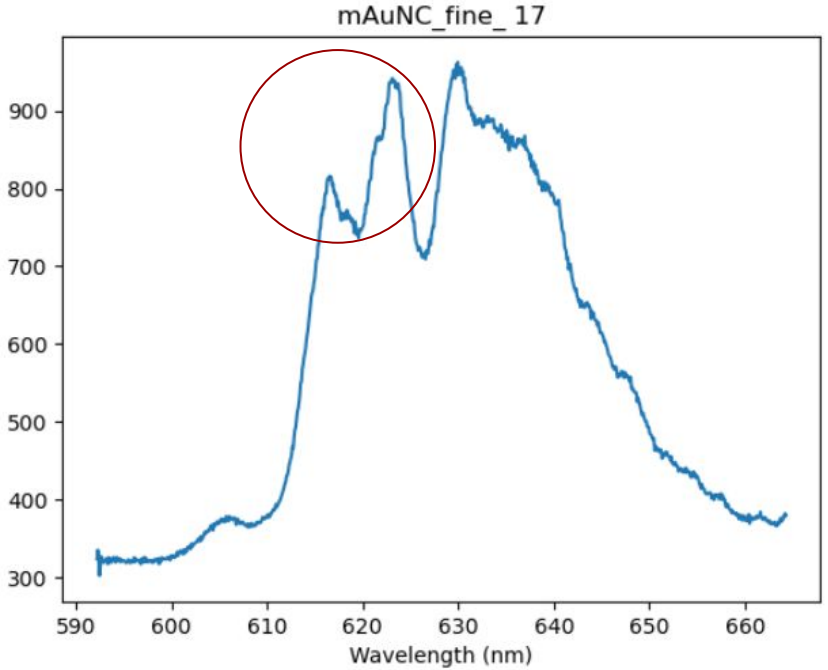

cryo_mAgNC_ 21
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 9
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

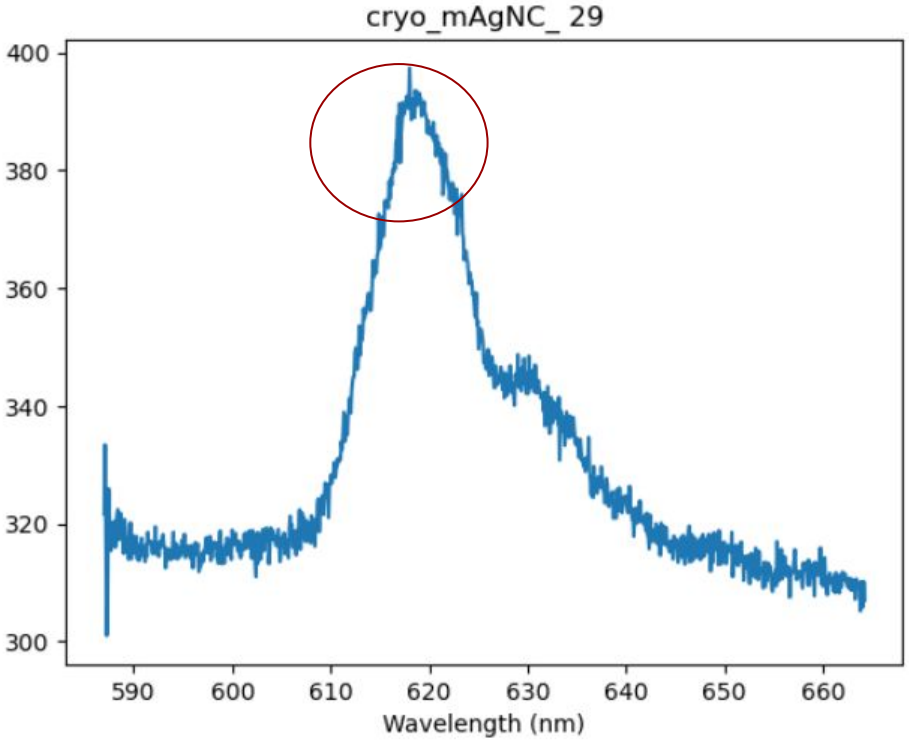

cryo_mAgNC_ 22
440
420
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

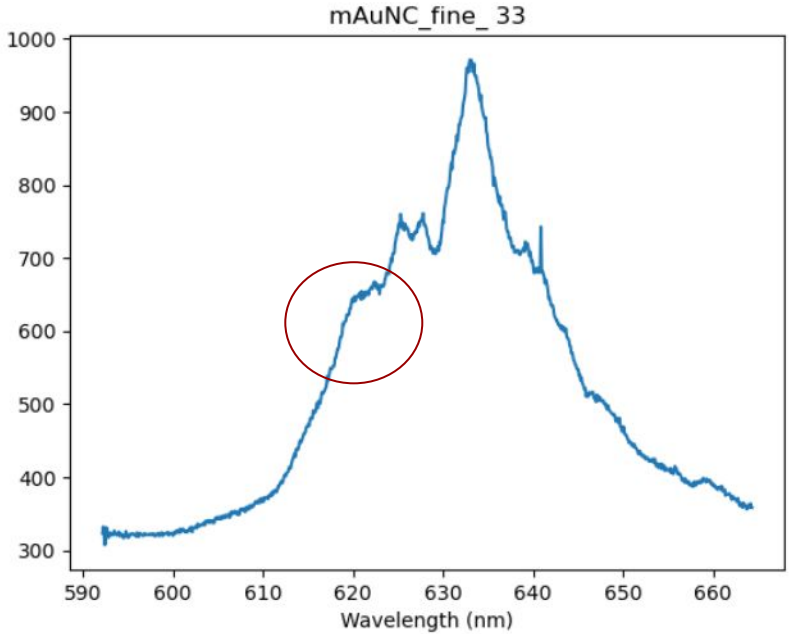

mAuNC_fine_ 6
1800
1600
1400
1200
1000
800
600
400
590
600
610
620
630
640
650
660
Wavelength (nm)

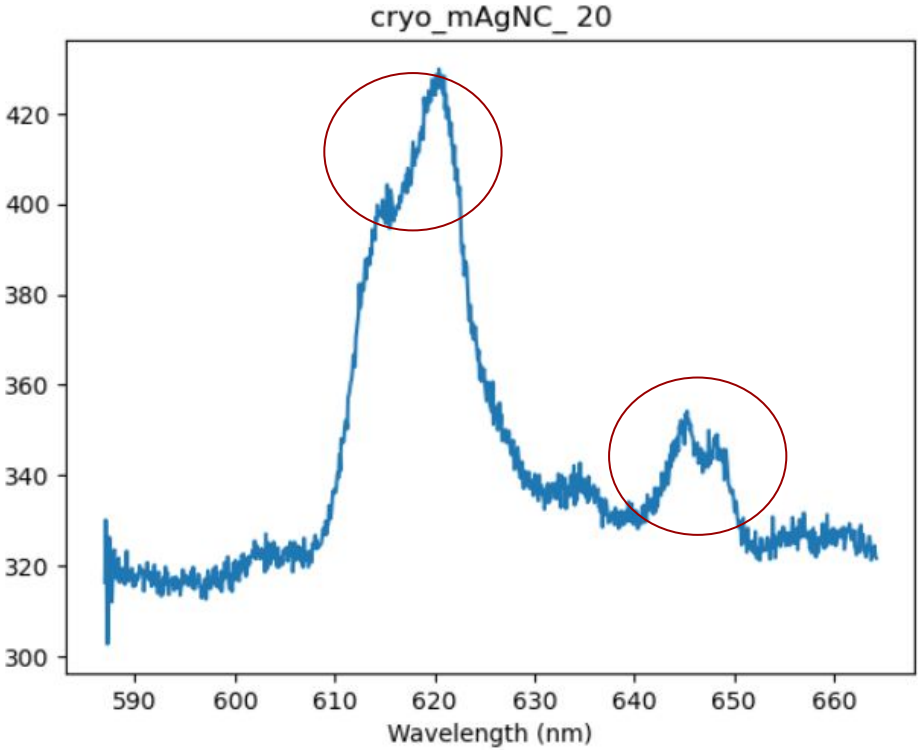


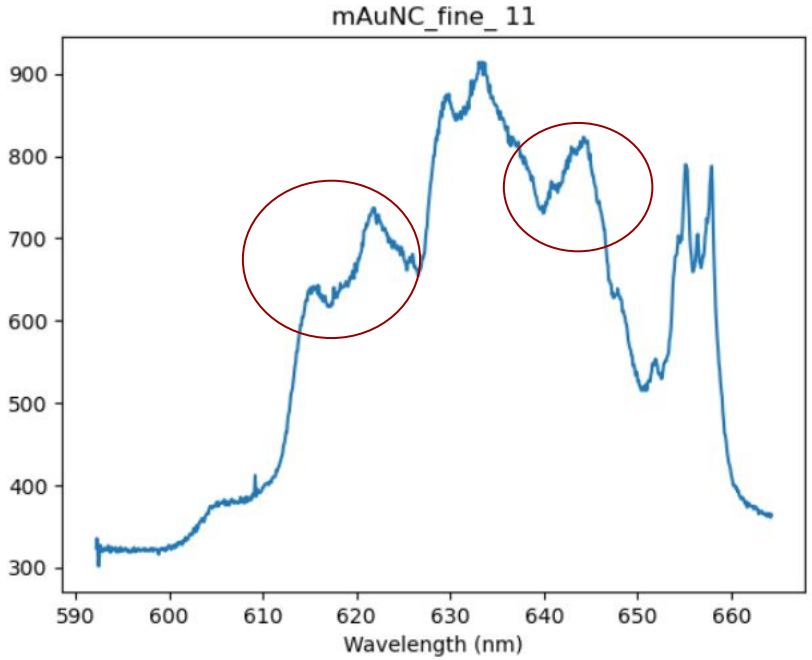


Definitely a gold nanocube point!

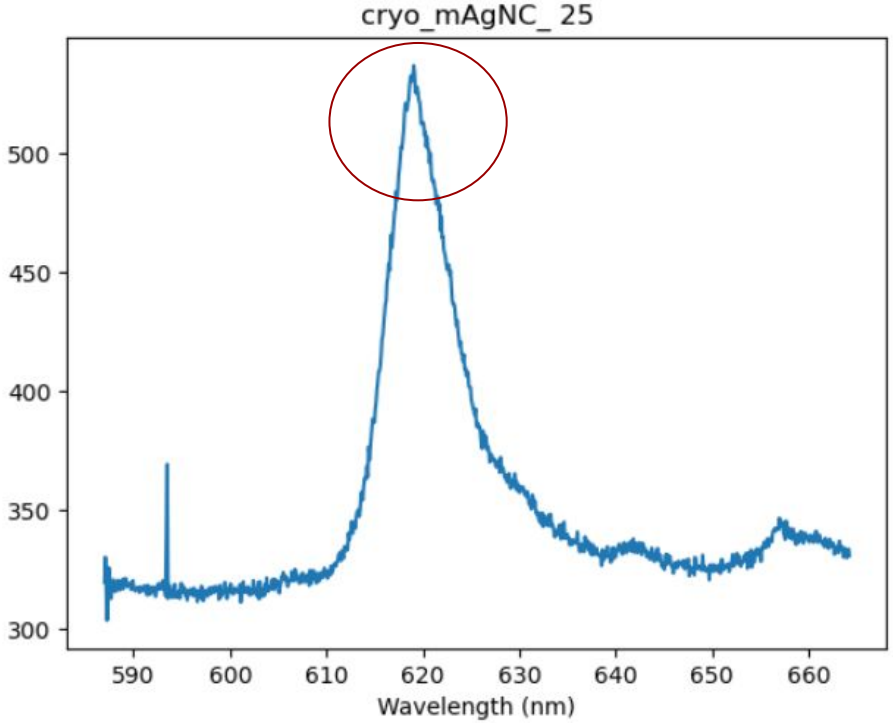

cryo_mAgNC_ 29
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

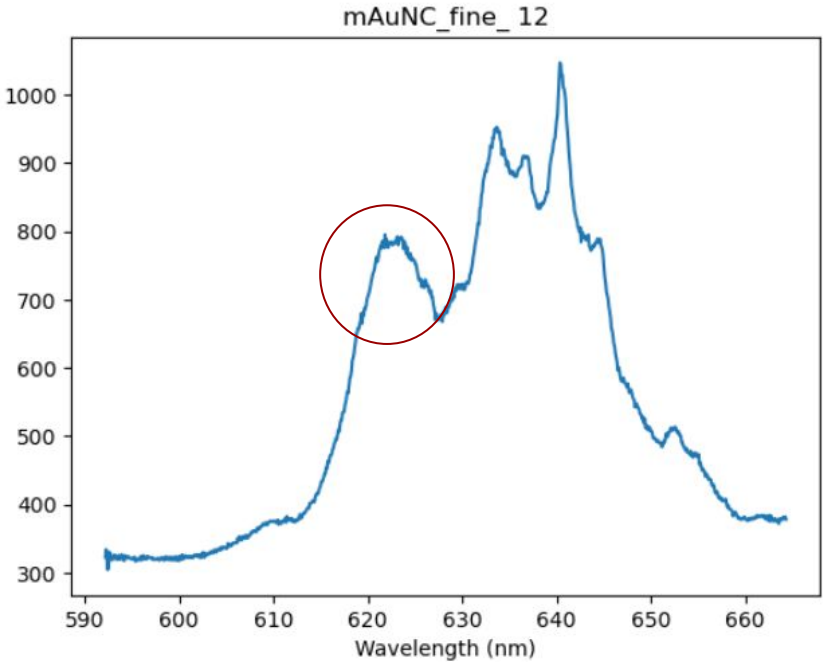

mAuNC_fine_ 33
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

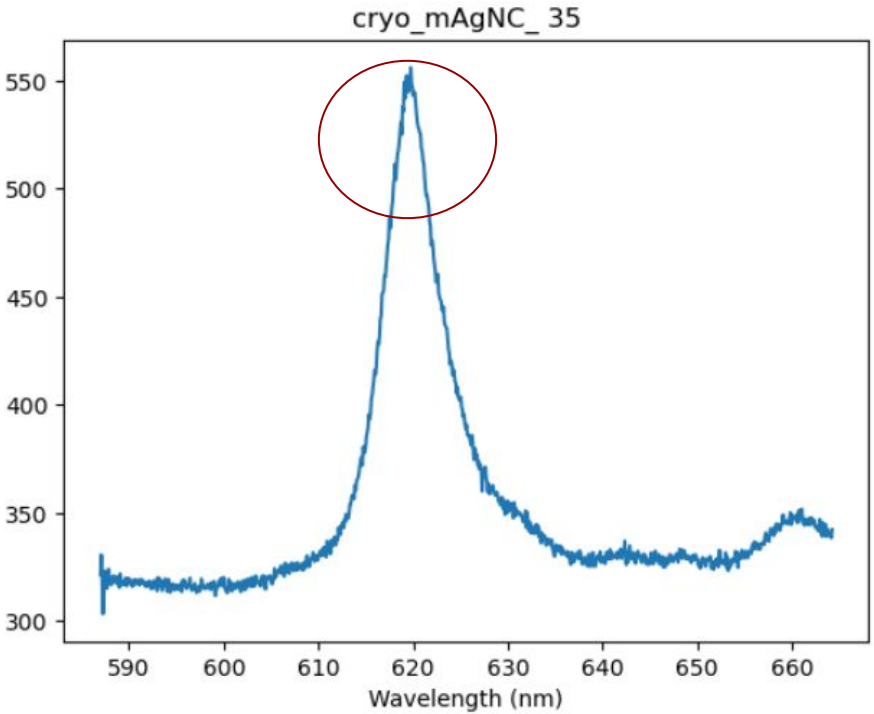

cryo_mAgNC_ 20
420
400
380
360
340
320
300
590
600
610
620
630
640
650
660
Wavelength (nm)

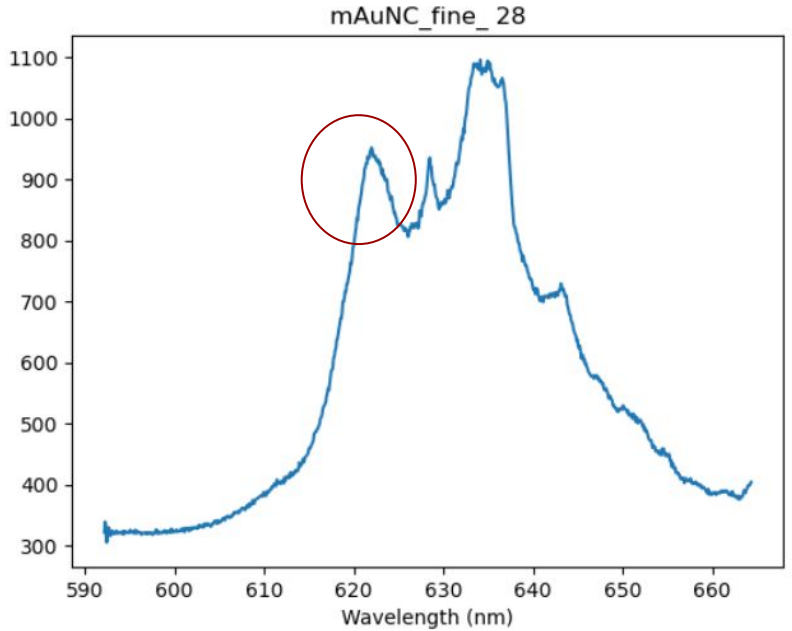

mAuNC_fine_ 11
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

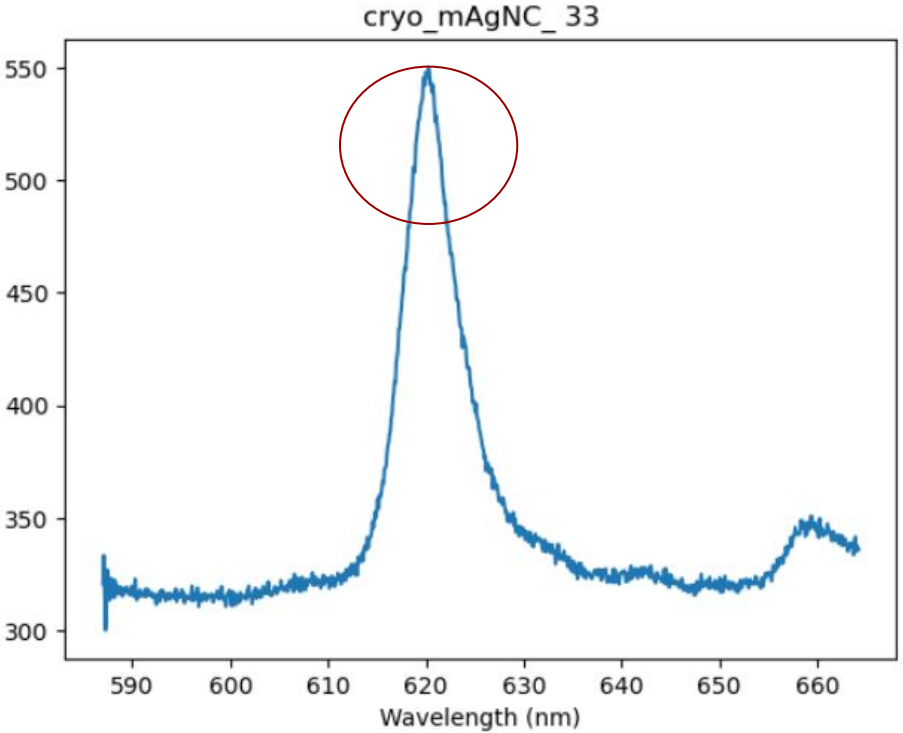
Group 3

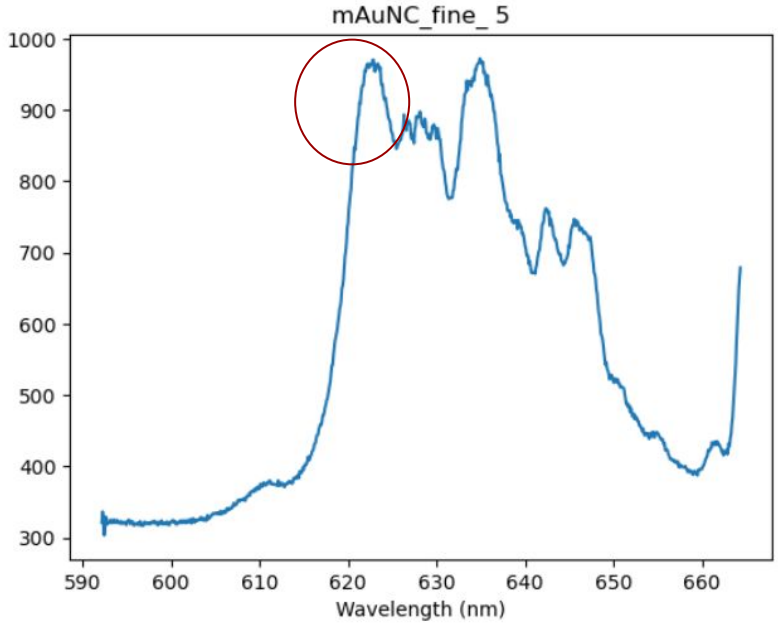

cryo_mAgNC_ 25
500
450
400
350
300
590
600
610
620
630
640
650
660
Wavelength (nm)

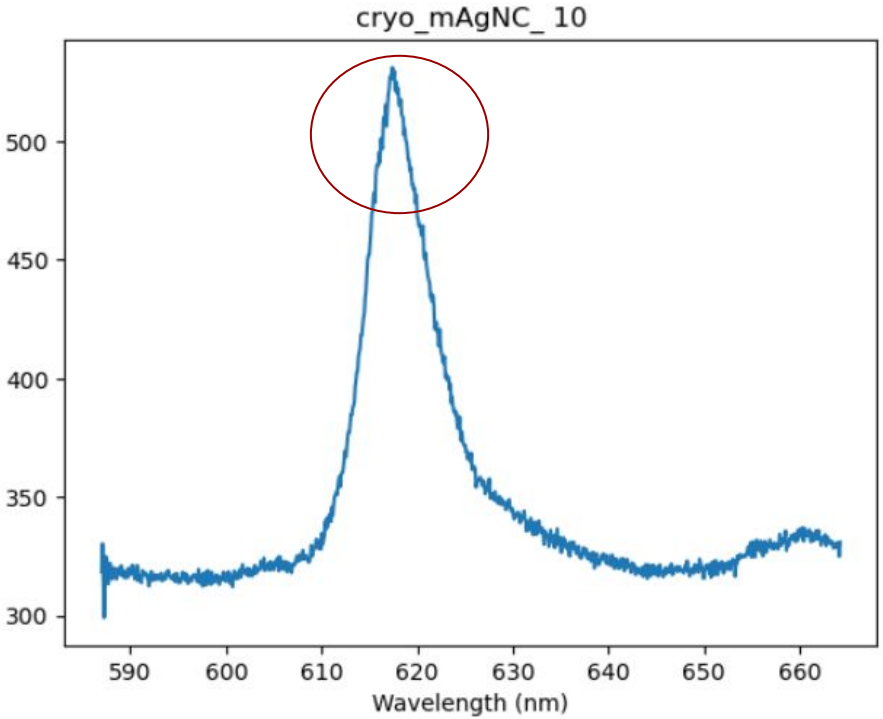

mAuNC_fine_ 12
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

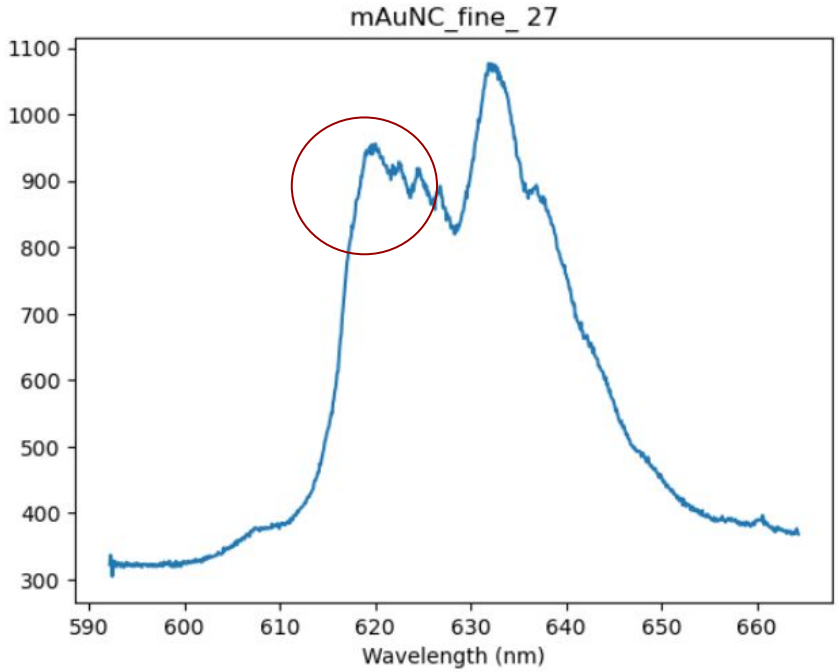

cryo_mAgNC_ 35
550
500
450
400
350
300
590
600
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 28
1100
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 33
550
500
450
400
350
300
590
600
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 5
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 10
500
450
400
350
300
590
600
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 27
1100
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

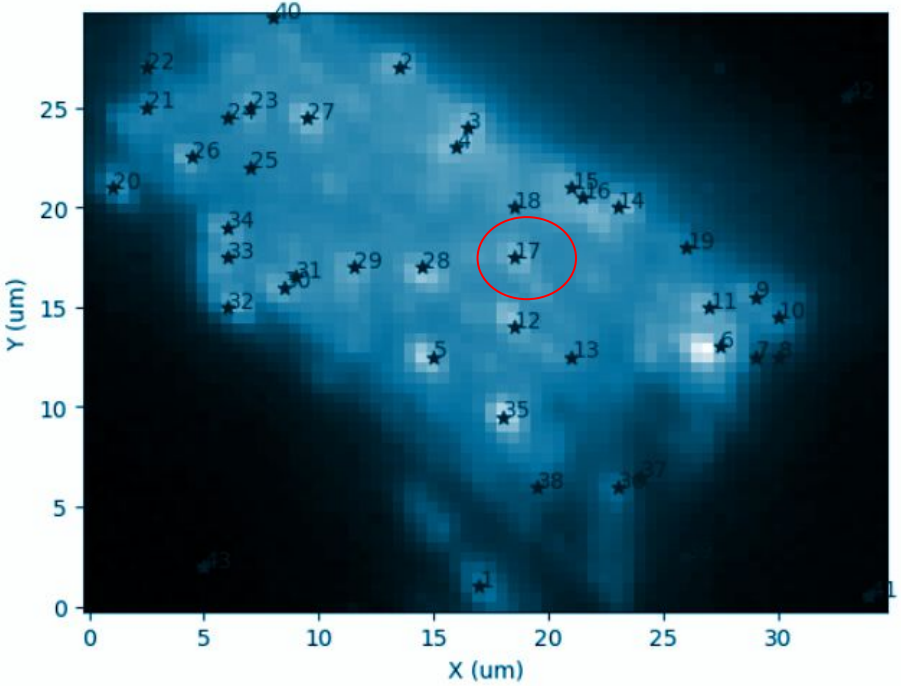
Do the spectra of the same points peak at the same wavelengths between the samples? How does the presence of silver nanocubes impact the measurements?

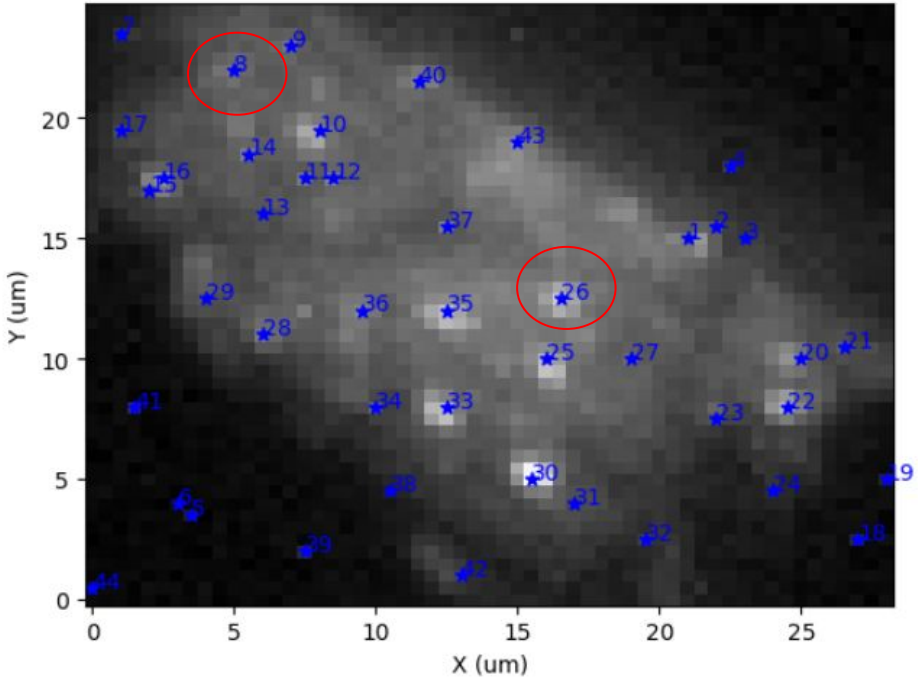
It seems like some peaks existed in both measurements between the same gold nanocube points. There may be some odd interactions between the gold and silver cubes.

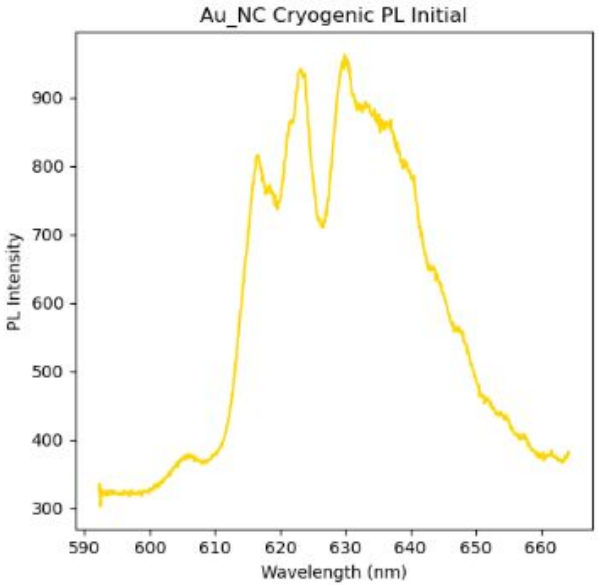
In general, how do the peaks differ between the gold and silver nanocubes? Does either provide more obvious enhancement?

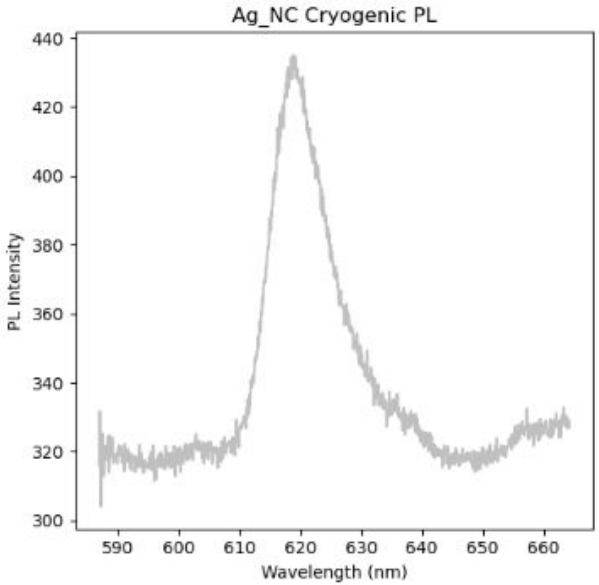
The gold nanocubes without any influence from the silver caused the highest peaks in the spectrum to be in the 615-620 nm range.

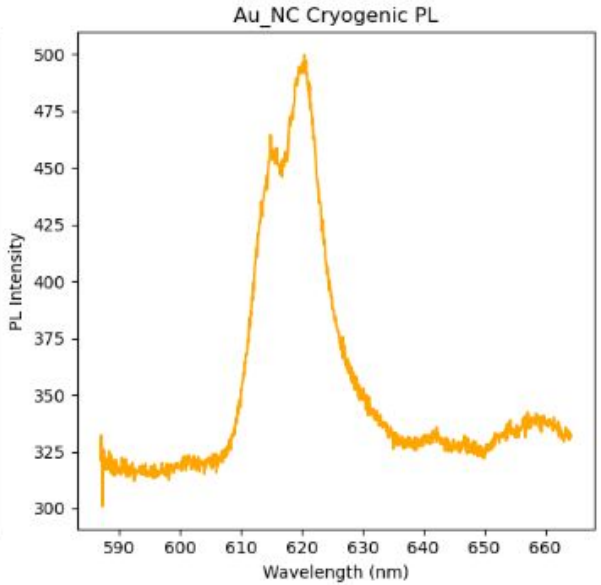
Overall, it appears the silver candidates have cleaner spectra and more obvious peaks than the gold nanocubes did. With the current data, there's no discernable difference in whether or not there's more obvious enhancement between the silver and gold nanocubes.

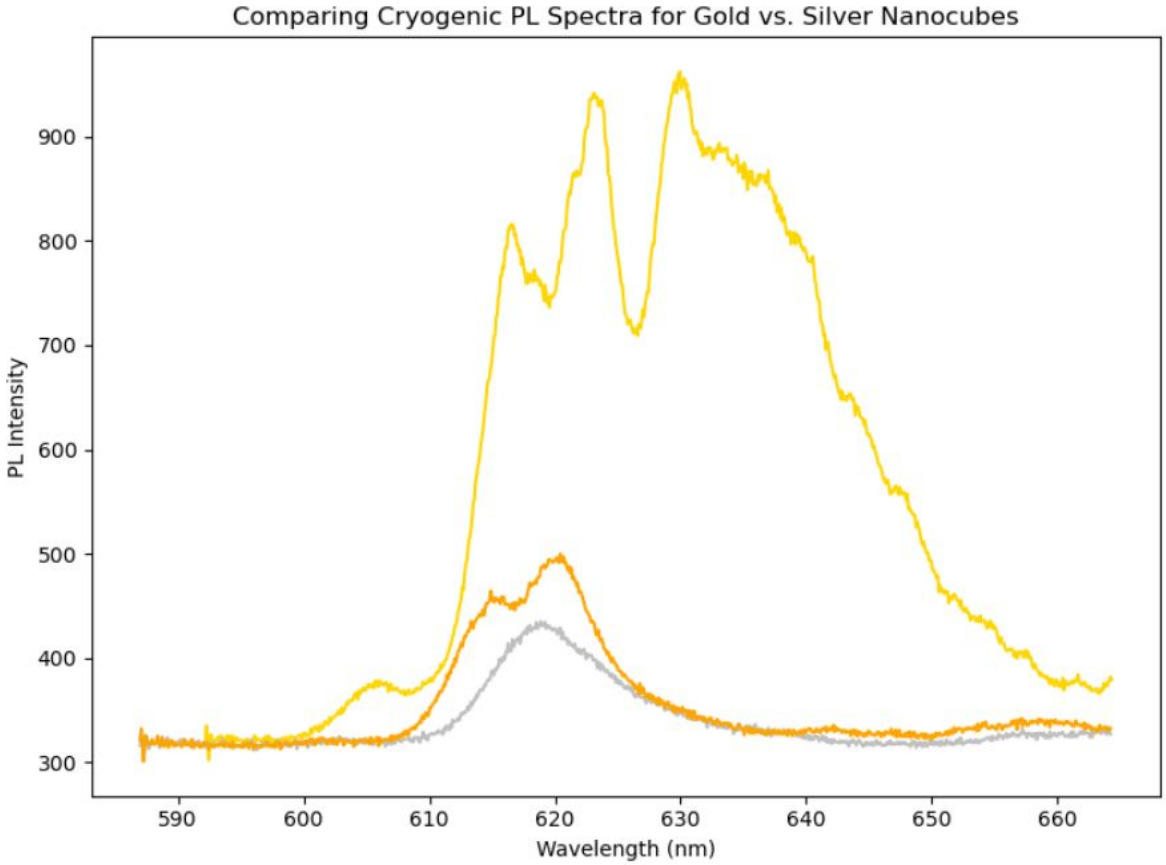

Y (um)
X (um)
0
5
10
15
20
25
30


Y (um)
X (um)
0
5
10
15
20
25


Au_NC Cryogenic PL Initial
PL Intensity
Wavelength (nm)
300
400
500
600
700
800
900
590
600
610
620
630
640
650
660


Ag_NC Cryogenic PL
PL Intensity
Wavelength (nm)
300
320
340
360
380
400
420
440
590
600
610
620
630
640
650
660


Au_NC Cryogenic PL
PL Intensity
Wavelength (nm)
300
325
350
375
400
425
450
475
500
590
600
610
620
630
640
650
660


Comparing Cryogenic PL Spectra for Gold vs. Silver Nanocubes
PL Intensity
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)