Enhancement Due to Photonic Coupling in Nanocavity Structures

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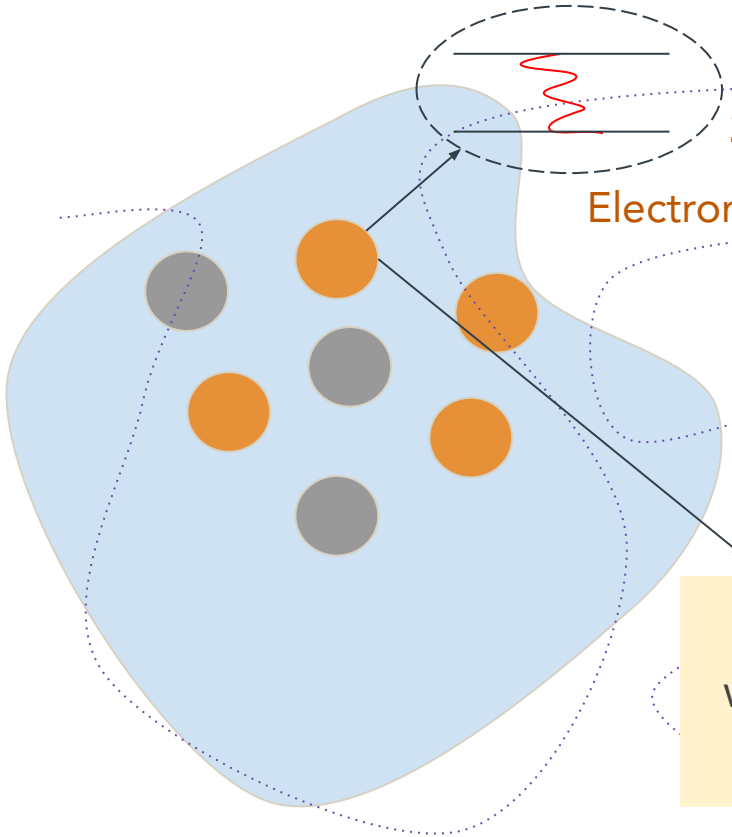
Background

How do quantum emitters work?



2-level Quantum emitter

Electromagnetic environment

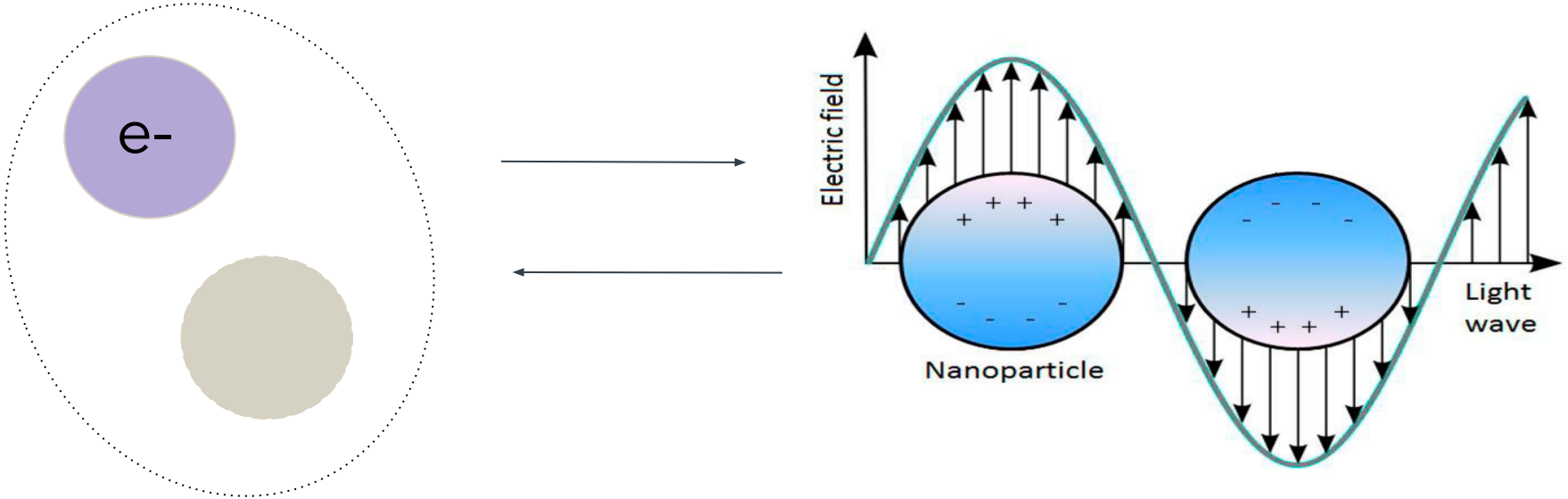
Optical properties!

Spontaneous photon emission!

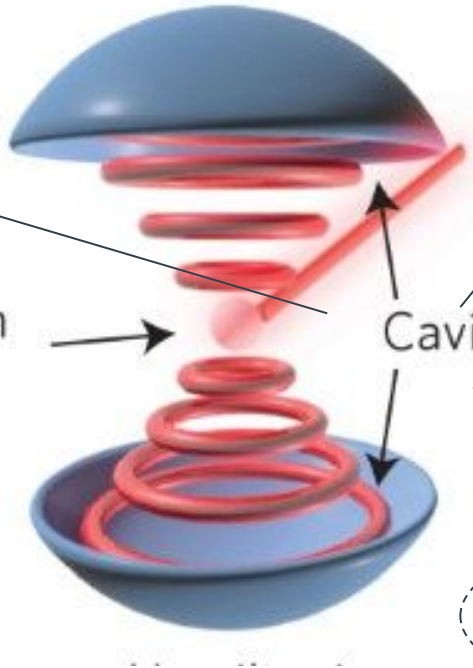
How can we alter this environment to favorably influence the decay rate of the system?

Multiple 2-level systems with differing polarization or wavelength

Why can a nanoscale quantum emitter can have emitting behavior?



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 --- The weak coupling of a 2-level emitter to a cavity leads to the modification of the spontaneous emission rate. The emitter can have multiple radiative modes and some nonradiative processes

- emission redirection --- the 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 $\eta _{NA}$. Different modes can have different efficiencies.

- absorption enhancement --- cavities alter the absorption cross-section of the emitter which causes absorption enhancement and lowers the saturation power needed from the pump laser.

Coupling parameters:

- emitter's decay in free space $\Gamma _0$\\
- cavity's loss rate κ\\
- emitter-cavity coupling coefficient g\\

This is dependent 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.

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 $F = \frac{3}{4\pi ^2} Q \frac{\lambda ^3}{V}$ (Purcell factor - represents the shortening of lifetimes; losses ensure that it is not directly related to the number of emitted photons), $Q \approx \frac{1}{\kappa}$ (cavity quality factor), and the cavity mode volume $\frac{V}{\lambda ^3}$.

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

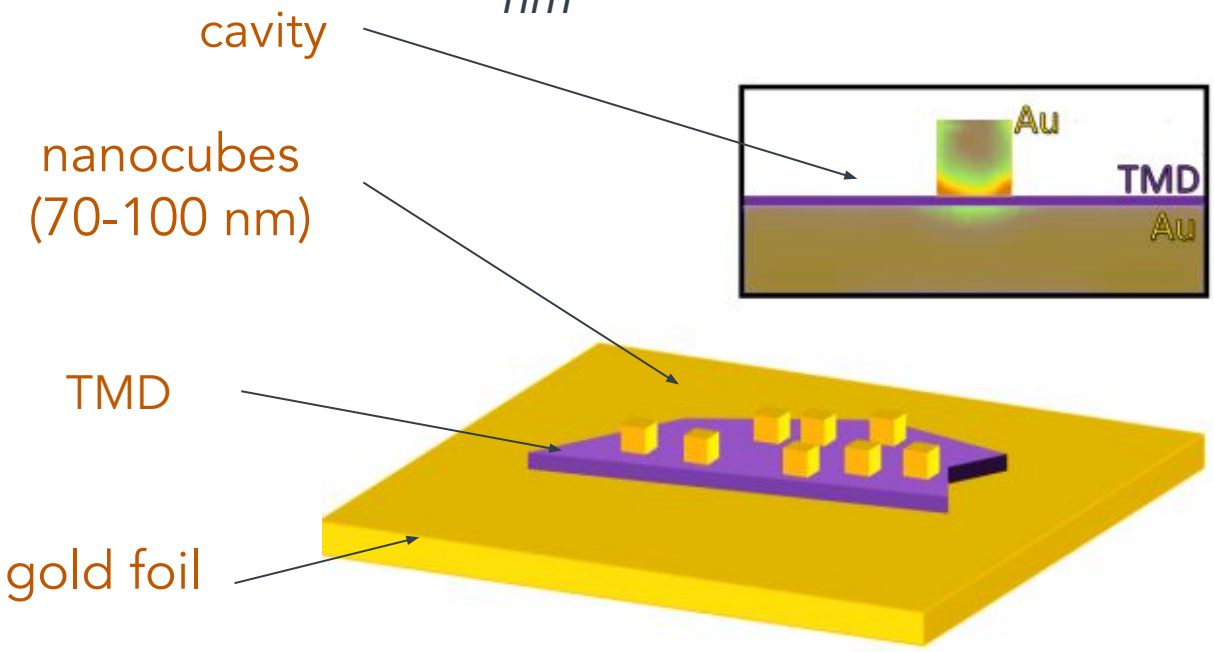
Sample 1 : Gold Nanocube

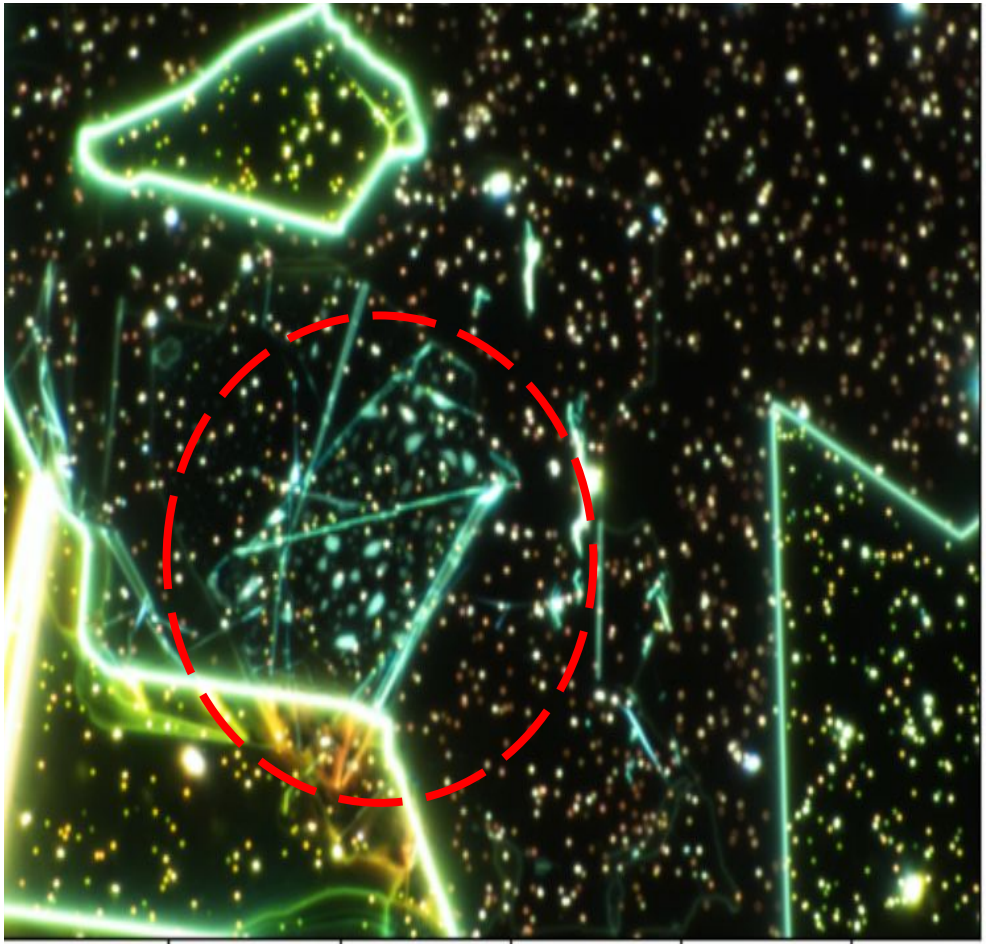
Sample 1: Gold Nanocube : Optical Data

The lightning-rod effect causes the corners of the cubes to be the regions of greatest enhancement

Material: *Monolayer TMD*

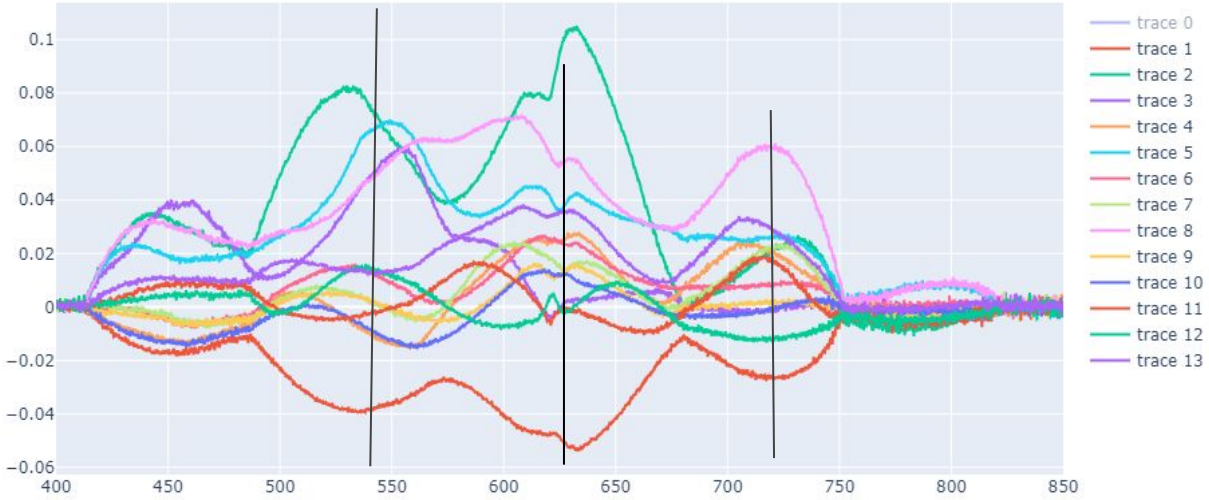
Film-Cube *Separation: 8-9 nm*





Darkfield Spectroscopy

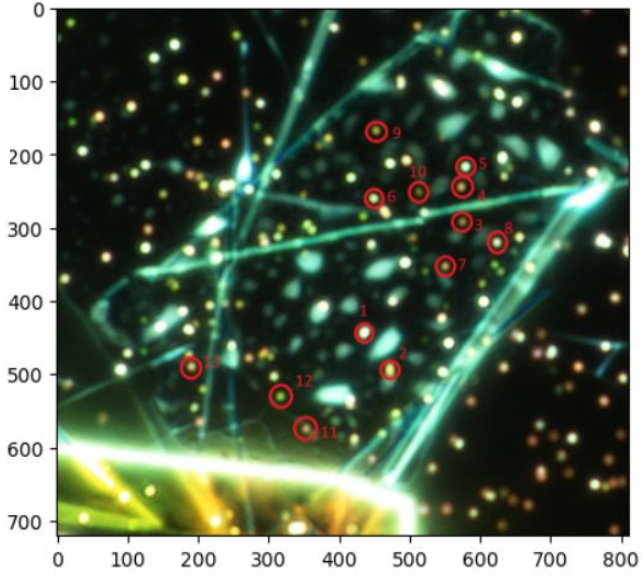
- white light scattering by nanocavities
- measure plasmonic resonances



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

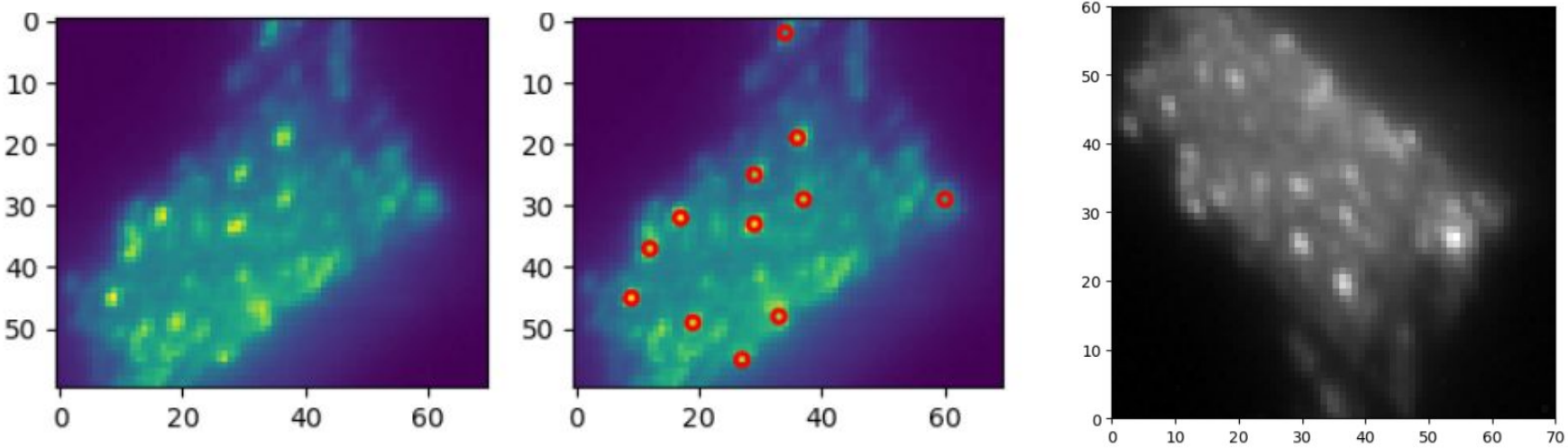
- 540 nm
- *625 nm !!!*
- 725 nm

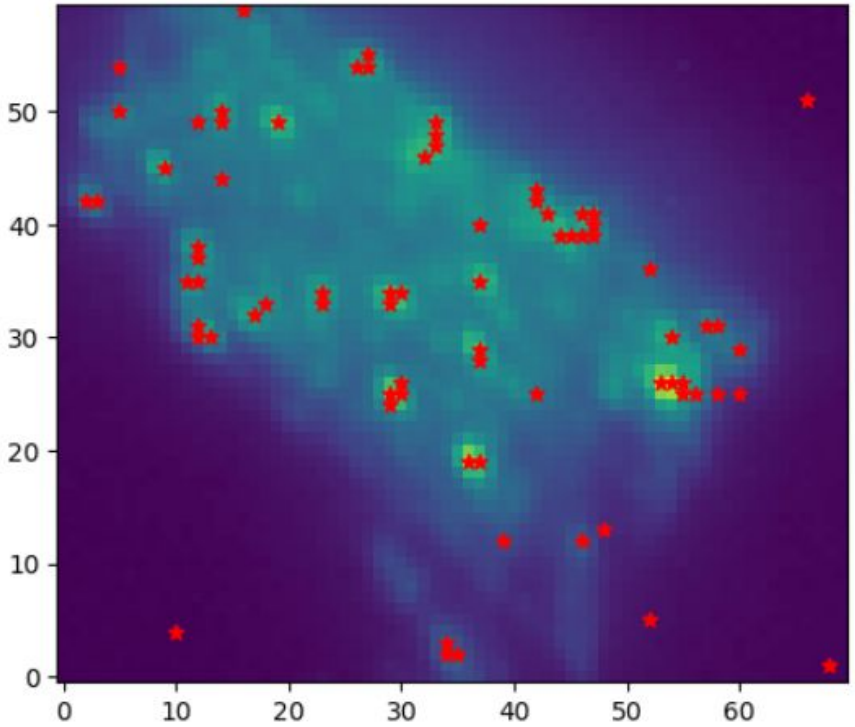
Labelled points on the darkfield image

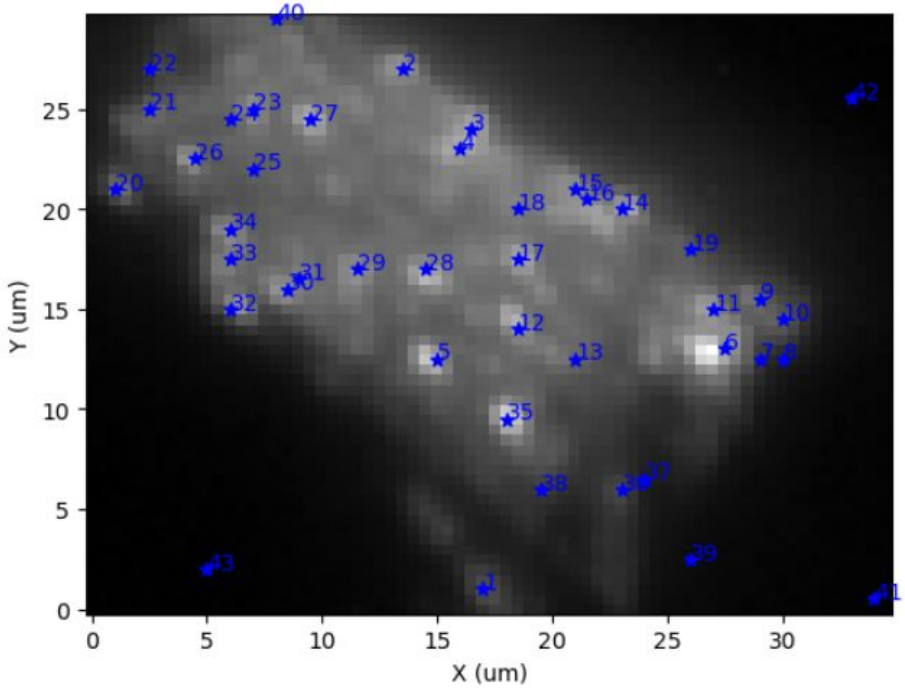


Hyperspectral Spatial Image

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

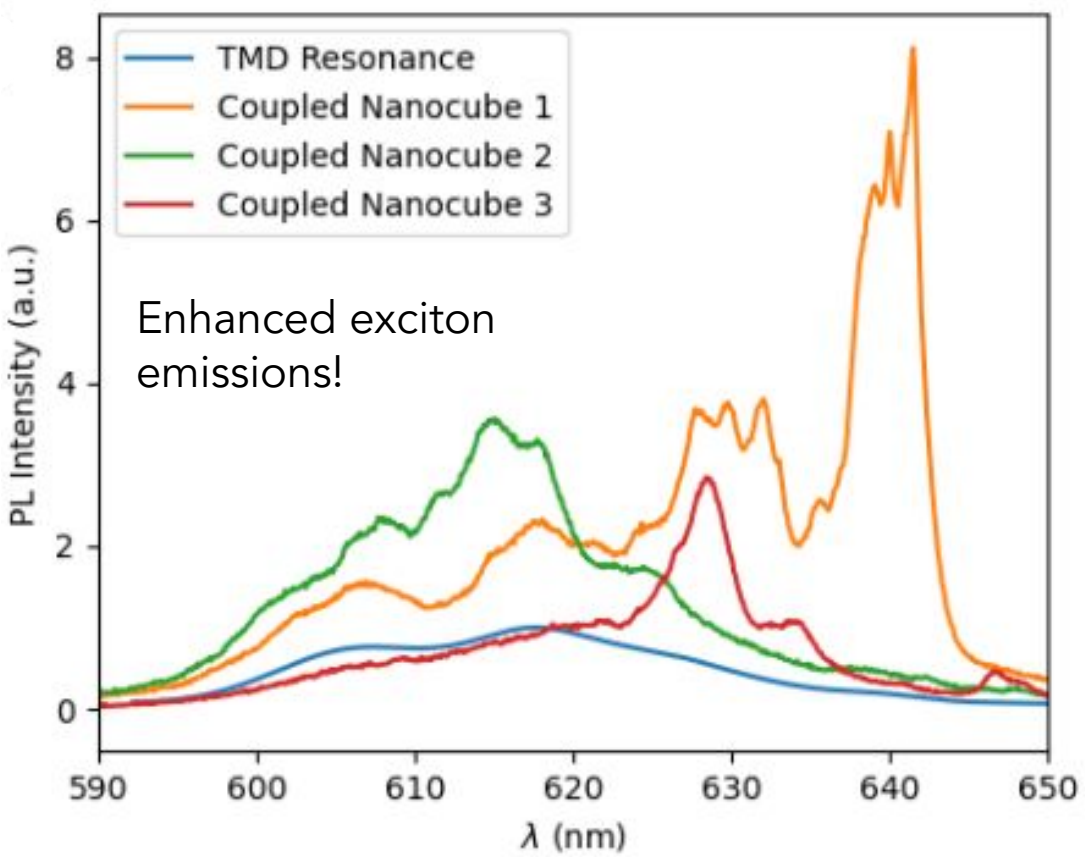


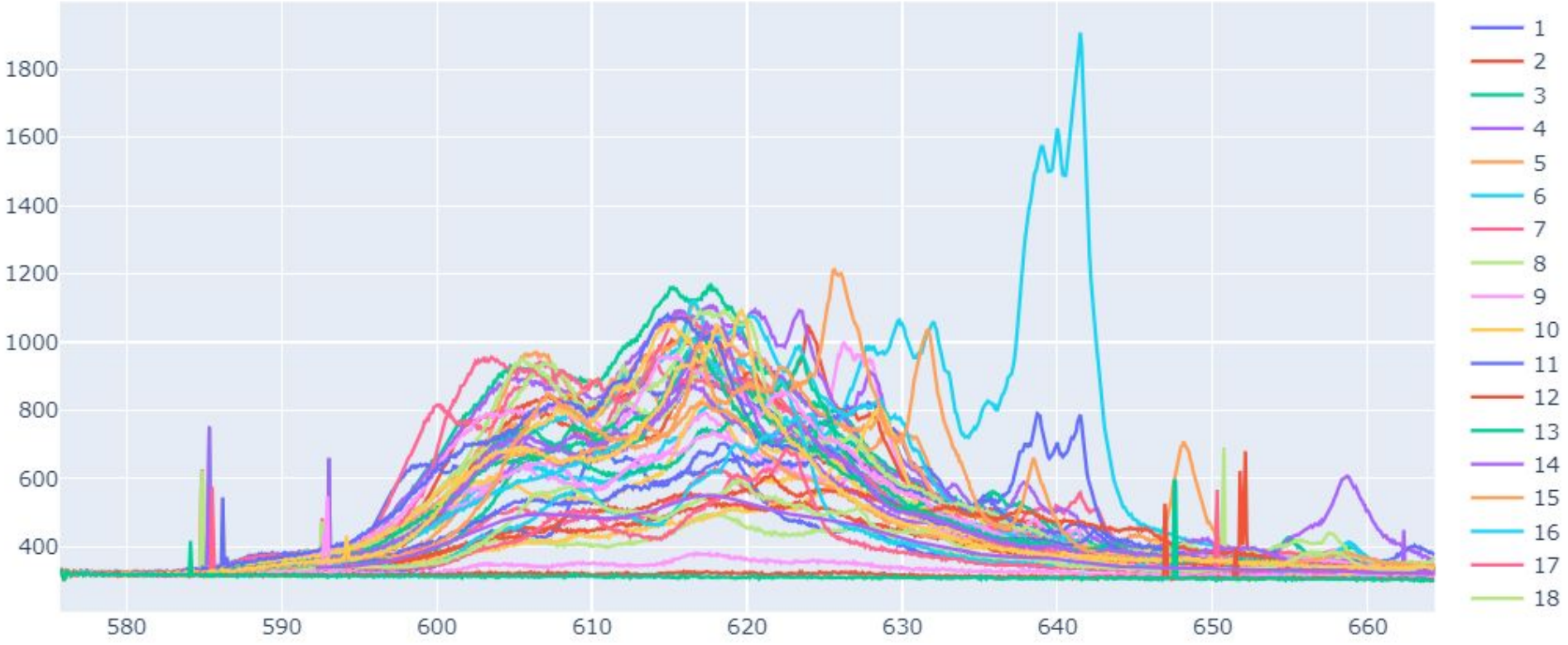

0
10
20
30
40
50
60


Y (um)
X (um)
0
5
10
15
20
25
30
40
22
2
42
21
24
23
27
3
26
25
20
15
16
18
14
34
19
33
31
29
28
17
30
11
9
32
12
10
6
5
13
35
38
37
39
43
1
41

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




1800
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610
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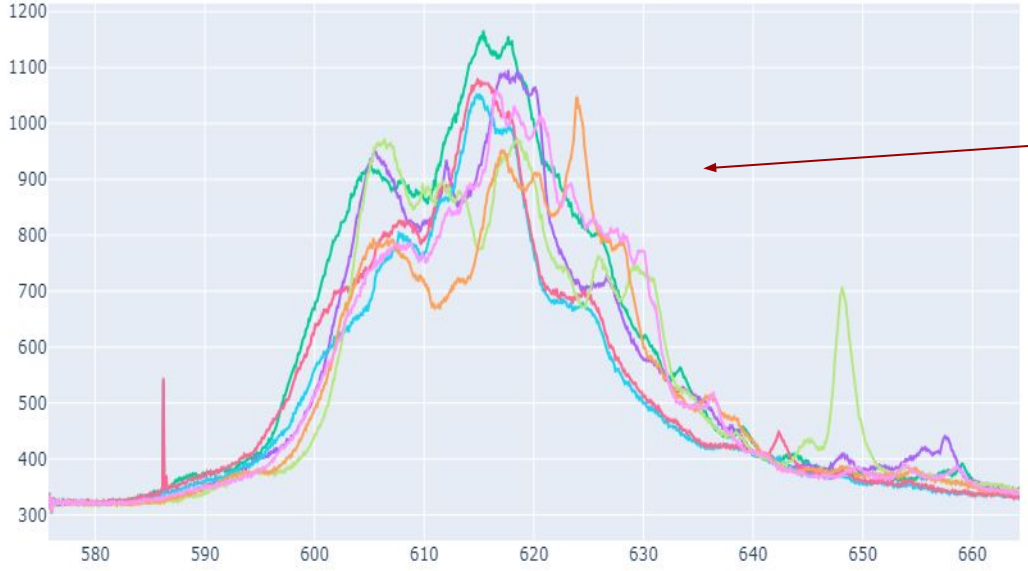
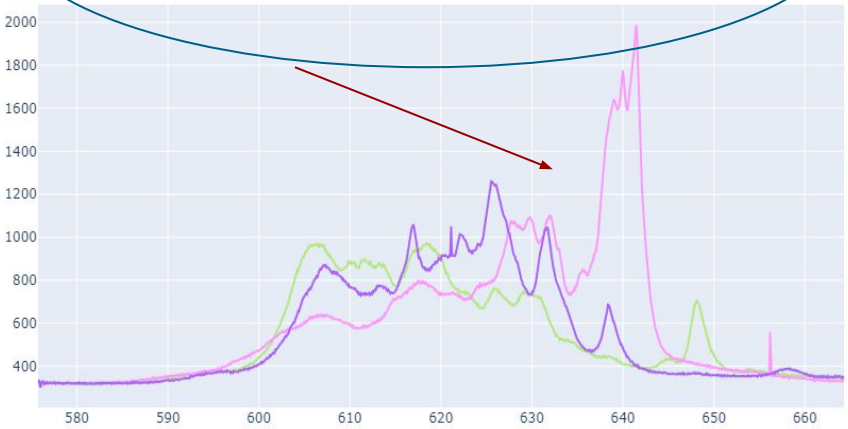
18



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

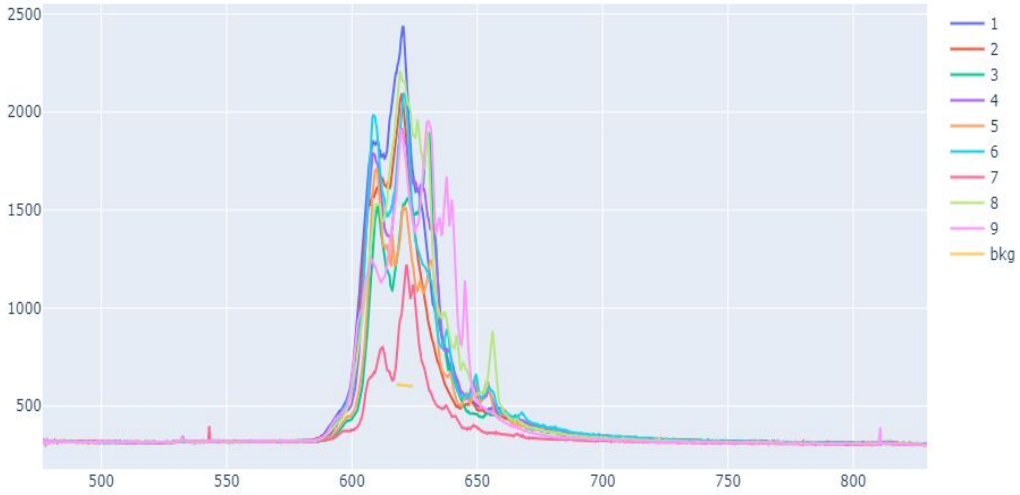
Sight trough at 608-611 nm and again at 620-625 nm

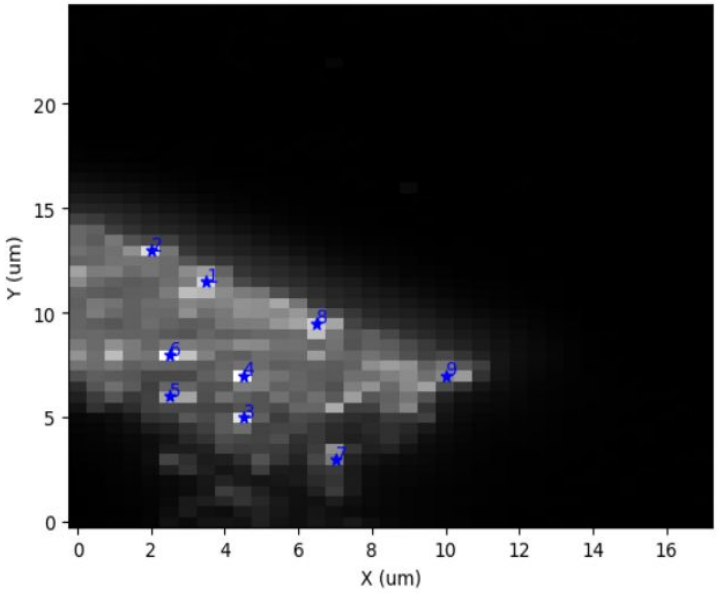
These were the brightest spots - less uniformity between them but we still have peaks at around 605, 630 , 640 and 615 nm



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

Room Temperature Higher Power PL





Peaks are largely concentrated at
610, 625, 640 nm

Background

Sample 1 : Gold Nanocube

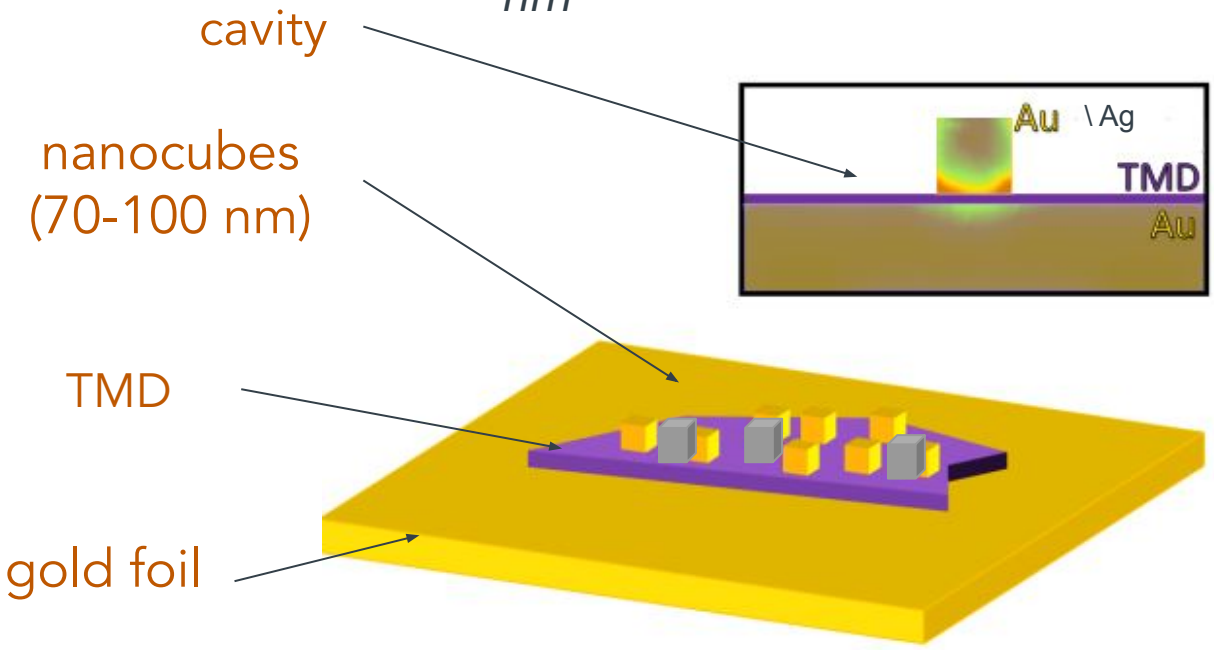
Sample 1 : Silver Nanocube

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.

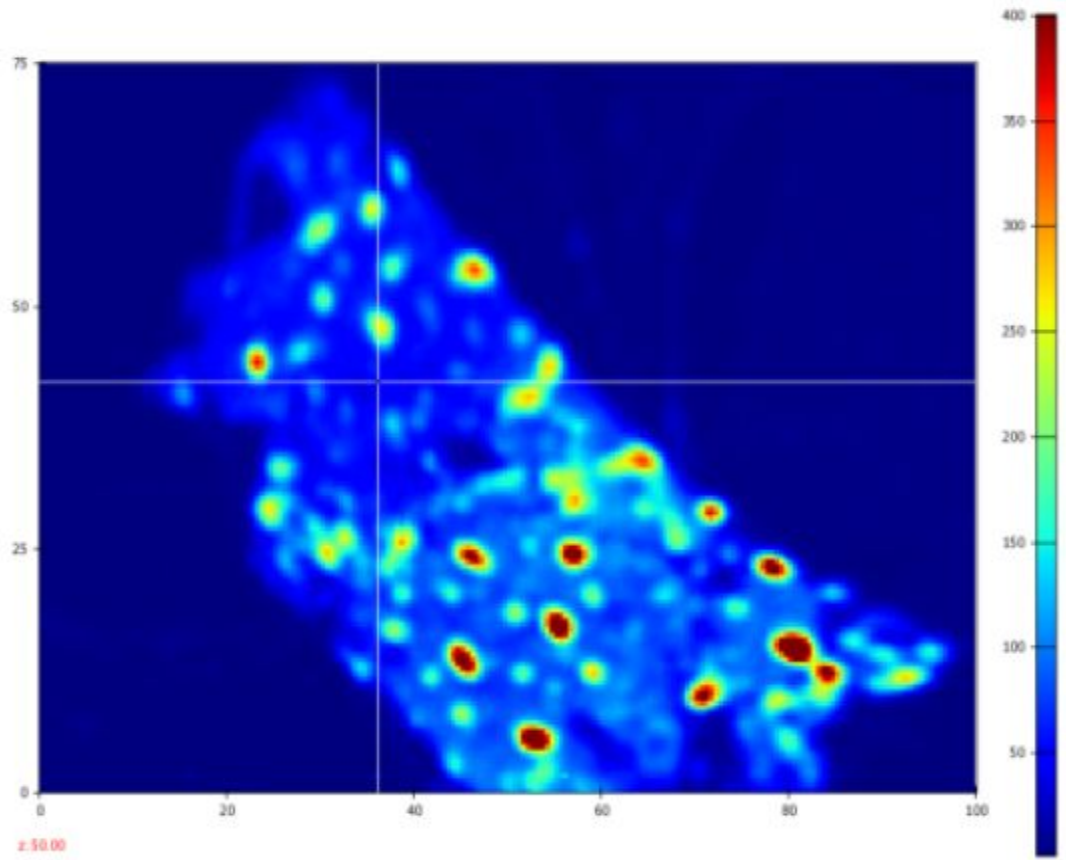
Material: *Monolayer TMD*

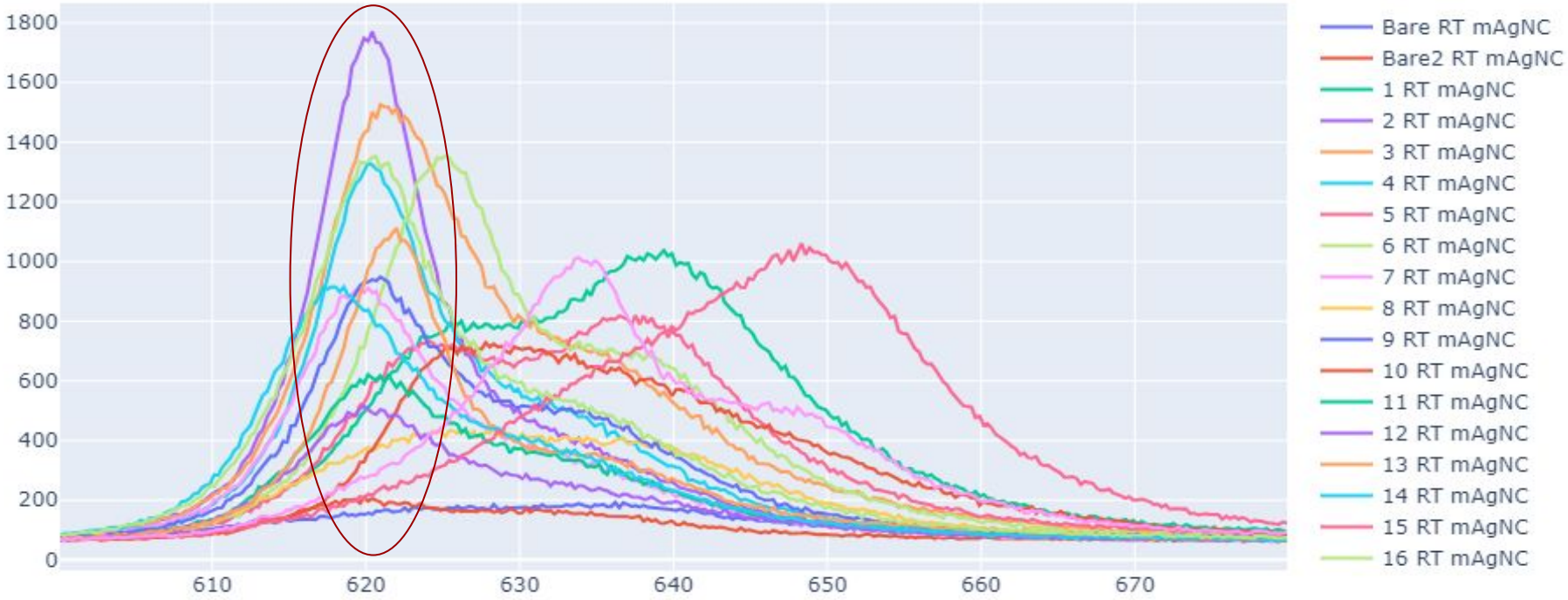
Film-Cube *Separation: 8-9 nm*



Photoluminescence (PL)

- Focus on each point individually to obtain a spectrum over a range of wavelengths
- This image is focused on a bare portion of the sample for control purposes




1800
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200
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620
630
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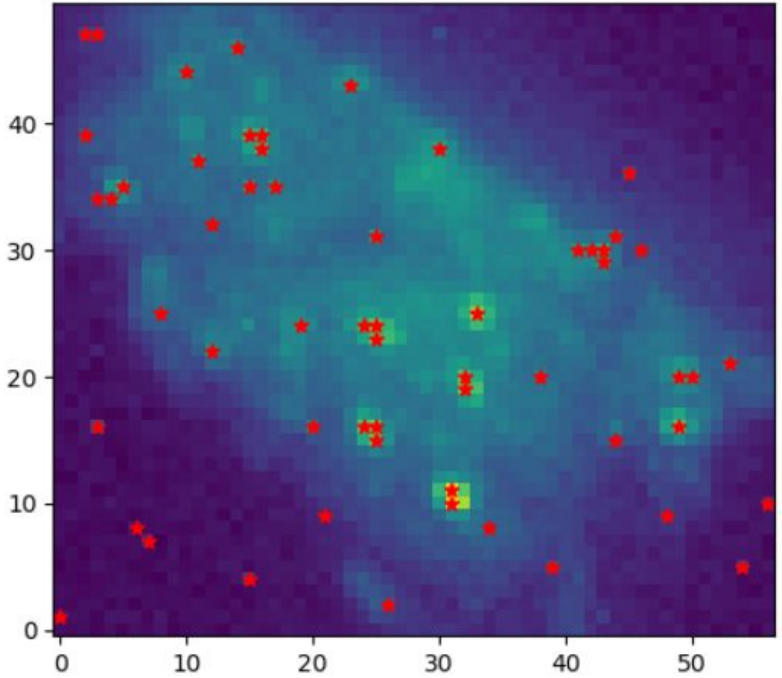
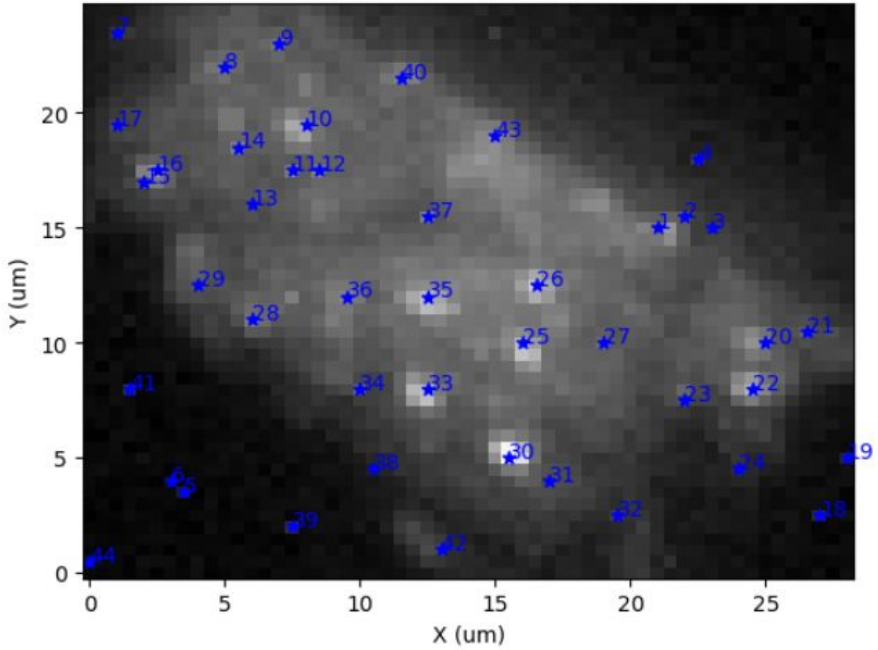
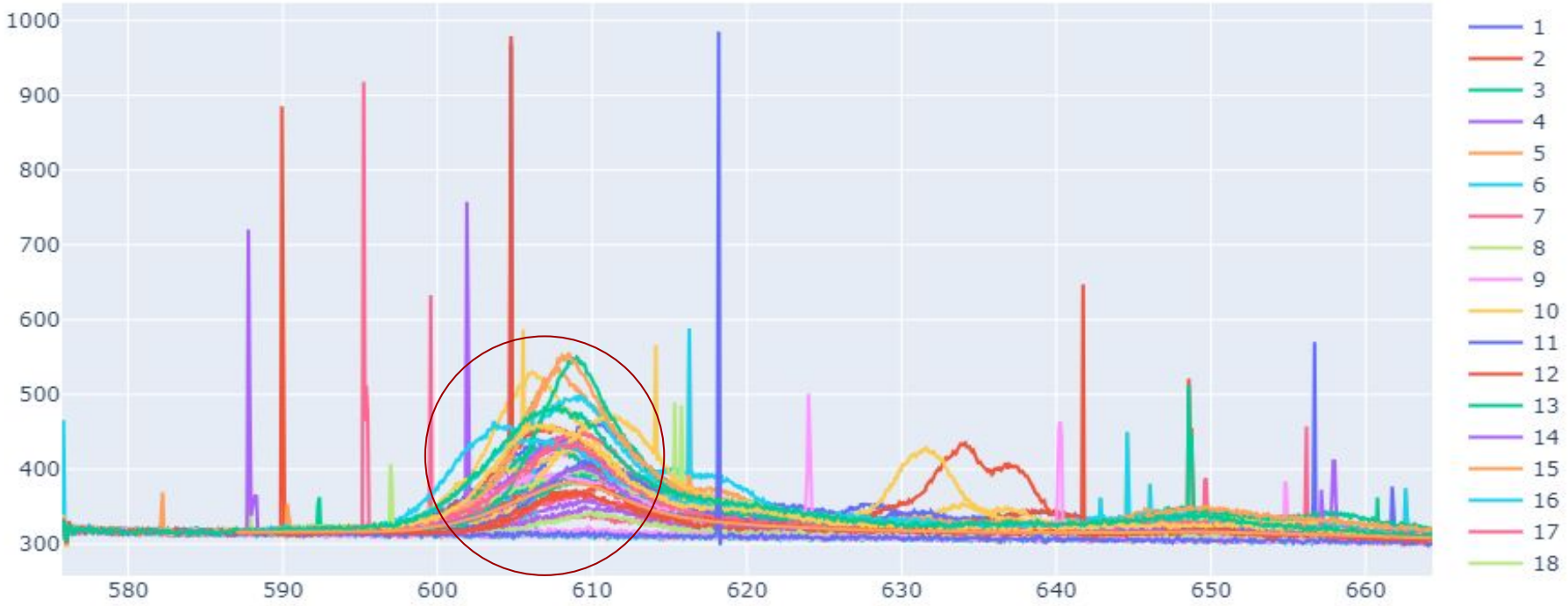
660
670
Bare RT mAgNC
Bare2 RT mAgNC
1 RT mAgNC
2 RT mAgNC
3 RT mAgNC
4 RT mAgNC
5 RT mAgNC
6 RT mAgNC
7 RT mAgNC
8 RT mAgNC
9 RT mAgNC
10 RT mAgNC
11 RT mAgNC
12 RT mAgNC
13 RT mAgNC
14 RT mAgNC
15 RT mAgNC
16 RT mAgNC

Hyperspectral Spatial Image - Cryogenic



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660
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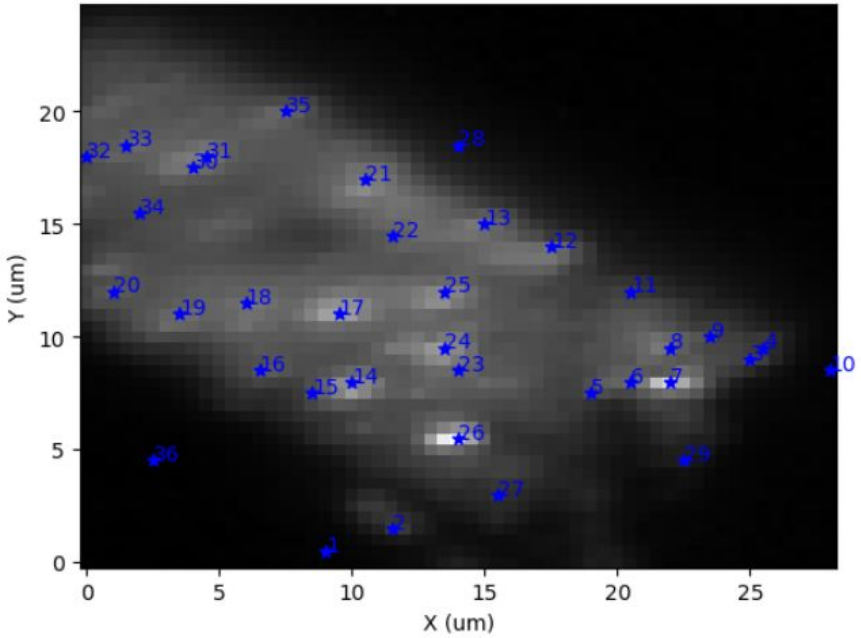
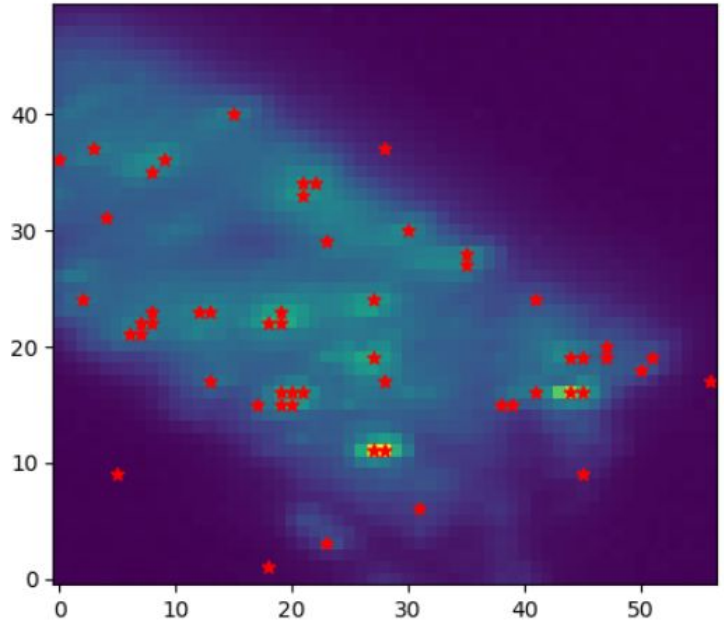
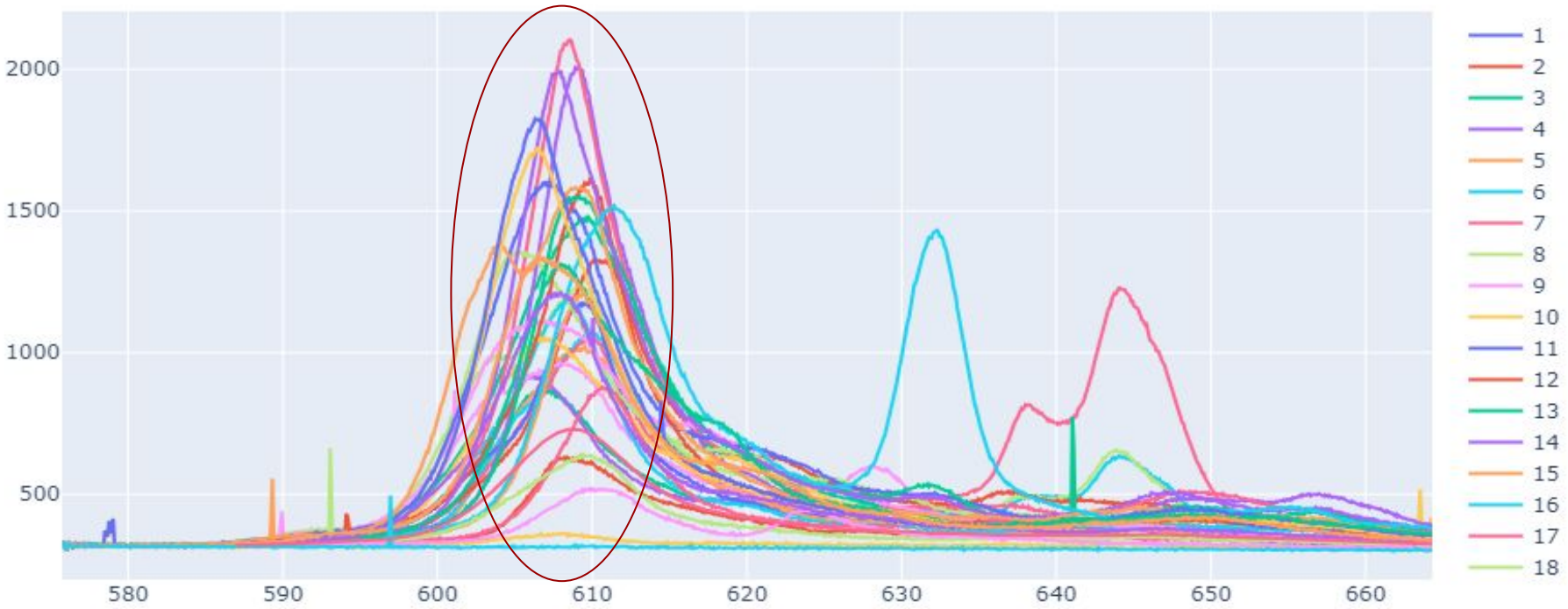
15
16
17
18

Hyperspectral Spatial Image - Cryogenic 20uW






2000
1500
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650
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4
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17
18

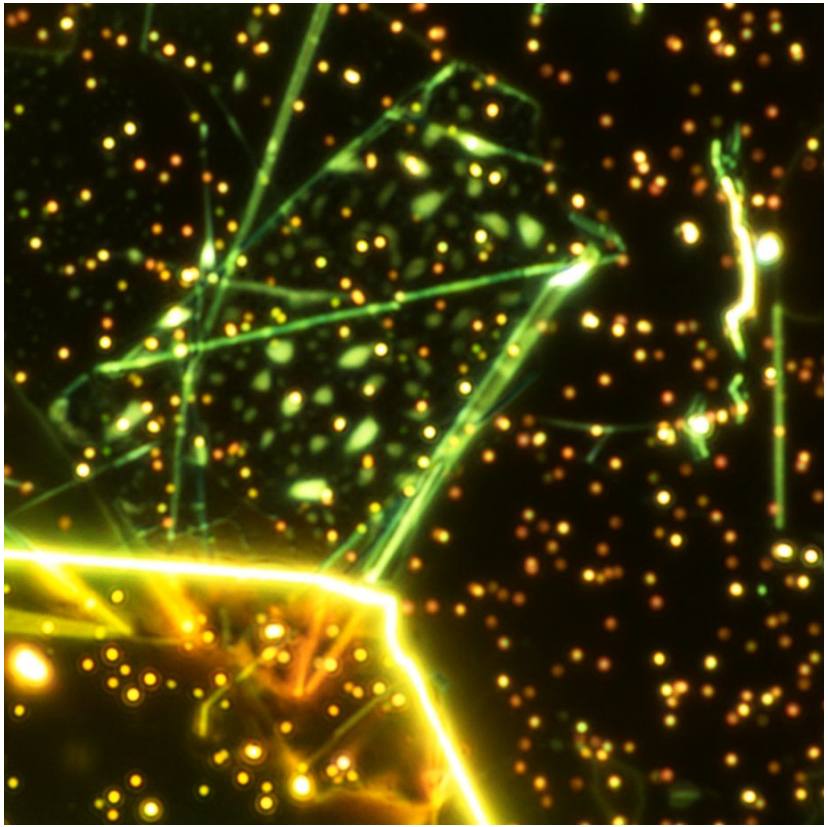
Comparing Results

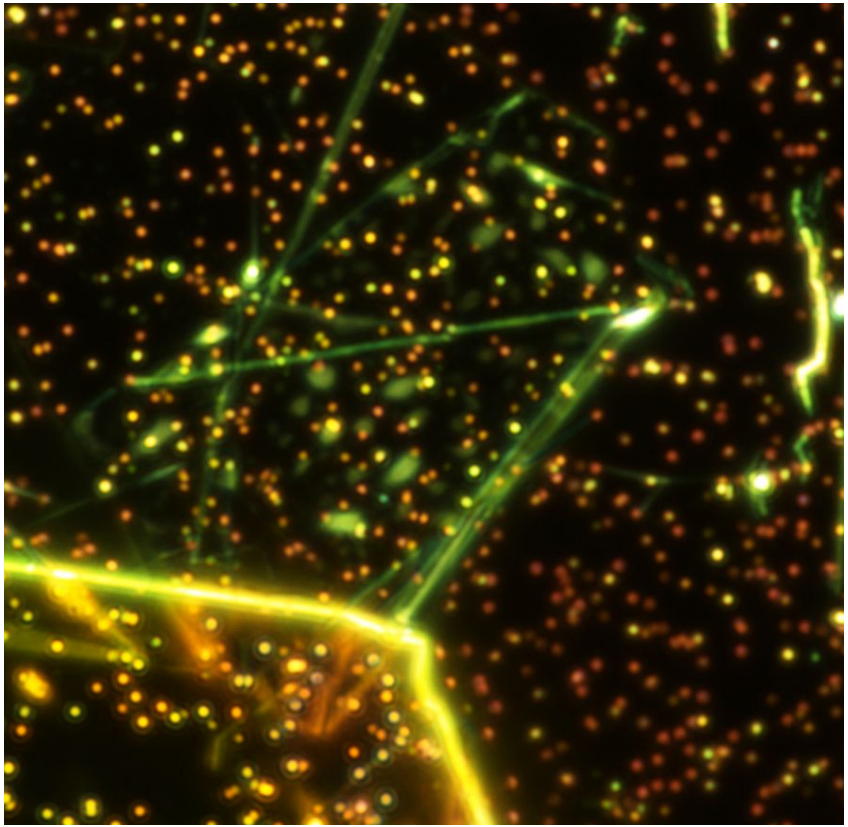
Do the spectra of the same points peak at the same wavelengths between the samples? How does the presence of silver nanocubes impact the measurements?

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



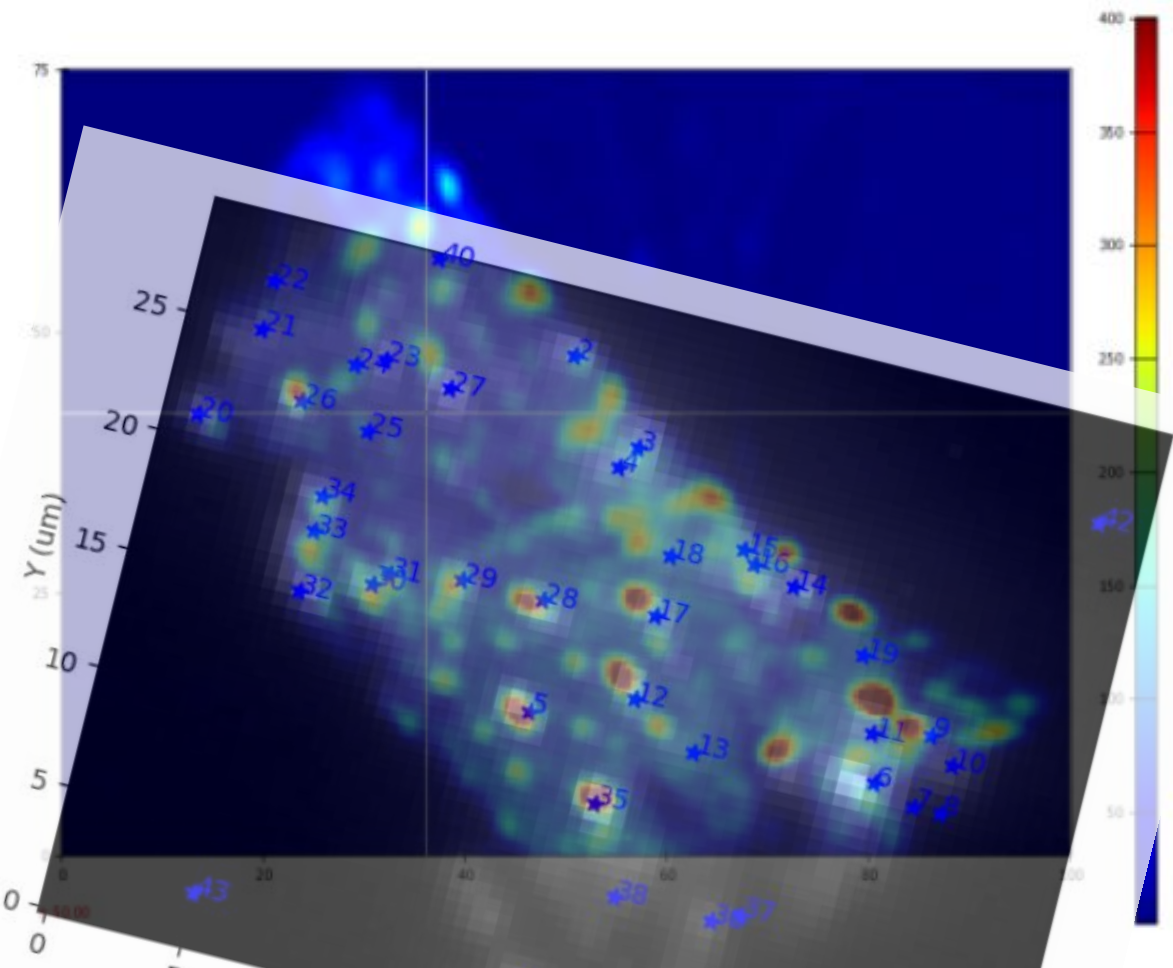


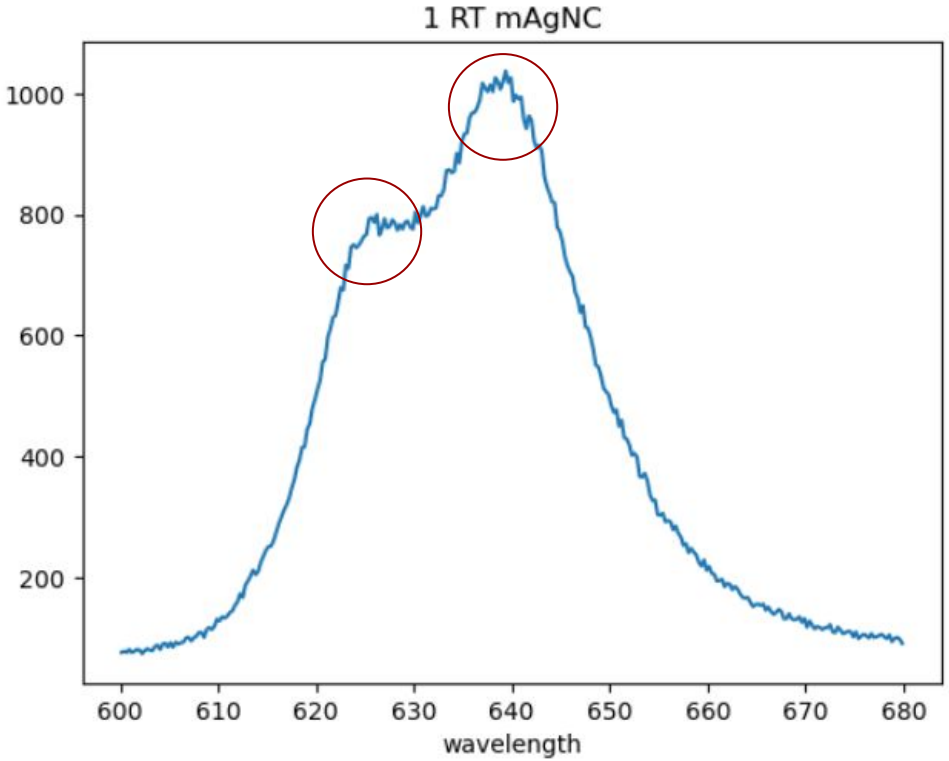
Room Temperature PL Gold v. Silver

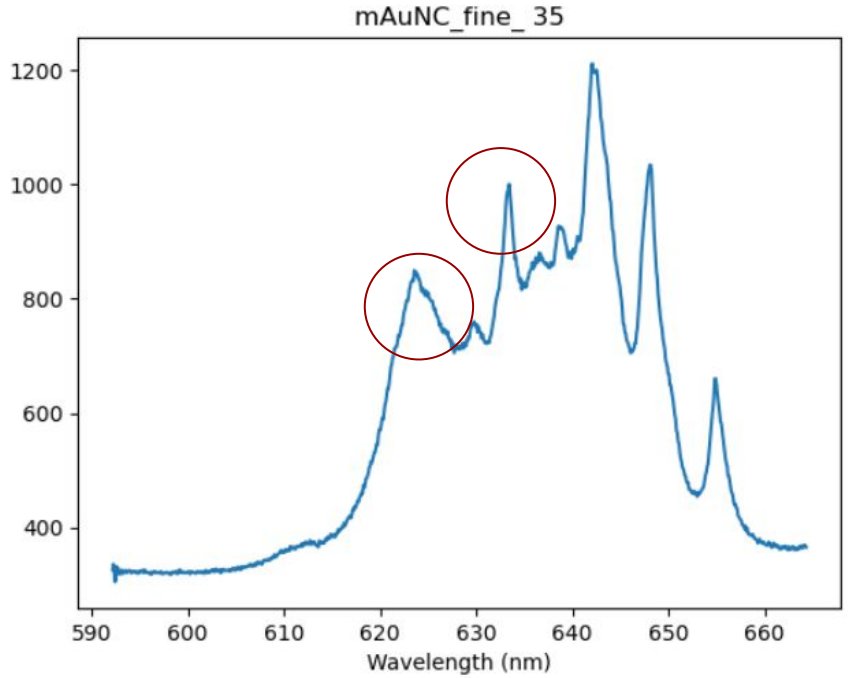
It appears that the correlations between the new and old points are as follows:

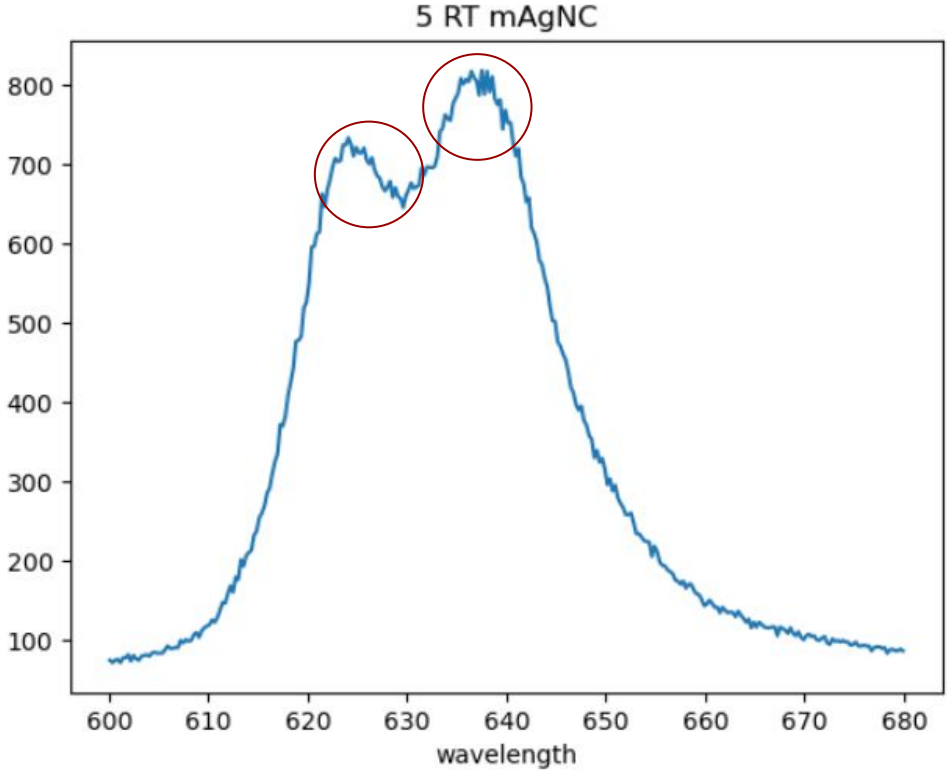
1 - 35
4 - 11
5 - 5
6 - 12
8 - 28
10 - 14
11 - 30
12 - 29
15 - 26

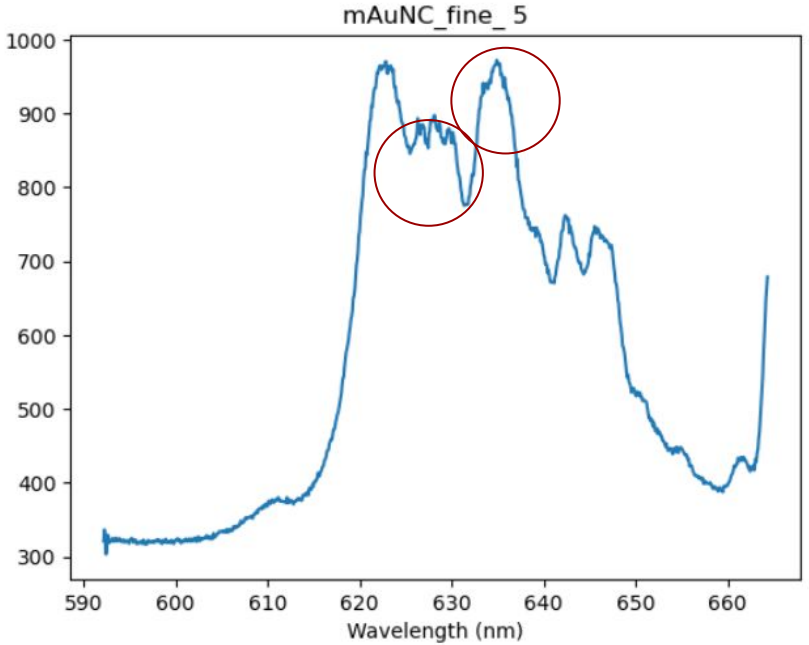
So we will compare the plots. Notice how the spectra will look very different but still preserve the location of certain peaks.

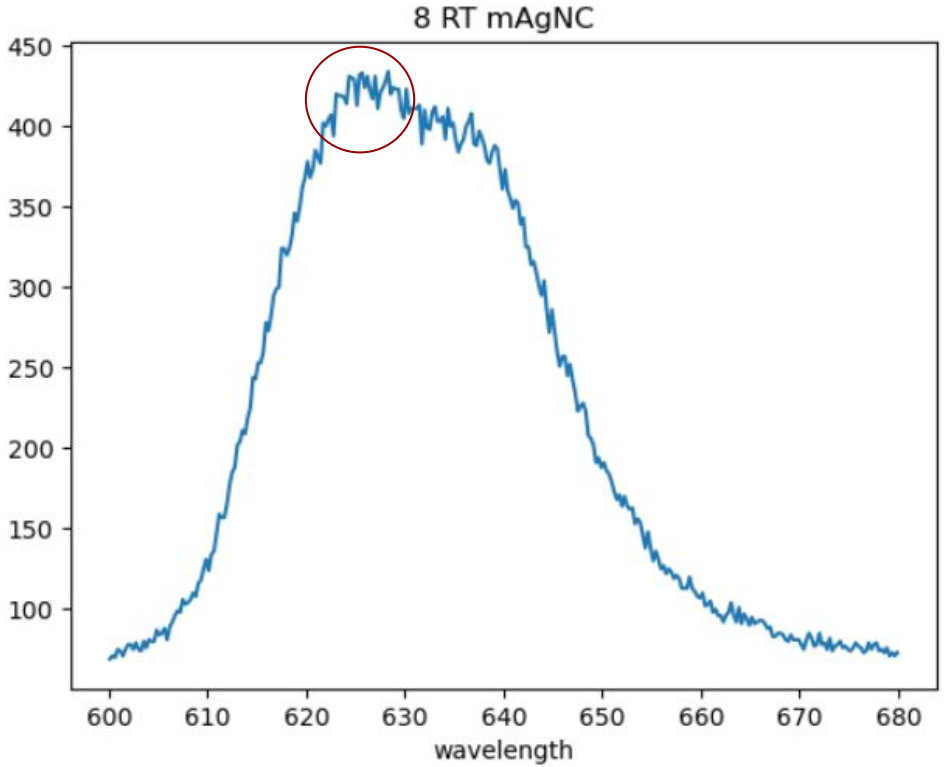


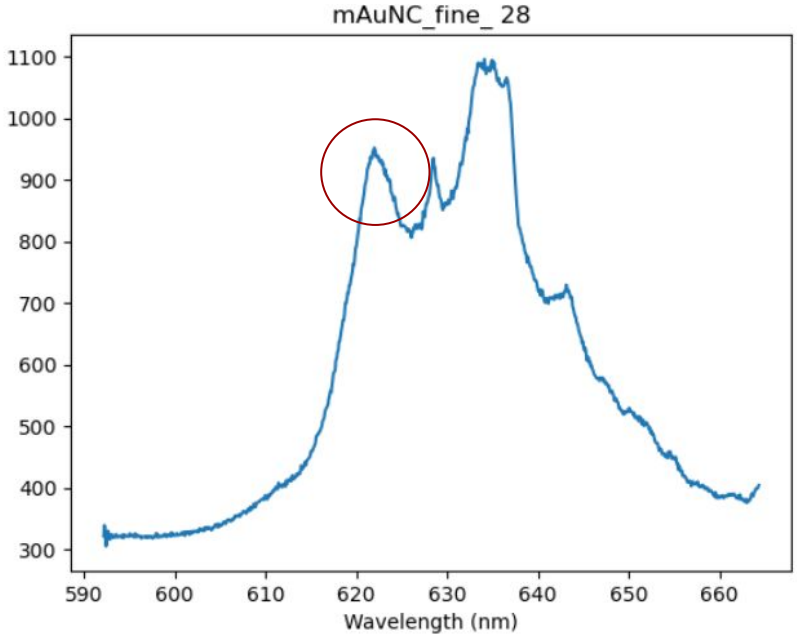

1 RT mAgNC
1000
800
600
400
200
600
610
620
630
640
650
660
670
680
wavelength

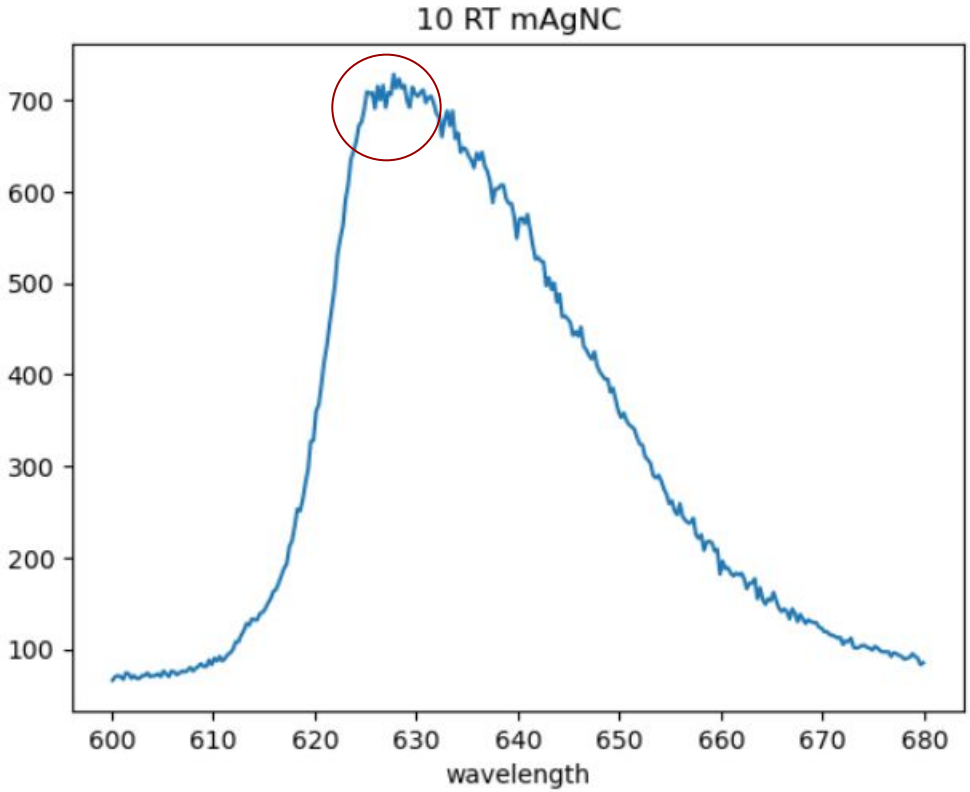

mAuNC_fine_ 35
1200
1000
800
600
400
590
600
610
620
630
640
650
660
Wavelength (nm)

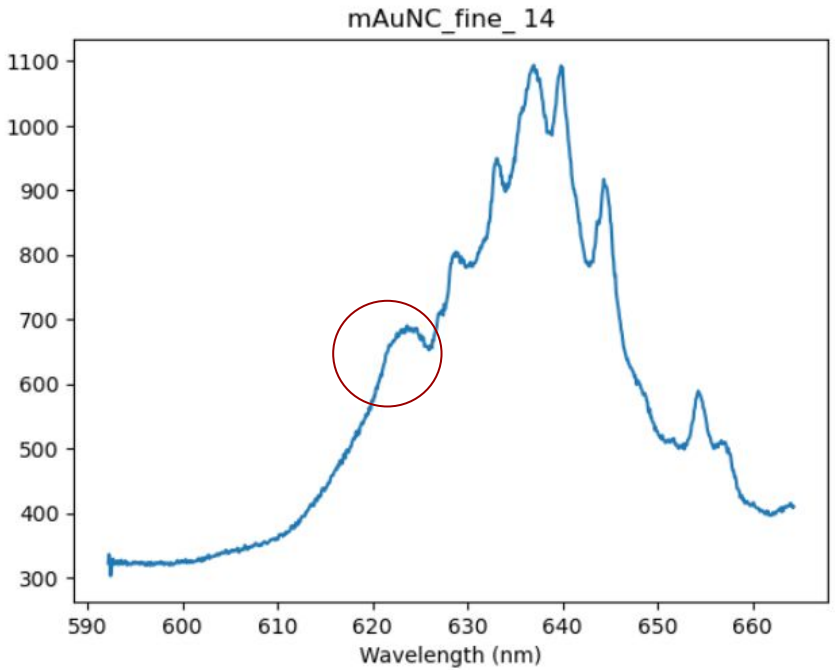

5 RT mAgNC
800
700
600
500
400
300
200
100
600
610
620
630
640
650
660
670
680
wavelength

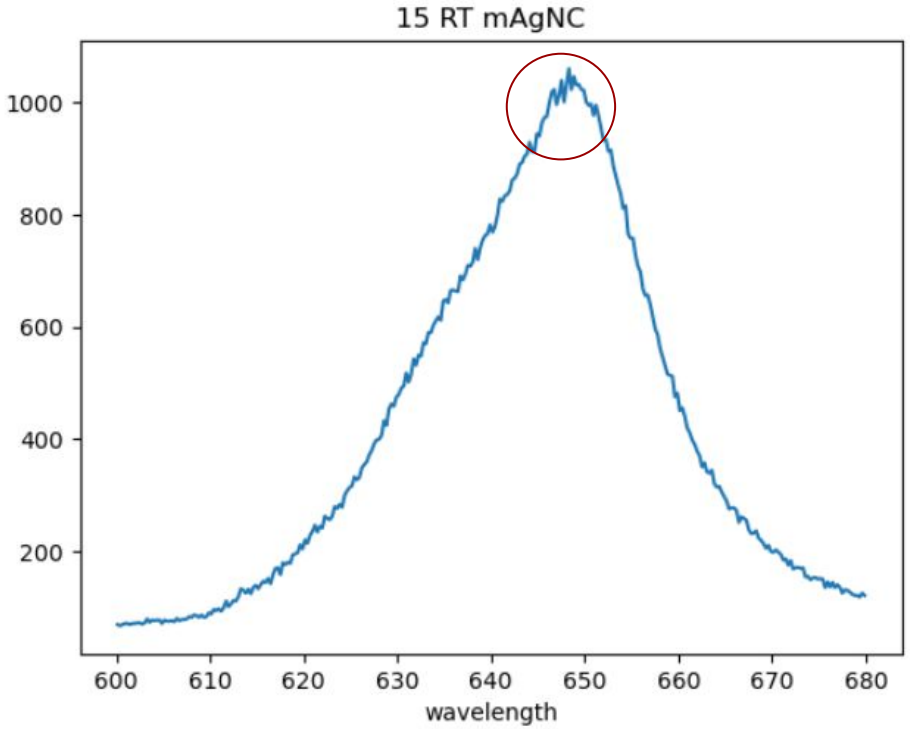

mAuNC_fine_ 5
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

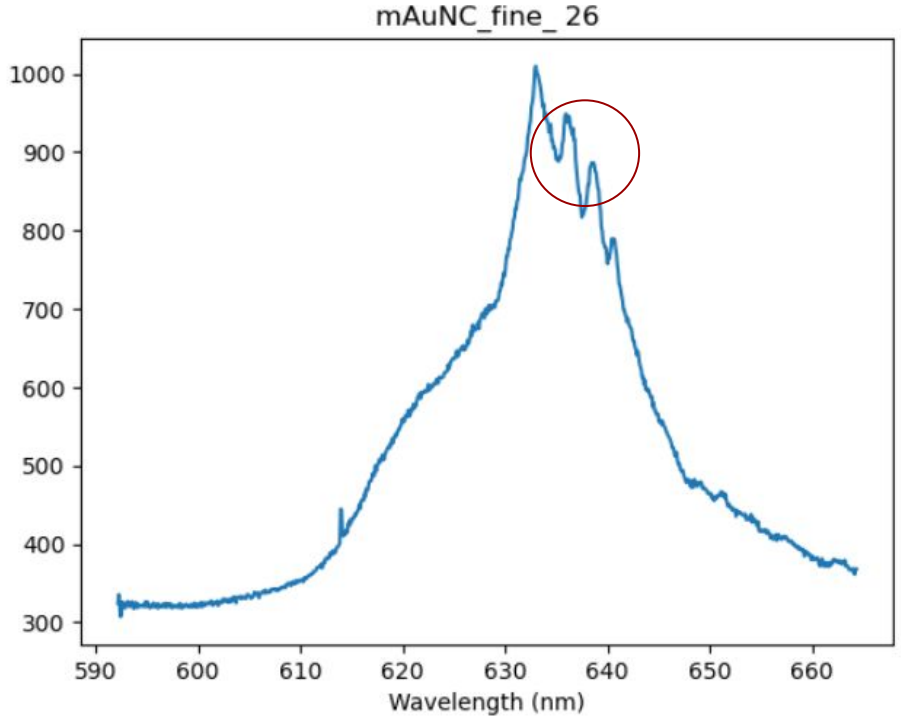

8 RT mAgNC
450
400
350
300
250
200
150
100
600
610
620
630
640
650
660
670
680
wavelength


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

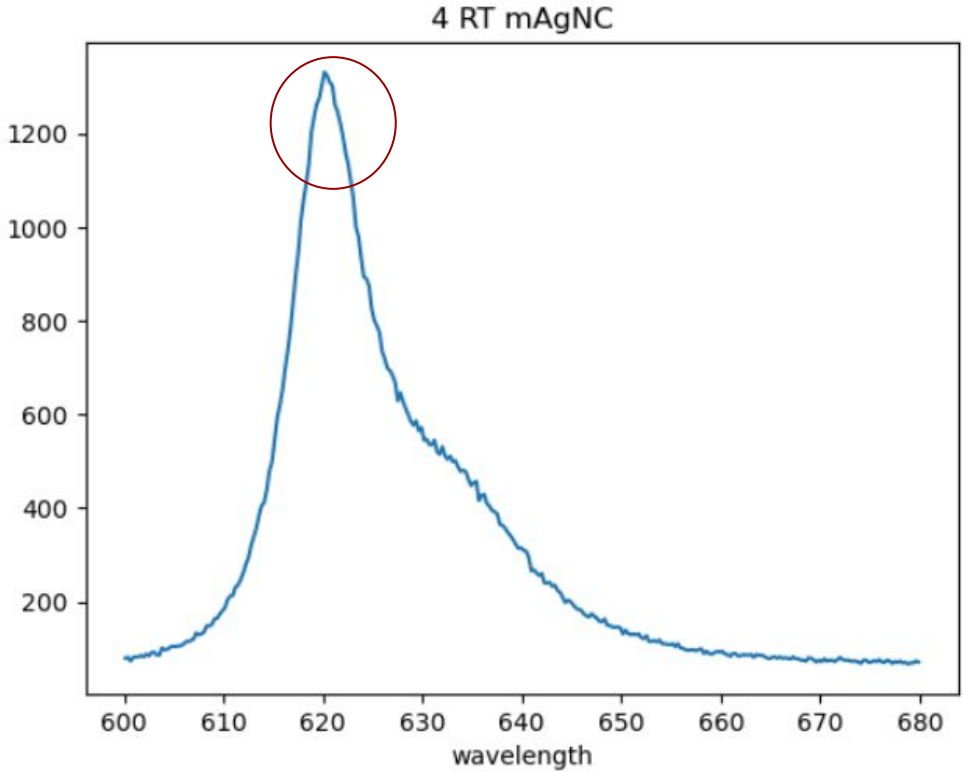

10 RT mAgNC
700
600
500
400
300
200
100
600
610
620
630
640
650
660
670
680
wavelength

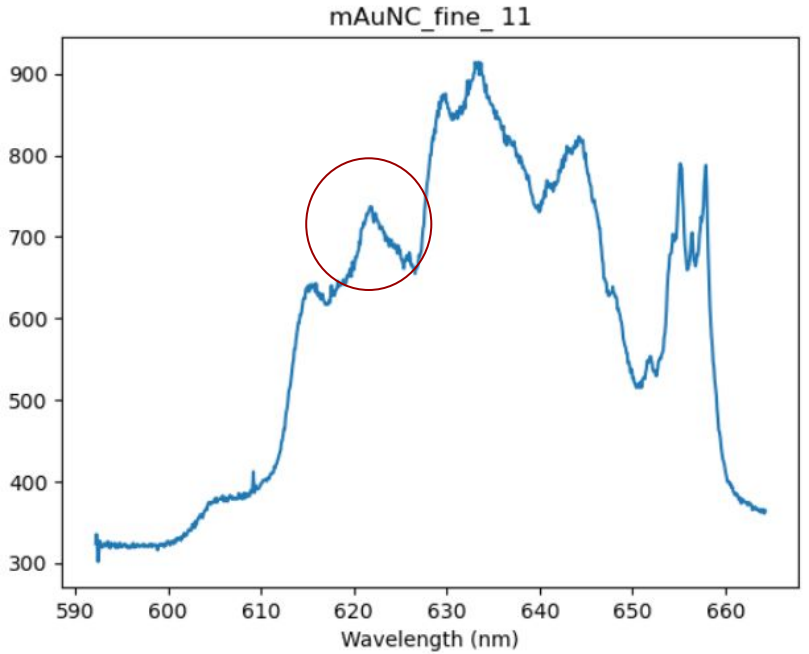

mAuNC_fine_ 14
1100
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)


15 RT mAgNC
1000
800
600
400
200
600
610
620
630
640
650
660
670
680
wavelength


mAuNC_fine_ 26
1000
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

The following comparisons are interesting because the PL spectrum from the points resemble each other, but the correlated points from prior measurements have even more drastically different spectra. They also resemble uncorrelated points…


4 RT mAgNC
1200
1000
800
600
400
200
600
610
620
630
640
650
660
670
680
wavelength


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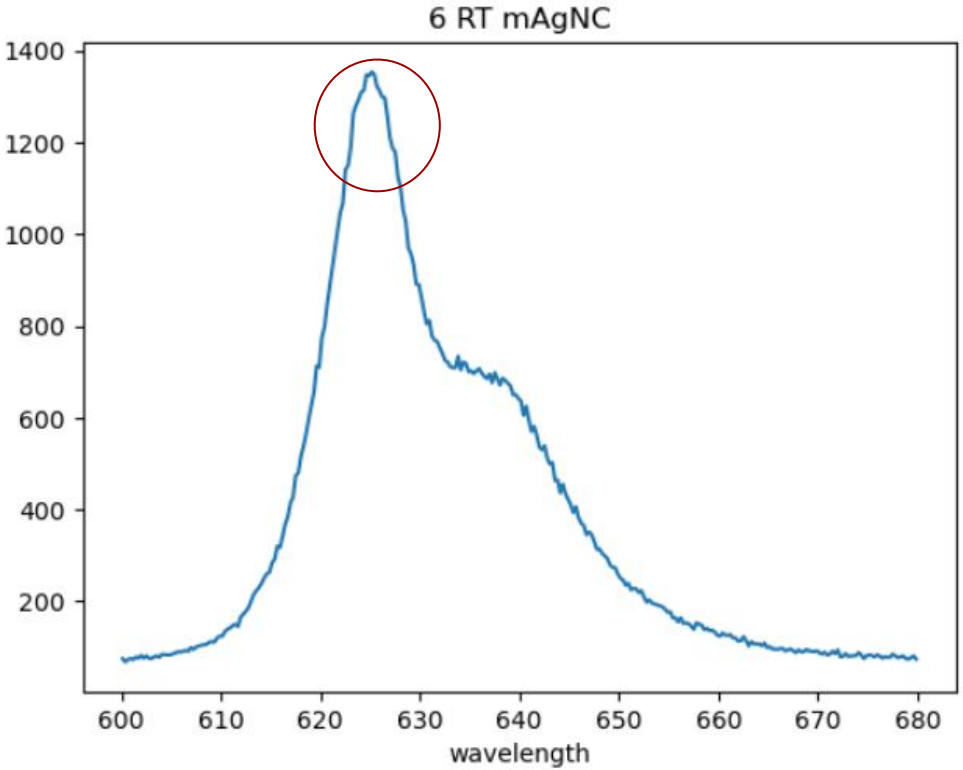
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)


6 RT mAgNC
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1200
1000
800
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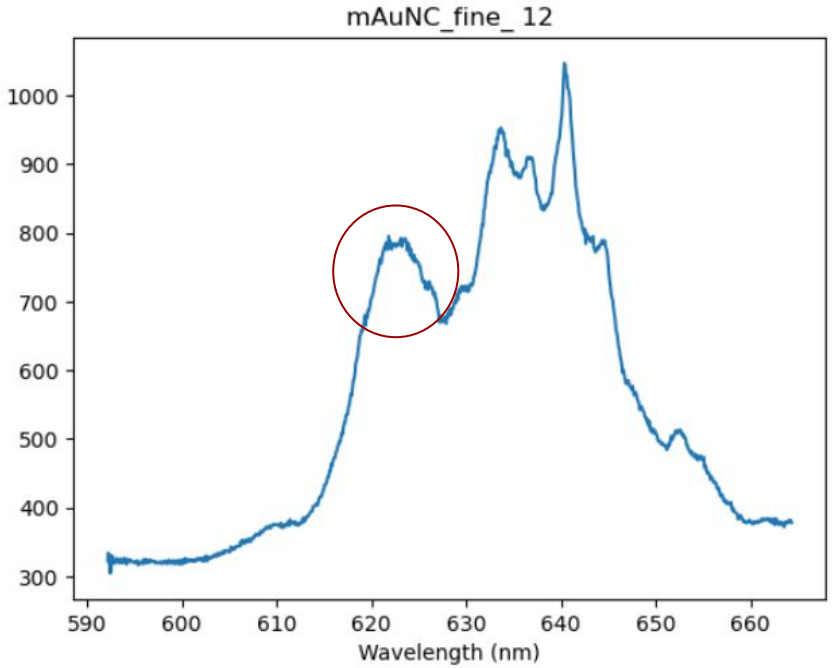
400
200
600
610
620
630
640
650
660
670
680
wavelength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600
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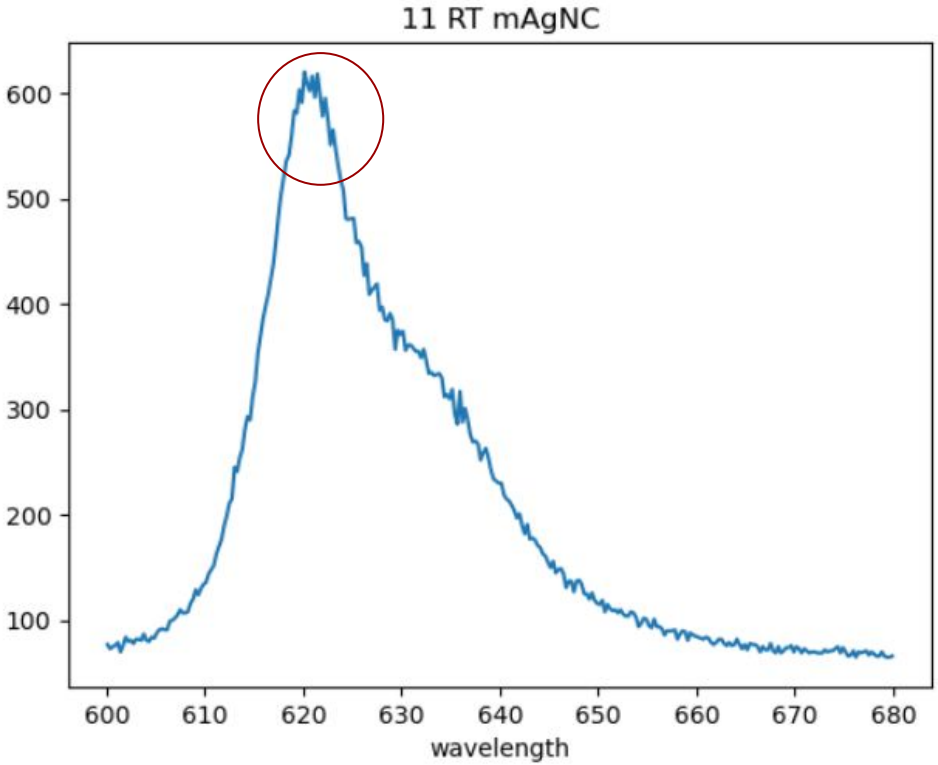
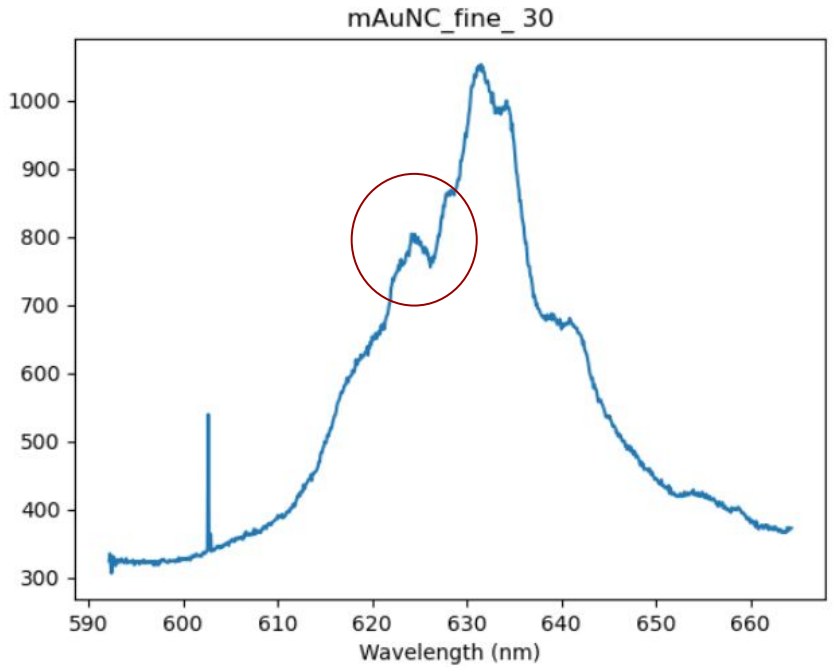
650
660
Wavelength (nm)


11 RT mAgNC
600
500
400
300
200
100
600
610
620
630
640
650
660
670
680
wavelength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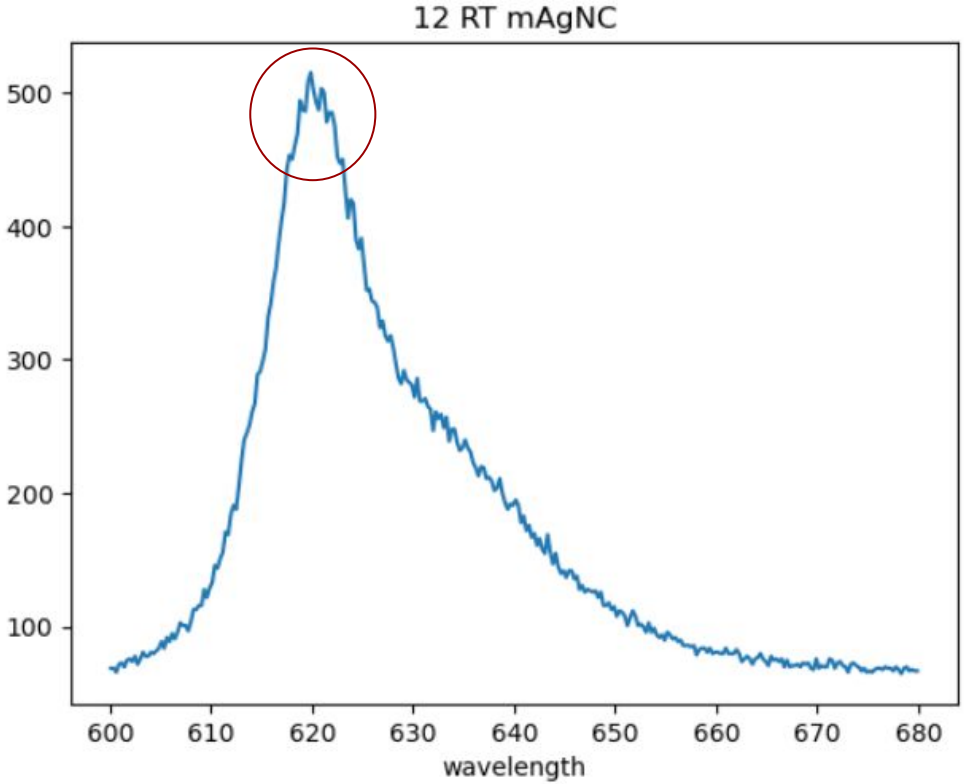
650
660
Wavelength (nm)


12 RT mAgNC
500
400
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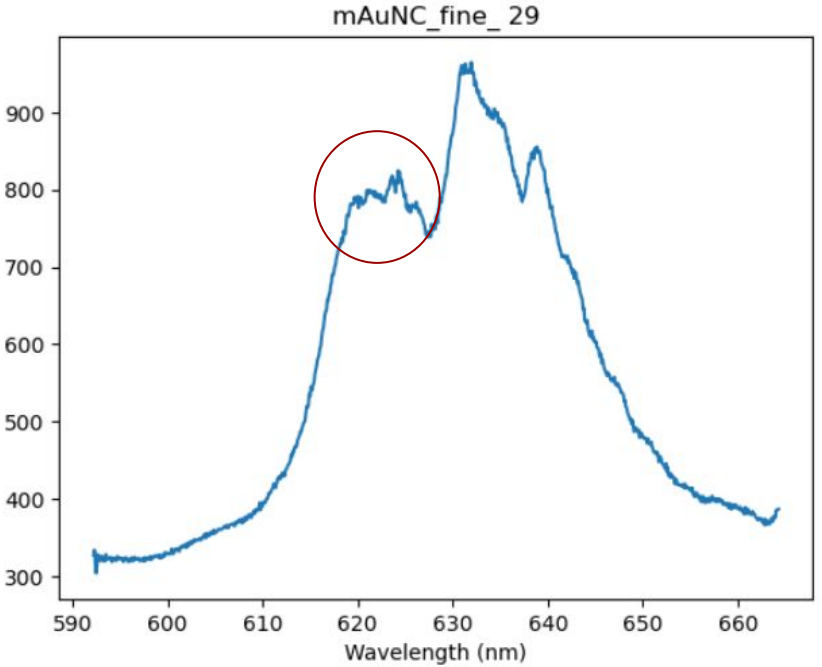
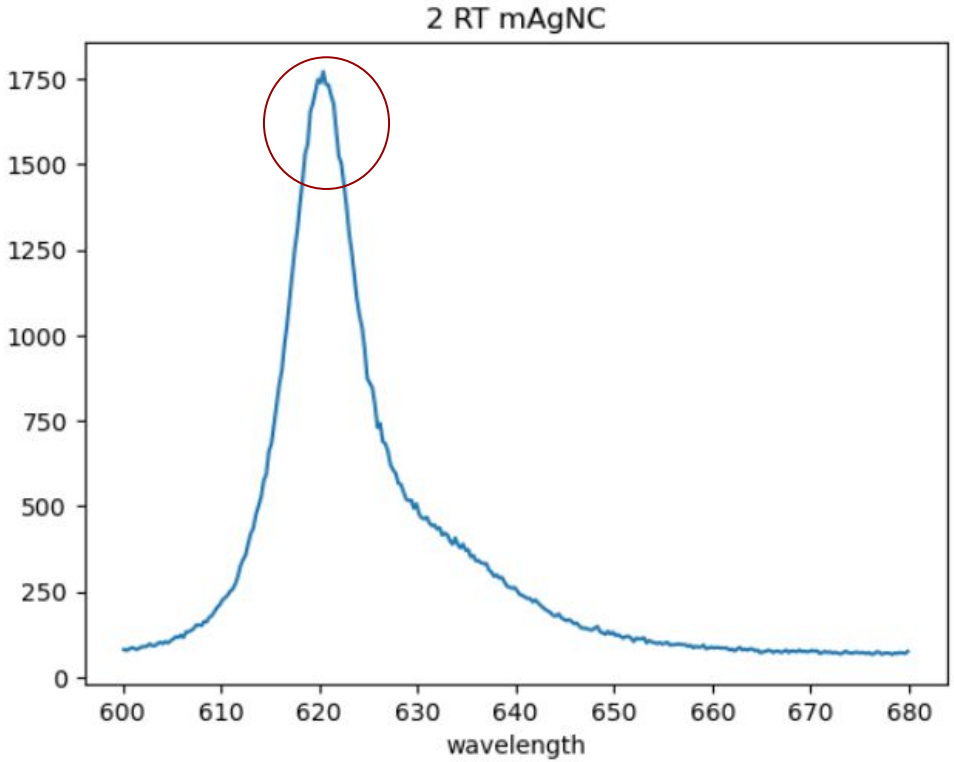
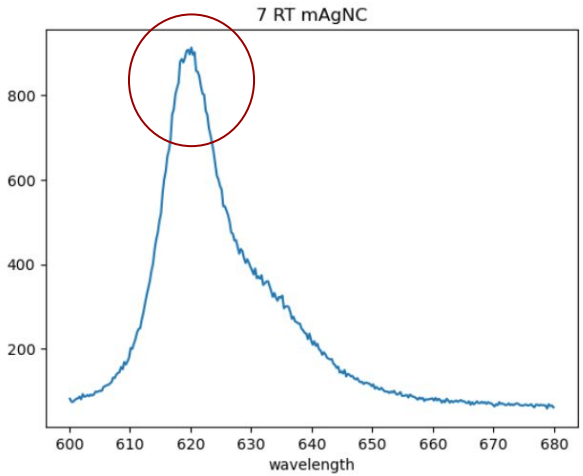
200
100
600
610
620
630
640
650
660
670
680
wavelength

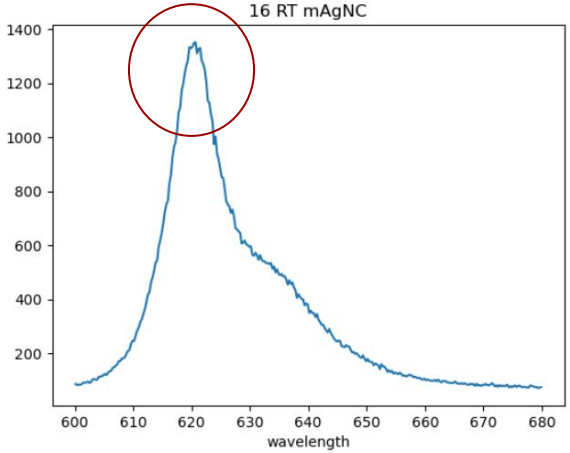

mAuNC_fine_ 29
900
800
700
600
500
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)

Point 2 is most likely a silver nanocube – out of all the new point measurements, it was the most isolated from old points.

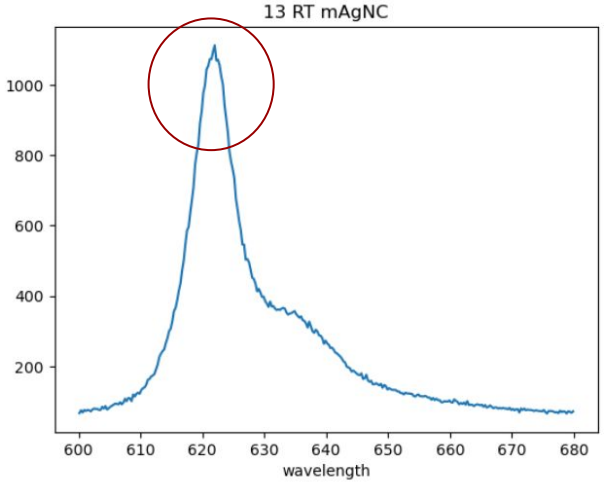
Notice the peak at around 620 nm corresponding to a very high intensity

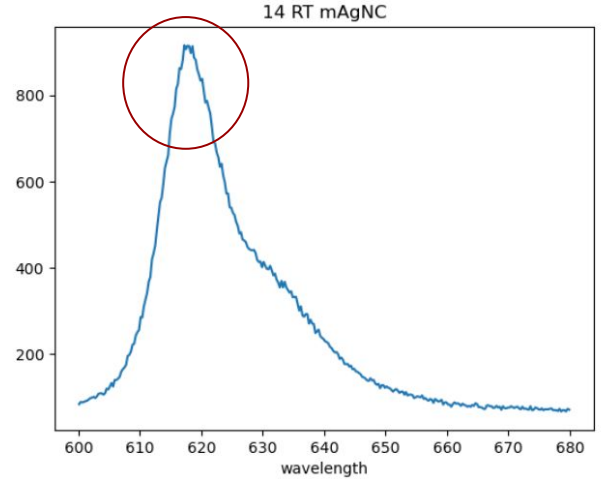






Notice how these spectra have a similar structure. Point 16 is notably outside of the general range of intensity for the peak. It is possible that these denote silver nanocubes.





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?

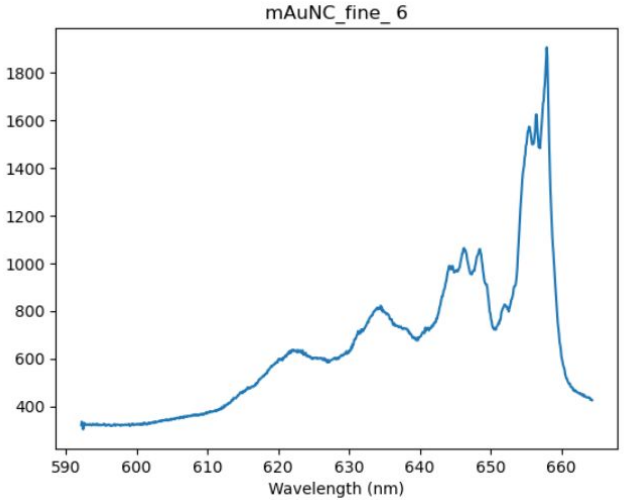
It seems like some peaks existed in both measurements between the same gold nanocube points, but the spectra of gold cubes from the sample with silver nanocubes had significantly more peaks at higher intensities. There may be some odd interactions between the gold and silver cubes.

Can we identify which points are likely silver nanocubes?

Point 2 is definitely silver, points 7, 13, 14, and 16 are also likely silver. Especially 7.

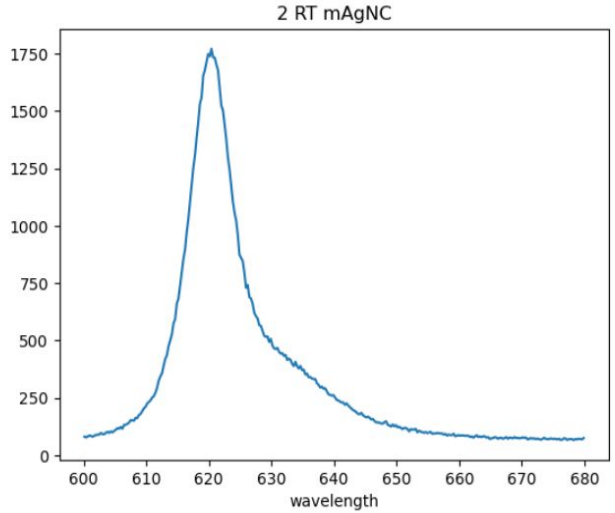
In general, how do the peaks differ between the gold and silver nanocubes? Does either provide more obvious enhancement?

The gold nanocubes without any influence from the silver caused the highest peaks in the spectrum to be in the 1000-1200 intensity range and 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.



Gold outlier

Silver outlier



Semester Progress

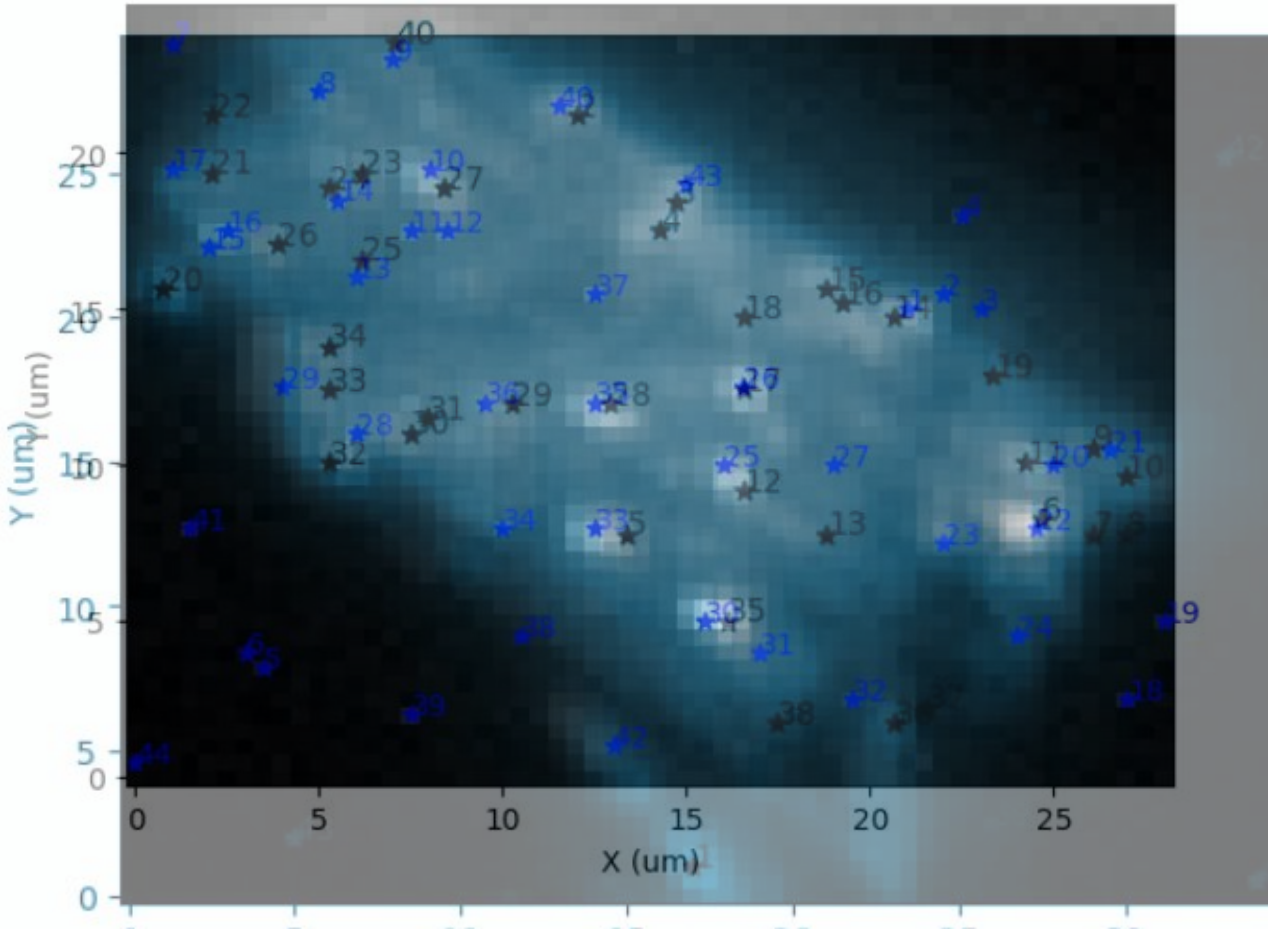
- Optical data analysis + summary for nanocube project (Sample 1)
- Noise reduction code using DMD algorithm
- Lots of graphene exfoliation + practice transfers with graphene and HBn during slower weeks

Grey points - Cryo Au
Blue points - Cryo Ag

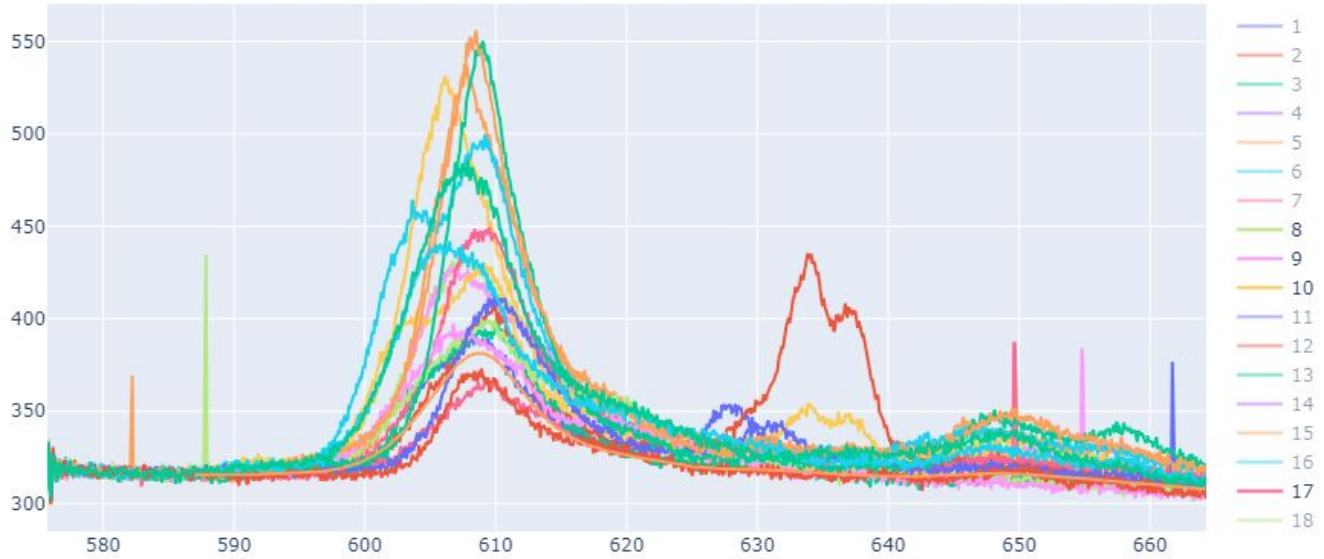
Ag — Au

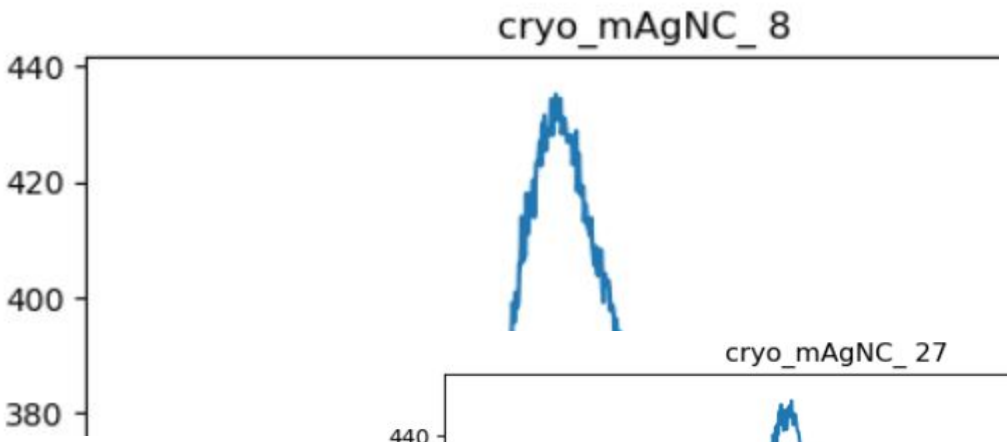
8 - new silver
10 - 27
17 - 21
21 - 9
22 - 6
23 - new silver
26 - 17
29 - 33
35 - 28
36 - 29
42 - new silver
43 - 3

9 - 40
13 - 25
20 - 11
25 - 12
27 - new silver
31 - new silver
33 - 5



Cryogenic mAgNC cleaned

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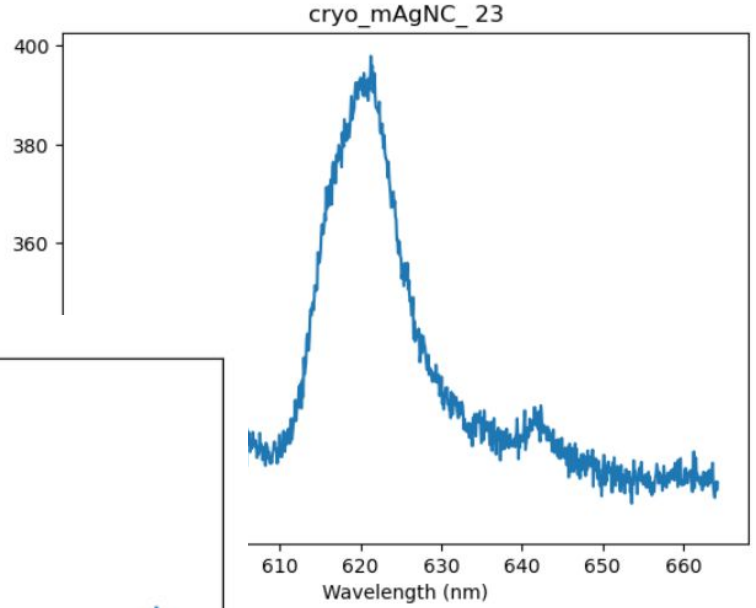
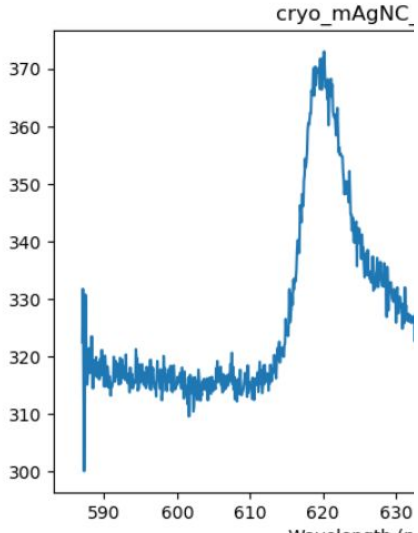
420
400
380


cryo_mAgNC_ 23
400
380
360
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_
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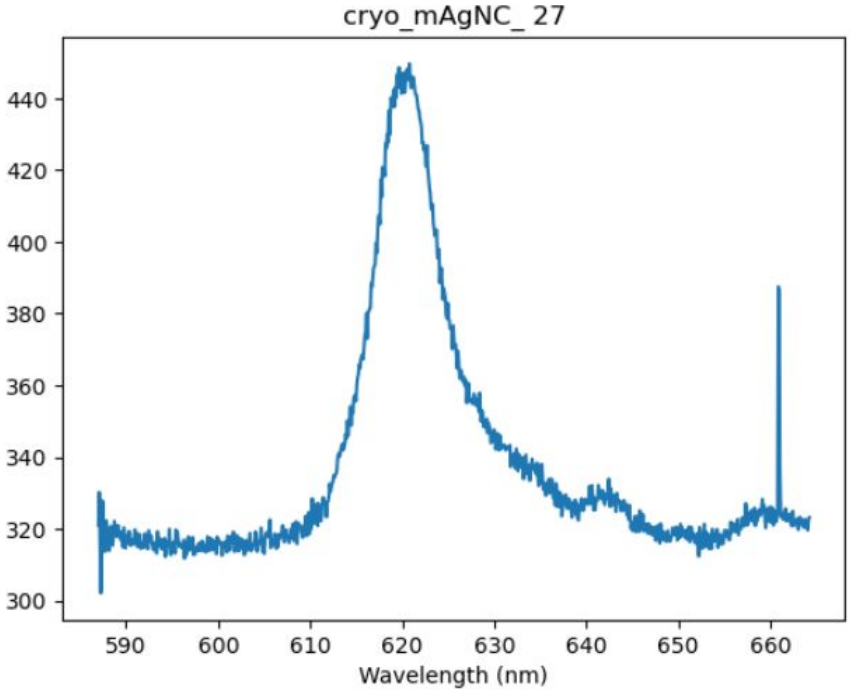
360
350
340
330
320
310
300
590
600
610
620
630
Wavelength (n


cryo_mAgNC_ 27
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380
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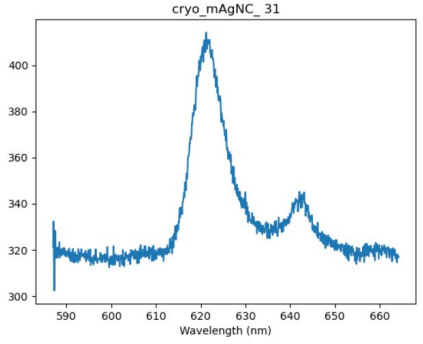
300
590
600
610
620
630
640
650
660
Wavelength (nm)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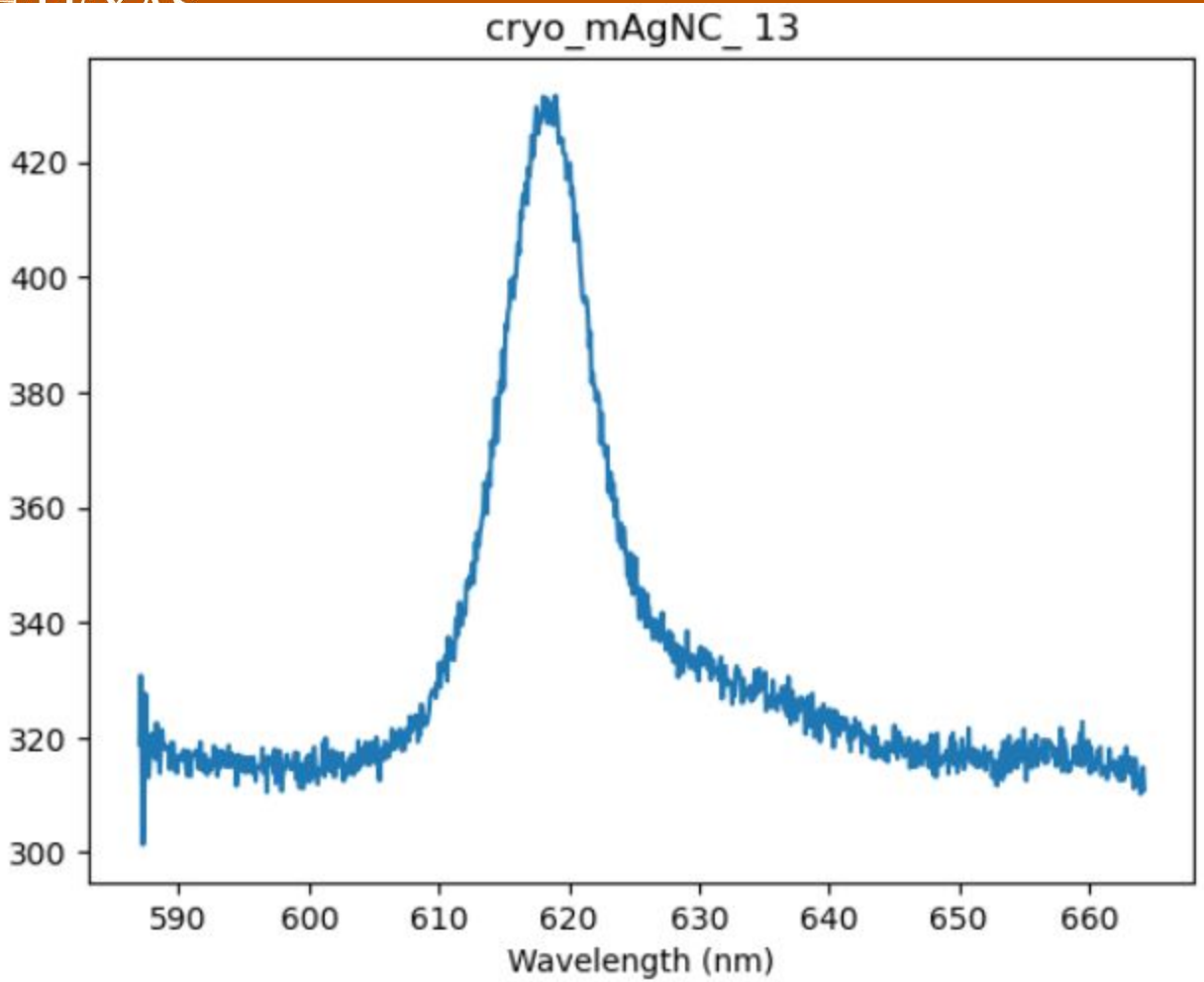
600
610
620
630
640
650
660
Wavelength (nm)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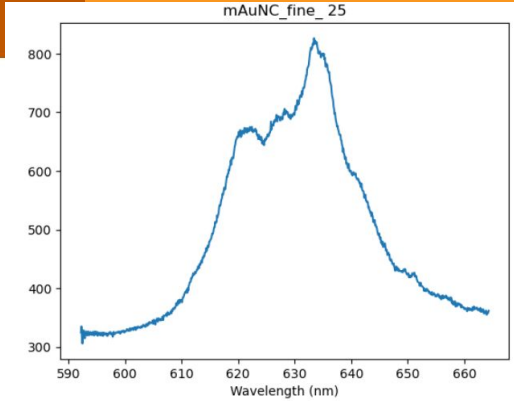
300
590
600
610
620
630
640
650
660
Wavelength (nm)


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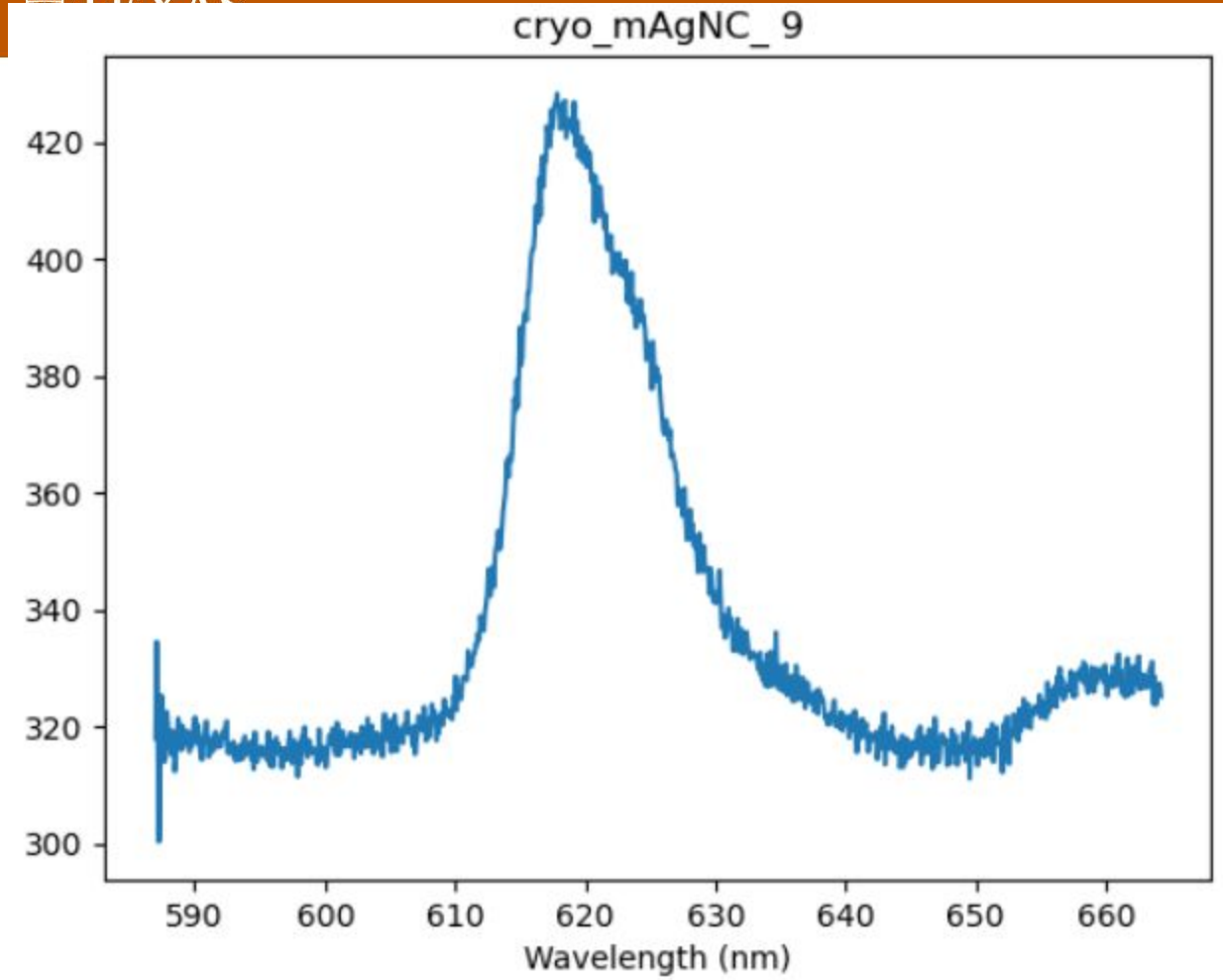
300
590
600
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 9
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300
590
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610
620
630
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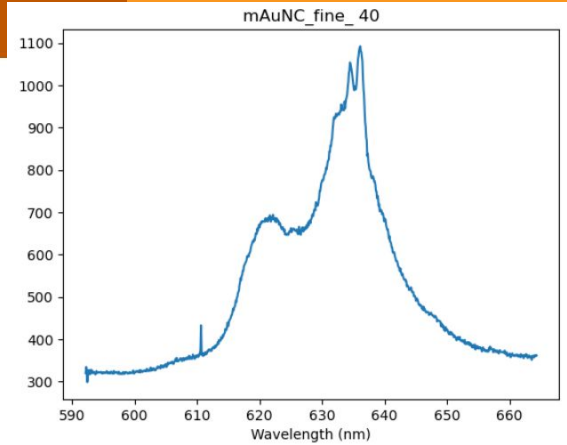
650
660
Wavelength (nm)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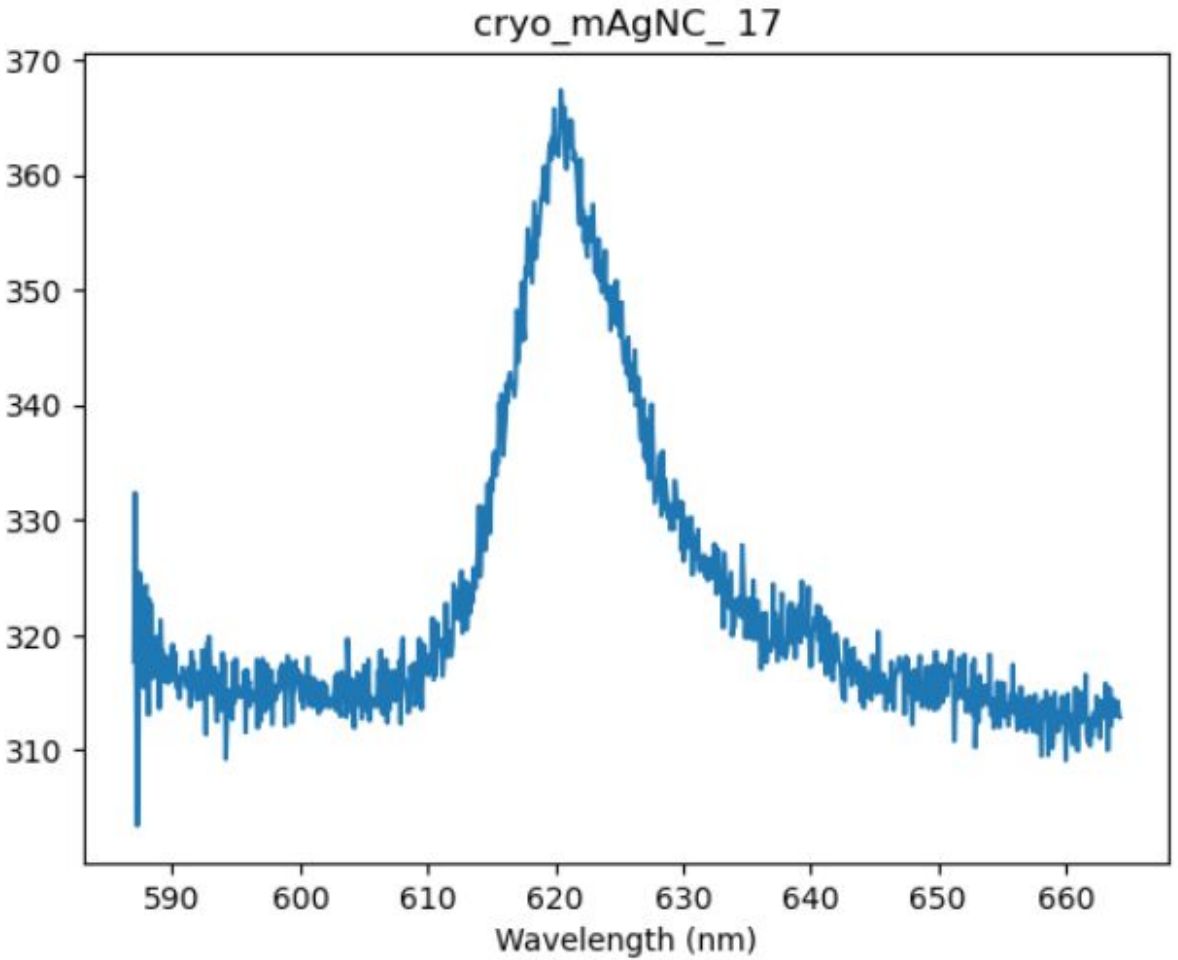
650
660
Wavelength (nm)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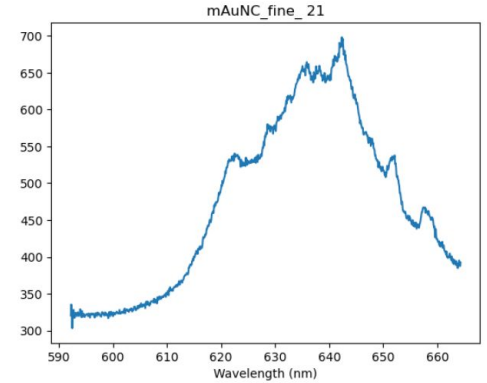
610
620
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 21
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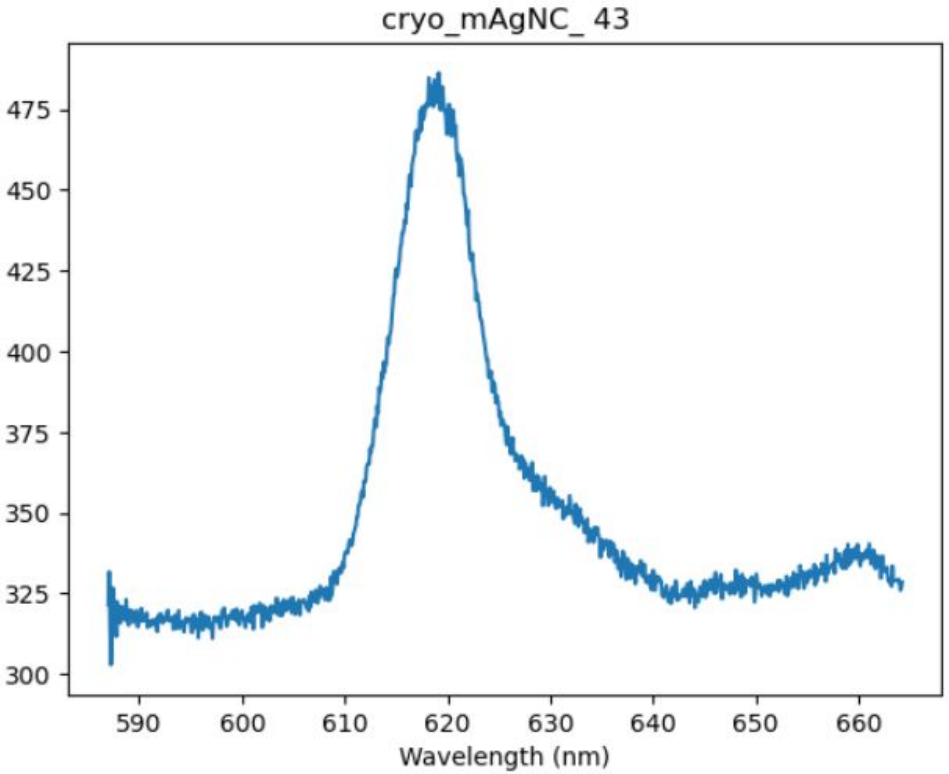
660
Wavelength (nm)


cryo_mAgNC_ 43
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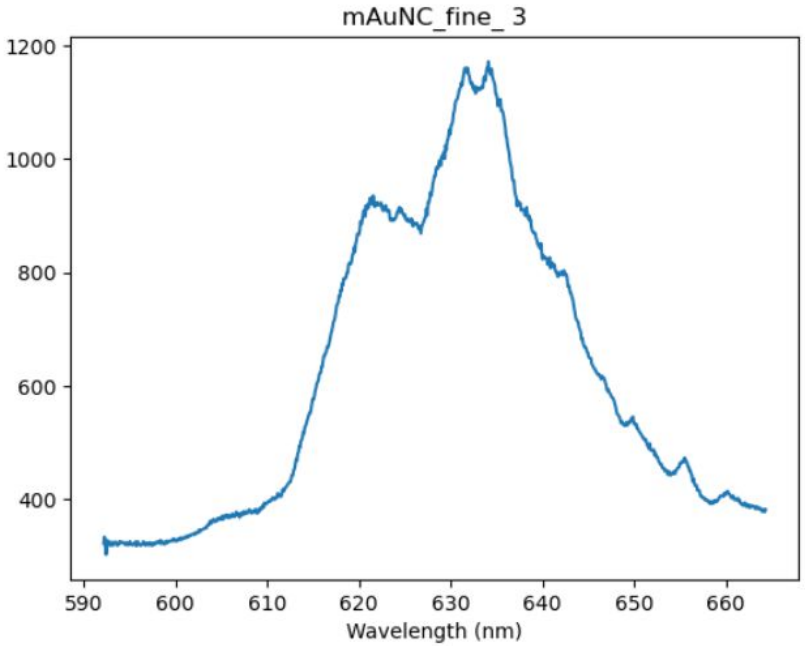
660
Wavelength (nm)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400
590
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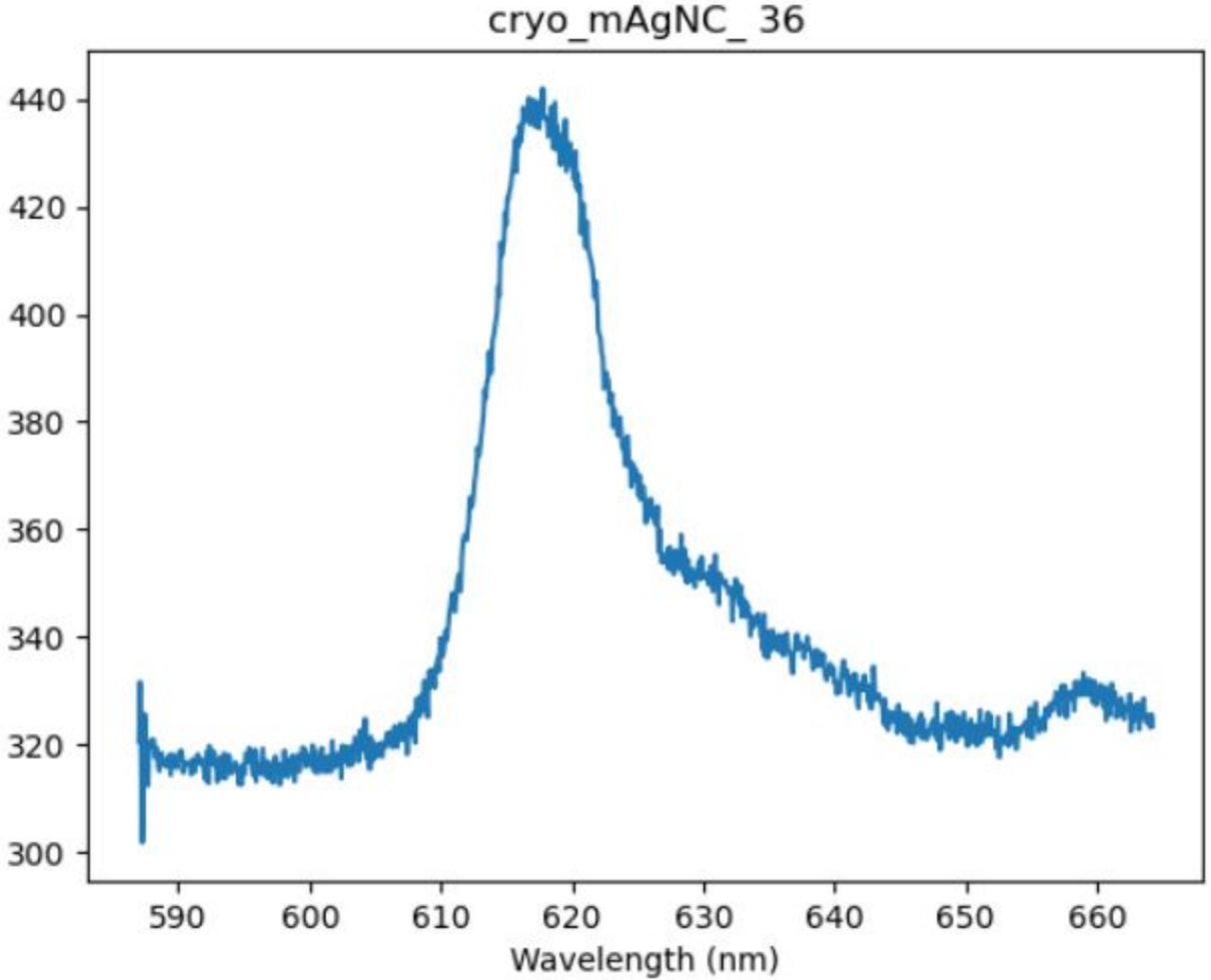
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 36
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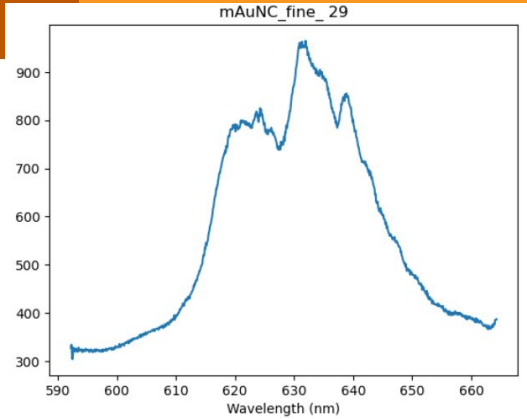
620
630
640
650
660
Wavelength (nm)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400
300
590
600
610
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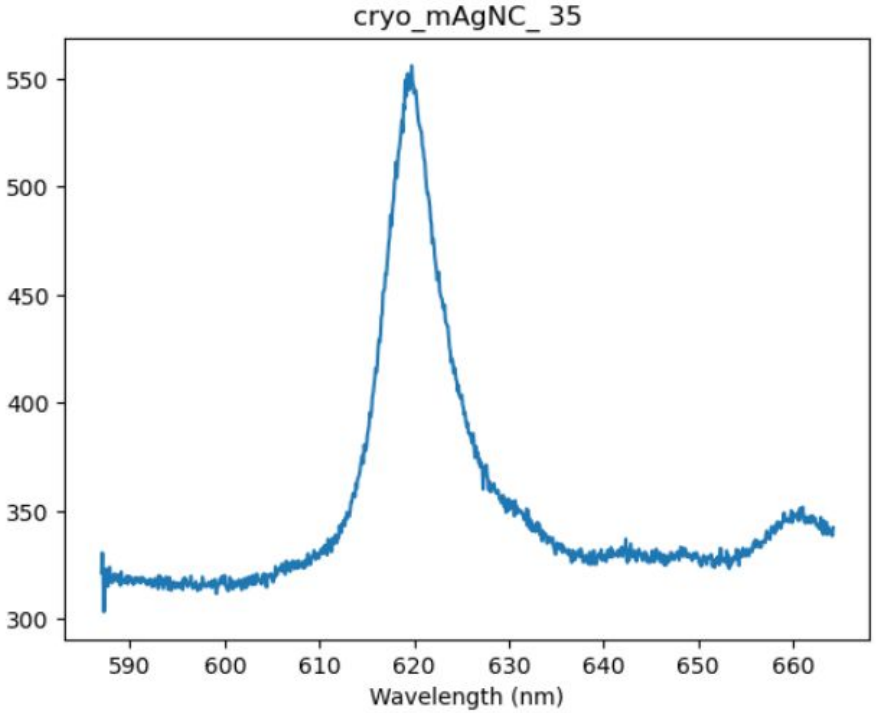
630
640
650
660
Wavelength (nm)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590
600
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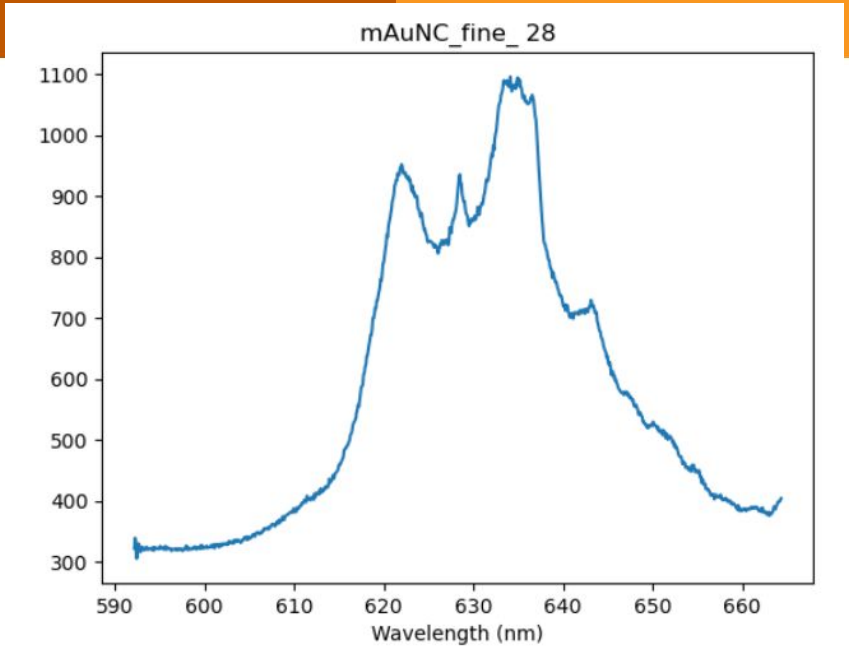
640
650
660
Wavelength (nm)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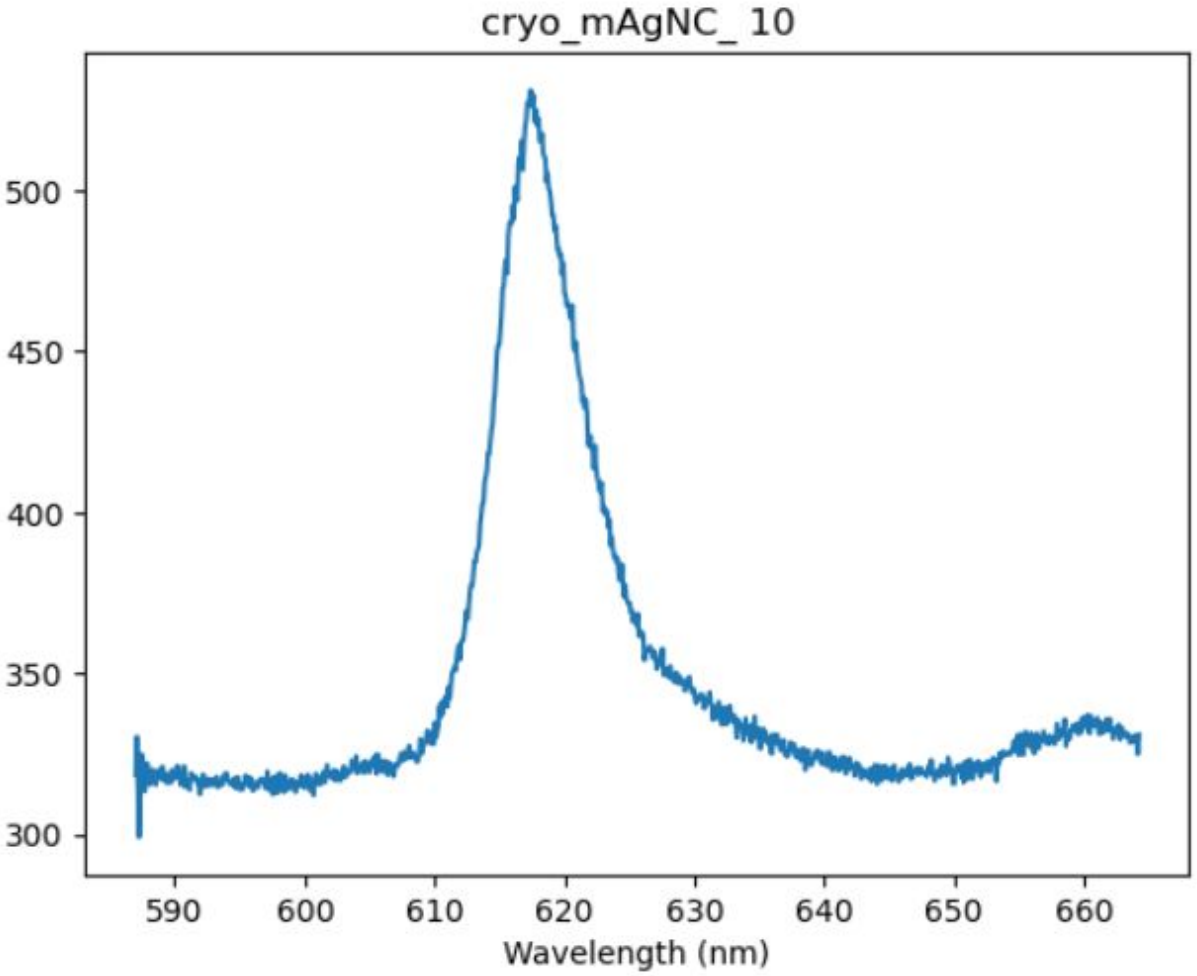
650
660
Wavelength (nm)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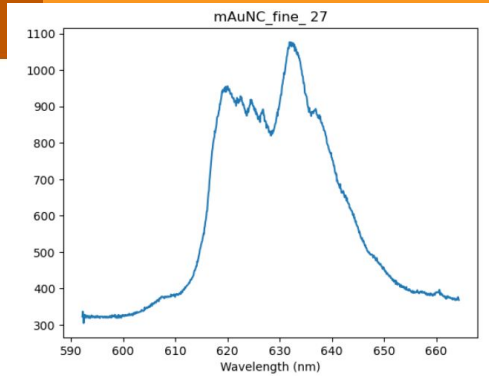
650
660
Wavelength (nm)


mAuNC_fine_ 27
1100
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800
700
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590
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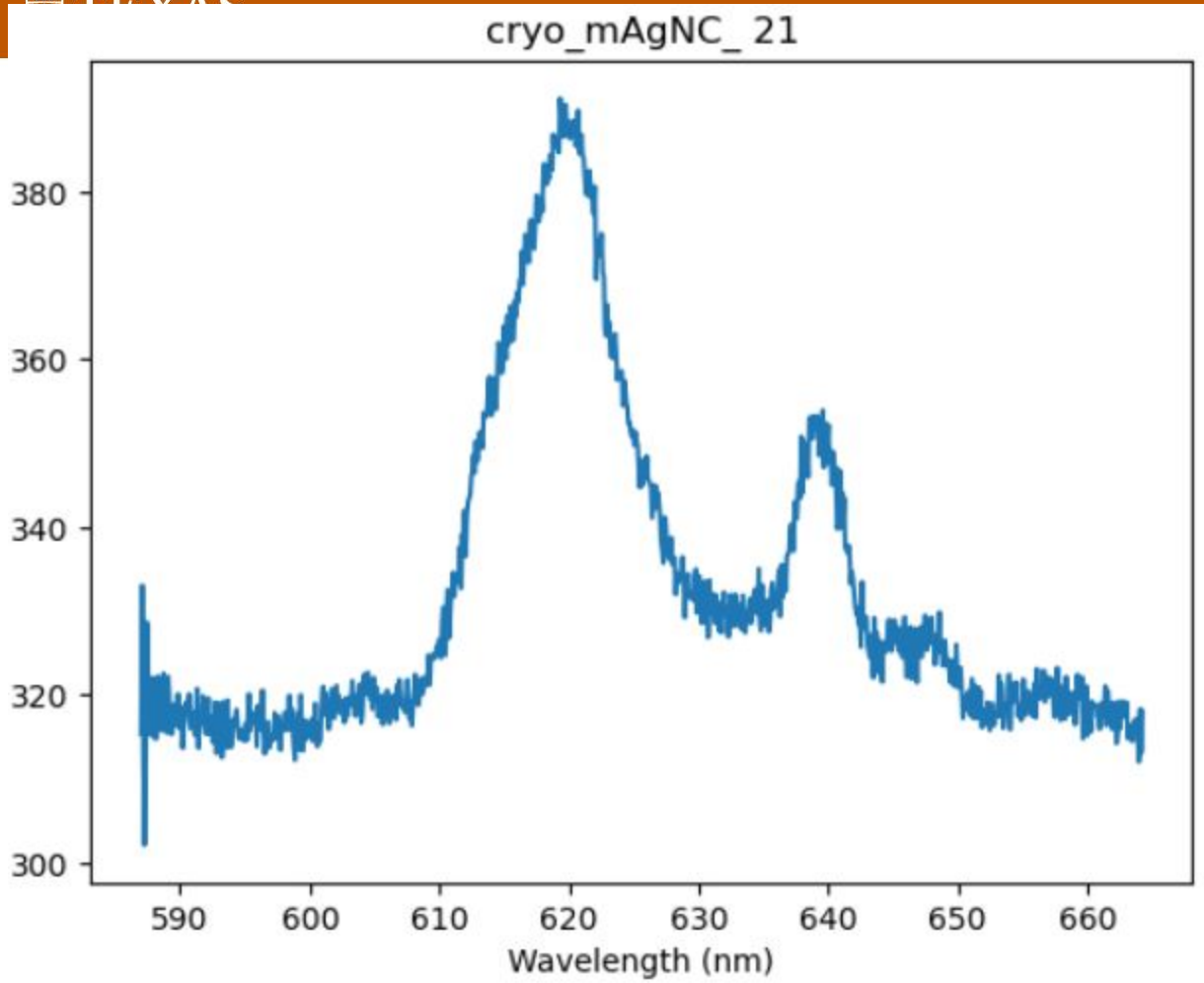
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 21
380
360
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590
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610
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640
650
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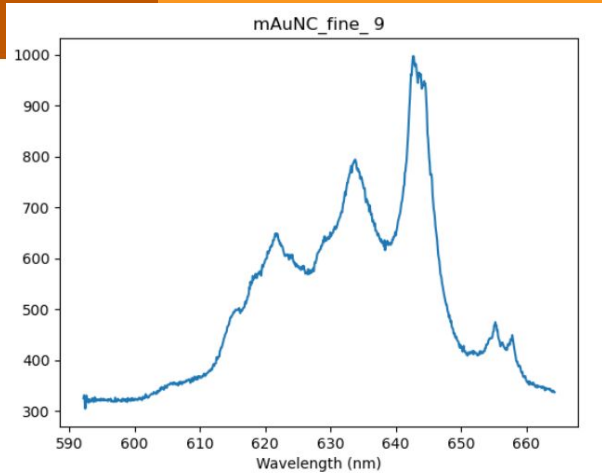
Wavelength (nm)


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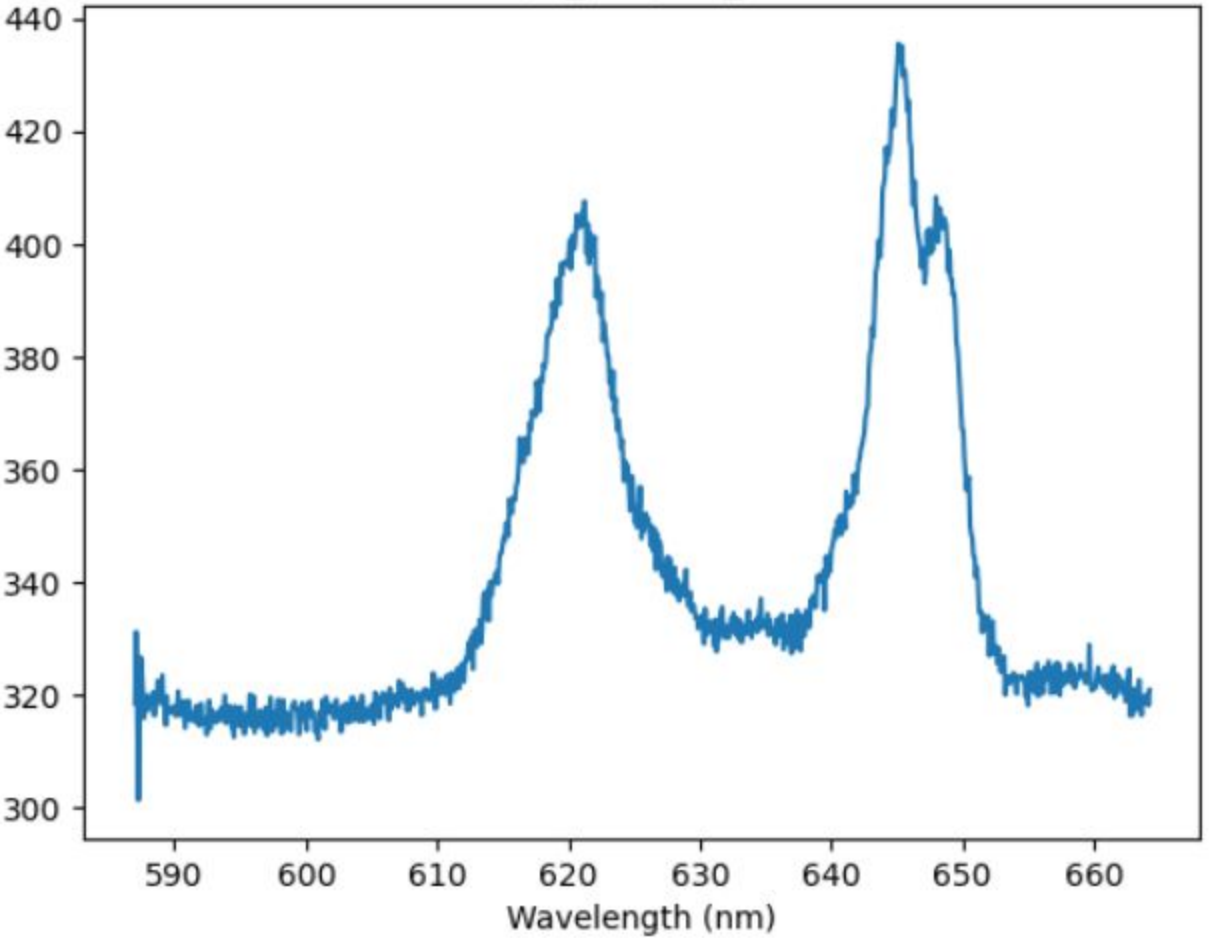
400
300
590
600
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 22
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300
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600
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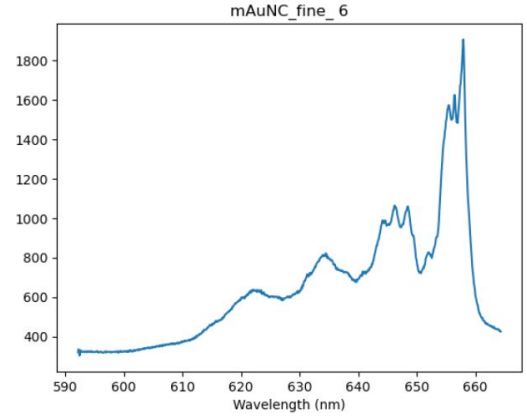
620
630
640
650
660
Wavelength (nm)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1400
1200
1000
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590
600
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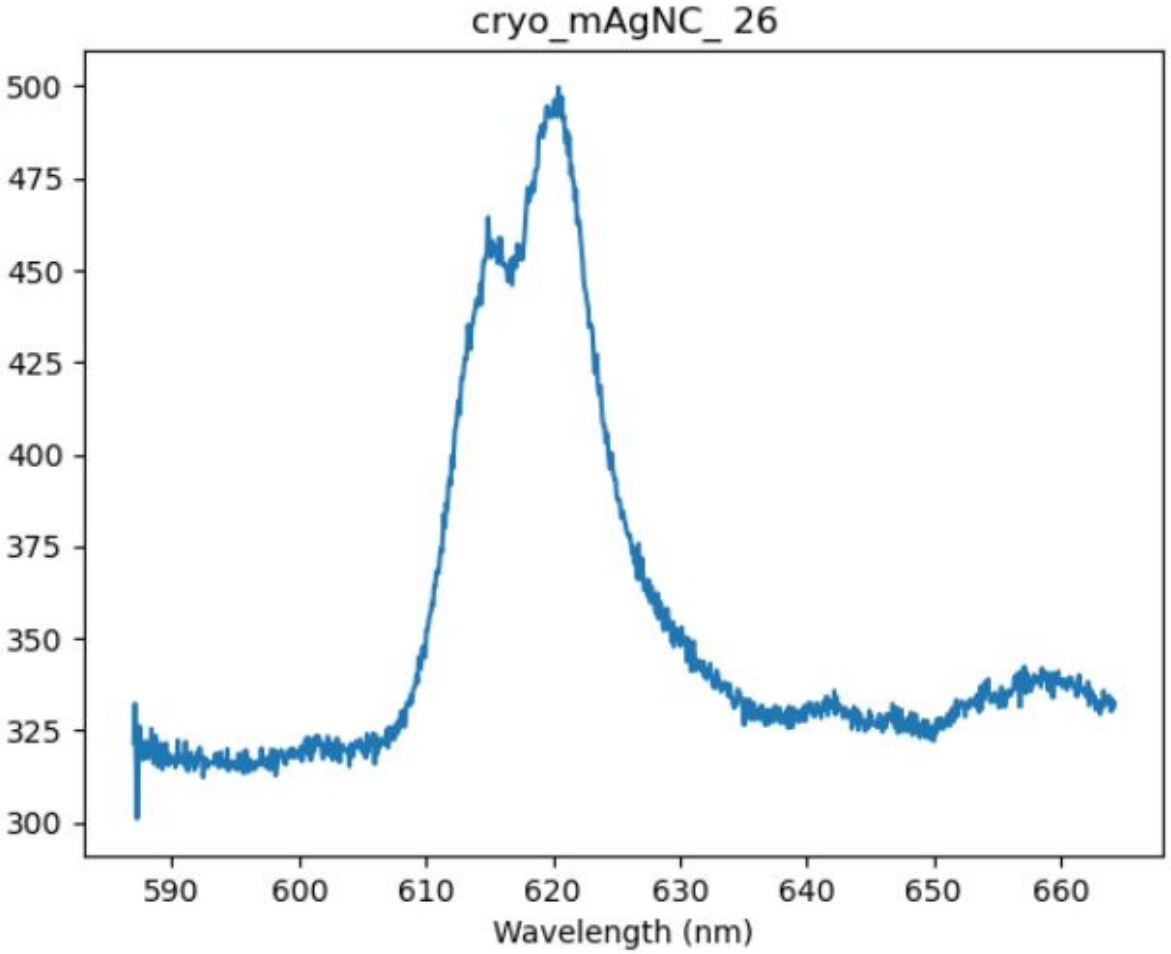
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 26
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590
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610
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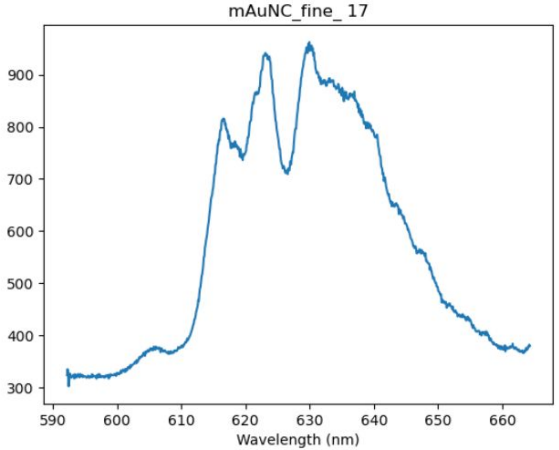
650
660
Wavelength (nm)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800
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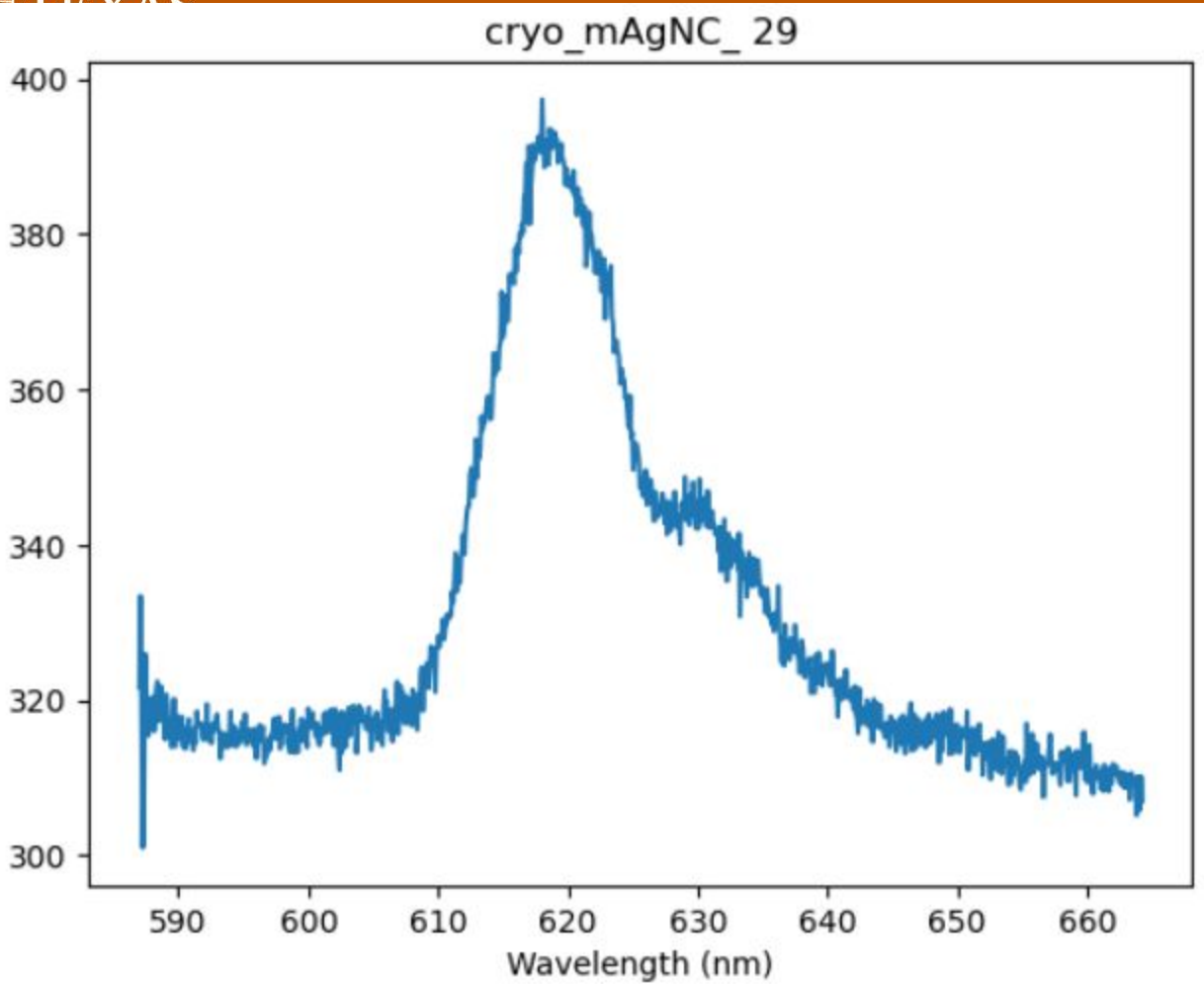
610
620
630
640
650
660
Wavelength (nm)


cryo_mAgNC_ 29
400
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320
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590
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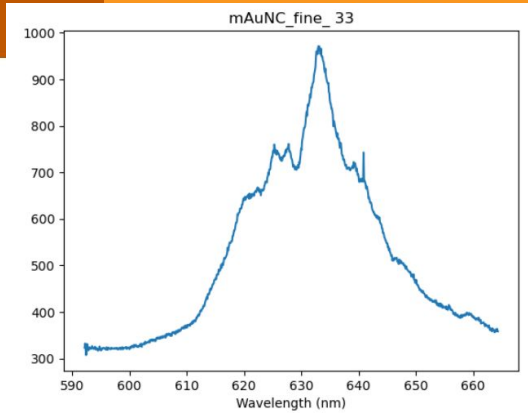
610
620
630
640
650
660
Wavelength (nm)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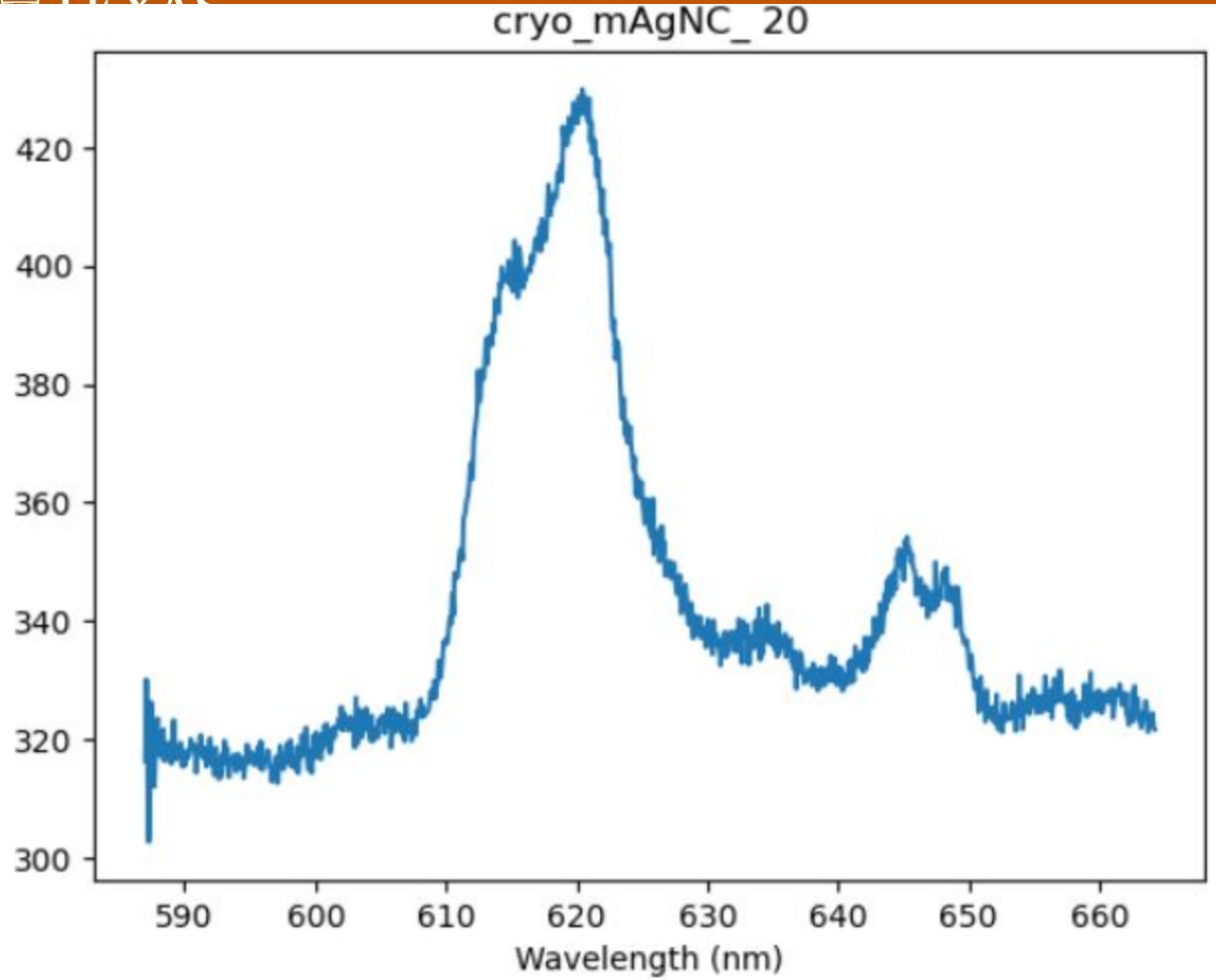
650
660
Wavelength (nm)


cryo_mAgNC_ 20
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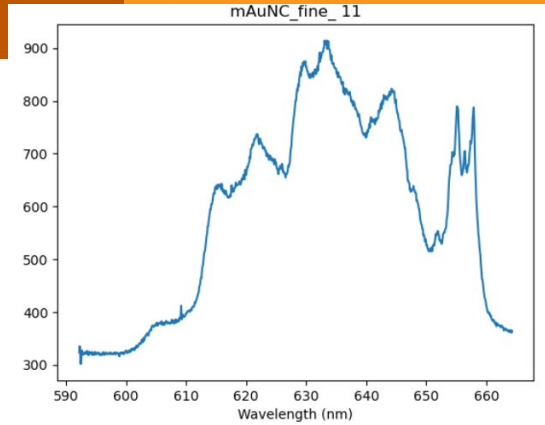
660
Wavelength (nm)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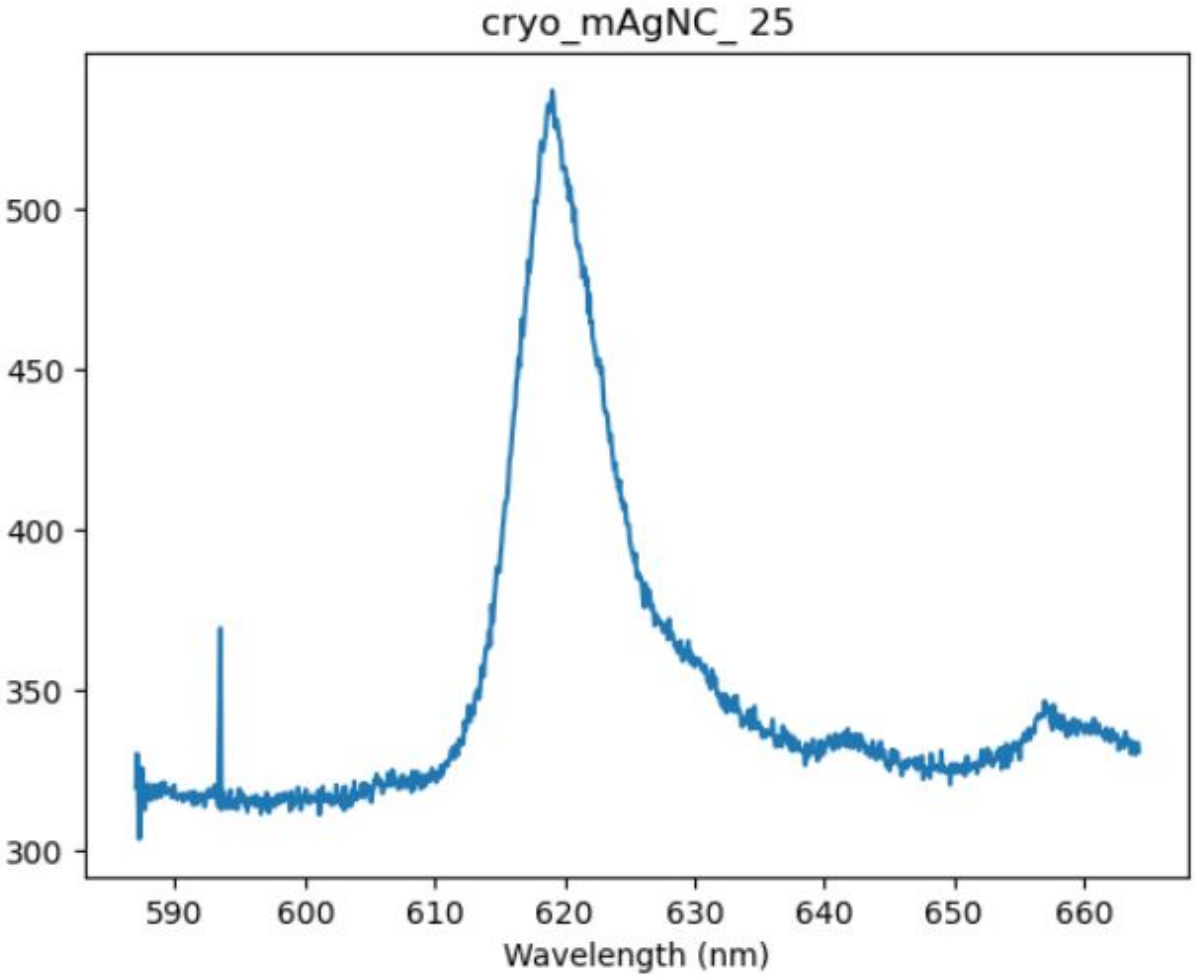
650
660
Wavelength (nm)


cryo_mAgNC_ 25
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610
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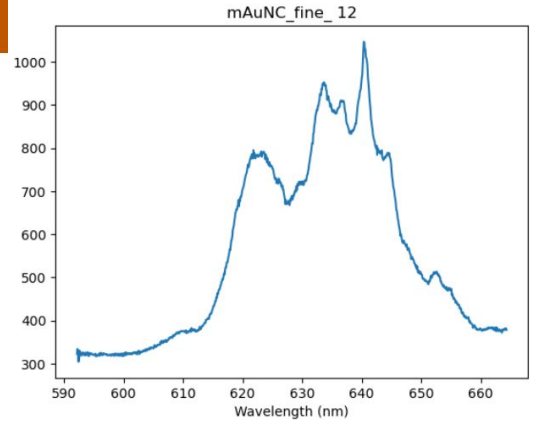
630
640
650
660
Wavelength (nm)


mAuNC_fine_ 12
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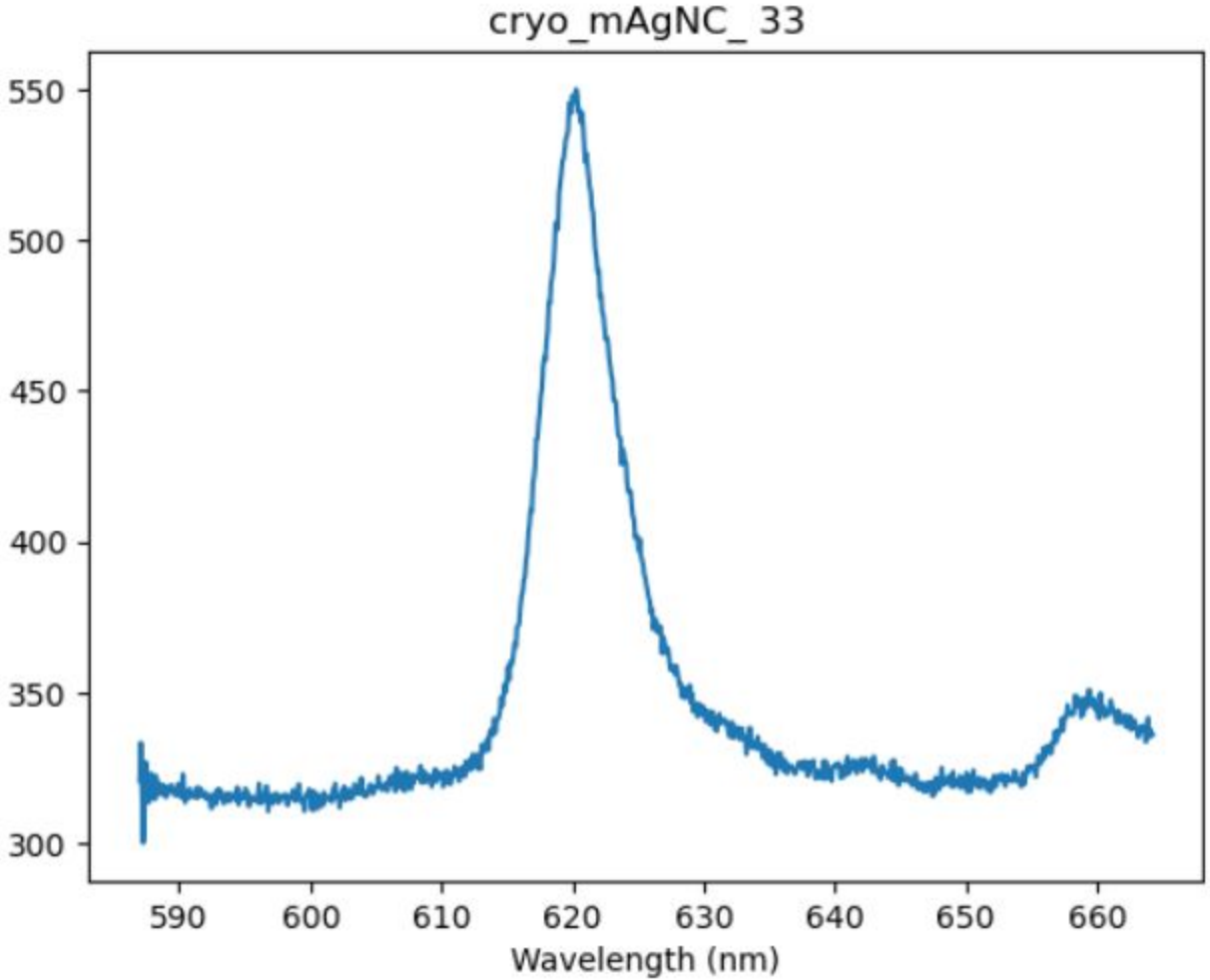
640
650
660
Wavelength (nm)


cryo_mAgNC_ 33
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620
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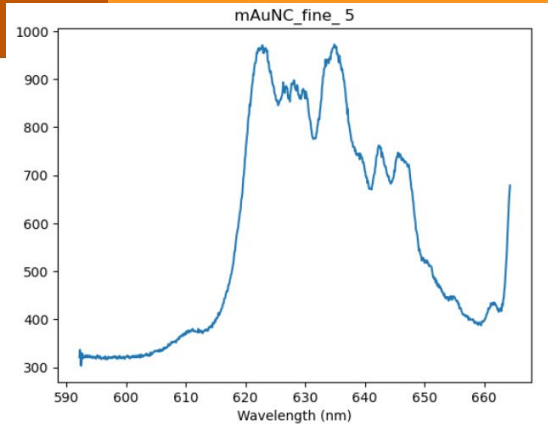
660
Wavelength (nm)


mAuNC_fine_ 5
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630
640
650
660
Wavelength (nm)