What we need to do to wrap up a basic set of measurements for Jialin and Qingyu (the only difference is in types of materials, not types of measurements; we will get a sample from Professor Chen Gang next week):

1. Improve the SNR. This has two components:
   1. We should try to reduce the scattered pump that is collected by the detectors. This will reduce the large vertically oriented noise signal that we saw yesterday.
   2. We should average longer. If longer averaging times are not possible in a single measurement scan, then we should collect data in several scans and average these. This will not only reduce any noise associated with the scattered pump but more fundamentally, it will give us improved SNR everywhere. The spectra we collected yesterday came rapidly, so we can do a lot more averaging.
2. We should make sure that we are in the single-exciton regime. There are two parts to this:
   1. We can estimate whether we are in the single-exciton regime by estimating the number of absorbed photons per QD per pulse based on the reported absorption cross sections of CdSe quantum dots and our measured pump fluence at the sample (using pinhole and assuming a Gaussian spatial mode). I attach a spreadsheet that does this calculation if you input our experimental parameters for pump transmission by a pinhole at the sample location.
   2. Measure a signal that depends on the excitation intensity and time after excitation. Typically, one does a TA measurement and looks at the absorption at the 1S peak. In the single-exciton regime, there will be dynamics associated with cooling (if one excites above the gap) and trapping. If one goes to lower fluences, then one is still in the single-exciton regime, so although the signal level will decrease, the shape should remain identical. However, if one increases the fluence and starts to excite multiple excitons per dot, then the dynamics will show a component associated with multi-exciton decay, typically due to Auger recombination (AR). This sort of measurement needs to be done even if the calculation in 2a suggests that one is in the single-exciton regime, since one needs to confirm this.

In our case, we could do a TA measurement, but that requires that we scan the delay stage, since AR will occur on tens to hundreds of picoseconds, much longer times than the AOM can access.

Alternatively, we can just do a short-range “TA” measurement using the AOM to just to an ON-OFF-ON-OFF sequence and adjust the pump delay, but we would need to do that at some short time (say 5 ps probe delay or so) and at some long time (say closer to 1 ns). We would do this measurement for two pump fluences. If the ratio of (or one can use a different transition if the pump scattering introduces too much noise) is the same at both fluences, then it means that at least the higher fluence is too high. One then needs to try a lower fluence and repeat the measurement until one finds a pair of fluences that yield the same traces (other than a scaling factor). Then both those fluences are in the single-exciton regime.

1. Once we have done that, we should collect 2D spectra like yesterday’s at, say, three different delays: a delay that is just beyond overlap of the probe with the second of the two pump pulses out of the AOM, a delay of 10 ps later, and a delay that is perhaps 100 ps.
2. The 2D spectra should then be compared to simple TA spectra at the same delays. If we are collecting the 2D spectra correctly, we should see the 2D spectra disappear rapidly (as a function of probe delay) compared to what is observed for the TA spectra. This is simply because the phase-cycled 2D spectra should just contain the coherent response.

A few other comments:

We should also record the probe spectrum itself. Without that, we only know the pump-induced changes in the probe, not the relative changes.

The focus should be on the cross peaks. From our data yesterday, we can see that we still have insufficient pump bandwidth for extracting homogeneous linewidths. For that we will probably have to wait until we build our NOPA.

That should be enough for Jialin’s thesis. However, there are other experiments that we could try if Jialin wants. For example, we could do 2D spectroscopy after first exciting the system with another pump pulse (split off part of the pump before the AOM system). That would allow us to look at couplings between different biexcitons. I don’t know what this would look like. It could be fun, but it is completely unnecessary.

For Qingyu, everything is the same as above, except that we will be looking at a different kind of sample and spectral range. We will get our samples from Professor Chen Gang’s students next week.