Standard Operation Procedure for Time-Correlated Single Photon Counting Spectroscopy Experiments Using Spectraphysics Tsunami Tunable Laser

Hazards Present: Class IV visible and invisible laser radiation capable of severe personal injury and starting a fire.

Hazards mitigation:

- **Administrative control:** Lasers Safety training classes offered by UCSB ES&H, orientation by OCF staff
- **Physical controls:** Inflammable, non-reflective metal beam blocks, screens and enclosures.
- **PPE required:** Laser safety glasses/goggles with OD>4 at the laser wavelength (LPA-ALEX for Tsunami beams (710-980 nm), KTP-Argon (180-540 nm) for Verdi G and the second harmonic of the NIR light, or similar), fire retardant lab coat.

Equipment required:

- Thermoelectric power meter
- NIR scope viewer
- Non-glossy paper white business card for beam tracing.

Startup sequence:

1. Turn on TE water chiller for Coherent Verdi G and Spectraphysics Tsunami lasers. The chiller setpoint is 18^{o}C.
2. Turn on the Verdi G power supply and Tsunami controller with a switch on the rear panel. For the former, wait until red "Fault" indicator on the front panel turns off (It turns on twice with ~2 seconds interval before turning off). Turn the safety key on the Verdi G power supply to on position.
3. Verify that the Tsunami output is blocked by an appropriate beam block. Start OPSL software on the controller PC. Once it connected to the Verdi laser, select remote control and set the output power to the level desired (6.2 W for 800 nm operation of Tsunami).
4. Tsunami normally should be lasing at this point, however, it takes about 20-30 minutes for the system to equilibrate and return the prealigned state. Thus, the laser should not be adjusted or used during this time.

5. While the laser is warming up, CCD camera and avalanche photodiode can be energized, along with the radio frequency (RF) driver, monitoring oscilloscope, and the pulse counter. The CCD camera cooling needs to be activated by starting Winspec software and activating the cooling process through hardware options (Setpoint T = -70°C).

The light source setup:

The laser light source layout is shown in Figure 1. It assumed that the Tsunami laser has been set up and optimized for specific wavelength previously by an OCF staff member. Tuning and optimization of the Tsunami is outside the scope of this SOP.

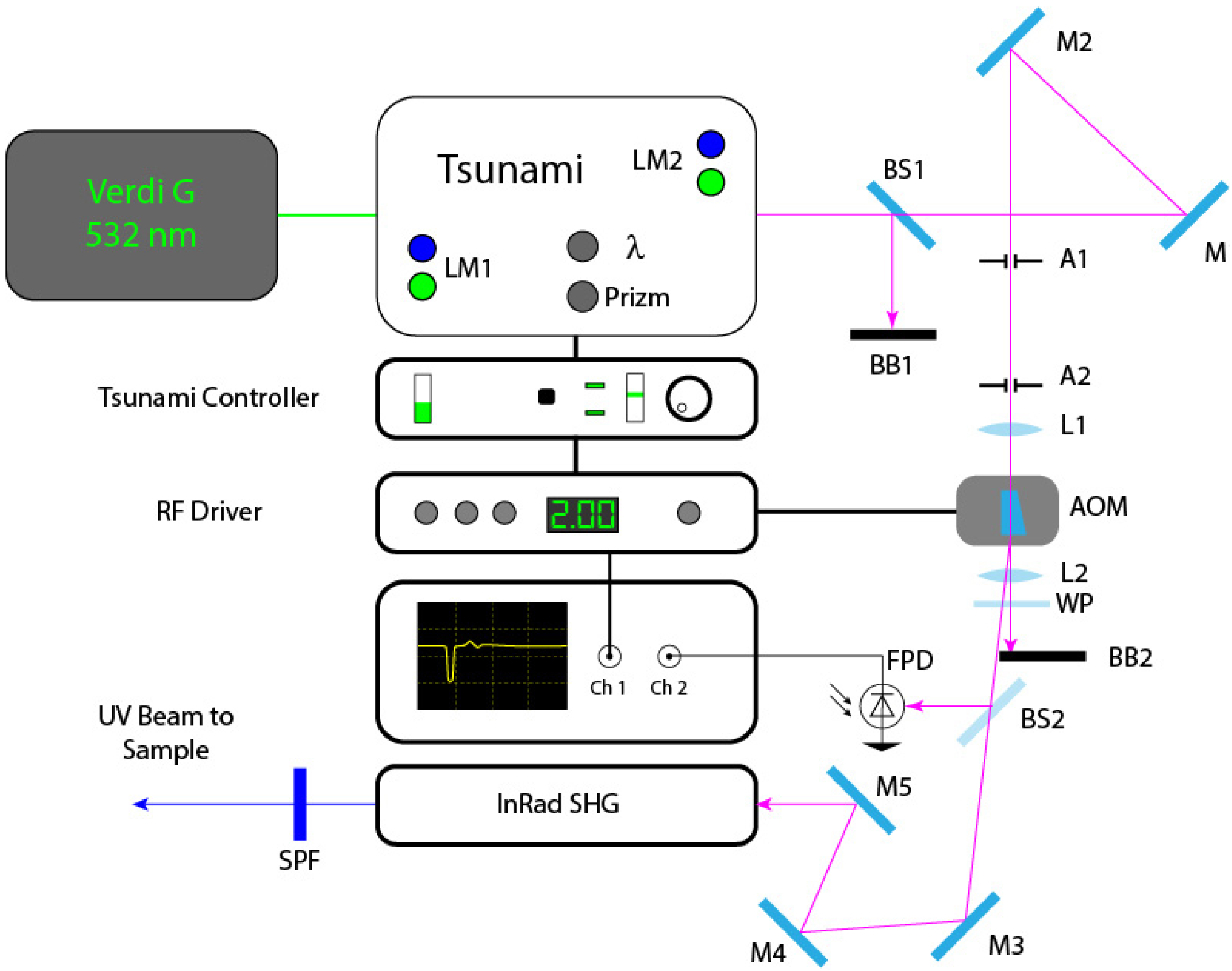

Figure 1. Layout of the femtosecond light source with pulse picker and optical harmonics generator.

1. Place the power meter between the output of Tsunami laser and the beamsplitter BS1. Place a metal screen after the iris aperture A2.

2. After warmup period and assuming that the laser has been aligned previously, the powermeter readings should be in the range 550-650 mW. The “Photodiode” LED bar on the Tsunami controller should be at least at 50% level. Press and release “Pulse Enable” button twice with ~1 second delay on the Tsunami controller and observe whether “Pulse” LED remains on while “Enable” LED is off. If pulsing does not start (which is indicated by solid green “Pulse” LED), repeat the procedure while adjusting the “Phase” knob on the controller.
3. If the Tsunami laser power is below 550 mW or stable pulsing mode cannot be achieved, peak it with laser cavity mirrors LM1 and LM2. **Adjust one knob at a time, always find the maximum power position before adjusting the next knob. If this rule is not followed, it is very easy to misalign the laser cavity and loose the lasing.** It is recommended to peak the power first with horizontal and vertical adjustment knobs of LM2 and then repeat the procedure with LM1 mirror. Several iterations may be required to find the peak power.
4. After peaking the laser power, start pulsed mode-locked operation as described in the paragraph 2. **If the desired power level and stable pulsing cannot be achieved, contact OCF staff for assistance.**
5. Place the powermeter sensor after beamsplitter BS1. The power level should be ~50% of the power before the beamspliter. **The laser power value after the beamsplitter should never exceed 350 mW otherwise AOM crystal can be damaged.** The power after BS1 can be tuned by changing BS1 angle of incidence. Make sure that the beam reflected by BS1 is blocked by the beam block BB1.
6. Remove the powermeter sensor. Using the NIR viewer scope, verify that the laser beam is centered on iris apertures A1 and A2. If necessary, recenter the laser beam using “dog leg” alignment protocol (Close down A1, use M1, to center the beam on it, open A1, close down A2, use M2 to center the beam on A2, repeat until the beam is dead centered on both A1 and A2).
7. Place the beam block BB2 after the waveplate WP. Verify that “Loop Lock” LED on the RF AOM driver is on. If not, verify that the Tsunami laser is pulsing, the “Photodiode” LED bar on the Tsunami controller is illuminated at least at 50% level, correct if necessary. Set the RF driver to continuous operation mode.
8. Remove the beam block placed after A2. Observe the laser light pattern on the beam block placed after WP with NIR viewer scope. One should see two (or three) elliptical spots, corresponding to direct and diffracted beams (See Figure 2).
9. It is possible that upon removal of the beam block, the Tsunami stops modelocking/pulsing. This happens due to destructive interference with the back reflection form the optics train. In this case, close down the iris aperture A2 so that the opening is just slightly bigger than the beam diameter. Block the beam before

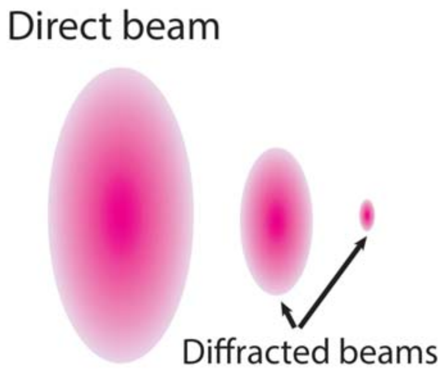

Figure 2. Diffracted beam intensity

BS1 and restart pulsing as described in paragraph 2. If the modelocking stops again after removal of the beam block, repeat steps 5 and 6. If this measure fails to maintain stable pulsing, misalign the laser beam *slightly* (about 5% of the beam diameter) vertically on the aperture A2. Contact OCF staff if the problem persists.

10. Move BB2 so that it blocks the direct beam and the diffracted beam propagates towards beamsplitter BS2. Using NIR scope viewer, center the beam on the fast photodiode FPD using BS2 alignment knobs (verify that FPD power switch is in "On" position).
11. Connect the FPD output to Channel 1 input of the digital oscilloscope with analog bandwidth >350 MHz. Channel 2 needs to be connected to the trigger output of the RF driver. Both channels must be set to 50 Ohm input impedance. Channel 1 sensitivity = 50 mV/div, Channel 2 sensitivity = 2 V/div, trigger source – Channel 2, trigger mode – normal, on the falling edge, time base – 10 ns/div. Adjust trigger level to achieve stable triggering of the oscilloscope.

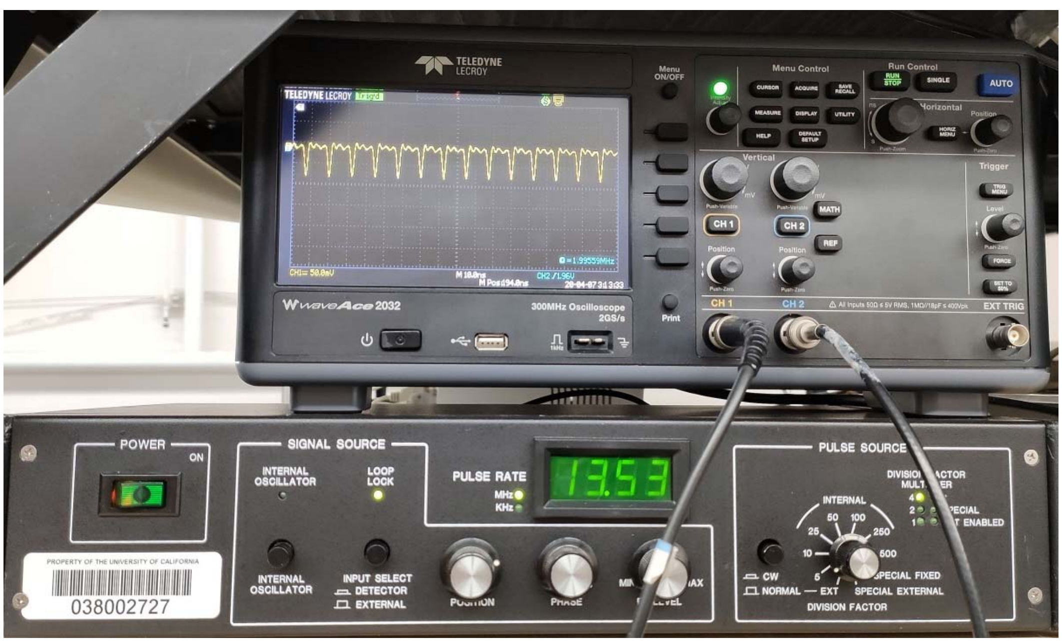

Figure 3. Laser pulse train in the CW diffraction mode.

12. At this point, a train of pulses should be seen on the oscilloscope screen (see Figure 3). Use BS2 alignment knobs to maximize the amplitude of pulses. If there is a noticeable low frequency modulation, make sure that the Tsunami laser power is peaked. To see the modulation better, one may need to increase the oscilloscope time base settings to 1 ms/div or so. If the modulation persists after the power peaking, one may try **small** adjustments of laser mirror LM2 or the laser prizm setting while observing the pulse train envelope. Adjust only one knob at a time and return it back to the initial position if the adjustment does not quench the modulation. Contact the OCF staff member if the problem persists.
13. This step may be omitted if the amplitude of diffracted pulses in continuous mode is greater than 40mV on the oscilloscope screen. Place the thermal powermeter after the beam block BB2 and note the power in the diffracted beam. Remove BB2 and measure the total power in diffracted and transmitted beams. The power in the diffracted beam should be in the range of 15-25% of the total power in the continuous diffraction mode (typically 40-50 mW in the diffracted beam, 200-250 mW total transmitted power when the power after BS1 is about 300 mW). If it is less than 15%, please refer to Appendix 1 or contact OCF staff member for adjustment of the AOM crystal. Reposition BB2 beam block, so that it blocks the direct laser beam.

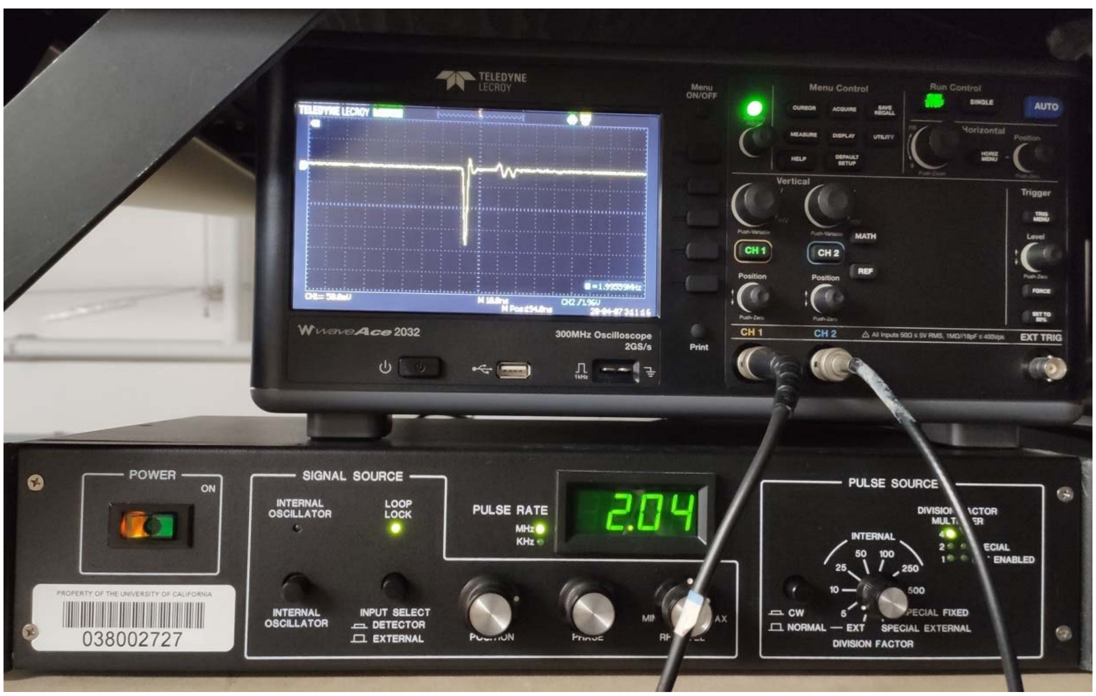

Figure 4. Laser pulse train in the normal diffraction mode.

14. Switch the RF driver to the normal operation mode. A single pulse with some afterpulses should be visible on the oscilloscope. Use “Delay” and “Phase” knobs to

maximize its magnitude. To set the RF power level, rotate “Power” knob clockwise until it stops and then turn it counterclockwise ¾ of the turn.

15. Verify that the beam is centered on the input orifice of Inrad 9300 second optical harmonics generator. If it is, switch the RF driver to continuous operation mode and observe the UV beam at the output of the second harmonic generator (SHG). One may optimize SHG process roughly by looking at the intensity of UV spot on a business card and adjusting the angle of SHG crystal with the corresponding knob on the SHG unit and rotating the waveplate WP. If there is no UV output, align the SHG unit by following directions in the manual or contact OCF staff.
16. Verify that the short-pass filter SPF is installed at the SHG generator output to block the residual NIR light. Normally Schott glass UG38 or similar is used for this purpose.

Optics Alignment

Typically, TCSPC experiments utilize either 90° or 0° geometries for collection of the photoluminescence. In the former case, the laser beam is aligned parallel to the optical table surface and perpendicular to the optical axis of the collection optics (Solid blue line in Figure 5). In the latter case, the laser beam is aligned coaxially with collection optics axis (Dashed blue line in the Figure 5). 90° geometry is used primarily for liquid samples or solid samples opaque at the laser wavelength. It allows to reduce the amount of the laser light scattered into the detector. The 0° geometry offers better PL collection efficiency but requires additional countermeasures to prevent the laser light from saturating or even damaging the detector. Detailed information on the detection optics alignment is provided in Appendix 2.

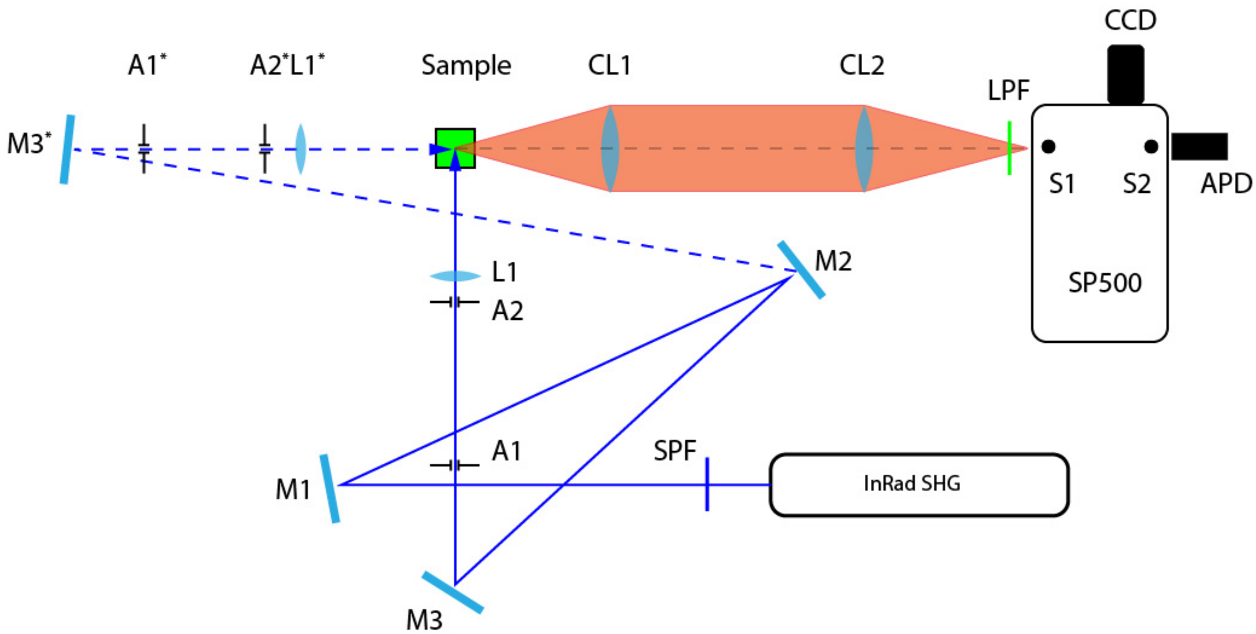

Figure 5. TCSPC measurements optics layout.

1. Set up the laser light source and the pulse picker as described in the previous chapter. Switch the pulse picker to CW operation mode to increase the visibility of the laser beam. Under normal conditions, the laser beam should be centered on mirror M1. Use a business card and NIR viewer scope to visualize the beam position. Normally, all beam steering mirrors (M1-M3) are broadband coated for the range of wavelength 380-700 nm. However, in many cases, they need not to be replaced for operation outside this spectral range, especially if the SHG unit is bypassed. Put a beam blocking metal screen anywhere between the sample holder and the SP500 spectrometer entrance slit.
2. Using the business card, center the laser beam on M2 by adjusting the angle of M1. Position M2 so that the reflected beam propagates towards M3 (or M3*, depending on

the geometry). The beam may need to be recentered on M2, if switching between 90^o and 0^O geometries.

3. There are two 4” postholders placed between M3 (M3*) and the sample holder: A1 and A2 (A1* and A2*). Also, an iris aperture on a post with alignment collar is available at the setup. When it is placed into a 4” postholder, the opening of the iris aperture is at the same height above the table surface as the optical axis of the spectrometer. Perform “dog leg” alignment of the laser by placing the alignment iris aperture at positions A1 and A2 (A1* and A2*) and recentering the beam on it by mirrors M2 and M3 (M3*), respectively. If 90^O geometry is used, make sure that the laser beam is blocked safely after the sample holder. Place the focusing lens in position L1 (L1*), so that the laser beam is perpendicular to it and centered.
4. Install an appropriate long-pass filter LPF in front of the spectrometer entrance slit. Use of interference filters is recommended due the absence of autofluorescnce and sharp transition from rejection to transmission. Typically, such filters should have 25-30 nm offset from the laser wavelength (i.e. for 400 nm laser, an interference filter with 430 nm cutoff wavelength is recommended). OCF has a set of filters for popular laser wavelengths.
5. Install an “alignment sample” into the sample holder. Any sample which produces visible, bright, and stable PL under the laser excitation can be used for alignment. Use of thin film samples is preferred because it enables one to achieve good confocality between excitation optics. Such samples are available at OCF or can be prepared by the staff. Set the spectrometer entrance slit to 500 μm and switch the output diverter mirror to the CCD camera. Open the LabView applet “SP500 control”, set the slew rate to 100 nm/min, select grating #3, and set the wavelength value to the center of emission spectrum of the alignment sample (otherwise, 600 nm is a reasonable setting for samples with visible PL). Make sure that the diverting mirror on the spectrometer is in the “CCD” position.
6. Set the pulse picker AOM driver to CW mode. In the Winspec software, set up full vertical binning acquisition mode (spectroscopy mode with the equivalent sensor dimension 1340x1), set the integration time to 0.3 seconds.
7. Position the sample so that the laser spot is approximately on the optical axis of the light collection system. Turn off the room lights (a desk light or a flash light can be used to navigate the lab). Start focusing (continuous) acquisition CCD camera mode in Winspec software and monito the signal magnitude.
8. Maximize the signal by adjusting the sample horizontal position in the direction perpendicular to the spectrometer optical axis using the micrometer on the sample stage (initial positioning may be done manually, and then the sample needs to be secured on the stage with a clamp). Once the maximum is found, adjust vertical

alignment knob of M3 (M3*), finally adjust the CL1 position along the optical axis of the spectrometer. Repeat the procedure several times. If the CCD camera saturates, one may use ND filters to attenuate the laser beam, reduce the SP500 input slit, or reduce the repetition rate of the laser by setting the pulse picker to the normal mode. Finally, the angle of the SHG crystal can be adjusted by the big knob on the SHG box while monitoring the fluorescence intensity.

9. Replace the alignment sample with the actual one and verify that the signal collection efficiency remains optimized.
10. Turn on the power supply for Advanced Micro Devices avalanche photodiode (APD). It will reach operating temperature in approximately 5 minutes. Energize the pulse counter. Enable APD with the switch located on the unistrat post. In the dark room or if the spectrometer diverter mirror is in “CCD” position, the pulse counter shoul read about 30-50 pulses per second. If the reading is exactly 0, the APD has not reached the operational temperature or the control switch has not been enabled.
11. Determine the wavelength where PL life-time has to be measured. Record the intensity value read by the CCD camera at this wavelength with 0.3 seconds exposure time. Set this wavelength in SP500 control applet. Put the spectrometer diverter mirror lever to “APD” position. Turn off the room overhead lights and unblock the laser when the APD is enabled. Observe the photon count rate (PCR) on the pulse counter display. The value should be of the same order of magnitude as the previously recorded spectral intensity value. If it is significantly lower, X and Y position of the APD may need to be adjusted.
12. APD position adjustment (to be performed by qualified personnel only, if the APD count rate is sufficient, do not do it.): Complete steps 1-11. Set the spectrometer wavelength to position corresponding to intense and stable emission in the sample. Observe PCR on the pulse counter display while adjusting APD X and Y translation knobs and maximize its value until it gets close to the spectral intensity value determined in the step 11. The knobs are located under light-blocking fabric on the top and left-hand side of the APD enclosure. They can be adjusted through the fabric, but this may be difficult. The fabric can be removed, but then the adjustments must be done in the completely dark lab. The fabric needs to be placed back after the adjustment. Adjust one knob at a time, switch to another knob only after finding the peak PR for the current one. Be careful not to interrupt the laser beam running under the APD enclosure with your hands or the fabric.

TCSPC Measurements:

1. Align the laser light source and the detection system as described in previous chapters.
2. TCSPC measurements can be done in the range ~1 ns – 5 μs. For longer time-scale, one needs to use photon counting multiscaler setup.
3. TCSPC measurements can be done in "reversed" and "standard" start-stop mode. The former mode has the maximum span of the transient of ~25 ns, which can be adjusted continuously. In contrast, the latter mode can have time spans equal to 500 ns, 1 μs, 2.5 μs, and 5 μs. Cabling configurations for these modes are shown in the Figure 6. Delay line – is about 30 feet long piece of RG-58 coaxial cable with BNC connectors. If it needs to be replaced any coaxial cable with 50 Ohm wave impedance will work. It has to be long enough to ensure about 25 ns delay of FPD signal relative to APD one.

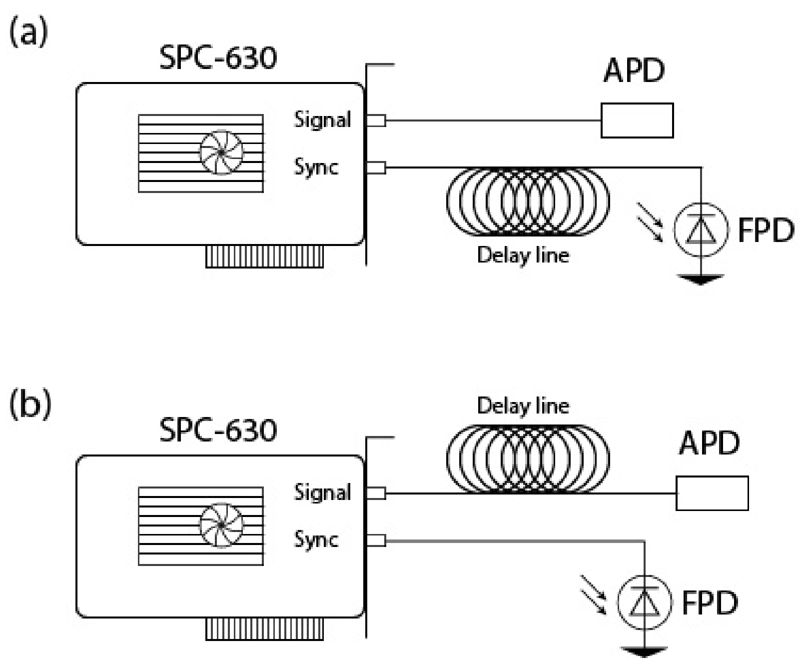

Figure 6. Configurations of TCSPC cabling: (a) "Reverse" and (b) "Direct" start-stop.

"Reverse" start-stop operation.

1. Set the cabling according to the Figure 6(a) after verifying that the pulse picker is operating correctly. Start SPCM software on the PC. Details on the SPCM software can be found in the Photon Counting Handbook by Becker and Hickl.
2. The screen shot of the SPCM software is shown in the Figure 7.

3. 4 bars in the lower left corner screen provide the key information regarding the system status:
   - The green bar (SYNC) displays the value of the trigger pulses rate. At all times, this value must be equal to the rate of laser pulses at the output of the pulse picker. If this value is different from the laser repetition rate, the sync threshold value needs to be adjusted. The typical range for this setting is -75 - -150 mV. If the sync rate stabilizes outside this range, verify that the amplitude of the SYNC pulse produced by the FPD is in this range, realign the beam splitter if necessary.
   - The black bar (CFD) displays the rate of photon counts selected by the constant fraction discriminator. The CFD value must be always less than that of SYNC rate. If the situation is the opposite, the APD may be exposed to intense light, malfunctioning, or the CFD limit is set too high (typical setting is about -120 mV). If CFD is configured correctly, the blue bar level should match the PCR counter readings. The fluorescence intensity must be attenuated so that the CFD rate is less than 2% of the SYNC rate.
   - The blue bar (TAC) shows the rate of pulses accepted by TAC converter. Under normal operations, its value should not exceed that of CFD. Adjust TAC settings to maximize it.
   - The red bar (ADC) indicates the number of photons falling into the range displayed in the window. Ideally, it should be close to TAC value. Adjust range, offset, and gain settings to optimize the ADC rate: With "reverse start-stop" configuration, initial range setting is usually 50 ns with gain setting equal to 2 which gives 25 ns temporal window width corresponding roughly to the start-stop delay provided by the delay cable. Adjust the offset value, so that the transient's onset is seen on the display and is close to the beginning of the time axis. If the transient duration is much shorter than the window width, the gain setting can be increased to increase the resolution of the measurement (The temporal window width decreases, the number of time bins does not change). Each time the gain is changed, the offset needs to be adjusted to return the onset of the transient to the beginning of the time axis. Optimize the gain and offset settings so that the transient spans over the most of the displayed temporal window.
   - Adjustment of the transient range, ADC offset and gain is performed visually by starting the transient acquisition with "Start" in the SPCM software. **<u>Pressing "Stop" interrupts the acquisition and the acquired transient will be destroyed if "Start" button is pressed again.</u>**
   - The TSCPC board setting are saved together with the data. One can recall the old settings by simply loading the saved transient.

- Usually the first and last 5% of the acquired transient are considered non-reliable and are blanked by TAC upper and lower settings (to 95% and 5% respectively) or during the data processing.

4. As mentioned in the previous paragraph, the fluorescence photon count rate in TCSPC experiments must be capped at 2% to avoid pile-up effects. There is no lower limit for the photon detection rate, for it can be compensated by increased acquisition time. However, practically the detected photon count rate should be at least similar to the dark/background signal of the APD. The latter can be evaluated by blocking the laser output and monitoring the APD counter/TAC signal. In long experiments, one may need to run the signal without sample with the same acquisition time to record the background noise profile.
5. The data collection time is determined by the desired signal-to-noise ratio and is limited by the photostability of the sample and the patience of the person collecting data.
6. The data are save in the *.sdt format which needs to be converted to an ACSII file for the processing. This is done by selecting "File -> Convert" menu items of the software. ASC output needs to be selected. It is recommended to use the LabView conversion software to normalize the data and correct the position of the time axis origin.

Shutdown procedures

1. Disable the APD output with the switch on the optical table.
2. Turn off the APD power supply.
3. Block the output of the Tsunami laser with a beam block.
4. Turn off the Verdi G laser in OPSL software, turn the key to the "off" position on its power supply, and turn off the main power switch on the back of the power supply.
5. Turn off the water cooling circulator.
6. Turn off Tsunami laser controller.
7. Turn off the pulse picker RF driver.
8. Turn off other equipment (oscilloscope, digital counter, CCD camera power supply).

Appendix 1.

NEOS AOM alignment for the pulse picker operation

Full alignment procedure is required if the AOM crystal has been removed or the pulse picker needs to be reset at a new location. Items in red are required only for the full alignment procedure. See Figure 1 in the SOP for the layout of optical components.

1. Set reference iris apertures A1 and A2 which define the laser beam direction and height relative to the optical table surface. Set 2 generic mirrors in a dog leg configuration and align a low power red laser (He-Ne) through them.
2. Set up another iris aperture A3 at about 30” from A2, so that the reference laser beam is centered on it. Using it as a reference, position a 50 mm focal lense biconvex lens L1 after A2, so that the laser beam is centered on it and it is perpendicular to the laser beam.
3. Position the AOM 4 axis stage, so that the focal point of the L1 is inside the TeO_2 crystal and the crystal face is perpendicular to the beam.
4. Position the biconvex lens L2 (f = 50 mm) after the AOM, so that it recollimates the beam passing through the AOM crystal and centers it on A3.

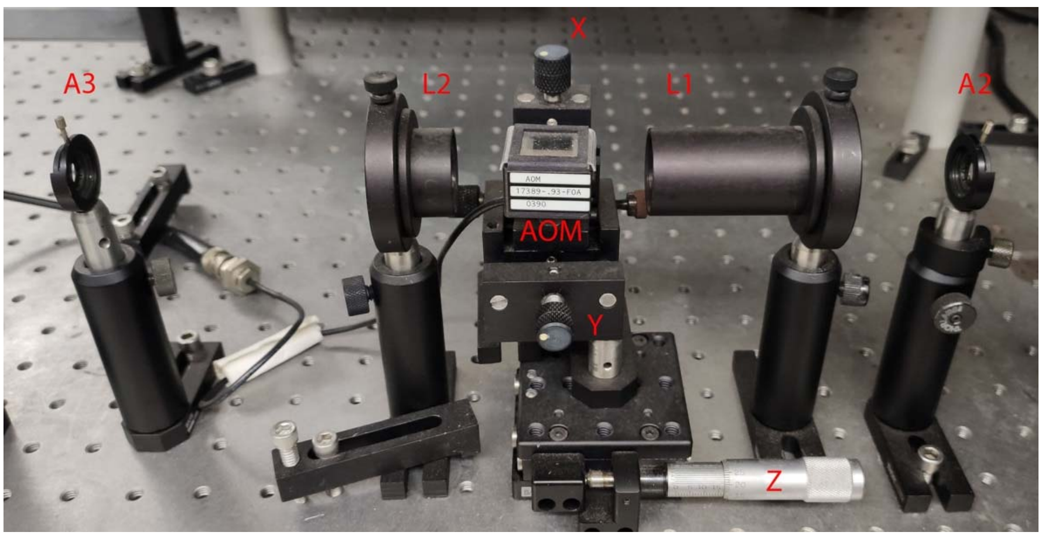

Figure A1. AOM positioning controls and side optics.

5. Turn on RF driver and set it to internal sync mode and enable continuous operation. Adjust X, Y, Z position of the AOM crystal to observe visually optimize the diffracted beam. Verify that the directly transmitted beam is not clipped by the AOM crystal.

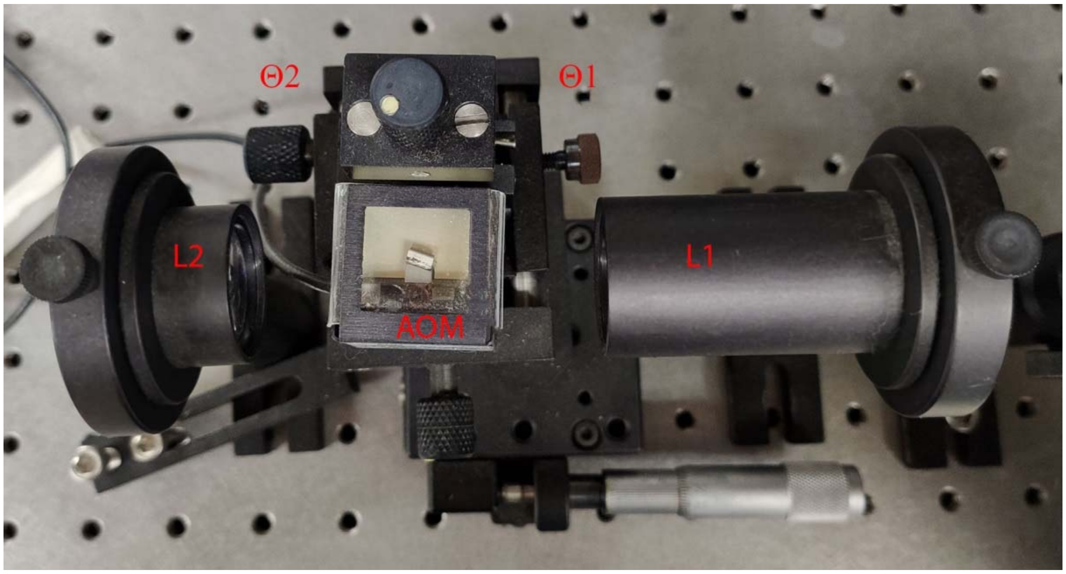

Figure A2. AOM top view.

6. Remove the alignment laser, place a beam block after A2 and align the Ti:Sapphire laser 800 nm beam through A1 and A2 using mirrors M1 and M2. The laser power before M1 should not exceed 350 mW.

7. Set the RF driver to synchronization with external detector and continuous diffraction mode. Verify that RF driver is in the "Loop-locked" mode with Tsunami laser. Attenuate the laser beam using OD1 neutral density filter. Place beam block BB2 after L2, so that it is blocking all beams passing through AOM crystal.
8. Remove the beam block placed after A2. Observe the shape of the beam transmitted through the AOM crystal after L2. Make **small** adjustments to X and Y position of the AOM crystal (in the plane perpendicular to the laser beam) and visually optimize the visibility of the diffracted beam. **Avoid hitting the edge of the crystal with the focused laser beam. This can damage crystal's electrical contacts which can be repaired only at the factory.** Make sure that the beam is not clipping inside the AOM cell. Remove the ND filter attenuating the laser beam.
9. Remove BB2 and place powermeter to measure the combined power of all beams coming from the AOM. Record the power value. Replace BB2 so that it blocks directly transmitted laser beam and does not block the diffracted beam. Place a powermeter after BB2 and measure the power of the diffracted beam. Calculate the diffraction efficiency. In the continious mode it should be in the range 15-25%.

10. Peak the power of the diffracted beam by carefully adjusting X and Y positions of the AOM crystal, then perform small adjustments of Z AOM's position (along the beam direction) and the crystal incidence angle. The latter is done by 2 screws which need to be rotated in opposite direction. During the alignment, monitor the beam shape on the power meter to avoid clipping of the beam. Iterate through XYZΘ adjustments until the target diffraction efficiency is achieved.
11. Place WP and BS2 after L2 and align the beam reflected from BS2 onto a fast photodiode FPD with bandwidth >200 MHz, NIR viewer scope is required for this. Using the fast oscilloscope triggered from RF driver optimize the FPD signal with BS2 alignment knobs.
12. Switch the RF driver to the single pulse mode, observe the single pulse electrical signal and optimize its magnitude by adjusting "Delay", "Phase", and "RF Power" knobs. Generally, the magnitude of the single pulse signal should be 2-3 times higher than that in the continuous operation mode.