4.4 ps IRF width of TCSPC with an NbN Superconducting Nanowire Single Photon Detector

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Abstract: An NbN superconducting nanowire single photon detector (SNSPD) was used to perform time-correlated single photon counting (TCSPC) with an instrument response function (IRF) width of 4.4 ps full width at half maximum (FWHM). The detector was developed collaboratively by MIT and the Jet Propulsion Laboratory (JPL), and the TCSPC module was a modified BH SPC-150 NX device. We demonstrate the resolution of the system by recording the fluorescence decay of IR 1061 (an infrared dye). The fluorescence lifetime of IR 1061 dissolved in dichloromethane was determined to be 43.7 ps.

Detector

Superconducting nanowire single photon detectors (SNSPDs) are high-performance single-photon counting detectors consisting of an ultra-thin superconducting strip biased by a DC current close the critical value. The absorption of a photon in the superconducting wire leads to a loss of superconductivity in the wire, increasing the resistance by several kilohms and shunting the bias current into a readout amplifier. More information about SNSPD technology and the detection mechanism can be found in [1]. SNSPDs have recently been demonstrated with a time resolution of 2.7 ps FWHM at 400 nm, and less than 5 ps FWHM time resolution at 1550 nm [2]. These ultra-low-jitter devices consist of NbN nanobridges which are 7 nm thick, 80 nm wide, and 5 µm long. While these proof-of-concept devices have negligble active area, it is predicted that larger devices which can be efficiently coupled to single-mode and multi-mode beams can still be engineered to have comparably low timing jitter by using a differential readout technique [3].

The detector described in reference [2] was used in a TCSPC experiment performed at JPL. In this experiment, the detector was cooled below 1 K using a closed-cycle Helium-4 sorption refrigerator, and read out using a low-noise SiGe cryogenic amplifier at the 4 K stage (Cosmic Microwave Technology CITLF1). The light from the experiment was coupled onto the detector through free-space windows and filters, using a lens located outside the cryostat, thereby avoiding the temporal dispersion introduced by fiber optics. SNSPDs have been used in the past for demonstrations of FCS [4] and singlet oxygen detection [5], but the detector-TCSPC combinations used in these experiments had timing jitter that limited the IRF width to 70 ps. Sub-30-ps IRF width of an SNSPD with a BH SPC-150 TCSPC device has been achieved by Toussaint et al. [8], 17 ps IRF width with a SNSPD from SCONTEL, Moscow [9] and a BH SPC-150N device [6].

TCSPC Device

For recording the shape of the light pulses we used a BH SPC-150 NX module [7]. The SPC-150 NX has input discriminators with 5 GHz bandwidth. Noise-optimized TAC-readout cricuitry results in an electrical IRF width of 3.5 ps FWHM. The minimum time channel width is about 400 fs. For the experiments described here, we modified the module by increasing the TAC transfer

ratio (time-to-voltage ratio) by a factor of two. This yields a minimum time channel width of 204 ps, and further reduces TAC readout noise. The electrical IRF of the module is shown in Fig. 1, left. The IRF width is 3 ps FWHM.

IRF Measurement

For the measurement of the optical IRF, we used a Calmar Mendocino sub-ps mode-locked fiber laser. The laser delivers pulses at 1064 nm wavelength, 50 MHz repetition rate, and an average power of about 1 mW. The pulse width is 800 fs. Photons reflected off a solid target were projected to the detector by a lens system. The light intensity was adjusted by a variable ND filter in the laser beam path. The photon pulses from the detector were connected to the 'CFD' (Start) input of the TCSPC module. Timing reference pulses were generated by a fast photodiode. The pulses were connected into the 'SYNC' (Stop) input of the TCSPC module. An IRF recorded this way is shown in Fig. 1, right. The IRF width is 4.4 ps.

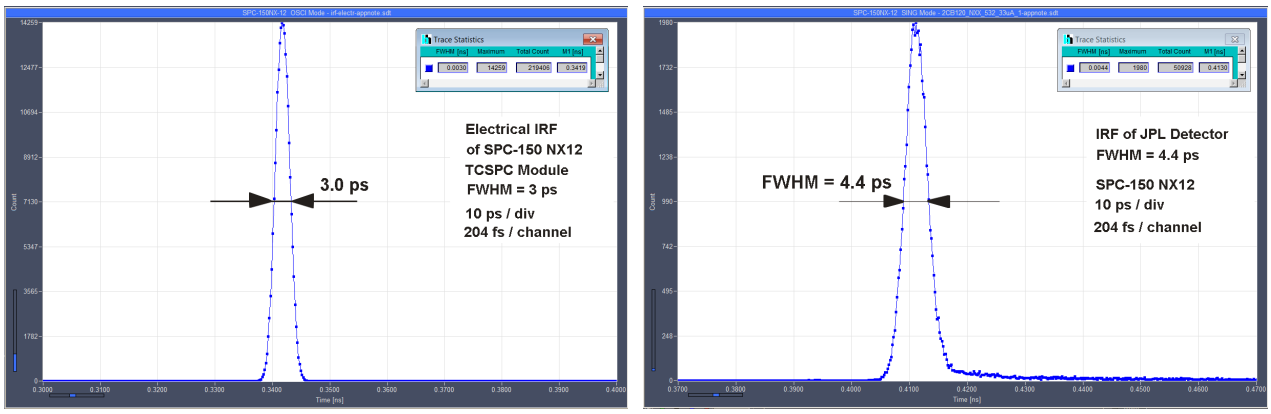

Fig. 1: Left: Electrical IRF of SPC-150 NX12 TCSPC module. Right: IRF of detector, including laser pulse width and possible synchronization jitter.

Fluorescence Decay Measurement

Fluorescence decay measurements were performed in the setup shown in Fig. 2. The dye solution was contained in a cuvette. We used a 10-mm glass cuvette that had walls of 4 mm thickness on two opposite sites. The horizontal cross section of the liquid volume is thus 2 mm by 8 mm. The laser beam was focused into the cuvette by an IR apochromate, L1. Fluorescence emitted by the dye solution is collected by another apochromate, L2. The beam is collimated by L2, and focused on the detector by an achromatic lens, L3. Two mirrors, M1 an M2, are used for alignment.

To avoid pulse broadening by transit time effects it is important that the laser beam is focused correctly into the center of the cuvette and that the fluorescence from exactly this spot is projected onto the detector. The detector then forms a confocal system with the excited spot. The effect is that out-of-focus detection is strongly suppressed. The suppression of out-of-focus light in this setup is in fact so good that a clean fluorescence signal is detected even without an emission filter.

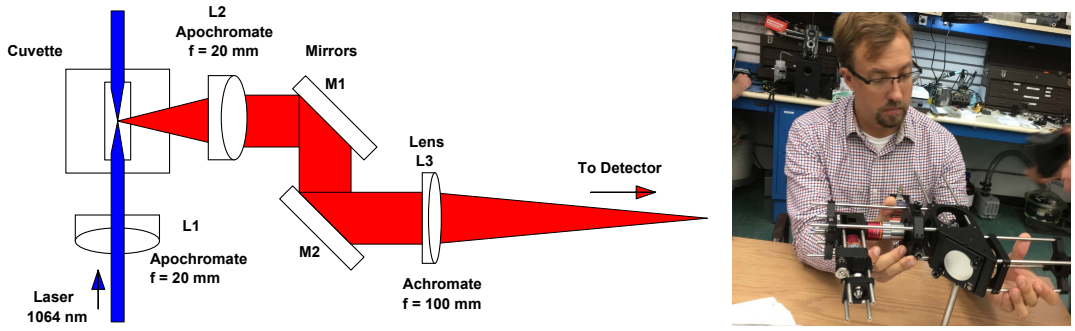

Fig. 2: Optical setup for fluorescence detection

For testing the setup we used a solution of IR 1061, an IR dye from Sigma Aldrich. The fluorescence lifetime of IR 1061 is shorter than 50 ps. With conventional fluorescence lifetime spectrometers the fluorescence decay cannot be reliably resolved. A decay curve recorded with the NbN detector described above is shown in Fig. 3. The time scale is 40 ps per division, or 400 ps over the entire observation-time interval. The resolution is so high that the curve almost looks like a nanosecond-fluorescence decay recorded by a 'normal' lifetime spectrometer.

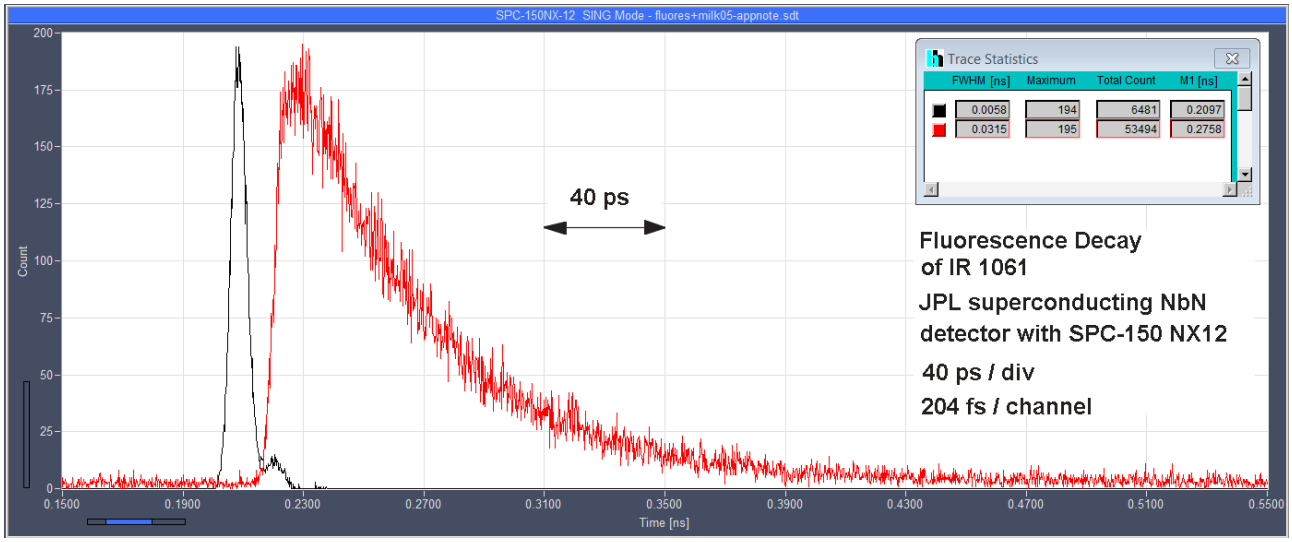

Fig. 3: Fluorescence decay of IR 1061. Black: IRF, measured in diluted milk. Red: Fluorescence

At the high time resolution of the system, transit time differences at the time scale of one ps and below become visible. For example, the fluorescence in Fig. 3 is visibly shifted against the IRF. The reason is that the IRF was determined in water with a refractive index of 1.33, but the dye was dissolved in dichloromethane, with a refractive index of 1.424. The transit times of the laser pulses over the path length of 5 mm within the solvent are 22.17 ps and 23.73 ps, respectively. The difference is 1.56 ps, which becomes visible in data with such high time resolution. Moreover, the IRF width measured in the cuvette is 5.8 ps, compared to 4.5 ps for the detection system itself. The broadening is explained by the fact that the fluorescence is detected from a non-zero sample volume: Already a longitudinal spread of 0.25 mm accounts to a pulse broadening of about 1 ps.

A single-exponential fit with bh SPCImage data analysis software is shown in Fig. 4. The fit delivers a fluorescence lifetime of 43.7 ps.

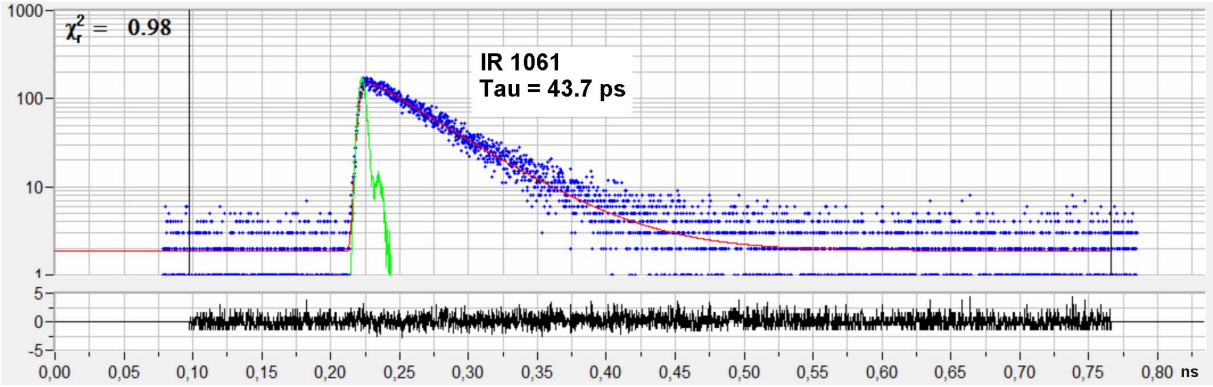

Fig. 4: Single-exponential fit with bh SPCImage data analysis software. Green curve shows IRF, blue dots data points of the fluorescence decay curve, red curve fit with single-exponential decay model. The fluorescence lifetime is 43.7 ps.

SPCImage includes a shift if the IRF against the decay data in the fit procedure. The fit of the IRF shift delivers a narrow χ^2 minimum because the fluorescence decay time is more than 7 times longer than the IRF width. It thus does not noticeably impair the fit accuracy. Under these conditions, a single-exponential fit with SPCImage delivers a (relative) standard deviation close to the theoretical value of $1/\sqrt{N}$ [71]. The standard deviation of the lifetime obtained from the IR 1061 data can thus be estimated from the number of photons in the photon trace. With 48.000 photons in the trace, the relative standard deviation is expected to be about 0.5 %.

Summary

To the best of our knowledge, the setup presented in this application note is the fastest TCSPC system ever described. With an IRF width of 4.4 ps, it easily resolves fluorescence lifetimes in the range of 50 ps and below. It also has the potential of resolving ultra-fast effects like protonation reactions or solvent-relaxation effects. Another application may be scanning of the surface profile of an object. The time resolution of 4.4 ps does, in principle, resolve depth differences of 1.3 mm with a single detected photon in each pixel. With 1000 photons per pixel the depth resolution would go down to about 40 µm. A frequent concern with NbN detectors is the low sensitivity due to the small detector active area. Although the quantum efficiency of the detector is close to one for the microbridge detector used in this experiment, only a fraction of the photons can be focused on the active area. The results presented above show the fluorescence decay functions are well detectable even with moderate focusing. Replacing the 100 mm lens in front of the cryostat by a 20 mm lens inside it would increase the efficiency by a factor of 25, which should be sufficient for a wide range of fluorescence experiments. Furthermore, it is anticipated that efficient, large-area SNSPDs with sub-5 ps time resolution can be engineered by using a differential readout to remove the geometric jitter [3].

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