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Tapered InP Nanowire Arrays for Broadband High-efficiency and High-speed Photodetection

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Detecting single photons is at the heart of quantum information and sensing technologies. Superconducting nanowire single-photon detectors have recently emerged with both broadband near-unity detection efficiency and unrivaled timing jitter; however, they are high cost and require cryogenic cooling. Here, we present a novel approach using tapered InP nanowire p-n junction arrays for high-efficiency, broadband and high-speed photodetection without the need of cryogenic cooling. Due to the truncated conical nanowire shape we achieve a broadband, linear photo-response from the UV to near-infrared with peak external quantum efficiencies exceeding 85%. In such devices ultrahigh gain beyond 10^5 is attained, such that a single photon per pulse can be distinguished over the dark noise, while simultaneously realizing a fast pulse rise time (<1 ns) and excellent timing jitter (<20 ps). With the flexibility to also easily change the nanowire material and

design for photodetection in the infrared in the future, such detectors open up new possibilities to enable a broader range of applications in remote sensing, dose monitoring in cancer treatment, 3D imaging and quantum communication.

Detection of light at the single photon level is advantageous for a wide range of applications in astronomy, sensing and biomedical engineering, as well as being a key driver of the rapidly developing field of quantum information science¹⁻³. Many photon starved situations benefit from high sensitivity detectors as well, for example in remote sensing of low power signals such as night time operation, identification and tracking of diffuse reflectors, communication of images from satellites at high speed and acquiring 3D images at long range with high resolution³⁻⁵. Indeed, the ability to detect light at the fundamental limit (i.e., single photons) enables observations over longer distances. Other possible applications in the longer term include singlet oxygen detection for dose monitoring in cancer treatment^{6,7}, quantum communication² and fault tolerant analysis testing of CMOS circuits^{8,9}.

Superconducting nanowire single-photon detectors have recently emerged to meet the demands of these applications for broadband high-efficiency, high-speed single-photon detection with ultra-precise timing resolution⁴; however, they are high cost and need to be cooled to cryogenic temperatures. We have developed a novel tapered nanowire p-n junction array that has the potential to overcome these drawbacks. Due to their unique size and shape, near-unity detection efficiency over a broad wavelength range from the UV to near-infrared at room temperature is possible. Operation at higher temperatures is advantageous for sensing applications requiring portability, for example, for use in the medical field or launching detectors on nanosatellites, where weight and space is at a premium¹⁰⁻¹².

A scanning electron microscope (SEM) image of an uncontacted nanowire detector array is shown in Fig. 1a, while a micrograph of a contacted nanowire array in cross-section is depicted in Fig. 1b. There are three important features of this nanowire detector design to achieve near-unity broadband photodetection efficiency. First, is the nanowire tapering; a conical nanowire structure with a smaller tip than the base. The tapering minimizes reflections of incoming light as a result of the adiabatic increase of the effective refractive index from vacuum ($n = 1$) at the nanowire tip towards bulk InP ($n = 3.4$) as the light propagates down the structure. Second, by gradually increasing the nanowire diameter towards the base, there is a continuum of available diameters from the top to bottom for the resonant coupling of guided modes over a broad wavelength range. Interestingly, this broadband absorption could be realized with a single nanowire. Finally, the nanowire pitch is optimized to maximize the absorption efficiency. The nanowire spacing should be far enough apart to ensure resonant waveguide modes are present, but close enough that light cannot pass through the nanowire array. This parameter requires optimization because if the nanowires are too close, reflections would increase due to an increase in the effective refractive index towards the bulk value, causing the high-efficiency broadband absorption to vanish.

Our InP tapered nanowire p-n junction detector devices were fabricated using a top-down dry etching approach which yields a uniform, ordered array of conical nanowires as shown in Fig. 1a. High quality planar p-n junction layers were first grown by metal-organic vapor phase epitaxy (MOVPE), followed by nanoimprint lithography and reactive ion etching (RIE). The shape of the top-down etched nanowires can be tuned by adjusting the dry etching parameters, such as the etchant flux, the ratio between the CH₄ and H₂ fluxes, the substrate temperature and the RF power. The optimized etching conditions resulted in a sloped sidewall profile, producing

truncated conical nanowires that are 1.6 μm long and are spaced by a 513 nm pitch, with a top diameter of 150 ± 10 nm and a bottom diameter of 350 ± 20 nm. Less than 20 % volume fraction as compared to the original planar film structure is occupied by the nanowires. A cross-section of the resulting tapered nanowire array with a top contact is shown in Fig. 1b. The black region visible between the nanowires is a layer of benzocyclobutene (BCB), which is used to planarize the sample prior to contacting the nanowires with indium tin oxide (ITO). This ITO layer is 300 nm thick and serves as the transparent top contact. Due to the deposition conditions the ITO layer naturally takes on a microstructure whereby the ITO increasingly accumulates around the nanowire tip. In addition, Fig. 1b shows how the space above the BCB is also filled with ITO, creating a continuous and conformal layer of ITO without spaces yielding improved contact resistance. The complete device structure from Fig. 1b is depicted schematically in Fig. 1c, while a picture of the completed detector package is shown in Figure 1d. Each black square corresponds to an independent 300 μm x 300 μm active device area which is surrounded by a 200 μm wide gold perimeter, serving as a ring contact used for wire bonding.

The dark visual appearance of the nanowire array (black square in Fig. 1d) indicates that the majority of light incident on the array is captured by the material. Indeed, vertically oriented nanowire arrays can be patterned and specifically arranged to act as near perfect absorbers^{13–17}. In a ray optics picture light should pass between the nanowires; however, experimental and theoretical studies have shown that given the correct choice of nanowire ensemble parameters such as length, diameter and pitch, near-unity absorption efficiency can be achieved. Moreover, arrays can be engineered such that the absorption is also wavelength, incident angle and polarization insensitive^{16–18}. Alternatively, device dimensions can be chosen such that the absorptance is highly wavelength selective. This wavelength selectivity is due to the fact that the

nanowires act as nanoscale dielectric waveguides. Light couples resonantly to guided modes of the nanowire depending on its diameter. Thus, by varying the nanowire diameter towards the base, multiple wavelengths can be absorbed with longer wavelengths being absorbed further down the nanowire as shown in Figs. 2a. This design results in near-unity absorption efficiency over a broad wavelength range, as well as locating the peak absorption position close to the embedded p-n junction (see Fig. S1 in the Supplementary Information).

This broadband high-efficiency absorption of the nanowire array is displayed by the solid green line in Fig. 2b, where using the fabricated device parameters including contacts in a finite-difference time-domain (FDTD) model predicts an average of 85 % absorption in the wavelength range from 450 to 900 nm. For comparison, without contacts, an average of 95 % absorption over the entire wavelength range is predicted. Only 6 % reflectance was measured from the uncontacted array (red curve, Fig. 2b), which confirms that reflected losses have also been minimized due to the tapered nanowire geometry. The device also exhibits excellent measured external quantum efficiency (blue curve, Fig. 2b), with a peak efficiency of 87 % at 860 nm and greater than 70 % throughout the entire wavelength range. These combined attributes of broad spectral coverage and high photoconversion efficiency also make these nanowire arrays an ideal platform for high performance multispectral photodetectors.

In the following, we investigate the photocurrent response of the nanowire avalanche photodiode and multiplication factor at room-temperature and the possibility of distinguishing a single photon per pulse over the dark noise. The results of this study are summarized in Figure 3. Fig. 3a shows the current-voltage characteristics of the device for varying incident optical power. The dark current is relatively low and is 25 pA at -1.0 V reverse bias, corresponding to 0.01 fA per nanowire or 10 nA/cm² normalized to the full contacted area. This dark current is at least six

orders of magnitude smaller as compared to bottom-up grown p⁺-i-n⁺ InP nanowire arrays¹⁹ or single wire devices which typically exhibit room-temperature dark currents on the order of tens to hundreds of pA per nanowire^{19–25}. The low dark current indicates the excellent quality of the array material, etched surface, and contacts. In avalanche photodetectors the shot noise from the dark current is a critical parameter as it can be the ultimate limiting factor for the device detectivity, which represents the smallest signal strength which can be successfully resolved. Considering the main contribution to the dark current is from thermally generated carriers, it can be significantly reduced by active thermoelectric cooling. In addition, the total dark current is proportional to the active device area, which can be minimized significantly in future work by reducing the detector size by two orders of magnitude, as our current device size is relatively large. Other strategies frequently employed in conventional thin-film InGaAs/InP APDs to reduce the dark current and afterpulsing effects can also be employed in future device designs, such as pulse or sinusoidal gating^{1,26,27}, fast active quenching^{28,29} or negative feedback structures^{30,31}.

With increasing reverse bias in Fig. 3a the current rises slowly until approximately -9.5 V where an abrupt threshold is reached and the current rapidly grows. The magnitude of the breakdown voltage ($V_B \sim -13$ V) signifies an avalanche breakdown mechanism in accordance with theory, whereby $V_B > 6E_g/q$ ³². This process occurs when the electric field in the junction reaches a high enough value such that photogenerated free carriers that are swept into the depletion region gain sufficient energy to cause impact ionization of bound carriers, effectively multiplying the current. Depending on the magnitude of the electric field and the impact ionization rates of the material, each carrier can undergo many subsequent ionization events, resulting in a self-sustaining avalanche. In most III-V materials, including InP, both carriers

participate in the multiplication process due to a small ionization rate ratio, $k=\alpha\beta$, between electrons and holes, denoted as α and β , respectively. We note that the avalanche breakdown mechanism was observed in 8 out of 12 of the devices studied. The other types of reverse bias characteristics that were measured are summarized in Supplementary Fig. S6.

The photocurrent response of the device in the photoconductive region, where there is no gain in Fig. 3b, exhibits a highly linear dependence in the average photocurrent as a function of excitation power at 700 nm. From the linear response exhibited by this device in Fig. 3b an internal quantum efficiency of approximately $\sim 73\%$ was extracted. Next, attenuating the incident optical power down to a single photon per pulse, the photocurrent remains easily distinguishable from the dark noise, as observed in Figure 3c. The photocurrent also remains linear in this low power regime, as shown in Fig. 3d, where the power density and photocurrent are presented as the number of photons and electrons per pulse. Remarkably, despite measuring at room temperature, more than 0.7 electrons per pulse are created in response to an illumination of a single photon per pulse. This result illustrates that even at room temperature the device demonstrates a remarkably high sensitivity, as required for single photon level detection.

The multiplication gain in the avalanche region can be estimated using:

$$M = \frac{I_{ph}(V) - I_{dark}(V)}{I_{ph(0)} - I_{(0)dark}}$$

where $I_{ph}(V)$ and $I_{dark}(V)$ are the measured excited and dark current, and $I_{ph(0)}$ and $I_{(0)dark}$ are the average measured excited and dark current in the low bias region with unity gain (*i.e.*, $M = 1$). The multiplication factor was calculated for an excitation of one photon per pulse and the result is given in Fig. 3e. A linear region exists from -10 V to -12 V where the gain rises steadily from a factor of 5 to 100. Approaching the breakdown threshold the gain rapidly increases,

reaching a remarkable current gain of greater than 10^5 at -12.75 V. This outstanding multiplication factor is to our knowledge the highest magnitude measured for a vertically oriented InP photodiode array and is consistent with the ultrahigh gain reported in previously horizontally contacted single InP nanowire devices^{21,23}.

The high multiplication gains reported here arise due to a reduction of dark current that develops with repeated measurement sweeps (see Supplementary Fig. S7). This is most likely caused by a charge accumulation layer at the nanowire surface, resulting in depletion of the nanowire core. This explanation is corroborated by recent work where deliberate electrostatic depletion of a single InP nanowire photodetector using ferroelectric enhanced side gates significantly reduced the dark current and improved the photoconductive gain²¹. Control over this process could prove highly advantageous to future device performance and stability.

A high-speed photodetector is an important metric for single-photon avalanche photodiodes as the rise and fall time determines the minimum intrinsic contribution to the total “dead time”. This is the amount of time required for the detector to return to a state where it is ready to register the arrival of another photon. In previous work of nanowire-based avalanche photodiodes, the high multiplication gain was not accompanied by a fast photocurrent response³³. Instead, single InP nanowire avalanche photodiodes with ultrahigh gain of 10^4 - 10^5 exhibited poor temporal response on the order of several to tens of milliseconds^{21,23}. This poor temporal response was attributed to a high overall RC time constant as a result of the high series device resistance and large parasitic capacitances in electrical contacting. In contrast, high bandwidth InP nanowire bipolar junction transistors demonstrated a fast photocurrent response with a full width at half maximum of 300 ps, but revealed a significantly lower gain factor of ~ 40 ²⁴.

Nanowires have also demonstrated persistent photoconductivity (PPC)^{33,34} in many materials including InP. This is the delayed relaxation of the photoexcited carriers due to charge trapping effects such that the device exhibits a long effective recombination time. However, we do not attribute our ultrahigh multiplication gain to a PPC effect as with several other reports^{33,35,36} since we have also simultaneously measured a rapid temporal response.

We now present the temporal photoresponse of our nanowire avalanche photodiode array as investigated in Figure 4. The 50 Ohm input on an oscilloscope with 500 MHz bandwidth was used to measure the photocurrent as a voltage pulse with no external amplification. In Fig. 4a, the photocurrent response is shown at an applied reverse bias of -10 V under pulsed excitation at 870 nm and power of 100 μ W with 76 MHz repetition rate. An apparent “ringing” in the output signal is observed, which may be solved in future work by maintaining radio-frequency impedance-matching standards throughout the circuit. A modified Gaussian fit of the pulse, shown in Fig. 4b, yields a 10 % - 90 % rise time of 0.6 ns and a full width at half maximum of 1.2 ns. This measured response time (τ_{meas}) is a slight overestimation of the intrinsic device temporal response (τ_{device}) since the response of the oscilloscope (τ_{scope}), the bias-T (τ_{bias-T}), and the laser pulse width (τ_{laser}) contributes to the total measured response time as: $\tau_{meas} = \sqrt{\tau_{device}^2 + \tau_{scope}^2 + \tau_{bias-T}^2 + \tau_{laser}^2}$. The temporal response also improves with increasing reverse bias as expected and is illustrated in Supplementary Fig. S9. This improvement is due to the increasing electric field and decreasing junction capacitance, which reduces the RC time constant of the device.

This rapid recovery time constant makes this nanowire array an excellent candidate for both high-speed avalanche photodetector functions such as optical communication interconnects and for applications in single photon counting modules operating in Geiger-mode exploiting the

ultrahigh multiplication gain regime. A low overall contribution from the contact resistance, estimated to be on the order of several Ohms³⁷, plays a major role in this favorable combination. In addition, the placement of the junction is close to the top of the nanowire, close to the area of highest optical absorption (see Figure 2a), significantly reducing the average transit time of the carriers in the low field drift region. It has also been shown that shrinking the volume of the avalanche region can improve the temporal response and dramatically reduce the excess noise^{38–40}. This is because a reduction in the multiplication volume results in a reduction in the effective ionization rate ratio, k_{eff} , of the microstructure as compared to the bulk material. Due to their nanoscale volume the k_{eff} extracted in measurements of nanowire avalanche detectors can be significantly smaller than that found in the bulk material^{41,42}. Furthermore, by employing a high-speed transmission line electrode design to contact the array, we expect this temporal response can be significantly improved. For example, a high-speed linear photodetector was fabricated from crossed InP nanowires contacted using an integrated coplanar waveguide signal line and a pulse FWHM of 14 ps was obtained²².

We highlight here that the speed of the detector is not the only parameter determining the ultimate temporal performance of the device. Another important consideration is the timing jitter, which represents the uncertainty in the arrival time of a detection event due to fluctuations in the output pulse. In a Geiger-mode photon counter this is generally discriminated using a fast voltage comparator, which signals when the leading edge of the avalanche current has passed a suitable threshold. The time from photon absorption to discrimination is not fixed due to spatial variation of the electric field and the finite transit time of the carriers within the volume of the device. A random equivalent-time sampling oscilloscope (LeCroy WaveExpert 100H) was used to acquire a persistence trace of the output pulse under the same excitation conditions.

Persistence mode superimposes many multiple subsequent waveforms on the same view, with more frequent data emphasised with deeper saturation or hotter colors as shown in Fig. 4c. This mode allows pulse noise and an upper bound on the intrinsic timing jitter of the photodiode to be visualized and estimated. A plot of the population histogram of the persistence data taken at 50% of the pulse rising edge was used to evaluate the inherent pulse jitter. A Gaussian fit to the histogram in Fig. 4d yields a FWHM value of 17 ps. The jitter in the output pulse also decreases with increasing reverse bias, as illustrated in Supplementary Figs. S10a-d. The inherent timing jitter of the APD is expected to depend strongly on the electric field, as high electric fields decrease the avalanche build-up time and the time uncertainty. This jitter is exceptionally low as compared to the total device jitter measured in InGaAs/InP APDs, which has more than 100 ps FWHM^{1,27,43}. We note that this quoted timing jitter in literature typically contains a contribution from the jitter of the quenching electronics as well. Therefore, our nanowire array detectors are well suited to use in high performance applications where precise photon arrival time is crucial, such as in quantum information science or time-of-flight surveillance measurements.

In conclusion, we have demonstrated the first tapered nanowire avalanche photodetector to demonstrate a combined near-unity multispectral absorption, ultrahigh multiplication gain, fast time response, and high timing resolution. Furthermore, this method can straightforwardly accommodate material and array engineering to optimize performance and extend operation to the infrared and beyond (illustrated for InGaAs in Supplementary Fig. S5). For example, InGaAs/InP^{1,44,45} or InGaAs/Si separate absorption and multiplication structures could be fabricated. In the latter case we envision that III-V nanowires on silicon benefits from the best of both materials whereby near-unity absorption in the infrared is achieved in the nanowire region and the multiplication occurs in silicon with ultra-low noise. Another method to investigate for

further improving excess noise and the time response are superlattice APD structures⁴⁶. Finally, distributed Bragg reflector structures⁴⁷ could further improve responsivity and act to filter unwanted wavelengths. These tapered nanowire avalanche detectors have the potential to be operated as fast linear avalanche photodetectors or Geiger-mode single photon counting modules with the addition of thermoelectric cooling.

Methods:

The III-V material stack, including p-n junction layers was grown on a Zn-doped (100) InP wafer by MOVPE. The nanowire array was then defined using a combined nanoimprint lithography and RIE technique. Particular effort was committed to developing additional process steps to ensure excellent sidewall smoothness and surface quality. See the Supplementary Information 1 for further details on the fabrication process. The reflectivity of the bare nanowire array was measured using an integrating sphere. The device was then planarized using a BCB layer and contacted using 300 nm of ITO. The completed device was epoxied to a standard ceramic chip carrier and the active areas were wire bonded to the carrier pins. The absorptance of the completed device structure was simulated using the finite-difference time-domain (FDTD) method via Lumerical optical software. Further details on the simulation and optimization process can be found in Supplementary S2.

The absorbed photon number was estimated as $\frac{P}{\frac{hc}{\lambda}} \times A \times \frac{1}{f}$ where P is the excitation power, A is the (simulated) absorption coefficient, f is the laser repetition rate and λ is the wavelength. The incident optical power was attenuated to the single photon per pulse level using a combination of stacked neutral density filters and a continuous variable filter wheel to achieve an effective power of 15, 30, 45, 60 and 75 pW focused on the device. The power density was monitored using a 90:10 fiber optic beam splitter installed in the optical path after the filter

wheel, and the power ratio between the two paths was first calibrated at higher incident power levels.

The chip carrier was mounted on a printed circuit board adapter with 50 Ohm SMA connectors. The assembly was then mounted onto an XYZ optical stage, and a 50 mm focal length lens was used to focus the laser excitation onto the active area of the device. The stage was then fine adjusted to give the maximum photocurrent at 0 V. A Keithly 6485 picoammeter was used to perform the dark current and photocurrent measurements.

Current-voltage measurements were taken in single sweeps, sequentially from low to high power without repeated sweeps in between in an effort to minimize dark current shifts due to either light exposure or multiple sweeps and yield the best comparison. The sweep direction was always from positive to negative voltage 0.75 V to -13.0V. The reading rate of the instrument was set to 1 power line cycle or 16.6 msec with a measurement delay of 10 msec. For temporal measurements with pulsed excitation, an RF bias tee (MiniCircuits ZX85-126-5+) was used to apply the DC reverse bias to the photodiode and extract the photocurrent pulse, which was measured using SMA cables connected to the 50 ohm input of a LeCroy WaveExpert 100H random equivalent time sampling oscilloscope.

Photocurrent measurements were performed using a Picoquant PDL800D pulse laser driver with a LDH-P-690 head operated at 690 nm and 40 MHz or a Coherent Mira 900 Ti:Sapphire 3 ps pulsed laser operated at 870 nm and 76 MHz , as specified in the text. The laser power was calibrated using a 90:10 fiber beam splitter with neutral density filters on the signal arm and a power meter monitoring the 90 % arm.

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Supplementary Information is linked to the online version of the paper at www.nature.com/nature.

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Author contributions

Y. Cui fabricated the devices. S.J. Gibson performed the photocurrent testing. S.J. Gibson and M.E. Reimer wrote the manuscript. D. Van Dam and B. Van Kasteren performed the optical measurements and simulations. M.E. Reimer supervised the work. All authors have given approval to the final version of the manuscript.

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Competing financial interests

The authors have no competing financial interests to declare.

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Figures

Figure 1. Tapered nanowire array device design. a) A scanning electron micrograph of the InP nanowire array after etching, imaged at a tilt angle of 30° from the substrate normal. b) Cross-sectional view of the completed device, showing the nanowire tapering, planarizing benzocyclobutene (BCB) layer (black region between the nanowires) and the deposited ITO top contact layer. c) A schematic of the full device structure from Fig. 1b also illustrating the rear contacting layers. d) Photograph of the device mounted and wire bonded to a standard ceramic chip carrier. There are approximately 35 devices (5 x 7) on this chip with individual gold contacts surrounding the nanowire array with an active device area of $300 \times 300 \mu\text{m}$.

Figure 2. Near-unity absorption. a) The FDTD simulated absorbance located along a contacted nanowire for I) 500 nm, II) 700 nm and III) 890 nm wavelength excitation, showing the position of peak absorbance close to the p-n junction location, 200 nm from the nanowire tip. b) Simulated absorbance (green curve) of the completed nanowire ensemble device. The patterned InP nanowire array with conical shape exhibits high absorbance over a broad spectral range from 400-900 nm (peak efficiency of 87 %, $\sim 85\%$ average from 450 nm to 900 nm). Reflected losses (red curve), measured using an integrating sphere, averaged less than 6% in an uncontacted nanowire array. The measured external quantum efficiency (blue curve) at 0 V shows a similar qualitative behavior to the absorbance, maintaining a value higher than 70%. Short wavelengths are absorbed in the ITO contact layer, while wavelengths longer than the InP bandgap are transmitted.

Figure 3. Photocurrent response and avalanche photodetection. a) Avalanche threshold voltages have been measured at 300 K in reverse bias under continuous wave excitation at 700 nm. b) The sub-avalanche photocurrent response, measured at -1.0 V bias, is highly linear over a large dynamic range from microwatts down to picowatts. c) By attenuating a 40 MHz pulsed laser excitation (3 ps pulsewidth) at 690 nm down to a single photon per pulse, the average

photocurrent signal can be distinguished above the dark current. d) Sub-avalanche photocurrent response measured under pulsed excitation at 690 nm and -1.0V reverse bias in the single photon regime. For this device, an internal quantum efficiency (IQE) of 73% was achieved at room temperature, corresponding to more than 0.7 electrons per pulse under an illumination of a single photon per pulse. e) The calculated multiplication gain in the avalanche regime at ambient room temperature. This curve corresponds to an average of one photon per excitation pulse.

Figure 4. Temporal response. a) Output voltage pulse measured at a reverse bias of -10 V under pulsed excitation at 76 MHz repetition with 870 nm and power of 100 μ W. b) Fitting the pulses using an exponentially modified Gaussian function, rise times of less than 0.6 ns and FWHM of less than 1.2 ns were obtained. c) A “persistence” plot of the photocurrent pulse

waveform under 76 MHz excitation at a power of 100 μ W is provided for a reverse bias of -10 V. d) The FWHM of the histogram, taken at 50% of the pulse rising edge from (c) was used to estimate an inherent timing jitter of 17 ps.