Ultrasensitive N-Photon Interferometric Autocorrelator

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We demonstrate a novel method to measure Nth-order (N = 1, 2, 3, 4) interferometric autocorrelation with high sensitivity and temporal resolution. It is based on the combination of linear absorption and nonlinear detection in a superconducting nanodetector, providing much higher efficiency than methods based on all-optical nonlinearities. Its temporal resolution is only limited by the quasiparticle energy relaxation time, which is directly measured to be in the 20 ps range for the NbN films used in this work. We present a general model of interferometric autocorrelation with these nonlinear detectors and discuss the comparison with other approaches and possible improvements.

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The temporal correlation functions of various orders are of fundamental importance in the classical and quantum description of optical fields. The first-order (field) autocorrelation function describes temporal coherence and therefore spectral linewidth, and second-order (intensity) autocorrelation is used to measure the temporal properties of pulsed sources and to distinguish quantum and classical fields, whereas the measurement of higher-order autocorrelation is more sensitive to coherence features (e.g., photon bunching) of the light field [1] and can be used to determine the asymmetry of light pulses [2]. Whereas the first-order autocorrelation function is easily measured using an interferometer and a linear detector [3], the measurement of higher-order correlation functions requires a process that is nonlinear in the intensity I(t). In interferometric autocorrelators, the normalized second-order correlation function g(2)(τ) = ⟨I(t)I(t + τ)/I(t)⟩ is usually measured by using either second-harmonic generation (SHG) in a nonlinear crystal, followed by a linear detector [4], or two-photon absorption (TPA) in the detector itself [5,6]. In both cases, the detector measures the square of the total intensity at the output of the interferometer ⟨I^2(t)⟩ = ⟨I(t)⟩ + ⟨I(t)⟩ [2].

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The device used in this work is based on a 4.3 nm-thick NbN film (critical temperature T_c = 10.2 K) sputtered on GaAs substrate and has a constriction size of about 150 nm, patterned by electron beam lithography and reactive-ion etching. The nanodetector is biased with a current I_b smaller than the superconducting critical current I_c. Similarly to nanowire superconducting single-photon detectors [12], the absorption of one or more photons produces a nonequilibrium population of QPs in the nanodetector’s active region, locally suppressing the superconductivity and increasing the probability of vortex crossing, which can result in the transition to the normal state [13,14].
the absorbed energy (i.e., to the number of photons) and of the bias current \( I_b \) so that the nanodetector can be set to respond to \( \geq N \) photons by choosing \( I_b \) [15,16].

In this study, we focus on the temporal characteristics of the nanodetector’s multiphoton response and on its application as an interferometric autocorrelator. As we focus here on the application to the characterization of classical light sources, we describe the detection process as a sequence of single-photon absorption events using the semiclassical photodetection theory. Assuming an incident light pulse with cycle-average intensity \( I_m(t) \) on the nanodetector, the probability of creating a hot spot within a time interval \( (t, t + dt) \) is equal to \( \zeta \Sigma I_m(t) dt \), where \( \zeta \) is related to the absorbance \( \eta_{ab} \) by \( \zeta = \eta_{ab} / h \nu \) and \( S \) is the active area.

In the two-photon regime, for example, the click probability \( P_{\text{click}} \) (assumed \( \ll 1 \)) is equal to the probability that two photons are absorbed, weighted by a function \( \eta(\tau_{12}) \) of the time difference \( \tau_{12} \) between absorption of the first and the second photon at times \( t_1 \) and \( t_2 \), respectively,

\[
P_{\text{click}} = \int_{-\infty}^{+\infty} \int_{-\infty}^{+\infty} \eta(t_2 - t_1) \zeta \Sigma I_m(t_1) \zeta \Sigma I_m(t_2) dt_1 dt_2 = \zeta^2 S^2 \int_{-\infty}^{+\infty} \eta(\tau_{12}) \int_{-\infty}^{+\infty} I_m(t) I_m(t + \tau_{12}) dt d\tau_{12}.
\]

(1)

The \( \eta(\tau_{12}) \) function, named the nonlinear response function (NRF) in the following, depends on the QP dynamics in the superconductor and determines the intrinsic response time \( \tau_{ND} \) of the nanodetector. The value of \( \eta(\tau_{12}) \) is expected to decay from a maximum value of \( \eta(0) \) (which depends on \( I_b \)) to 0 for \( \tau_{12} \gg \tau_{ND} \). In general, the detection probability in the \( N \)-photon regime (\( N \geq 2 \)) is

\[
P_{\text{click}} = \zeta^N S^N \int_{-\infty}^{+\infty} \eta_N(\tau_{1N}, \tau_{2N}, \ldots, \tau_{N-1,N})
\times \int_{-\infty}^{+\infty} I_m(t) I_m(t + \tau_{1N}) I_m(t + \tau_{2N}) \ldots
\times I_m(t + \tau_{N-1,N}) dt d\tau_{1N} d\tau_{2N} \ldots d\tau_{N-1,N}.
\]

(2)

where \( \tau_{iN} = t_N - t_i \) denotes the difference between the absorption times of the \( i \)th and the last photon.

When the incident pulse width is much larger than \( \tau_{ND} \), Eq. (2) is approximated as

\[
P_{\text{click}} \approx \int_{-\infty}^{+\infty} \eta_N(\tau_{1N}, \tau_{2N}, \ldots, \tau_{N-1,N})
\times \int_{-\infty}^{+\infty} I_m(t) I_m(t + \tau_{1N}) I_m(t + \tau_{2N}) \ldots
\times I_m(t + \tau_{N-1,N}) dt d\tau_{1N} d\tau_{2N} \ldots d\tau_{N-1,N}.
\]

In the two-photon regime, for example, after filtering out the interference terms, Eq. (1) becomes

\[
P_{\text{click}}(\tau_d) \approx \eta(0) + \eta(\tau_d) + f(\tau_d) \text{ (see Supplemental Material [17]).}
\]

The NRF can be measured by probing the autocorrelator with short pulses. For a pulse duration much shorter than \( \tau_{ND} \), the response of the nanodetector placed at the output of a Michelson interferometer can be found from Eq. (2).

The NRF was first measured by sending 1.6 ps pulses from an optical parametric oscillator (OPO) at \( \lambda = 1.13 \mu m \) into a fiber-based Michelson interferometer and then to a nanodetector held at a temperature of 1.2 K using a lensed fiber producing a spot with an \( e^{-2} \) diameter of \( 5 \mu m \). The delay \( \tau_d \) in the interferometer is controlled by a motorized delay line (coarse control) and a fiber stretcher (fine control). The \( I_c \) of the device was about 26 \( \mu A \). During the measurement, the nanodetector was set in different photon regimes by choosing a proper \( I_b \). As shown in Fig. 2(a), the count rate (CR) was measured as a function of the light power at different \( I_b \) values. The solid lines with slopes of 1.04, 2.06, 3.06, and 3.99 are the fits to the measured data.
in log-log scale in the power ranges where the one-, two-, three-, and four-photon response was dominant [15]. We chose one point at each of the four photon regimes and measured the CR as a function of \( \tau_d \) normalized by its values at long delays, as shown in Fig. 2(b). The data points near \( \tau_d = 0 \), where the measured autocorrelation is sensitive to the first-order coherence, are not shown in the plot and were not considered in the fit since they introduce additional fitting error (see Supplemental Material [17]). At \( I_p = 18.0 \) \( \mu \)A, the CR is independent of the delay since the detector is working in the linear regime. When the \( I_p \) was lowered to 12.5, 9.7, and 8.4 \( \mu \)A (corresponding to the two-, three-, and four-photon regimes, respectively), a maximum was observed at zero delay. Since the width of these peaks is much wider than the OPO pulse width, the measurement probes the intrinsic response of the detector. In particular, the two-photon normalized response in Fig. 2(b) directly provides the \( \tau_{\text{ND}} \) value in the \( N \)-photon regime, since the \( \tau_{\text{ND}} \) is determined by the thermalization and relaxation processes of the photocreated QP and by the functional dependence of the CR [13,14]. Indeed, the QP population first grows as the electron population thermalizes via electron-electron scattering, in a time scale of a few ps, then decays due to electron-photon interaction and phonons escaping to the substrate [18]. This QP decay is expected to determine the \( \tau_{\text{ND}} \) value in the \( N \)-photon regime, since the \( I_p \) is chosen so that \( P_{\text{click}} \) is high only when all QP produced by the \( N \) photons are present at the same time. As \( P_{\text{click}} \) is a strong function of the \( N \) number, involving many microscopic parameters, a fit of the measured \( \eta(\tau) \) using a microscopic model would not be reliable. Instead, we introduce an empirical Gaussian NRF defined as

\[
\eta(\tau) = \eta(0) \exp\left(-\frac{(\tau_{12}/\tau_{\text{ND}})^2}{2}\right).
\]

The fits to 10 measured two-photon autocorrelation traces provide a \( \tau_{\text{ND}} \) value of 20.4 \( \pm \) 0.8 ps [19]. To fit the \( N > 2 \) traces, we further assume that the multiphoton response factorizes as

\[
\eta_N(\tau_1, \tau_2, \ldots, \tau_{N-1}, N) = \eta(\tau_1)\eta(\tau_2) \ldots \eta(\tau_{N-1}, N).
\]

This is reasonable if one assumes that \( \eta_N \) has an approximately exponential dependence on the total QP concentration after absorption of the \( N \)th photon, as suggested by the vortex-assisted photodetection model [13,14], and that the QP relaxation time does not depend on QP concentration. Using the \( \tau_{\text{ND}} \) value extracted from the two-photon autocorrelation traces as described above, \( P_{\text{click}}(\tau_d) \) was calculated for the three- and four-photon regimes from Eq. (2) without additional fitting parameters and shows excellent agreement with the experiment [Fig. 2(b)], which provides strong experimental support to our model.

With knowledge of the temporal resolution, autocorrelation experiments were performed on pulses generated by a gain-switched 1.3 \( \mu \)m diode laser with 10 MHz repetition rate and about 70 ps pulse width. The one-, two-, three-, and four-photon regimes were first found by choosing different \( I_p \) values. At each \( I_p \), CRs were recorded as a function of \( \tau_d \) in the \( N \)-photon \( (N = 1, 2, 3, 4) \) regime as shown in Figs. 3(a)–3(d), respectively. The fringe contrast ratio is observed to increase with \( N \), in good agreement with the theoretical values of 2, 8, 32, and 128 for \( N = 1, 2, 3, \) and 4, respectively. In the one-photon regime [Fig. 3(a)], the normalized first-order autocorrelation function \( g^{(1)}(\tau_d) \) was calculated from the visibility of the interference fringes [3]. For \( N > 1 \), higher-order intensity autocorrelation traces were obtained by applying a low-pass filter to the interferograms in Figs. 3(b)–3(d). In order to explain the experimental results, we assumed that the incident light was a Gaussian pulse with a linear chirp. The electric field is modeled as

\[
E(t) = E_0 \exp\left[-4\ln(2)(1 + iA)t^2/\tau_p^2\right],
\]

where \( E_0 \) is the field amplitude, \( A \) is the linear chirp parameter [22], and \( \tau_p \) is the full width at half maximum (FWHM) of the pulse. By fitting the measured \( g^{(1)}(\tau_d) \) in Fig. 3(a) together with the second-order intensity autocorrelation (low-pass trace) in Fig. 3(b), \( A \) and \( \tau_p \) were determined to be 5.3 and 70.6 ps. The third- and fourth-order interferometric autocorrelations were then calculated based on
Eq. (2) without additional fitting parameters. Their low-pass traces show a good agreement with the experiments, considering the very simplified assumption for the chirp. An enlarged view of each interferogram is shown as an upper-right inset in each panel of Fig. 3, showing a clear narrowing of the fringes for increasing $N$. As shown in the lower-right inset of Fig. 3(d), the FWHM of the fringes normalized by their period scales as approximately $1/\sqrt{N}$, which is a characteristic of multiphoton interferometry [23] and further confirms our conclusions.

The nanodetector-based autocorrelator provides much higher sensitivity as compared to conventional autocorrelators. The two-photon autocorrelation trace in Fig. 3(b) was taken at $P_{pk}P_{av} = 5.6 \times 10^{-17} \text{W}^2$ ($P_{pk}$ is the peak power, and $P_{av}$ is the average power), about 7 orders of magnitude lower than the minimum reported $P_{pk}P_{av}$ based on TPA [24] and about 2 orders of magnitude lower than the lowest $P_{pk}P_{av}$ using SHG [25]. For input pulses longer than $\tau_{ND}$, Eq. (2) can be written as $P_{lick} = C_{ND} \int_{-\infty}^{+\infty} P_{in}^2(t)dt$, where $P_{in} = I_{in}S$ is the incident power and $C_{ND} = \sqrt{\pi \eta_{abs}} S C_{pop}/(h\nu)^2$ represents a nonlinear response efficiency. Using the measured values of $\eta_{abs} = 1.5 \times 10^{-4}$, $\eta(0) = 0.5$ [16], and dark count rate $R_{dark} = 1 \text{ Hz}$, we derive a sensitivity of $I_{pk}^{-1}$ $P_{av}^{-1} = 5.8 \times 10^{-20} \text{W}^2$ (see Supplemental Material [17]), corresponding to $\sim 4$ photons/pulse in our experiment. In a higher-photon regime, the advantage of using linear absorption is even larger. Indeed, the three-photon autocorrelation shown in Fig. 3(c) was performed at an average power of about 1 nW, corresponding to $P_{pk}P_{av} = 2.0 \times 10^{-21} \text{W}^3$, an improvement of about 21 orders of magnitude over that in Ref. [26]. To the best of our knowledge, $N$-photon interferometric autocorrelation for $N > 3$ has not been reported before. As compared to autocorrelators based on spatial coupling of the optical beam to multiple single-photon detectors and electronic correlation [27], our nanodetector provides higher temporal resolution, much easier readout, and phase information.

The very high nonlinear response of the nanodetector can be directly traced to the finite size and time duration of the hot spot created by the real absorption of one photon, as compared to the virtual transitions involved in TPA. Indeed, a two-photon detection is triggered if the second photon is absorbed within the volume and time duration of the hot spot created by the first photon. This shows that a compromise exists between $C_{ND}$ and $\tau_{ND}$: for the nanodetector, the $\tau_{ND}$ value is determined by the QP relaxation time while in TPA it is related to the lifetime of the virtual states associated to the TPA transition, on the order of femtoseconds [6]. A similar compromise exists in SHG-based autocorrelators, where higher conversion efficiency requires a longer SHG crystal translating into a smaller phase-matching bandwidth and lower temporal resolution $\tau_{res}$ [28,29]. Defining $P_{pk}P_{av}/\tau_{res}$ as a figure of merit, the present nanodetector is about 2 orders of magnitude better than the record using TPA [24] and comparable with the record based on SHG, where much higher experimental complexity is required [25]. We note that a key advantage as compared to SHG-based autocorrelators is the nanodetector’s large wavelength range, limited only by the requirement to operate in the desired $N$-photon regime, which can be easily adjusted by varying the $I_b$. This, in principle, enables the measurement of interferometric autocorrelation from the visible to the mid-infrared wavelengths.

The sensitivity of our autocorrelator is presently limited by the low $\eta_{abs}$ value related to the spatial mismatch between the incoming beam and the nanodetector’s active area and to the small thickness of the NbN film. By focusing the beam with a high-numerical aperture lens, we can achieve a much higher absorbance $\eta_{abs} = 10^{-2}$ [30]. The integration of a plasmonic antenna and a bottom reflector [31] could increase $\eta_{abs}$ to the $10^{-1}$ range, leading to $P_{min} = 10^{-25} \text{W}^2$ for the two-photon autocorrelation. On the other hand, increasing the detector area (as done in meander nanowire detectors [32]) results in $\eta_{abs} \propto L$ and $\eta(0) \propto \lambda_{bs}/L$ ($L$ is the nanowire length, and $\lambda_{bs}$ is the hot spot length) so that $C_{ND}$ scales linearly with $\eta_{abs}$. The nanoscale nature of the detector is therefore crucial to reaching the ultimate sensitivity. Finally, we note that the 20 ps temporal resolution in our experiments is limited to the QP relaxation time in NbN films and could be much improved using different superconducting materials, such as high-$T_c$ Y-Ba-Cu-O films, where relaxation times $\sim 1 \text{ps}$ were observed [33], opening the way to the characterization of $N$th-order correlation functions in a few-ps range with unprecedented sensitivity.

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*Note added.—* After submission of this Letter, a QP relaxation time of about 15 ps, measured using a different experimental technique in NbN meander nanowire detectors on a different substrate, was reported in Ref. [34].

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[10] The fast correlation technique demonstrated in Refs. [1,11] also belongs to this category but suffers from the low sensitivity of streak cameras in the near-infrared region.
[19] This value is comparable but shorter than the hot-electron energy relaxation time in a NbN microbridge measured by electro-optic sampling [20] and by terahertz spectroscopy [21]; this difference may be related to the difference in film thickness and film properties.
[30] This value is obtained by assuming that the diameter of the Gaussian beam spot decreases from 5 to 0.5 μm by using a high-numerical aperture lens and the nanodetector’s active area is 150 nm × 150 nm.