Photon correlation in single-photon frequency upconversion

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Abstract: We experimentally investigated the intensity cross-correlation between the upconverted photons and the unconverted photons in the single-photon frequency upconversion process with multi-longitudinal mode pump and signal sources. In theoretical analysis, with this multi-longitudinal mode of both signal and pump sources system, the properties of the signal photons could also be maintained as in the single-mode frequency upconversion system. Experimentally, based on the conversion efficiency of 80.5%, the joint probability of simultaneously detecting at upconverted and unconverted photons showed an anti-correlation as a function of conversion efficiency which indicated the upconverted photons were one-to-one from the signal photons. While due to the coherent state of the signal photons, the intensity cross-correlation function $g^{(2)}(0)$ was shown to be equal to unity at any conversion efficiency, agreeing with the theoretical prediction. This study will benefit the high-speed wavelength-tunable quantum state translation or photonic quantum interface together with the mature frequency tuning or longitudinal mode selection techniques.

OCIS codes: (040.3060) Infrared; (190.7220) Upconversion; (230.0040) Optical devices.

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1. Introduction

Quantum manipulation of photonic qubits by nonlinear frequency conversion has been the research focus for a broad range to fundamental and practical applications in quantum information technology. For example, entangled photon pairs or mid-infrared sources can be generated by spontaneous frequency down-conversion [1–7], and four-wave mixing in photonic crystal fiber is convenient for telecom wavelength correlated photon pair generation [8–12]. Especially, single-photon frequency upconversion provides a new method for sensitive detection of infrared photons based on the sum-frequency generation (SFG) [13–16]. Theoretically, the infrared single-photon level signal could be upconverted to visible region with near unity conversion efficiency by strong pump field and high nonlinear coefficient crystal [17]. Then the visible single-photon detectors can handle with the infrared photons with high detection efficiency. Recently, the researches realized high efficient single photon frequency upconversion system by several schemes and led to various applications. The frequency upconversion detectors have provided the technical support for the long distance quantum key distribution over 100 km [18]. Meanwhile, the tunable upconversion photon detectors were expected to apply to the DWDM optical communication systems [19]. And the ultrasensitive spectrometer based on the upconversion detection was also developed for spectrum analysis of infrared signals at single photon level [20]. More significantly, this nonlinear sum-frequency generation process at single-photon level drew attention to the quantum features of the signal photons. It has been demonstrated that the quantum character of single photons from a semiconductor quantum dot could be maintained in the upconversion process [13]. And the photon number distribution of the infrared signal photons were analyzed by the visible photon number resolving detectors with frequency upconversion technique [21,22].

In this paper, we investigated the photon correlation in the single-photon frequency upconversion process with multi-longitudinal mode of both signal and pump sources which were provided by two synchronized mode-locked fiber lasers [23,24]. With these two multi-longitudinal mode sources, high conversion efficiency of 80.5% was achieved. And with this system, we further studied the intensity cross-correlation of the unconverted infrared signal photons and the upconverted SFG photons. At the output of the frequency converter, the unconverted infrared signal photons showed intensity anti-correlation with the SFG visible photons as a function of the conversion efficiency. However, since the input signal photons

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were in a coherent state, the intensity cross-correlation function $g^{(2)}(0)$ was equal to 1 for all conversion efficiency, indicating that the frequency upconversion process was a random event for the individual incident signal photons. The observation would help to better understand the quantum feature of the single-photon frequency upconversion. And with the help of the mature frequency tuning or longitudinal mode selection techniques, the multi-longitudinal mode upconversion system may support tunable multi-mode photonic quantum-information interface and precise wavelength control on single-photon manipulation.

2. Theoretic model

The single-photon frequency upconversion process in a nonlinear medium can be described by

$$\hat{H} = i\hbar\chi E_p(\hat{a}_1\hat{a}_2^\dagger + H.c.), \tag{1}$$

where $\hat{a}_1$ is the annihilation operator corresponding to the infrared signal photons at frequency ω_1 and $\hat{a}_2^\dagger$ is the creation operator corresponding to the upconverted photons at frequency ω_2 [25, 26], χ is the coupling constant which is determined by the second-order susceptibility of the nonlinear medium, and $H.c.$ denotes a Hermitian conjugate. In the case of a strong pump, there is negligible depletion of the pump field and its amplitude can be treated classically as E_p.

The initial condition at the input facet of the nonlinear medium could be written as

$$\left|\phi\right\rangle = \left|\psi_1, 0_2\right\rangle, \tag{2}$$

where $\left|\psi_1\right\rangle$ represents the input signal photon state at frequency ω_1 and $\left|0_2\right\rangle$ represents the vacuum state at frequency ω_2. The dynamics of the input and output quantum fields in phase-matched SFG process can be described by the coupled-mode equations as

$$\hat{a}_1(L) = \hat{a}_1(0)\cos(|gE_p|L) - \hat{a}_2(0)\sin(|gE_p|L), \tag{3}$$

$$\hat{a}_2(L) = \hat{a}_2(0)\cos(|gE_p|L) + \hat{a}_1(0)\sin(|gE_p|L), \tag{4}$$

Where g denotes the nonlinear coupling coefficient, L is the interaction length in the nonlinear crystal. The corresponding creation operators can be found by taking the Hermitian conjugates of these equations.

At the output of the nonlinear medium, the expected value of the photon number operator of the infrared signal photon is given by

$$\begin{aligned} \left\langle \hat{n}_1 \right\rangle &= \left\langle \hat{a}_1^\dagger(L)\hat{a}_1(L) \right\rangle \\ &= \langle \phi | [\hat{a}_1^\dagger(0)\cos(|gE_p|L) - \hat{a}_2^\dagger(0)\sin(|gE_p|L)] \\ &\quad [\hat{a}_1(0)\cos(|gE_p|L) - \hat{a}_2(0)\sin(|gE_p|L)] \left|\phi\right\rangle, \end{aligned} \tag{5}$$

Since the mode at frequency ω_2 is assumed in the vacuum state, we can write

$$\left\langle 0_2 \right| \hat{a}_2^\dagger(0) = \hat{a}_2(0)\left|0_2\right\rangle = 0. \tag{6}$$

Then we readily obtain for the average number of infrared unconverted photons at the output facet of the nonlinear medium:

$$\begin{aligned} \left\langle \hat{n}_1 \right\rangle &= \left\langle \psi_1 \right| \hat{a}_1^\dagger(0)\hat{a}_1(0) \left|\psi_1\right\rangle \cos^2(|gE_p|L) \\ &= \left\langle \psi_1 \right| \hat{n}_0 \left|\psi_1\right\rangle \cos^2(|gE_p|L), \end{aligned} \tag{7}$$

Where $\hat{n}_0$ means the photon number operator of the input infrared signal photon.

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Meanwhile, the average number of the SFG photons can be likewise given by

$$\langle \hat{n}_2 \rangle = \langle \hat{a}_2^\dagger(L)\hat{a}_2(L) \rangle = \langle \psi_1 | \hat{n}_0 | \psi_1 \rangle \sin^2(|gE_p|L), \tag{8}$$

The average numbers of the unconverted infrared photons and the SFG photons are both correlated to the intensity of the pump field.

The joint probability of simultaneously detecting a photon at both ω_1 and ω_2 at the output of the frequency upconverter P_{12} could be proportional to $\langle \hat{n}_1 \hat{n}_2 \rangle$. With the input $\hat{a}_2(0)$ in the vacuum state, we readily obtain with the help of Eq. (6) that

$$\begin{aligned} \langle \hat{n}_1 \hat{n}_2 \rangle &= \langle \hat{a}_1^\dagger(L)\hat{a}_2^\dagger(L)\hat{a}_2(L)\hat{a}_1(L) \rangle \\ &= [\langle \hat{n}_0^2 \rangle - \langle \hat{n}_0 \rangle] \cos^2(|gE_p|L)\sin^2(|gE_p|L), \end{aligned} \tag{9}$$

On writing the single-photon frequency upconversion efficiency η as

$$\eta = \sin^2(|gE_p|L), \tag{10}$$

We have

$$\langle \hat{n}_1 \hat{n}_2 \rangle = [\langle \hat{n}_0^2 \rangle - \langle \hat{n}_0 \rangle]\eta(1-\eta), \tag{11}$$

which is dependent on the conversion efficiency of the frequency upconversion process.

Moreover, the intensity cross-correlation function $g^{(2)}(\tau)$ at $\tau = 0$ is then obtained from

$$g^{(2)}(0) = \frac{\langle \hat{n}_1 \hat{n}_2 \rangle}{\langle \hat{n}_1 \rangle \langle \hat{n}_2 \rangle} = \frac{\langle \hat{n}_0^2 \rangle - \langle \hat{n}_0 \rangle}{\langle \hat{n}_0 \rangle^2}, \tag{12}$$

When the incident photons are in one-photon Fock state, meaning $\langle \hat{n}_0^2 \rangle = \langle \hat{n}_0 \rangle$, the probability of detecting a photon both in frequency ω_1 and ω_2 is then zero. The unconverted infrared photons and SFG photons are anti-correlated. When the incident photons are in a coherent state, meaning $\langle \hat{n}_0^2 \rangle - \langle \hat{n}_0 \rangle = \langle \hat{n}_0 \rangle^2$, the intensity cross-correlation equals 1, which means no anti-correlation will be observed, independent from the conversion efficiency. It indicates that the frequency upconversion process is a random event for the individual incident signal photons.

Further, in a standard single-photon upconversion scheme, either the infrared signal source or the strong pump field is operated in a single-longitudinal mode to make sure the one-to-one upconverted replication. If the strong pump comes from a multi-longitudinal mode, the frequency conversion process becomes complicated. In our previous work, we demonstrated that quantum features of the incident photons could be preserved in the multi-longitudinal mode pump with suitable line-width of the pump source [25]. But in this upconversion system, the signal and the pump source were both in the multi-longitudinal mode, then the frequency conversion process became more complicated. Under this circumstance the Hamiltonian should be rewritten as

$$\hat{H} = i\hbar \sum_{ij} \chi E_{pi} (\hat{a}_{1j} \hat{a}_{2ij}^\dagger - H.c.), \tag{13}$$

where the E_{pi} is the pump electric field related to each longitudinal mode numbered by i, and the $\hat{a}_{1j}$ is the annihilation operator of the infrared signal photons related to each longitudinal mode numbered by j, the $\hat{a}_{2ij}^\dagger$ is the creation operator of the SFG photons corresponding to the longitudinal mode of both pump field and infrared signal photons. Then the dynamics of the

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input and output quantum fields in phase-matched SFG processes can be described by the coupled-mode equations as

$$\begin{aligned} \frac{d\hat{a}_{1j}}{dt} &= \frac{1}{ih}[\hat{a}_{1j}, \hat{a}_{1j}\hat{a}_2\hat{H}] = -g\sum_i E_{pi}\hat{a}_{2ij} \\ \frac{d\hat{a}_{2ij}}{dt} &= \frac{1}{ih}[\hat{a}_{2ij}, \hat{H}] = gE_{pi}\hat{a}_{1j}. \end{aligned} \tag{14}$$

The superposition state of infrared signal photons and SFG photons are represented by the operators $\hat{a}_1$, $\hat{a}_2$, which are defined as

$$\begin{aligned} \hat{a}_1 &= \sum_i \hat{a}_{1j} \\ \hat{a}_2 &= \sum_j \sum_i C_i \hat{a}_{2ij}, (\sum_i C_i^2 = 1), \end{aligned} \tag{15}$$

where $E_p^2 = \sum_i |C_i|^2$ and $C_i = E_{pi} / E_p$ denotes the probability amplitude of each longitudinal mode. Then the coupled-mode equation can be trivially solved by using the initial condition at the input facet of the nonlinear medium to yield

$$\hat{a}_1(L) = \hat{a}_1(0)\cos(|gE_p|L) - \hat{a}_2(0)\sin(|gE_p|L), \tag{16}$$

$$\hat{a}_2(L) = \hat{a}_2(0)\cos(|gE_p|L) + \hat{a}_1(0)\sin(|gE_p|L). \tag{17}$$

We get the same results as in a single-longitudinal mode situation (Eq. (3), Eq. (4).

And the joint probability $\langle \hat{n}_1\hat{n}_2 \rangle$ and the second-order correlation faction $g^2(0)$ in the multi-mode give the same results as that in the single-mode. The coherence properties of the incident signal photons have been maintained in this multi-mode system.

3. Experiment realization of multi-mode single-photon frequency upconversion

The single-photon frequency upconversion was realized with a synchronous pulsed pump source. The scheme used in our experiment is illustrated in Fig. 1. The upconversion system was composed of two master-slave synchronized fiber lasers (for the signal photons and escort pump source) and the frequency upconversion detection. The master was an etterbium-doped fiber laser passively mode-locked by nonlinear polarization rotation effect with a repetition rate of 17.6 MHz. In order to get a narrow spectrum for approaching the quasi-phase-matching bandwidth of the periodically poled lithium niobate crystal (PPLN) which was 0.3 nm, a reflective fiber Bragg grating (FBG) was used as a bandpass filter outside the laser cavity. The output was divided spectrally into the reflection and transmission parts by the FBG. The spectrum of the reflection part was centered at 1.04 μm with 0.27 nm bandwidth. The full-width of half-maximum (FWHM) of the pulse duration was about 11 ps. After attenuation, this part was prepared as the signal single-photon source. While the transmission part of the FBG used as a trigger by means of cross-phase modulation effect to initiate the mode-locking of the slave fiber laser. Without the master injection, the slave Erbium-doped fiber laser was self- mode-locked by nonlinear polarization rotation effect at its fundamental repetition rate of 4.4 MHz, which was a quarter of the master laser's repetition rate. Then with the master laser injection, the slave laser operated at its fourth harmonic repetition rate (17.6 MHz) in accordance with the master laser. Similarly, in order to get a narrow spectrum for the quasi-phase matching bandwidth, a bandpass filter was placed in the slave laser cavity. When synchronized, the spectrum of the slave laser was centered at 1.55 μm with 0.18 nm bandwidth. The output of the slave laser was further amplified and then used as the pump source. After dual-stage Erbium-doped fiber amplifier, the maximum pump power reached about 86.9 mW, with the FWHM pulse duration about 14 ps. The peak power of the pump source was up to 350 W to fulfill the strong nonlinear interaction. The stable

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synchronized system provided a perfect condition to observe the photon correlation in single-photon frequency upconversion.

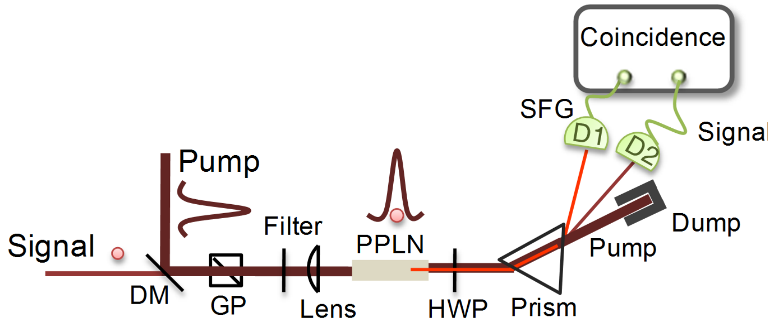

Fig. 1. Experimental setup of the correlation detection in single-photon frequency upconversion. DM, dichroic mirror with HR at 1.04 μm and AR at 1.55 μm; GP, Glan prism; PPLN, periodically poled lithium niobate crystal; HWP, half-wave plate; D1, D2, Si-APD single-photon detectors.

As shown in Fig. 1, the infrared signal photons at 1.04 μm were combined with the pump pulses at 1.55 μm by a dichroic mirror. The mixed beams were focused at the center of the PPLN crystal by a lens of 100-mm focal length after a long-pass filter cutting off at 1000 nm for blocking the noise from the LD pump of the fiber laser and fiber amplifier. The polarization of the infrared signal photons and the pump pulses were enforced to be the same vertical polarization by a Glan prism. The SFG photons at 0.62 μm appeared when the maximum temporal overlap achieved by finely tuning the time delay between the signal and the pump pulses. The operation temperature of the crystal was controlled around 109.0 °C within a fluctuation less than 0.1 °C, which allowed a quasi-phase-matching frequency upconversion for the PPLN with a grating period of 11.0 μm.

4 Experimental results and discussion

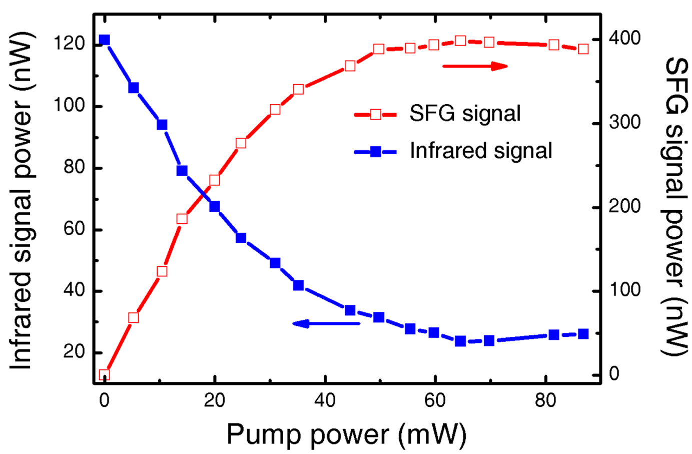

Fig. 2. SFG signal and the unconverted infrared signal intensity as a function of the pump power.

In this experiment, the unconverted infrared signal photons were separated from the mixed beams for the coincidence detection with the SFG photons at the output of the PPLN. The power meters monitored the output intensity of the two channels when the input infrared signal power was at μW order. We measured the SFG signal power and the unconverted infrared signal power as a function of the pump power as shown in Fig. 2. When the pump

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power was low, the SFG signal power increased remarkably with the pump power, while the infrared signal power decreased sharply. When the pump power was over 69.8 mW, the SFG signal power reached the maximum value and the infrared signal power reached the minimum value, indicating that the conversion efficiency reached the maximum value of 80.5%. With further increasing of the pump power, the intensity of the SFG signal dropped down and that of the infrared signal augmented, showing the Rabi oscillation-like nature of upconverison. It can be concluded that in the experiment, the maximum conversion efficiency did not reach 100% was not because of lack of pump power but maybe due to the slight spatial mismatch in the PPLN crystal. In the whole process, the power of the unconverted infrared signal and the SFG signal showed a relation of ebb and flow, indicating the anti-correlation of the two channels.

Then, we attenuated the incident infrared signal to few-photon level (10 photons/pluse) and measured the coincidence counts between the SFG photons and the unconverted infrared photons to observe the photon correlation in the single-photon frequency upconversion process. The power-meters were replaced by the single-photon detectors. The quantum efficiency of the single photon detector was 70% at the SFG photon wavelength, and 6% at the unconverted infrared photon wavelength. The dark counts were around 200 s^{-1}. In this experiment, the SFG photons passed through a group of spectral and spatial filters with total transmittance about 50.8% while the total transmittance for filtering the unconverted infrared signal photons was about 5%. In order to compensate the large different counts of the two channels, an attenuator with 13 dB was inserted before the detection part of the SFG photons. The outputs of the single-photon detectors for the SFG photons and the infrared photons were connected to a coincidence counter. The time window of the coincidence counter was set at 2 ns to include all the photon counts within the pump pulse envelop.

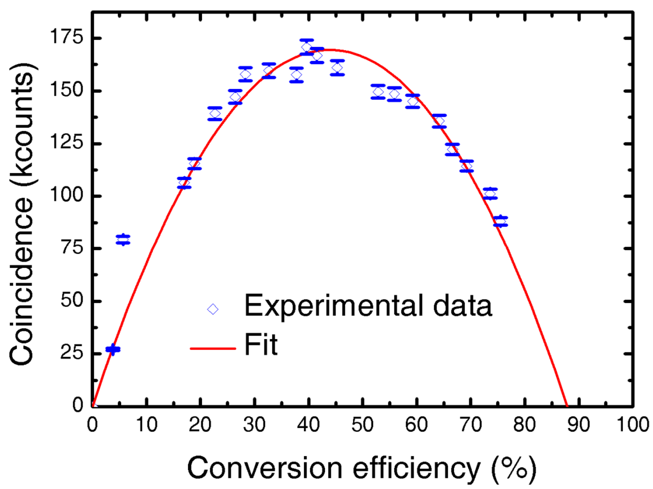

Fig. 3. Coincidence measurement between the SFG photons and the unconverted infrared photons dependent on the conversion efficiency.

Figure 3 shows the experiment record of coincidence counts as a function of the conversion efficiency. The counts increased sharply at the low conversion efficiency and reached the maximum at about 40%, just half of the maximum conversion efficiency. Then the counts dropped down showing a symmetrical curve. The coincidence counts represented the joint probability of simultaneously detecting at the SFG and unconverted photons, which showed an intensity anti-correlation as predicted by Eq. (11). It indicated that the SFG photons came from the initial infrared photons one-to-one in the upconversion process.

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The intensity cross-correlation function $g^{(2)}(0)$ could be calculated by normalizing the joint detection probability by the total number of counts registered by each detector at the output of the upconverter,

$$g^{(2)}(0) = \frac{N_C}{N_1 N_2 R T_{acq}}. \tag{18}$$

Where N_1 and N_2 are the counting rate on each detector at ω_1 and ω_2 at the output of the frequency upconverter, respectively [27].R is the repetition period, and T is the acquisition time. For the attenuated laser pulses, $g^{(2)}(0)$ is equal to 1 for all conversion efficiency.

In the frequency upconversion process, the SFG photons should maintain the coherence properties of the initial infrared photons. Since the single photon source from the attenuated fiber laser which was a coherent light source, the photons obeyed the Poissonian distribution. The SFG photons would follow Poisson distribution as well [26]. Although the joint probability of the simultaneously detecting the signal infrared photons and the SFG photons showed an anti-correlation as the function of the conversion efficiency, the normalized value of $g^{(2)}(0)$ was always close to 1 at all conversion efficiencies, as shown in Fig. 4. According to the theoretical analysis, it indicated that the SFG photons and infrared photons were both in the coherent light states. And the frequency upconversion process was completely a random event for each individual incident signal photons independent on the conversion efficiency.

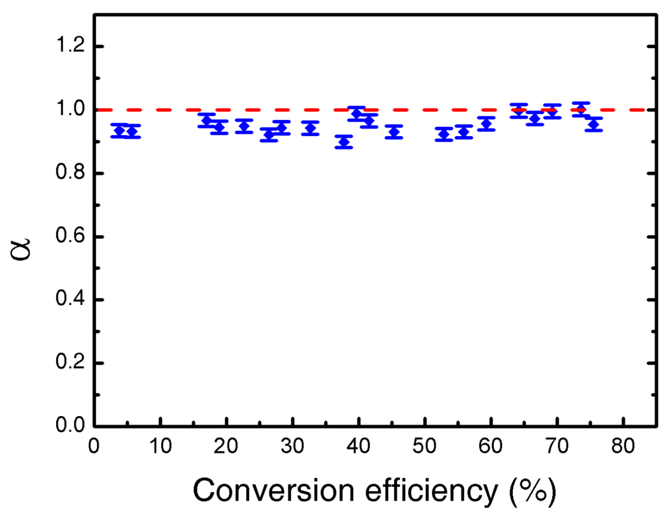

Fig. 4. Intensity cross-correlation of the SFG photons and unconverted infrared photons at different conversion efficiencies.

5. Conclusion

In conclusion, a near unity single-photon frequency upconversion system was realized based on two multi-mode synchronized fiber lasers. In this system, the anti-correlation as a function of the conversion efficiency was observed in the joint probability of the simultaneously detection at the upconverted and unconverted photons despite of the multi-mode operation of the signal and the pump source. Since the infrared signal photons were from a coherent light source and upconversion process maintained the coherence properties of the initial infrared photons, the normalized intensity cross-correlation function $g^{(2)}(0)$ was equal to 1 at all conversion efficiencies. This multi-mode upconversion system further with spectrally engineered is expected to be harnessed for applications in quantum information processing,

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such as the preparation of pure heralded single photons or a de-multiplexer of multiple quantum channels in the pulsed QKD schemes.

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