Toward quantum frequency combs: Boosting the generation of highly nonclassical light states by cavity-enhanced parametric down-conversion at high repetition rates

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We demonstrate the generation of multiphoton quantum states of light by cavity-enhanced parametric down-conversion in the high-repetition-rate pulsed regime. An external enhancement cavity resonant with the spectral comb of modes of a mode-locked pump laser provides a coherent buildup of the pump intensity and greatly enhances the parametric gain without sacrificing its high repetition rate and comb structure. We probe the parametric gain enhancement by the conditional generation and tomographic analysis of two-photon Fock states. Besides its potential impact for efficiently generating highly nonclassical or entangled multiphoton states in many existing experimental setups, this scheme opens exciting perspectives toward the combination of quantum and comb technologies for enhanced measurements and advanced quantum computation protocols.

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I. INTRODUCTION

Nonclassical and entangled multiphoton states are gaining ever-increasing attention as fundamental resources to probe the quantum world, to implement quantum communication and computation protocols, and to develop promising future quantum technologies. In particular, multiphoton highly nonclassical states are key resources for quantum information processing using continuous variables [1]. Furthermore, universal quantum computation based on photonic qubits has recourse to multiphoton entanglement to implement quantum algorithms with linear optics [2], and multiphoton entangled states are also crucial for applications in quantum metrology and cryptography [3,4].

Currently, most of the methods used to generate nonclassical and entangled multiphoton states are based on spontaneous parametric down-conversion (SPDC) processes producing two-mode squeezed states of the form

$$|\psi\rangle = \sqrt{1-|\lambda|^2}\sum_{n=0}^{\infty}\lambda^n|n\rangle_i|n\rangle_s \quad (1)$$

in noncollinear signal and idler modes. The parametric gain λ depends on the crystal nonlinearity and is proportional to the amplitude of the pump field. In the low-gain limit, the probability of producing entangled photon pairs scales linearly with the pump intensity, while the generation of n-photon states depends on the nth power of it. Dramatic enhancements of the multiphoton production rates can thus be achieved even with relatively modest increases in the pump intensity. Great efforts have been recently made in order to increase the parametric gain λ and thus enhance the weight of multiphoton contributions to the emission, both in the continuous-wave and in the pulsed regimes. While, in the former case, optical parametric resonators are adopted to overcome the limits connected to the low intensity of cw pumps (see, for example, Refs. [5–7]), ultrashort and ultraintense pump laser sources are also being actively used [8–10], albeit with complex amplified laser systems and at low repetition rates.

Another recent revolution in modern physics has been triggered by the advent of femtosecond frequency combs. Their invention has redefined the entire field of precision measurements, demonstrating an enormous potential in accurate frequency determination and space-time positioning [11,12]. Even if frequency-comb techniques have so far belonged to the realm of classical physics, it is foreseen that the ever-increasing accuracy in comb stabilization will soon get close to measurement quantum limits, and quantum-enhanced comb measurements will then require the generation of nonclassical light with a comb structure [13]. From an entirely different perspective, the possibility of nonlinearly coupling the huge number of modes of a frequency comb has also been recently proposed as a promising way to realize large and arbitrarily scalable cluster states for one-way quantum computing [14].

It is therefore of high interest to transpose the many advantages offered by frequency combs to the quantum domain. This clearly requires the generation of highly nonclassical and entangled states possessing a comb spectral structure; hence in high-repetition-rate pulsed schemes. Although a few experiments have already succeeded in producing nonclassical states from pulsed laser systems at high repetition rates [15–17], only very low parametric gains are normally obtained in such cases.

Here we demonstrate the use of an external enhancement cavity to coherently boost the intensity of pump pulses and thus greatly increase the SPDC gain. Differently from previous approaches, this is now obtained without additional costs in terms of laser power and, more importantly, without compromising the high repetition rates required for comb applications. We verify the generation of multiphoton nonclassical radiation by producing and tomographically analyzing two-photon Fock states.

Although resonant enhancement cavities are quite common in the continuous-wave regime, there are only a few

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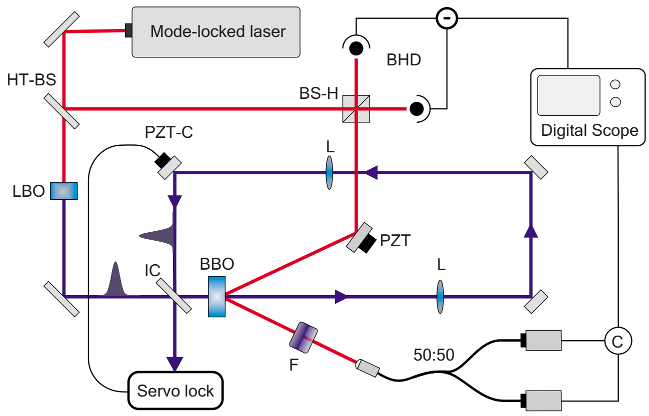

FIG. 1. (Color online) Experimental setup. The pump enhancement cavity (a simplified version is shown here for clarity) is built around the SPDC crystal [β–barium borate (BBO)]. Along the idler channel two on-off detectors are connected to a 3 dB single-mode fiber coupler after a series of spectral and spatial filters (F). The high-transmission beam splitter (HT-BS) is used to extract a small portion of the main laser source to be used as a reference beam (local oscillator) in the balanced homodyne detector (BHD) after mode-matching optics (not shown). The phase between the local oscillator and the analyzed state is adjusted by a mirror mounted on a piezoelectric stage (PZT). Once one of the two (or both) on-off detectors clicks, the homodyne signal is acquired by a digital scope and then analyzed.

applications in combination with pulsed laser sources. Early experiments with picosecond pulses demonstrated high-efficiency generation of low-order harmonics [18,19]; recent approaches have concentrated on the generation of high-order harmonics in a gas jet at high repetition rates [20,21] in an effort to transpose the femtosecond frequency-comb structure of the pump laser to the extreme ultraviolet. Indeed, a pulsed enhancement cavity is essentially made of a ring resonator whose longitudinal mode structure exactly matches the comb of modes (spaced by the pulse repetition rate) of the mode-locked source laser. This is accomplished by carefully adjusting and locking the external cavity length to that of the laser cavity. In the time domain this condition is seen to give rise to a constructive interference between the pulse circulating in the enhancement cavity and those coming from the laser. The coherent addition of the energy from many successive pulses of the laser pulse train can thus result in a significant buildup of the intracavity energy.

II. EXPERIMENTAL SETUP

The experimental setup shown in Fig. 1 consists in a second harmonic generation crystal [lithium triborate (LBO)] which generates radiation at 393 nm from a 1.5 ps mode-locked laser with a repetition rate of $\mathcal{R}=82$ MHz. The uv beam pumps a 3 mm type-I BBO crystal and produces SPDC into well-defined idler and signal spatial modes. In order to conditionally generate photon Fock states, a pair of on-off detectors (single-photon-counting modules Perkin-Elmer model AQR 14) is placed after a 50% beam splitter in the idler channel. When one detector or both click, a single- or two-photon Fock state, respectively, is prepared in a well-defined spatiotemporal mode along the signal channel. In principle, using this setup, it is possible to prepare Fock states of any order n depending on the preparation measurement performed on the idler channel. Up to now, Fock states with $n=1,2$ only [16,22,23] have been generated and characterized by quantum tomography, due to the very low gain in the SPDC process. In this configuration the expected rates of single- and two-photon state production ($\mathcal{R}_1$ and $\mathcal{R}_2$, respectively) are simply related as $\mathcal{R}_2=\mathcal{R}_1^2/2\mathcal{R}$, implying that any enhancement in the single-photon production rate scales quadratically in the two-photon one.

After the preparation of a given state, balanced homodyne detection is performed in the signal channel by mixing the signal state with a strong reference beam called the local oscillator using a 50% beam splitter (BS-H). The outputs of the beam splitter are then detected by proportional photodiodes connected to a home-made wide-bandwidth amplifier [24]. From the acquisition of many homodyne data for states prepared in the same way one can obtain the quadrature distribution of the state for a given phase between the local oscillator and the signal. A complete set of quadrature distributions at different phases allows one to reconstruct the density matrix and Wigner function of the analyzed signal state.

An enhancement cavity with a length of 3.6 m, corresponding to the 12 ns time delay between successive pulses from the laser, has been built around the SPDC crystal. It uses seven low-loss plane mirrors with reflectivity $R\approx 99.95\%$ for the pump pulses at 393 nm. Two lenses (L) with a focal length of 600 mm are carefully positioned in the cavity in order to produce a beam waist of about 250 μm inside the SPDC crystal. In this configuration the measured cavity losses (indicated by $1-R_m$, where R_m is the overall effective cavity reflectivity) amount to about 7%, mainly due to the residual reflections and absorptions on the crystal and the lenses. An input coupler (IC) with a reflectivity $R_i=90\%$ is used (undercoupled cavity configuration). A portion of the beam reflected from the input coupler is used to lock the cavity to the resonance peak by using the method proposed by Hänsch and Couillaud [25]. The expected cavity power enhancement

$$\mathcal{E}=\frac{1-R_i}{(1-\sqrt{R_iR_m})^2} \tag{2}$$

is then calculated to be $\mathcal{E}=14$, with a cavity finesse of $\mathcal{F}=35$, which is in very good agreement with the measured one.

Differently from the schemes used for intracavity second harmonic generation [18,19], here pump depletion plays no role in limiting the enhancement factor by losses, because of the very low parametric gain, which allows an almost complete recycling of the pulse energy after interaction with the crystal. Moreover, the use of a noncollinear SPDC configuration does not impose an output coupler for the down-converted light, and thus eliminates another important source of losses (like those experienced when trying to couple xuv light out of the cavity in [20,21]). Finally, the use of a pico-

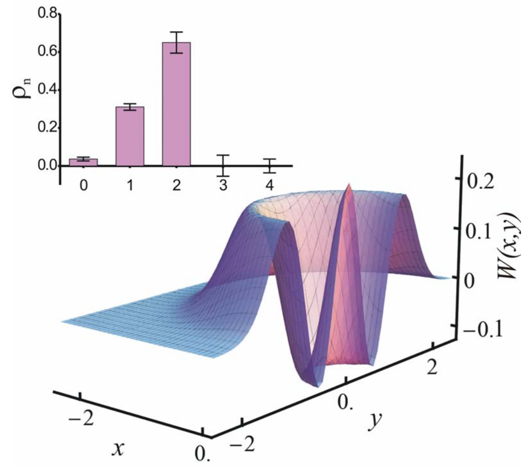

FIG. 2. (Color online) Experimentally reconstructed density matrix diagonal elements and corresponding Wigner function for the two-photon Fock state generated by cavity-enhanced SPDC.

second pulse source allows us to avoid the problems connected to intracavity dispersion when working with ultrashort femtosecond pulses. However, these will have to be taken into account when dealing with femtosecond frequency combs, since carrier phase stabilization is possible only in the ultrashort-pulse regime.

In order to verify the production of nonclassical multiphoton states we first check the generation efficiency of single-photon Fock states by using a single on-off detector in the idler channel. The preparation rate in this case is 5.8 kHz, to be compared to the 500 Hz of previous experiments performed by our group with the same pump power [15,17,26]. The single-photon production rate is thus enhanced by a factor of about 12, in good agreement with the expected increase of pump energy. Another particular advantage of using an enhancement cavity is that it works as a Gaussian spatial filter for the pump pulses. This has allowed us to simplify a part of the setup and further decrease the overall system losses by removing the pinhole-based spatial filtering of the second harmonic pump light that was present in our previous experiments [27].

III. GENERATION OF TWO-PHOTON FOCK STATES

Using the enhancement cavity we are now also able to generate two-photon Fock states at a sufficient rate without any increase in the pump laser power. We have acquired about 7000 quadrature measurements with a mean rate of 0.14 ± 0.05 Hz in about 14 h of experimental run. This rate represents a 150-fold increase over the exceedingly low value found for a single-pass configuration and, as expected, the two-photon enhancement factor is approximately the square of the one measured for the single-photon case. In Fig. 2 we show the reconstructed density matrix elements and the resulting Wigner function for the two-photon Fock state. The maximum likelihood method [28,29] has been used to retrieve the five diagonal elements of the density matrix and the contribution of the inefficient detection ($\eta_d = 0.67$) has been taken into account in the reconstruction procedure. A clear central peak and a negative ring region around the origin of the quadrature axis space are evident (and are still present even without correcting for detection losses) in the Wigner function, the latter being a sign of the highly nonclassical character of the generated state. From the reconstructed elements of the density matrix (inset of Fig. 2) the contribution of the residual $|0\rangle\langle 0|$ and $|1\rangle\langle 1|$ terms is still evident. These are due to residual impurities in the state preparation, which contribute as losses with $\eta_{p_2} = 0.81$. On the contrary, no higher-order contributions are visible in the density matrix.

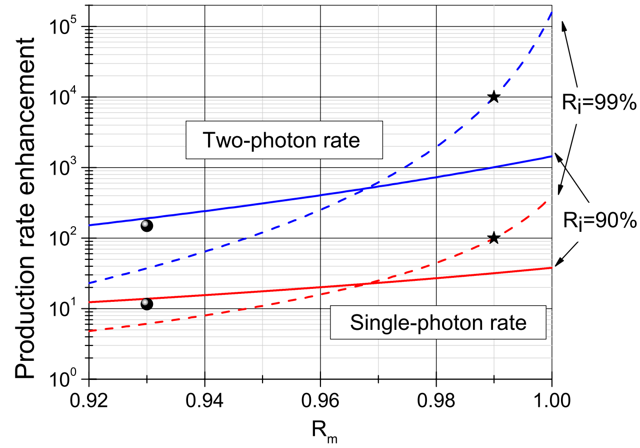

FIG. 3. (Color online) Cavity-enhanced production rates for the single- [red (lower) curves] and two-photon [blue (upper) curves] Fock states. The horizontal axis represents the overall effective reflectivity R_m of the cavity, and two curves are calculated for different reflectivities R_i of the input coupler (R_i=99%, dashed curves; R_i=90%, solid curves). Current experimentally measured enhancement factors are indicated by solid circles corresponding to $R_m = 0.93$ and R_i=90%. The expected enhancements for an optimized, impedance-matched cavity with $R_i = R_m = 0.99$ are indicated by stars.

IV. DISCUSSION AND CONCLUSIONS

It is worth noting that cavity losses can be easily reduced to below 1% by improving the design of the enhancement cavity: this involves the use of fewer mirrors and of concave ones to replace the lenses, and the application of an ultrahigh-quality antireflection coating to the SPDC crystal faces. In such a case, and in the impedance matching condition (with $R_i = R_m = 0.99$), we can expect a cavity finesse of $\mathcal{F} = 300$ and an enhancement factor up to about $\mathcal{E} = 100$. The resulting increase in the parametric gain would thus reflect in a 10^4 enhancement with respect to the two-photon preparation rate without the cavity (see Fig. 3). The enhancement-cavity approach would thus allow us to achieve the same rates as obtained by Ourjoumtsev *et al.* [23] with a cavity-dumped femtosecond laser working at a repetition rate which is a factor of 10^2 smaller than ours.

In conclusion, we have demonstrated a simple technique to greatly enhance the gain of pulsed parametric down-conversion and the production of nonclassical multiphoton states while preserving the high pump repetition rate and its comb spectral structure. A picosecond pump enhancement cavity built around the SPDC crystal is shown to provide an enhancement factor of about 15 (see [27]) in the production rate of single-photon Fock states, and an increase of more than two orders of magnitude in the rate of two-photon state generation. With a further slight reduction of cavity losses the enhancement factor would still greatly increase. Furthermore, if issues connected to cavity dispersion are properly taken into account, the use of intracavity SPDC with phase-stabilized femtosecond pump pulses will provide much higher gains and a stable frequency-comb structure.

The extension of this approach with the adoption of a synchronously pumped optical parametric oscillator configuration, where collinear degenerate parametric down-conversion is used in a cavity resonant also for the signal and idler modes, would give access to multimode squeezing [30] and multipartite continuous-variable entanglement, as recently proposed for the realization of cluster states for one-way quantum computation [14,31].

We believe that this technique will have a strong impact in a more widespread production of highly nonclassical states, giving the opportunity to investigate higher-dimensional Hilbert spaces and multiphoton entanglement even with modest available pump powers. Moreover, it will help in combining two of the most intriguing and promising avenues in modern physics, opening the way toward quantum-enhanced frequency-comb technologies and to appealing schemes for quantum-information processing.

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