P H Y S I C A L   R E V I E W   L E T T E R S

Nonlinear Interactions with an Ultrahigh Flux of Broadband Entangled Photons

Barak Dayan, Avi Pe'er, Asher A. Friesem, and Yaron Silberberg

Department of Physics of Complex Systems, Weizmann Institute of Science, Rehovot 76100, Israel

We experimentally demonstrate sum-frequency generation with entangled photon pairs, generating as many as 40 000 photons per second, visible even to the naked eye. The nonclassical nature of the interaction is exhibited by a linear intensity dependence of the nonlinear process. The key element in our scheme is the generation of an ultrahigh flux of entangled photons while maintaining their nonclassical properties. This is made possible by generating the down-converted photons as broadband as possible, orders of magnitude wider than the pump. This approach can be applied to other nonlinear interactions, and may become useful for various quantum-measurement tasks.

Entangled photons play a dominant role in quantum communication science due to their inherent nonclassical correlations [1–5]. Considerable effort has been invested towards achieving nonlinear interactions with entangled photons, since such interactions are significant for all-optical quantum computation and quantum metrology [6–9]. Most works have focused on enhancing the non-linear photon-photon coupling in order to achieve conditional phase shifts with single photons. Several strategies aimed at increasing the low efficiencies of nonlinear interactions with single photons (typically below 10^{-10}) were developed. One such strategy is increasing the nonlinearity through strong photon-atom coupling in a high-finesse cavity [9–11]; another uses mixing with a strong pump that is coherent with the interacting photons [12]. Other proposed schemes rely on enhanced nonlinearities obtained through electromagnetically induced transparency [13–15]. Resonantly enhanced two-photon absorption was performed with narrowband down-converted light at power levels that approached the entangled-photons regime, demonstrating a nonclassical departure from quadratic intensity dependence [16]. At lower power levels, where down-converted light can be considered as composed of separated photon pairs, a completely linear intensity dependence of two-photon absorption and sum-frequency generation (SFG) is predicted [17–20]. Such a linear dependence can be shown to break the Cauchy-Schwartz inequality for classical fields [21].

Here we propose an alternative approach towards achieving nonlinear interactions with entangled photons, which does not focus on increasing the nonlinear coupling, but rather on obtaining an ultrahigh flux of entangled photon pairs. In our setup we generate a flux of about 10^{12} entangled pairs per second, a flux that corresponds to a classically high power level of 0.3 μW. This flux is orders of magnitude greater than is typically utilized in quantum-optics experiments, which are usually limited to electronic detection rates.

The key element in achieving such a high photon flux while maintaining its nonclassical properties is generating

the entangled pairs as broadband as possible. The physical reason for this is that the arrival times of the two photons are correlated to within a time scale that is inversely proportional to their bandwidth [1]. Thus, the maximal flux $\Phi_{\max}$ of down-converted photons that can still be considered as composed of distinct photon pairs scales linearly with the down-converted bandwidth Δ_{DC} [18,22]:

$$\Phi_{\max} \approx \Delta_{\mathrm{DC}}. \tag{1}$$

This maximal flux, which corresponds to a mean spectral photon density of $n = 1$ (one photon per spectral mode), is the crossover point between the classical high power regime and the nonclassical low power regime.

A complete quantum-mechanical analysis [22] shows that the rate of SFG events with correlated pairs (i.e., photons generated from the same pump mode) indeed includes a term that depends linearly on the intensity:

$$R_c \propto \Delta_{\mathrm{DC}}(n^2 + n). \tag{2}$$

The dependence on Δ_{DC} indicates that the probability for up-conversion is inversely proportional to the temporal separation between the photons. This dependence on the bandwidth is not unique to entangled photons; the rate of SFG induced by classical pulses demonstrates the same dependence on their bandwidth, for the same reason. It is only the linear dependence on n in Eq. (2) that is unique to entangled photons. Note that all up-converted correlated pairs produce SFG photons back at the wavelength of the pump. Accordingly, in Eq. (2) it was assumed that the pump bandwidth δ_p is completely included within the available up-converted bandwidth δ_{UC} (i.e., the bandwidth that is phase matched for up-conversion in the nonlinear crystal).

Apart from the SFG events that are generated by correlated pairs there is always a classical background noise due to "accidental" SFG of uncorrelated photons. Unlike correlated pairs, uncorrelated pairs can be up-converted to any wavelength in the range of $2\Delta_{\mathrm{DC}}$ around the pump frequency. The actual rate $R_{u.c.}$ of such SFG events is

therefore proportional to the available up-converted bandwidth δ_{UC} [22]:

$$R_{u.c.} \propto \delta_{UC} n^2. \tag{3}$$

Like in many other detection schemes, this expression simply measures the background noise that falls within the spectrum of the "receiver."

To clarify the role of entanglement in the SFG process, let us first consider the narrowband case, where only one pair of signal and idler modes is involved in the down-conversion process. Assuming a low down-conversion efficiency that yields a mean spectral photon density of $n \ll 1$, the state $|\psi\rangle$ of the down-converted light can be described by

$$|\psi\rangle \approx M|0\rangle + \sqrt{n}|1\rangle_s|1\rangle_i, \tag{4}$$

with $M \sim 1$. The subscripts s, i denote the signal and idler modes, whose frequencies sum to the pump frequency. Note that the spectral width of the modes can be defined as the spectral width of the pump mode δ_p (this means we quantize the fields according to the longest relevant time scale, which is the coherence time of the pump, and so the number of photons is defined over a time period of δ_p^{-1} [23]). The state $|\psi\rangle$ clearly describes an entanglement between the signal and idler modes, which are both either in the vacuum state $|0\rangle$ or in the one-photon Fock state $|1\rangle$. It is quite straightforward to show that this entanglement is the source for the linear term in Eq. (2). When the down-converted bandwidth is significantly larger than the pump bandwidth, time and energy entanglement is created between the photons. This actually means that the photon pairs can be generated in $N = \Delta_{DC}/\delta_p$ different entangled mode pairs:

$$|\psi\rangle \approx M|0\rangle + \sum_{j=1}^{N} \sqrt{n}\,|1\rangle_{s_j}|1\rangle_{i_j}. \tag{5}$$

Although the phase of each signal or idler mode separately is inherently uncertain, the excitations of these mode pairs are mutually coherent, all having a combined phase that is shifted by $\pi/2$ from the pump phase. Thus, the SFG probability amplitudes induced by all these pairs add coherently, resulting in an amplification by N^2 of the correlated SFG rate, compared to the narrowband case. Intuitively speaking, one factor of N comes from having N more photon pairs, and the second one comes from the fact that the temporal separation between the photons of each pair is smaller by a factor of N. On the other hand, The SFG induced by up-conversion of uncorrelated pairs is summed incoherently, resulting in an enhancement only by a factor of N. Indeed, by dividing Eqs. (2) and (3), we obtain that the ratio between the correlated and uncorrelated rates is

$$\frac{R_c}{R_{u.c.}} \approx \frac{\Delta_{DC}}{\delta_{UC}}\left(\frac{n+1}{n}\right) \leq N\left(\frac{n+1}{n}\right), \tag{6}$$

where the inequality results from the condition of $\delta_{UC} \geq \delta_p$. As is evident, this ratio is decreased when the up-converted bandwidth is larger than the pump bandwidth; hence the importance of performing a narrowband SFG. Note that this gain of the correlated process over the uncorrelated one holds at high powers as well, and affects not only SFG, but any other nonlinear mixing between the signal and idler fields (e.g., two-photon absorption [24,25]).

It is important to point out that while the gain described in Eq. (6) results from correlations, such correlations do not necessarily require entanglement. Coherent correlations between two broadband fields are the basic principle of all spread-spectrum communication schemes [26,27]. In fact, without the nonclassical linear term, Eqs. (2), (3), and (6) are identical to the equations for the signal and noise in spread-spectrum communication performed with shaped classical pulses [28]; one has only to replace Δ_{DC} with the bandwidth of the pulses and δ_p with the spectral resolution of the phase modulations performed on the pulses. However, while coherent pulses can be easily shaped to exhibit spectral phase and amplitude correlations, such correlations between Fock states are inherently nonclassical and imply entanglement. This entanglement is manifested in the fact that the gain in Eq. (6) is stronger by a factor of $(n + 1)/n$ than is classically achievable, due to the additional, nonclassical linear term in the correlated SFG process. At high power levels the linear term becomes negligible and the correlations between the fields are almost identical to the correlations that can be obtained by shaping classical pulses. This is not to say that the increased precision of the correlations (i.e., the squeezing) vanishes at high powers, yet its influence on the SFG process becomes negligible. All previous experiments that involved SFG with down-converted light [29–31] were performed at power levels that greatly exceeded Φ_{max}, where the intensity dependence was completely quadratic. The correlation effects observed in these experiments are well described within the classical framework, and are identical to those demonstrated with shaped classical pulses [28,32].

To conclude this discussion we summarize that all the nonclassical properties exhibited in SFG of down-converted light result from the part of the process that has a linear intensity dependence. This part exists due to the entanglement between signal and idler modes, and becomes negligible in high powers. Although time and energy entanglement is not required for the nonclassical behavior, it amplifies its effect with respect to the classical, uncorrelated SFG process.

Despite the fact that the SFG process exhibits a linear intensity dependence at $n \ll 1$, it is nonetheless a two-photon process. Thus, a random loss of either a signal or an

idler photon is equivalent to the loss of the entire pair, and will lead to a ''classical'' quadratic reduction of the SFG signal. We expect therefore two different intensity dependencies of the SFG process: linear dependence, when the two-photon production rate is changed; quadratic dependence, when the entangled photon flux is attenuated by any linear-optics mechanism.

In our experiment we used a single-frequency ($\delta_p \approx$ 5 MHz around 532 nm) doubled Nd:YAG laser to pump a 12 mm long periodically poled KTiOPO$_4$ (PPKTP) crystal, generating infrared (IR) entangled photons with a broad bandwidth of $\Delta_{DC} = 31$ nm around 1064 nm. According to Eq. (1), this bandwidth implies a crossover flux of $\Phi_{max} = 8.2 \times 10^{12}$ photons per second, i.e., about 1.5 μW. A similar PPKTP crystal was used for the SFG process. The phase-matching conditions for the up-conversion process in this crystal were tuned to obtain an up-converted bandwidth of $\delta_{UC} \approx 100$ GHz around 532 nm. According to Eq. (6), these bandwidths ensure that the correlated SFG process dominates at any power.

The experimental setup is depicted in Fig. 1. Basically, entangled photons down-converted in one crystal were up-converted in the other crystal to produce the SFG photons. The entire layout was designed to maximize the interaction and collection efficiencies. The noncritical phase matching of the PPKTP crystals eliminates the ''walk-off'' between the down-converted photons and the pump, and allows optimal focusing [33] of the pump on the first crystal and of the entangled photons on the second one. The optimal focusing and the high nonlinear coefficient of the crystals (~ 9 pm/V) yielded high nonlinear interaction efficiencies of up to 10^{-7}. Both crystals were temperature stabilized to control their phase-matching properties.

For the SFG photons to be detected distinctly, any residue of the pump had to be filtered out from the down-converted photons by a factor of at least 10^{-18}. We chose to do so with a set of four dispersion prisms, designed for refraction at the appropriate Brewster angle. This arrangement had two major advantages over schemes which rely on harmonic filters for filtering out the pump. First,

Brewster-angle prisms enabled a very low loss of down-converted photons. Second, this layout enabled a tunable compensation of dispersion (mainly from the crystals), thereby avoiding a significant reduction of the bandwidth effective for the SFG process. The prisms were made of highly dispersive SF6 glass, in order to minimize the dimensions of our setup. The entangled-photon beam was filtered out from the SFG photons by a harmonic-separator mirror, and its power was measured by a sensitive InGaAs detector. The SFG photons were further filtered by line filters for 532 nm and counted with a single-photon counting module (SPCM) from EG&G (model AQR-15).

In order to verify that any photon detected by the SPCM was the result of the SFG process, and not a residue of the pump, we destroyed the phase matching in the second crystal by changing its temperature, and observed the SPCM's count drop to its dark count (~ 50 s^{-1}), even with the pump running at full power (5 W). This dark-count level was subtracted from all the subsequent measurements, which were performed with integration times of 2–5 s, and with pump powers that did not exceed 2.5 W.

We measured the power dependence of the SFG process in two ways: one, by attenuating the entangled-photon beam with optical attenuators; the other by reducing the power of the pump laser. Figure 2 depicts the results of these two measurements. As expected, attenuation resulted in a classical, purely quadratic decrease of the SFG counts. However, reducing the entangled-pair flux by decreasing the pump power resulted in a nearly linear decrease of the
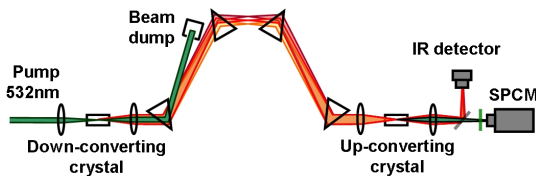

FIG. 1 (color online). Experimental layout. Entangled photons generated by down-conversion of a pump laser in one crystal are imaged through a set of four dispersion prisms onto a second crystal to generate the SFG photons. The entangled-photon beam is separated from the SFG photons by a harmonic-separator mirror and its power is measured by an InGaAs detector. The SFG photons are further filtered by 532 nm line filters and are counted with a single-photon counting module.
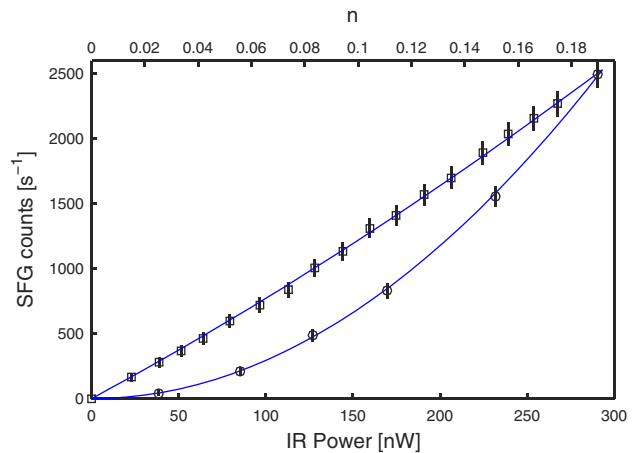

FIG. 2 (color online). Power dependence of SFG with entangled photons. When the down-converted IR power is reduced by optical attenuators, the SFG rate drops quadratically: circles, experimental; line, quadratic fit, exactly like classical light sources. However, when the IR power is reduced by decreasing the pump-laser power, thus reducing the photon-pair production rate, the SFG rate decreases in a close-to-linear manner, as quantum mechanically predicted: squares, experimental; line, calculated $\alpha(n + n^2)$, with α chosen to fit. The log-log slope of these measurements varies from 1.01 at the lowest powers to 1.14 at the highest powers.

SFG signal. The slight deviation from linearity at high powers is in complete agreement with our calculations, validating that the entire measurement was performed in the regime of $0 < n < 0.185$, i.e., well below the crossover flux. The maximal SFG count in this experiment was about 2500 s^{-1}. Taking into account that the collection efficiency of the SFG photons (limited by the transmission of the filters and the detector's efficiency) was about 6%, the number of generated SFG photons actually reached $40\,000 \text{ s}^{-1}$, a flux that was visible even to the naked eye.

While our approach still suffers from a low nonlinear efficiency, and thus is not readily applicable for quantum computation, it nonetheless introduces new capabilities for quantum metrology, since it reveals the nonclassical correlations between entangled photons with fidelity that exceeds that of electronic coincidences. Specifically, the SFG process simultaneously measures both the time difference and the energy sum of the photons, without measuring their individual energies or times of arrival. The large gain of $N = \Delta_{\mathrm{DC}}/\delta_{\mathrm{UC}}$ over the uncorrelated SFG rate depends directly on this effect, which implies that narrowband SFG of time and energy entangled photons automatically rejects coincidences of nonentangled photons. This valuable property is unattainable with electronic coincidence detection, since electronic photodetectors detect the actual arrival time of each photon separately, and therefore must be as broadband as the photons themselves.

Our work points at the possibility of maintaining the nonclassical properties of entangled photons even at classically high powers by utilizing broadband, continuous down-conversion [34]. We believe this ability to perform nonlinear interactions with ultrahigh fluxes of broadband entangled photons holds promise in quantum-measurement science, in particular, for phase measurements at the Heisenberg limit [35].

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