Resonant Sum Frequency Generation with Time-Energy Entangled Photons

S. Sensarn,* Irfan Ali-Khan, G. Y. Yin, and S. E. Harris

Edward L. Ginzton Laboratory, Stanford University, Stanford, California 94305, USA

(Received 18 September 2008; published 6 February 2009)

We describe a technique for enhancing the efficiency of sum frequency generation using entangled signal and idler photons. By resonating the sum frequency field, we observe that the generated power varies linearly with input power and is increased by a factor of 12.

DOI: 10.1103/PhysRevLett.102.053602 PACS numbers: 42.50.−p, 42.65.−k

As is now well understood, a phase-matched nonlinear crystal pumped by a monochromatic beam will generate pairs of time-energy entangled photons often termed as the signal and the idler and which, taken together, are often referred to as a biphoton [1]. Silberberg and colleagues have shown that, if these photons enter a nonlinear crystal phase-matched so as to generate the sum frequency field, the resulting field can be monochromatic and scale linearly with input power [2–4]. With the rate of generated biphotons denoted as $\mathcal{R}_{\mathrm{pair}}$ and the characteristic temporal length of the biphoton as T, this nonclassical behavior occurs when $\mathcal{R}_{\mathrm{pair}}T < 1$. When $\mathcal{R}_{\mathrm{pair}}T > 1$, sum frequency generation (SFG) is in the classical regime and is characterized by generated power proportional to $\mathcal{R}_{\mathrm{pair}}^2$ and a linewidth of SFG that is equal to the cross correlation of the signal and idler line shapes. In the quantum regime, time-energy entangled photons with broad individual bandwidths can be summed to produce a monochromatic field. We note earlier theory and experiments describing nonlinear processes with nonclassical light and, in particular, the linear dependence of two photon processes on the incident power [5–7].

This Letter describes, both theoretically and with a proof-of-principle experiment, how one may resonate the sum frequency field (Fig. 1) so as to enhance the rate of quantum SFG and the ratio of the quantum to the classical component. Time-energy entangled signal and idler photons are generated by a parametric down-converter in the low gain regime (not shown) with a monochromatic pump so that $\omega_p = \omega_s + \omega_i$. These photons are summed in a nonlinear crystal placed inside a cavity whose qth longitudinal mode (at frequency ω_q) is detuned from the pump frequency by $\delta\omega = \omega_p - \omega_q$. The input and output mirrors of the cavity are assumed to be transparent over the broad biphoton bandwidth and highly reflecting at the sum frequency. The essence of the quantum enhancement is that the work done by a time-varying dipole moment against an electric field at its own frequency is increased by the presence of the resonator. This enhancement occurs pair by pair, thus resulting in the linear dependence of sum-frequency-generated power on the incident biphoton rate.

There is considerable motivation for improving the efficiency of SFG in the quantum regime. For example, quantum SFG may be used as an ultrafast correlator of broadband biphotons [2,8–11]. However, ultrafast quantum correlation requires the use of short crystals that result in impractically low SFG rates. In the following, we show theoretically that, for a lossless crystal and an output mirror reflectivity R, the sum frequency power is enhanced relative to the same crystal without a resonator by a factor of $4/(1-R)$. The quantum case has the unusual property that the conversion efficiency is proportional to the effective peak power of the biphoton $\cong \hbar\omega/T$ while retaining the same resonance enhancement as a classical monochromatic field.

We work in the Heisenberg picture with traveling-wave signal and idler beams denoted by the annihilation operators $a_s(t,z) = \tilde{a}_s(t,z)\exp[-i(\omega_s t - k_s z)]$ and $a_i(t,z) = \tilde{a}_i(t,z)\exp[-i(\omega_i t - k_i z)]$, respectively, where operators with a tilde vary slowly with time and distance. We use a slowly varying envelope formalism with the standing-wave cavity mode operator written as $a_q(t,z) = \tilde{a}_q(t)\times\exp(-i\omega_q t)\sin(k_q z)$, where $k_q = q\pi/L$. We project the broadband generated dipole moment operator, proportional to $\tilde{a}_s(t,z)\tilde{a}_i(t,z)$, against the cavity mode. With the traveling-wave SFG field emitted from mode q denoted by $a_{\mathrm{sum}}(t,z) = \tilde{a}_{\mathrm{sum}}(t)\exp[-i(\omega_p t - k_p z)]$, the equation for the evolution of $\tilde{a}_q(t)$ and its relation to the SFG field is

$$
\begin{aligned}
\frac{\partial \tilde{a}_q(t)}{\partial t} + \frac{\Gamma}{2}\tilde{a}_q(t) &= \tilde{P}_q(t)\exp(-i\delta\omega t) + \tilde{\mathcal{F}}(t),\\
\tilde{P}_q(t) &= i\kappa_c \frac{2}{L}\int_0^L \tilde{a}_s(t,z)\tilde{a}_i(t,z)\\
&\quad\times \exp(ik_p z)\sin\left(\frac{q\pi z}{L}\right)dz,\\
\tilde{a}_{\mathrm{sum}}(t) &= \sqrt{\gamma}\,\tilde{a}_q(t).
\end{aligned} \tag{1}
$$

Denoting the spacing of the cavity modes at the sum frequency by $\Delta = c/(2Ln)$, the decay rate Γ of photons in the cavity is determined by the mirror reflectivity R and the single-pass power loss ξ. With the output coupling rate $\gamma = \Delta(1-R)$, the total (power) decay rate is $\Gamma = 2\xi\Delta + \gamma$. We take all fields to be plane waves with cross sectional area A and take the refractive index n and nonlinearity d to be independent of frequency. With the cavity and summing

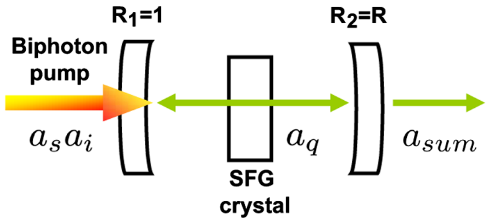

FIG. 1 (color online). Schematic of resonant enhancement. The cavity is transparent to the incoming signal and idler photons. The operator $a_s a_i$ denotes the incoming biphoton field; a_q is the resonant sum frequency field, and $a_{\rm sum}$ is the generated SFG output field.

crystal of the same length, the coupling constant $\kappa_c = (d/n^2)(\mu_0 \hbar \omega_p \omega_s \omega_i L/A)^{1/2}$. The quantity $\tilde{\mathcal{F}}(t)$ is a Langevin noise operator that has contributions from an incoming wave at the right-hand mirror (not shown) and a macroscopic loss term. Both are negligible at room temperature. The normalization is such that $\langle \tilde{a}_q^\dagger(t)\tilde{a}_q(t)\rangle$ is the total number of photons in the cavity mode and $\langle a_{\rm sum}^\dagger(t) a_{\rm sum}(t)\rangle$ is the rate of sum photons exiting the cavity.

The solution of Eqs. (1) for the SFG field operator is

$$\tilde{a}_{\rm sum}(t) = \sqrt{\gamma}\exp\left(-\frac{\Gamma}{2}t\right)\int_{-\infty}^{t}\exp\left(\frac{\Gamma}{2}t'\right)\tilde{P}_q(t') \times \exp(-i\delta\omega t')dt'. \quad (2)$$

We assume that the signal and idler fields are not depleted in the summing crystal and transform to the frequency domain using $a_s(t,z) = \int a_s(\omega,z)\exp(-i\omega t)d\omega$ and $a_i(t,z) = \int a_i(\omega,z)\exp(-i\omega t)d\omega$. $\tilde{P}_q(t)$ becomes

$$\tilde{P}_q(t) = -\kappa_c \int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\Phi(\omega_1,\omega_2)a_s(\omega_1)a_i(\omega_2) \times \exp[-i(\omega_1+\omega_2-\omega_p)]d\omega_1 d\omega_2,$$

$$\Phi(\omega_1,\omega_2) = \exp\left[-i\frac{\Delta k(\omega_1,\omega_2)L}{2}\right]\mathrm{sinc}\left[\frac{\Delta k(\omega_1,\omega_2)L}{2}\right], \quad (3)$$

where $\Phi(\omega_1,\omega_2)$ is a phase matching factor with $\mathrm{sinc}(x) = \sin(x)/x$ and $\Delta k(\omega_1,\omega_2) = k_q - [k(\omega_1)+k(\omega_2)]$. Equation (3) is substituted into Eq. (2) to obtain the SFG output field:

$$\tilde{a}_{\rm sum}(t) = -\kappa_c\int_{-\infty}^{\infty}\int_{-\infty}^{\infty}\frac{\sqrt{\gamma}\Phi(\omega_1,\omega_2)a_s(\omega_1)a_i(\omega_2)}{\Gamma/2 - i(\omega_1+\omega_2-\omega_p+\delta\omega)} \times \exp[-i(\omega_1+\omega_2-\omega_p+\delta\omega)t]d\omega_1 d\omega_2. \quad (4)$$

For monochromatically pumped down-conversion, the down-converted signal and idler fields at the input of the summing crystal [$a_s(\omega)$ and $a_i(\omega')$, respectively, where $\omega' = \omega_p - \omega$] can be described in terms of initial vacuum fields $a_{s0}(\omega)$ and $a_{i0}(\omega')$, respectively, as [8]

$$a_s(\omega) = A(\omega)a_{s0}(\omega) + B(\omega)a_{i0}^\dagger(\omega'),$$
$$a_i^\dagger(\omega') = C(\omega)a_{s0}(\omega) + D(\omega)a_{i0}^\dagger(\omega'). \quad (5)$$

Substituting Eqs. (5) into Eq. (4) and noting the commutator $[a_{i0}(\omega_1), a_{j0}^\dagger(\omega_2)] = \frac{1}{2\pi}\delta_{ij}\delta(\omega_1-\omega_2)$, we evaluate $\langle a_{\rm sum}^\dagger(t)a_{\rm sum}(t)\rangle$ and find the rate of SFG photons in mode q exiting the cavity to be

$$\mathcal{R}_{\rm cav} = \left(\frac{\kappa_c}{\pi}\right)^2\int_{-\infty}^{\infty}\left[\frac{\gamma}{\Gamma^2+4\delta\omega^2}f(\omega) + \frac{\gamma}{\Gamma^2+4(\omega-\omega_q)^2}g(\omega)\right]d\omega, \quad (6)$$

where $f(\omega)$ and $g(\omega)$ are

$$f(\omega) = \left|\int_{-\infty}^{\infty}A(\Omega)C^*(\Omega)\Phi(\Omega,\omega_p-\Omega)d\Omega\right|^2\delta(\omega-\omega_p),$$
$$g(\omega) = \int_{-\infty}^{\infty}|B(\Omega)|^2|C(\Omega-\omega+\omega_p)|^2|\Phi(\Omega,\omega-\Omega)|^2 d\Omega. \quad (7)$$

The SFG rate in Eq. (6) consists of a quantum term containing the function $f(\omega)$ and a classical term containing the function $g(\omega)$. The quantum term is the result of generation with correlated photons; it may be shown to scale linearly with input power and has a monochromatic spectrum [2,8]. The classical term comes from SFG with uncorrelated photons and varies quadratically with input power. Its spectrum is proportional to the cross correlation of the signal and idler spectra, multiplied by the cavity line shape.

In order to normalize the improvement that results from the use of a cavity, we write the SFG rate for the traveling-wave case with no cavity present. This rate is

$$\mathcal{R}_{\rm tw} = \left(\frac{\kappa_{\rm tw}}{2\pi}\right)^2\int_{-\infty}^{\infty}[f(\omega)+g(\omega)]d\omega, \quad (8)$$

where $\kappa_{\rm tw} = \kappa_c/\Delta^{1/2}$, and k_q in the function $\Phi(\omega_1,\omega_2)$ becomes $k(\omega_1+\omega_2)$. As in the cavity case, the SFG rate consists of a quantum term that varies linearly with input power and a classical term that varies quadratically [8].

The quantum SFG rate is enhanced by the presence of the cavity by an amount equal to the ratio of the terms containing $f(\omega)$ in Eqs. (6) and (8). When the cavity is tuned to resonate the pump frequency ($\delta\omega = 0$), the quantum SFG enhancement ratio is

$$\eta_q = \frac{4(1-R)}{[(1-R)+2\xi]^2}. \quad (9)$$

Maximizing with respect to R, the largest quantum SFG enhancement is obtained for $R = 1-2\xi$; for this reflectivity, the enhancement ratio is equal to the inverse of the round-trip power loss $1/(2\xi)$.

To study the effect of the cavity on the classical component of SFG, we assume that the signal and idler spectra are sufficiently broadband that the dipole moment at the sum frequency is constant over many cavity modes. In this case, $g(\omega)$ is a constant, and the generated SFG spectrum is periodic with resonant peaks separated from each other by the cavity free spectral range. To obtain the net enhancement of the classical term, we integrate over a single free spectral range centered at ω_q. The resulting classical SFG enhancement ratio is then

$$\eta_c = \frac{2}{\pi}\int_{\omega_q-\pi\Delta}^{\omega_q+\pi\Delta}\frac{\gamma}{\Gamma^2+4(\omega-\omega_q)^2}d\omega \cong \frac{\gamma}{\Gamma} = \frac{1-R}{(1-R)-2\xi}, \tag{10}$$

where the second equality follows for sufficiently high finesse that $\tan^{-1}(2\pi\Delta/\Gamma) \to \pi/2$. If the cavity is lossless, $\eta_c = 1$. Though generation at resonance is enhanced, generation off resonance is suppressed so that the integrated classical SFG enhancement ratio is unity.

We now describe a proof-of-principle experiment where quantum SFG with frequency-degenerate, time-energy entangled biphotons is enhanced by a factor of 12. A schematic of the experiment is shown in Fig. 2. Frequency-degenerate biphotons, with a calculated bandwidth of 32 nm, are generated by spontaneous parametric down-conversion in a 20 mm long periodically poled, magnesium oxide-doped, stoichiometric lithium tantalate crystal (MgO:PPSLT, HC Photonics Corp.) pumped by an 8 W, cw laser at 532 nm (Coherent Verdi V10). The pump, signal, and idler photons are polarized along the extraordinary axis of the crystal. A four-prism setup is used to filter out the strong pump and to provide dispersion compensation for the down-converted biphotons. SFG occurs inside a cavity that is reflecting at 532 nm and transparent at 1064 nm.

The SFG crystal is a 20 mm long MgO:PPSLT crystal (HC Photonics Corp.) antireflection coated at 1064 and 532 nm. The confocal cavity consists of two 20 mm radius of curvature spherical mirrors, one of which is mounted onto a piezoelectric crystal. The input mirror has a reflectance of $R_{\text{in}} > 99.5\%$, and the output mirror has a reflectance of $R_{\text{out}} = 95.5\%$. (The measured single-pass crystal loss is $\xi \sim 2\%$, so that the optimum reflectivity of the output mirror is $R_{\text{out}} = 1\text{–}2\xi \cong 96\%$.) The SFG signal is filtered, collected into a multimode fiber, and detected with a single photon counting module (SPCM, PerkinElmer SPCM-AQR-16-FC).

The cavity is aligned in three stages. Initial alignment is performed by optimizing the coupling of a 532 nm reference beam into the TEM_{00} cavity mode. Further alignment is performed using the SFG signal by scanning the cavity over a free spectral range at 50 Hz and maximizing the average detected SFG signal. Final adjustments are made by slowly scanning the cavity and optimizing the detected resonance peaks.

A typical scan of the cavity over a free spectral range is shown in Fig. 3. The resonant behavior of the SFG process is clearly observed, with peak generation rates on resonance that are significantly higher than the traveling-wave rate of ~3200 counts/s. The solid theoretical curve is a plot of $\mathcal{R}_{\text{cav}}$ in Eq. (6) normalized to best fit the average of 26 measurements of the resonant SFG rate.

To better quantify the enhancement, we measure the resonantly enhanced SFG rate at various down-converted infrared input powers. For each input power, the cavity is repeatedly scanned over a single free spectral range to obtain 22–26 samples of the resonant SFG rate. These are averaged and plotted in Fig. 4. The large error bars (standard deviations) are caused by air currents and temperature instability in the cavity. Also shown is the traveling-wave SFG rate obtained by removing the out-coupling mirror.

The solid curves in Fig. 4 are theoretical fits to the cavity data using Eq. (6) and to the traveling-wave data using Eq. (8). Both are scaled to minimize the mean-square error. This scaling includes the effects of SPCM detection efficiency, crystal nonlinearity, periodic poling errors, and transverse mode overlap. Based on the fits, the enhancement ratio is 12 and is about a factor of 2 less than the theoretical prediction using Eq. (9). The discrepancy may be due to imperfect mode matching or cavity instability.

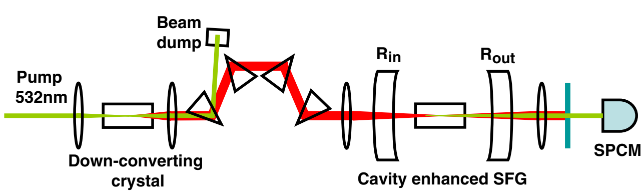

FIG. 2 (color online). Schematic of the experiment. Details are in the text.

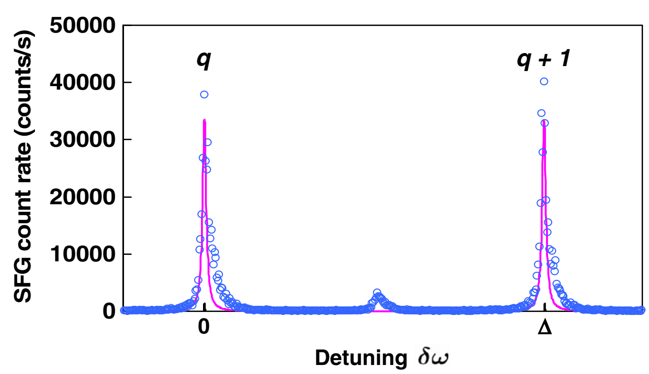

FIG. 3 (color online). SFG count rate versus detuning. Circles are data points from a single scan. The solid curve is a theoretical fit using Eq. (6) (see the text for normalization). The small peak at $\Delta/2$ comes from odd-parity transverse modes in the confocal resonator [12].

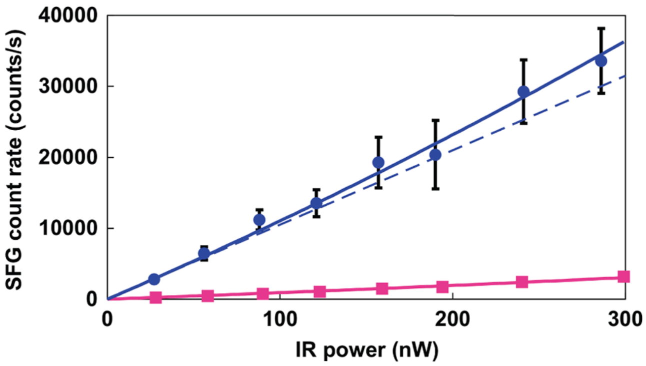

FIG. 4 (color online). SFG count rate versus infrared input power. Circles are resonant SFG rates. Squares are traveling-wave SFG rates. Curves are theoretical fits (see text). The SFG enhancement ratio is 12.

We note that, in our experiment, the classical terms containing $g(\omega)$ in Eqs. (6) and (8) are about 3 orders of magnitude smaller than the quantum terms containing $f(\omega)$. Because a long summing crystal is used, the phase matching factor $\Phi(\omega_1, \omega_2)$ greatly restricts the bandwidth of the generated dipole moment, suppressing classical SFG. The dashed line in Fig. 4 is tangent to the cavity theory curve at zero input power. The small deviation from linearity of the theory curve comes entirely from the quantum term in Eq. (6).

It seems likely that, by using crystals with lower bulk absorption loss, higher-quality AR coatings, or perhaps by using a microtoroidal crystal cavity [13], much larger enhancement factors can be obtained. This may be sufficient to allow a homodyning technique where the output of the resonant sum generator is mixed with the original pumping laser and the relative path length of the signal and idler beams is varied. One thereby obtains the complete biphoton waveform, including phase information [10].

In summary, we have shown how the quantum nature of sum frequency generation with time-energy entangled photons allows resonant enhancement. Experimentally, we obtain a twelvefold enhancement of the near-monochromatic sum frequency radiation that is generated by summing signal and idler radiation with a bandwidth of about 32 nm.

The authors thank Joseph Schaar, Shengwang Du, and Martin Fejer for helpful discussions. This work was supported by the U.S. Army Research Office and the U.S. Air Force Office of Scientific Research.

*sensarn@stanford.edu

[1] Y. Shih, Rep. Prog. Phys. **66**, 1009 (2003).
[2] B. Dayan, A. Pe'er, A. Friesem, and Y. Silberberg, Phys. Rev. Lett. **94**, 043602 (2005).
[3] B. Dayan, Phys. Rev. A **76**, 043813 (2007).
[4] A. Pe'er, B. Dayan, A. Friesem, and Y. Silberberg, Phys. Rev. Lett. **94**, 073601 (2005).
[5] J. Gea-Banacloche, Phys. Rev. Lett. **62**, 1603 (1989).
[6] N. Georgiades *et al.*, Phys. Rev. Lett. **75**, 3426 (1995).
[7] D. V. Strekalov, M. C. Stowe, M. V. Chekhova, and J. P. Dowling, J. Mod. Opt. **49**, 2349 (2002).
[8] S. E. Harris, Phys. Rev. Lett. **98**, 063602 (2007).
[9] D. Strekalov, A. B. Matsko, A. A. Savchenkov, and L. Maleki, Phys. Rev. A **71**, 041803(R) (2005).
[10] S. E. Harris and S. Sensarn, in *Coherence and Quantum Optics IX: Proceedings of the Ninth Rochester Conference on Coherence and Quantum Optics*, edited by N. P. Bigelow, J. H. Eberly, and C. R. Stroud, Jr. (University of Rochester, Rochester, NY, 2007), pp. 65–71.
[11] M. B. Nasr *et al.*, Phys. Rev. Lett. **100**, 183601 (2008).
[12] M. Hercher, Appl. Opt. **7**, 951 (1968).
[13] V. S. Ilchenko, A. A. Savchenkov, A. B. Matsko, and L. Maleki, Phys. Rev. Lett. **92**, 043903 (2004).