Design of a high-power continuous source of broadband down-converted light

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We present the design and experimental proof of principle of a low threshold optical parametric oscillator (OPO) that continuously oscillates over a large bandwidth allowed by phase matching. The large oscillation bandwidth is achieved with a selective two-photon loss that suppresses the inherent mode competition, which tends to narrow the bandwidth in conventional OPOs. Our design performs pairwise mode locking of many frequency pairs, in direct equivalence to passive mode locking of ultrashort pulsed lasers. The ability to obtain high powers of continuous *and* broadband down-converted light enables the optimal exploitation of the correlations within the down-converted spectrum, thereby strongly affecting two-photon interactions even at classically high power levels, and opening new venues for applications such as two-photon spectroscopy and microscopy and optical spread spectrum communication.

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I. INTRODUCTION

When two-photon interactions are induced by down-converted light with a bandwidth that exceeds the pump bandwidth, they behave as a pulse temporally, but as a narrow continuous-wave (CW) field spectrally [1,2]. At low photon fluxes this behavior accounts for the time and energy entanglement between the down-converted photons [3–6]. Two-photon interactions, such as two-photon absorption (TPA) and sum-frequency generation (SFG), can also exhibit such a behavior even at high power levels, when the atomic level in TPA or the generated light in SFG have a sufficiently narrow bandwidth [1,2]. This behavior, which results from the correlations within the down-converted spectrum, does not depend on the squeezing properties of the light and is insensitive to linear losses. Accordingly, it has potential applications such as two-photon microscopy, optical spread spectrum communication [7] and subdiffraction limit lithography [8], where to be viable, an efficient, low threshold source that generates high power broadband down-converted light in a continuous mode of operation is necessary.

In the past, the only way to generate high power broadband down-converted light was to pump strongly a nonlinear crystal, enough for the down-conversion process to become stimulated in a single pass through the crystal. Since the nonlinear interaction is weak, only pulsed down conversion could overcome the inherent inefficiency, since the pump power threshold is impractical for CW operation. This threshold can be significantly lowered by resorting to an oscillator with a high finesse cavity, but then mode competition narrows drastically the bandwidth of the actual oscillation even though phase matching allows broad oscillation. Specifically, since all the down-converted mode pairs compete for the energy of the pump, the mode pair with the highest gain is dominant, while all other pairs are suppressed.

Here we present the design and experimental proof of principle for a low-threshold optical parametric oscillator (OPO) that generates broadband, continuous, steady-state oscillations. We exploit the special properties of two-photon excitation with broadband down-converted light to cause loss only to narrowband oscillations and not to broadband oscillations. Specifically, since such two-photon excitations are induced in a pulse-like manner, they can be coherently controlled by tailoring the level of dispersion at different locations within the OPO cavity, leading to a situation where narrowband oscillations, which are insensitive to dispersion, suffer two-photon loss but broadband oscillations do not. Thus, since the dominant oscillation in a cavity is always the one with the best gain-loss relation, i.e., the highest conversion efficiency, only broadband oscillations build up.

This paper is organized as follows: First we review the two-photon coherence properties of broadband down-converted light. Then we present the basic principles of our OPO cavity design, and analyze in some detail the expected spectrum, threshold and performance. Finally, we describe an experiment that supports our proposed approach, and present some concluding remarks.

II. TWO-PHOTON COHERENCE

We use the term "two-photon coherence" to describe the inherent phase and amplitude correlation between the signal $A_s(\omega)$ and the idler $A_i(\omega)$ fields of down-converted light. Although each field in itself is incoherent thermal noise, the two fields are complex conjugates of each other [7,9], as

$$A_s(\omega) = A_i^*(\omega_p - \omega), \tag{1}$$

where ω_p is the pump frequency. Such symmetry in the spectrum indicates that temporally, the slow varying envelope of the total down-converted field $A_{\rm DC}(t)=A_s(t)+A_i(t)$ around the center frequency $\omega_p/2$ is real; i.e., $A_{\rm DC}(t)$ has only one quadrature in the complex plane, when compared to the pump as the phase reference [10].

It should be noted that the exact precision of the spectral correlation is *not* crucial. It is true that quantum mechanically the precision of the correlations between the down-converted modes can exceed the shot-noise level, resulting in squeezing of the electromagnetic quadratures of the generated light. However, while the high precision of the correlations (i.e., the squeezing) is easily destroyed by linear losses, their effect on two-photon interactions can still be dramatic. Specifically, when the down-converted bandwidth is significantly larger than the pump bandwidth, the down-converted light can induce two-photon interactions with the same efficiency and sharp temporal behavior as ultrashort pulses, while exhibiting high spectral resolutions and low peak powers as those of the narrowband pump. In the high-power regime, these seemingly nonclassical properties are completely described within the classical framework, and do not depend on the squeezing degree of the down-converted light. This equivalence of broadband down-converted light to coherent ultrashort pulses also implies an ability to coherently control and shape the induced two-photon interactions with pulse-shaping techniques, although the down-converted light may be neither coherent, nor pulsed [2].

These special properties of TPA and SFG induced by down-converted light can be easily understood from the symmetry of Eq. (1). In general, the TPA probability (or SFG intensity) R at frequency Ω is given by

$$R(\Omega) \propto \left| \int d\omega A_{\mathrm{DC}}(\omega) A_{\mathrm{DC}}(\Omega - \omega) \right|^2. \tag{2}$$

When we substitute the expression for the down-converted field

$$A_{\mathrm{DC}}(\omega) = A_s(\omega) + A_i(\omega) = A_s(\omega) + A_s^*(\omega_p - \omega), \tag{3}$$

we obtain four terms in the integrand: signal-signal (s-s) term, idler-idler (i-i) term, and two cross terms (s-i). Since both the signal and the idler are incoherent noisy fields, the spectral phase of the s-s and i-i term is random, so their contribution after summation can be neglected. The mixed terms however contain correlated phases. Thus, when $\Omega = \omega_p$, the random phase of the integrand cancels out, leading to a fully constructive interference, exactly as if we used coherent transform limited pulses. Accordingly,

$$R(\Omega) \propto \left| \int_0^{\omega_p/2} d\omega |A_s(\omega)|^2 \right|^2. \tag{4}$$

When $\Omega \neq \omega_p$, this perfect phase correlation no longer holds, so its contribution can be neglected, similar to the s-s (i-i) terms. As a result, most of the spectral TPA (SFG) response is concentrated in a very narrow peak at the pump frequency (as narrow as the pump laser). The peak to background ratio is equal to the ratio of bandwidths between the down-converted light and the pump $N = \Delta_{\mathrm{DC}}/(2\delta_p)$, so it can reach many orders of magnitude with broadband down conversion from a narrow CW pump.

A salient feature in our approach is that the narrow spectral peak in the TPA (SFG) response can be controlled by simple spectral phase manipulations within the spectrum of the down-converted light. When a spectral phase $\phi(\omega)$ is applied to the down-converted field, the TPA (SFG) response becomes

$$R(\Omega) \propto \left| \int_0^{\omega_p/2} d\omega |A_s(\omega)|^2 e^{i[\phi(\omega) + \phi(\omega_p - \omega)]} \right|^2, \tag{5}$$

indicating that the total response is unaffected by antisymmetric phase functions. However, with symmetric spectral phase functions, such as a small relative delay or material dispersion, the spectral peak can be completely suppressed [2].

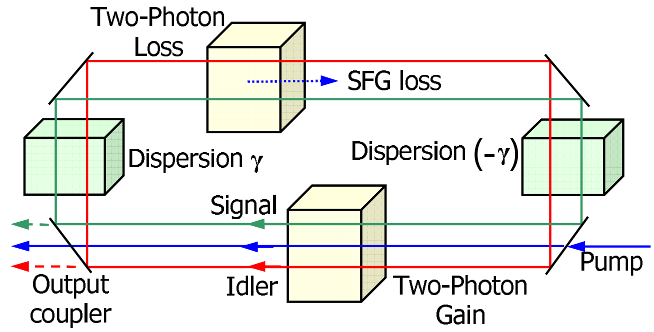

FIG. 1. (Color online) A pairwise mode-locked OPO cavity configuration.

It is important to understand the effect of a resonant cavity on the two-photon coherence properties of the generated light. Since a cavity is perfectly transparent at the resonant frequencies, where the amplitude or the phase of the resonant mode are not affected, the symmetry between the matching signal and idler cavity modes is preserved. Theoretically, in the absence of intracavity linear loss (apart from the output coupling), the squeezing properties of the light emitted from a cavity also remain intact. In practice, the actual degree of squeezing at high power can reach $\sqrt{T/L}$, where L is the linear intracavity loss and T is the output coupler transmission [11]. Due to the discrete nature of the spectrum in a cavity, the signal-idler temporal correlation is *periodic* with a periodicity of the cavity round-trip time. Yet, when many signal-idler mode-pairs oscillate, the temporal correlation is still transform limited by the broad oscillation spectrum (with periodic reoccurrence).

III. DESIGN PRINCIPLES OF THE OPO CAVITY

We consider the cavity configuration of a doubly resonant optical parametric oscillator, schematically shown in Fig. 1, where both the signal and the idler resonate. In addition to the gain medium, the cavity also includes a two-photon loss medium, inserted between two opposite dispersions. The two-photon loss medium can be, for example, a two-photon absorber or sum-frequency generator (it's exact properties are discussed later on). The main thrust of our cavity design is to exploit the dispersion in the cavity to control the two-photon loss, so that narrowband oscillations suffer loss, while broadband oscillations do not, thus becoming dominant. Specifically, a small amount of dispersion introduces a quadratic spectral phase, which according to Eq. (5), can reduce drastically the efficiency of two-photon processes. Thus, broadband oscillations can avoid the two-photon loss while narrow oscillations cannot.

In order for the dispersion not to affect down conversion at the gain medium, the phase relations should be restored after passage through the loss medium. This is achieved by introducing the inverse dispersion. Since this scheme inherently relies on the existence of a coherent phase relation between the fields to be manipulated, it is specific to broadband down conversion and not suitable to other broadband lasers. It is interesting to note that active mode locking of an

OPO (as opposed to passive here) was pursued in the past [12] as a source for coherent short pulses (frequency comb).

Our approach for obtaining broadband oscillations is inherently equivalent to passive mode locking for obtaining ultrashort pulses. In both approaches a nonlinear loss is inserted inside the cavity to enforce broadband oscillations; in passive mode locking it is Kerr lensing or saturable absorption [13,14], while in our approach it is a two-photon loss. In a mode-locked laser, ultrashort pulses are generated by locking the phases of all the single frequency modes to be equal, whereas in our approach ultrashort signal-idler temporal correlations are generated by locking the phases of all the frequency pairs to be equal, namely “pairwise mode locking.” Accordingly, one can view the mode-locked laser as a broadband one-photon coherent source and a pairwise mode-locked OPO as a broadband two-photon coherent source.

Just as in ultrafast mode locking, it is also necessary to compensate for the dispersion in our OPO cavity, so that the total dispersion per pass is zero. A time domain explanation for this is that efficient broadband down-conversion requires the signal-idler phase relations to be maintained after every pass in the cavity. A frequency domain explanation is that since $\omega_s+\omega_i=\omega_p$, the signal and the idler frequencies are exactly symmetric around the center frequency $\omega_p/2$. Yet, in a doubly resonant OPO, both the signal and the idler frequencies are chosen from the frequency comb of the passive cavity. Consequently, in order to enable a broadband oscillation, the passive frequency comb should be as symmetric as possible. Since the comb is distorted by the total dispersion in the cavity, a symmetric comb requires that all even orders of dispersion be zero [15]. Therefore, the dispersion γ introduced before the two-photon loss medium, must later be compensated. Odd orders of dispersion do not affect the interaction since they distort the comb symmetrically.

One of the most important properties of passively mode-locked lasers is that the frequency comb is exactly equally spaced, which is very appealing for precise measurements of optical frequencies [16,17]. Although the frequency comb of our OPO cavity is not necessarily equally spaced, symmetry around the center frequency is guaranteed with essentially the same precision (or even better, due to potential squeezing).

IV. ANALYSIS OF EXPECTED SPECTRUM, THRESHOLD AND PERFORMANCE

Let us now examine how the spectrum emitted from the OPO is affected by the properties of the two-photon loss medium. If the loss medium is nondispersive, this spectrum is influenced only by the dispersion γ between the gain and the loss media, which introduces a parabolic spectral phase $\exp(i\gamma\omega^2)$ onto the spectrum. We expect the OPO oscillation to develop such that the two-photon loss is minimal. The resulting mechanism of the oscillating frequency pairs in the OPO cavity is illustrated in Fig. 2. As shown in Fig. 2(a), the two-photon loss can even be completely eliminated when the oscillation is composed of two frequency pairs. The dispersion shifts the phase of the outer frequency pair by π compared to the inner pair ($\pi/2$ for every single frequency). As a result, the inner pair interferes destructively with the outer pair in the SFG/TPA process and there is no need for additional frequency pairs. However, for a truly broad oscillation, two frequency pairs are obviously not enough. To show how more pairs can be generated, we first consider having two loss media in the cavity at two different dispersion points γ_1 and γ_2 ($\gamma_2\approx 2\gamma_1$). In this case, any two frequency pairs selected to minimize the loss at the first medium according to γ_1 will suffer high losses at the second medium according to γ_2. In order to minimize both losses simultaneously, the OPO must double the number of frequency pairs to four, as shown in Fig. 2(b). Following this reasoning, three different dispersion values will lead to eight frequency pairs and so on. Consequently, the ideal two-photon loss medium is a highly dispersive two-photon absorber, where dispersion is continuously accumulated along the loss medium.

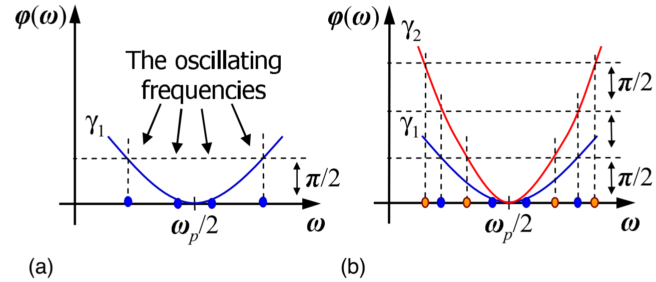

FIG. 2. (Color online) The selection mechanism of the oscillating frequency pairs in the OPO cavity. (a) Assuming a single two-photon loss medium at a dispersion value γ_1 and (b) assuming two media for two-photon loss at two different dispersion values (γ_1,γ_2).

Note, that if we use SFG in a long dispersive crystal for the loss process, the net result is just equivalent to having a single dispersion value (the middle value). This is because the SFG *amplitude* generated at the beginning of the crystal propagate through and coherently interferes with that generated at the end. Thus, in order to use SFG for the two-photon loss, it is necessary to somehow incoherently “dispose” of the generated SFG photons; for example, by absorbing the SFG photons within the crystal with some doping. A more practical alternative is to use the walk-off between the down converted and SFG beams. Specifically, in birefringent phase matching, where the polarizations of the down converted and the SFG photons are orthogonal, the SFG beam would usually be deflected by a walk-off angle. If the down-converted beam is tightly focused into the birefringent crystal, the SFG photons will leave the propagation path after a short distance, leading to a desired incoherence between losses at different positions in the crystal. For example, the walk-off angle in a type-I phase matched β-barium borate (BBO) crystal (pump wavelengths of 400–700 nm) is $\sim$65 mrad, indicating that when focused to 30 μm diameter the beams will not overlap after 0.45 mm. Thus, a 10 mm long crystal will be equivalent to $>$20 independent two-photon absorbers at different dispersion values.

In order to analyze the dependence of the conversion efficiency on the bandwidth of oscillations for the pairwise mode locked OPO (assuming steady state operation), we extend the analysis of the threshold pump intensity and conversion efficiency, previously done for the case of monochromatic signal and idler [9]. Specifically, we performed a similar analysis for broadband signal and idler with the mode competition suppression scheme. For simplicity, we assumed at the beginning just one nondispersive SFG two-photon loss medium in the cavity. Later we generalize the result for several (or even a continuum of) dispersion values.

Assuming that the depletion of the pump is relatively low and that the gain per pass in the cavity is not very high, it is valid to assume that the intensities of the signal and idler fields are constant throughout the cavity. Accordingly, the pump amplitude after the nonlinear gain medium A_p^+ can be written as

$$A_p^+ = A_p^0 - \chi l \int d\omega A_s(\omega) A_i(\omega_p - \omega), \tag{6}$$

where A_p^0 is the pump amplitude entering the medium, l the length and χ the nonlinear coupling constant is assumed to be independent of frequency, which is a reasonable assumption for frequencies close to the degeneracy point. Similarly, the amplitude of the SFG two-photon loss A_{TPL} is

$$A_{\mathrm{TPL}} = \chi l \int d\omega A_s(\omega) A_i(\omega_p - \omega) \exp(i\gamma\omega^2), \tag{7}$$

where γ denotes the dispersion accumulated during propagation from the gain medium to the two-photon loss medium. When other phase control mechanisms are used, Eq. (7) should be modified accordingly (without affecting the analysis). We considered for the loss only the correlated signal-idler mixing that sums back to the pump frequency, and disregarded uncorrelated terms at other frequencies. The justification for this is twofold: First, the uncorrelated terms are unaffected by a spectral phase, so they are "indifferent" to our manipulations [2]. Second, since the SFG process occurs in a long crystal, efficient SFG is possible only for a very narrow bandwidth around the pump frequency due to phase matching. Thus, contributions from uncorrelated terms are negligible.

Now, assuming the output coupler reflectivity to be equal for both the signal and the idler, the cavity conditions are symmetric, so the idler and the signal are complex conjugates. We can therefore incorporate Eq. (1) into Eqs. (6) and (7), to obtain

$$A_p^+ = A_p^0 - \chi l \int d\omega |A_s(\omega)|^2,$$

$$A_{\mathrm{TPL}} = \chi l \int d\omega |A_s(\omega)|^2 \exp(i\gamma\omega^2). \tag{8}$$

Defining the loss amplitude function $F(\gamma)$ as

$$F(\gamma) \equiv \int d\omega |A_s(\omega)|^2 \exp[i\gamma\omega^2], \tag{9}$$

yields

$$A_p^+ = A_p^0 - \chi l F(0), \quad A_{\mathrm{TPL}} = \chi l F(\gamma). \tag{10}$$

Note that $F(0)$ is proportional to the number of signal photons in the cavity, which is equal to the number of down-converted photon pairs.

It is now possible to write an energy conservation equation, where in steady state the number of photons per second lost from the pump is equal to the number of signal-idler photon pairs leaving the cavity per second, as

$$|A_p^0|^2 - |A_p^+|^2 - |A_{\mathrm{TPL}}|^2 = TF(0), \tag{11}$$

where T is the loss in the cavity, which is equal to the output coupler transmission in an ideal cavity. Substituting Eq. (10) into Eq. (11) and performing some algebra, yields

$$\frac{TF(0)}{|A_p^0|^2} = \frac{1}{1 + |F(\gamma)/F(0)|^2}\left(\frac{2T}{\chi l A_p^0} - \frac{T^2}{\chi^2 l^2 |A_p^0|^2}\right). \tag{12}$$

The left-hand side of Eq. (12) can be identified as the conversion efficiency η, since it is just the number of down-converted signal-idler photon pairs leaving the cavity per second divided by the number of incident pump photons per second. Since we assume perfect phase matching, the pump field can be taken as real, and together with the expression for the threshold pump intensity [9], of

$$|A_{p\text{-}th}|^2 = \frac{T^2}{4\chi^2 l^2}, \tag{13}$$

we can rewrite Eq. (12) as

$$\eta = \frac{4}{1 + |F(\gamma)/F(0)|^2}\left(\left|\frac{A_{p-th}}{A_p^0}\right| - \left|\frac{A_{p-th}}{A_p^0}\right|^2\right). \tag{14}$$

For convenience, we now define N as the ratio between the actual pump intensity (I_p) and the threshold pump intensity (I_{th})

$$N \equiv \frac{I_p}{I_{p-th}} = \left|\frac{A_p^0}{A_{p-th}}\right|^2. \tag{15}$$

Substituting Eq. (15) into Eq. (14), yields

$$\eta = \frac{4}{1 + |F(\gamma)/F(0)|^2}(\sqrt{N} - 1)\frac{2}{N}. \tag{16}$$

Equation (16) indicates that for equal thresholds (equal N), the dominant oscillations will be those that minimize the two-photon "tax" $|F(\gamma)|^2$ for any pumping power (for any N); i.e., a broad oscillation. As explained above, several (preferably a continuum of) independent loss media at different dispersion values are required for a stable broad oscillation. Then, the dominant oscillations would be those that minimize the total "tax," specifically

$$\sum |F(\gamma_n)|^2 \to \int d\gamma |F(\gamma)|^2, \tag{17}$$

where the integral expression stands for the continuum limit.

Comparing the two limiting possibilities of very narrow oscillations, where the two-photon "tax" is essentially independent of γ, and very broad oscillations, where the two-

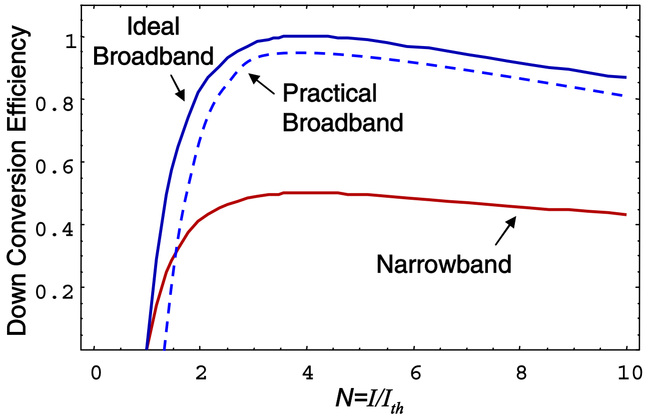

FIG. 3. (Color online) Calculated conversion efficiency as a function of $N=I_p/I_{th}$ for a narrow oscillation, ideal broadband oscillation and practical broadband oscillation (dashed). I_{th} is taken to be the minimum threshold intensity among all possible narrowband oscillations.

photon loss tends to zero for a large enough dispersion, it is evident that the improvement in conversion efficiency approaches a factor of 2. Such an improvement is significant, indicating why broad oscillations would be highly favored. In practice, it is expected that broad oscillations will have a slightly higher threshold. Thus, when the pump power is low, narrow oscillations will dominate. But, as the pump power is increased well above threshold, the situation becomes more and more favorable for the broader oscillations. The conversion efficiency as a function of N is given in Fig. 3 for three cases: (1) very narrow oscillations, (2) ideal broad oscillations, where the threshold is as low as for narrow oscillations, and (3) practical broad oscillations, where the threshold is slightly higher. Note that for very broadband oscillations, where the two-photon loss is negligible, the conversion efficiency can approach unity around $N=4$. This, of course, is most desirable for applications.

Figure 3 indicates that in order to obtain broadband oscillations, it is desirable that the threshold for broad oscillations will be equal to the minimum threshold of the narrowband oscillations. In other words, since broadband oscillations can be decomposed into many signal-idler frequency pairs, it is desired that all these pairs will have the same threshold —i.e., that the threshold intensity will be independent of wavelength. This requirement can be met to a high degree as depicted in Fig. 4, in which the calculated threshold intensity as a function of wavelength is presented for two cases of broad phase matching. One case, shown in Fig. 4(a), involves a PPKTP crystal pumped at 532 nm. The other case, shown in Fig. 4(b), involves a BBO crystal pumped at 728 nm. As evident, the threshold intensity is essentially constant (up to 15%) over the entire phase matching bandwidth.

V. EXPERIMENTAL PROOF OF PRINCIPLE

In order to demonstrate experimentally the underlying principles of pairwise mode locking, we constructed the linear OPO cavity schematically depicted in Fig. 5. Gain in

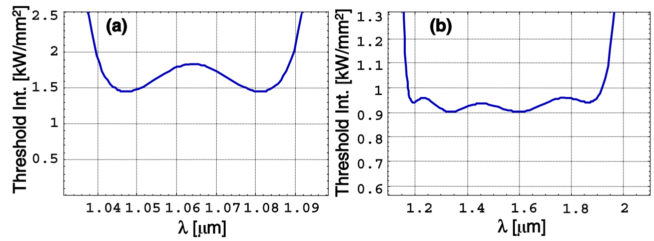

FIG. 4. (Color online) Threshold pump intensity (I_{th}) as function of signal wavelength. (a) For a 10 mm long periodically polled KTP crystal with 4% loss in the cavity pumped by 532 nm in a broad phase matching configuration. (b) For a 14 mm long BBO crystal with 1% loss in the cavity pumped by 728 nm in an ultrabroad, zero dispersion, phase matching configuration.

such a linear cavity OPO exists only when the down-converted light propagates through the medium along the direction of the pump. Therefore, for the two-photon loss, we exploited SFG that occurs during backward propagation. Due to this SFG loss, the conversion efficiency for narrowband oscillations of a doubly resonant OPO cannot exceed 50% in a linear cavity configuration [9], so a ring cavity would be required in order for narrowband oscillations to overcome this limit. Since for broadband oscillations the SFG loss can be eliminated, the conversion efficiency with our OPO can exceed 50% even with a linear configuration. Moreover, since the SFG photons are emitted backward out of the OPO cavity, it is possible to isolate and detect them in order to measure the two-photon loss for both narrowband and broadband oscillations. A disadvantage of the configuration in Fig. 5 is that both the gain and the loss occur in the same crystal, so it is impossible to exploit the walk-off effect in order to obtain a continuum of dispersion values; effectively the configuration is equivalent to having one nondispersive two-photon loss at a specific dispersion value. Accordingly, the oscillation spectrum is expected to contain four strong lobes instead of a truly broad spectrum. This configuration, can therefore demonstrate only how a two-photon loss in the cavity leads to the first step of pairwise mode locking (four frequencies), but not full scale pairwise mode locking.

As noted earlier, it is necessary to match the cavity frequency comb to the pump frequency in a doubly resonant OPO. In our experimental configuration, this was achieved by inserting an electro-optic phase modulator (EOM) into the OPO cavity [a 10 mm long $RbTiPO_4$ (RTP) crystal]. The positive dispersion in the cavity, mainly from the two crystals (EOM+gain medium), was calculated to be 3800 fs^2 and the prism pair, made of highly dispersive SF57 glass and separated by a distance of 85 cm, introduces the opposite dispersion. An intracavity lens ($f=125$ mm) and the curved output coupler ($R=75$ mm) focused the beams to about 30 μm diameter inside the gain medium - a 12 mm long periodically polled $KTiPO_4$ (PPKTP) crystal, pumped at a wavelength of 532 nm from a single-frequency doubled Nd:Yag laser (Verdi from Coherent). The bandwidth allowed by phase matching for this crystal is ~50 nm around 1064 nm. The output coupler transmission was 2%. Under these conditions, a threshold of ~0.3 W for oscillations was measured.

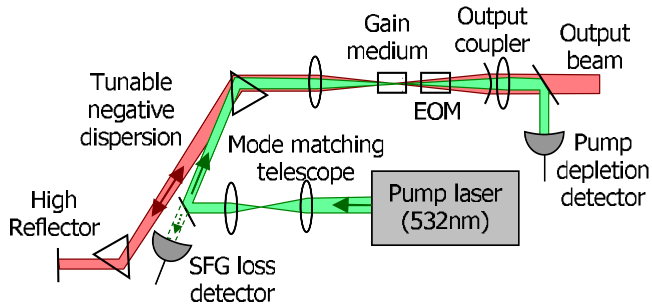

FIG. 5. (Color online) Experimental OPO cavity configuration.

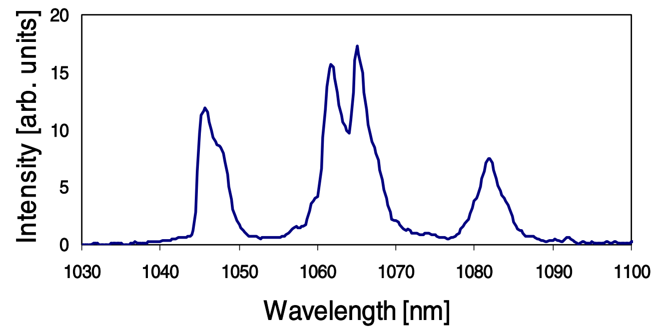

FIG. 7. (Color online) A typical oscillation spectrum of the OPO configuration in Fig. 5.

The experimental measurements of the pump depletion and the SFG two-photon loss are presented in Fig. 6. All measurements were taken at a pump power of 1 W, and an output power of ~150 mW emitted from the OPO. When the voltage applied to the EOM is varied, the length of the cavity is linearly scanned. Accordingly, Figs. 6(a) and 6(c) represent two different measurements of the pump intensity after passage through the cavity as a function of the differential cavity length in units of the pump wavelength. Obviously, the pump is strongly depleted whenever the cavity length satisfies the oscillation condition (twice in every scan). Figures 6(b) and 6(d) are the corresponding measurements of the SFG two-photon loss, showing the loss that appears whenever an oscillation develops. Using the prism pair to tune the dispersion we performed these measurements twice: First, when a residual dispersion (~600 fs^2) exists in the cavity [Figs. 6(a) and 6(b)] and then, when the dispersion is optimally compensated [Figs. 6(c) and 6(d)].

When dispersion exists in the cavity, a broadband oscillation cannot develop since the oscillation condition is different for different frequency pairs. As a result, only a narrowband oscillation develops at any specific cavity length, and as expected, the measured SFG loss is high and the pump depletion low. However, when the dispersion is compensated, a broadband oscillation is allowed and indeed the SFG loss is negligible and the pump depletion very high. The results in Fig. 6(b) were obtained at the center of the pump beam with a small detector (0.8 mm^2) and indicate depletion of >85%. The total depletion, measured with a large detector, was >60% probably because of imperfect spatial overlap between the pump and the cavity mode due to residual astigmatism from the prism pair. Since the measured SFG loss was negligible, this indicates a down-conversion efficiency of more than 50%. Due to imperfect polarization control in this preliminary configuration, linear intracavity losses were relatively high, yielding an output coupling efficiency of only 25% and a total emitted power of only 150 mW.

We then added a feedback loop to actively lock the OPO cavity length to the pump frequency in order to obtain stable oscillations. We first verified that the cavity emits CW light with a fast detector and then measured the emitted spectrum with a fast spectrometer (4 ms integration time). A typical spectrum is shown in Fig. 7. The spectrum was unstable and varied on a time scale of ~0.5 s. Yet, it was indeed composed of four lobes, with a spacing that agrees well with the

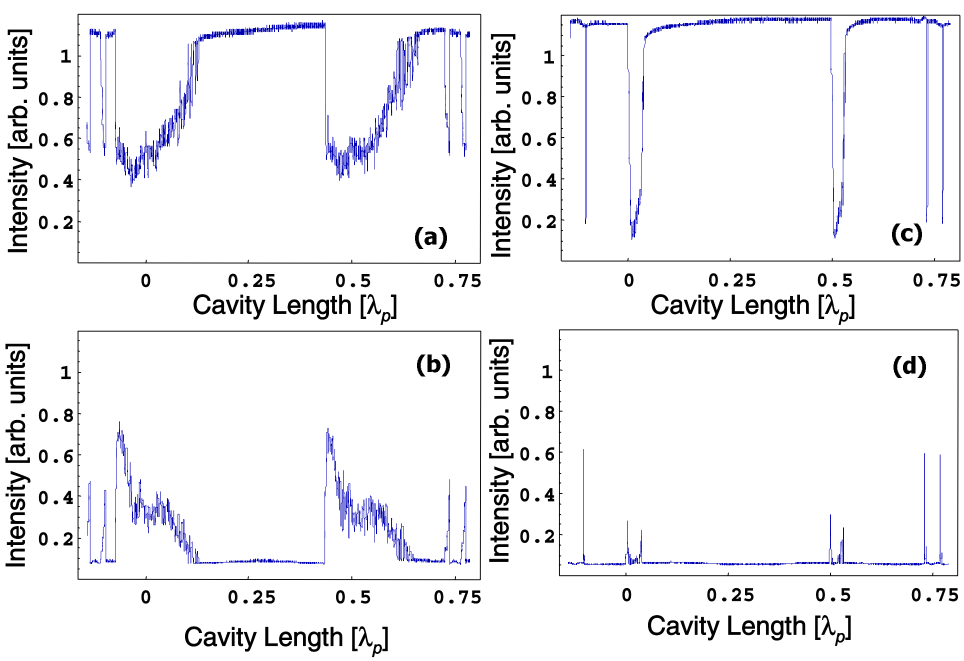

FIG. 6. (Color online) Measurements of pump depletion and SFG loss as a function of the differential OPO cavity length over a range of one pump wavelength. (a) Pump depletion and (b) the corresponding SFG loss when dispersion is not optimally compensated (~600 fs^2 residual dispersion). (c) Pump depletion and (d) corresponding SFG loss when dispersion is optimally compensated.

above dispersion analysis, taking into account the actual dispersion in the cavity. The reason for the instability is that the four oscillating frequencies are not unique, so minute changes in the cavity, such as uncompensated acoustic vibrations or air currents, may cause the frequencies to spontaneously hop.

In order to demonstrate the two-photon coherence of the light it is necessary to show that TPA or SFG with the generated light can be controlled by spectral phase manipulations. The measurements of the pump depletion and intracavity SFG loss in Fig. 6 serve this purpose directly. Efficient down conversion is possible only when two-photon coherence is satisfied, and the fact that the SFG loss is practically eliminated with the four-lobed spectrum is a direct manifestation of the destructive interference between different frequency pairs within the oscillation spectrum due to two-photon coherence.

VI. CONCLUDING REMARKS

We investigated both theoretically and experimentally a pairwise mode-locking approach, that should lead to a low threshold OPO source emitting a broad spectrum of down-converted light. In this paper we considered a doubly resonant OPO, but the concept of pairwise mode-locking applies equally to singly resonant OPOs (only one of the signal-idler fields resonates). Indeed, assuming both signal and idler traverse the two-photon loss medium with opposite dispersions, the resulting spectrum will be similar. Although the oscillation threshold of a singly resonant OPO is higher, the needed configuration can be much simpler, since it does not require active locking of the cavity to the pump frequency (the idler frequencies are not constrained by the cavity).

Due to the direct analogy between passive mode locking of ultrafast lasers and our pairwise mode locking, we believe that the precise symmetry in the frequency comb of a pairwise mode-locked OPO can be advantageous for precision two-photon spectroscopy, just as the precise comb of ultrafast lasers is currently a major tool for precision one-photon spectroscopy.

Finally, although our analysis was purely classical, the potential nonclassical properties of such an OPO are of great interest. As mentioned before, a major limitation in the generation of squeezed light are the linear losses in the OPO cavity. Since our pairwise mode locking involves only two-photon losses that do not affect the signal-idler photon-number correlation, our OPO can potentially generate broadband squeezed light, which can be applicable to suppression of spontaneous emission [18] as well as to optical phase measurements at the Heisenberg limit [19,20]. It is clear that due to high linear losses in our current experiment, substantial squeezing was not achieved so far.

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