Shaping the spectrum of downconverted photons through optimized custom poling

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We present a scheme for engineering the joint spectrum of photons created via spontaneous parametric down conversion. Our method relies on customizing the poling configuration of a quasi-phase-matched crystal. We use simulated annealing to find an optimized poling configuration which allows almost arbitrary shaping of the crystal’s phase-matching function. This has direct application in the creation of pure single photons—currently one of the most important goals of single-photon quantum optics. We describe the general algorithm and provide code, written in C++, that outputs an optimized poling configuration given specific experimental parameters.

I. INTRODUCTION

The generation of pure non-classical states of light is one of the most important goals of optical quantum information science. At present, the most popular method for generating quantum light relies on spontaneous parametric down conversion (SPDC) [1]—a nonlinear process that converts high-energy photons into pairs of lower energy photons. Photon sources based on SPDC have widespread application in quantum computation [2], quantum communication [3] and quantum metrology [4, 5], as well as in more specialized areas such as quantum imaging [6], quantum lithography [7] or optical coherence tomography [8]. The ability to control the characteristics of quantum states of light becomes increasingly important as these experiments evolve. In particular, the generation of high-purity heralded, or even near-deterministic, single photons will require careful engineering of the spectral shape and correlations of the generated photon pairs.

In general, photon-pairs generated via SPDC are correlated in frequency and are described by a joint spectral amplitude. The joint spectral amplitude is determined by the properties of the incident pump field and the material properties of the nonlinear crystal used to facilitate the downconversion process. Detection of a photon in one mode heralds the presence of another photon in the second mode. However, spectral correlations between the photons degrade the spectral purity of heralded photons.

The most common method for modifying the shape of the joint spectral amplitude is filtering, however, this can lead to photon loss, which can in turn degrade the purity of the generated quantum state [9, 10].

More sophisticated methods involve shaping the spectrum at the source using techniques such as quasi-phase-matching (QPM). In QPM, a nonlinear structure is constructed from individual domains of birefringent material with alternating orientation. Periodic QPM has been used to reduce the correlations in the joint spectral amplitude, leading to high-purity heralded single photons [11–15]. However, the extent to which spectral separability can be achieved is limited by the side lobes in the joint spectral amplitude that arise from the finite length and rectangular shape of the crystal. Modest filtering is still required to achieve high spectral purity.

QPM has also been used for the purposes of spectral shaping [19, 20]. For example, chirped gratings were employed for pulse compression in second-harmonic generation. Chirped gratings have also been used for the generation of ultra-broad-spectrum, top-hat shaped photons for optical coherence tomography [8]. However, most methods for spectral shaping involve changing the domain width, which may be incompatible with stringent phase-matching conditions.

Using non-trivial QPM for the generation of spectrally decorrelated photons was proposed by Brańczyk et al. [21], who showed that modulation of the nonlinearity profile of a downconversion crystal can be used to remove the side lobes and drastically improve the spectral purity of heralded single photons. In this method, a discretised approximation to the desired nonlinearity profile was achieved using higher-order poling. Dixon et al. [22] proposed an alternative method for spectral decorrelation, in which the crystal’s duty-cycle pattern is customized while the grating period is fixed.

In this paper, we present a new method, in which we directly manipulate the domain orientations. Our method has certain advantages over the methods in [21] and [22]. In particular, our method allows almost arbitrary shaping of the phase-matching function. This provides flexibility in designing the maximum nonlinearity for a given phase-matching function width—a property that is fixed in [22] and not easily modified in [21]. Furthermore, in contrast with the method in [22], we fix the width of each domain in the grating. This preserves the phase-matching properties of the crystal—a stringent requirement in many experiments.

II. SHAPING THE PHASE-MATCHING FUNCTION

Spontaneous parametric downconversion is a nonlinear process that facilitates the conversion of high-energy
 photons (typically referred to as pump photons in mode $p$) into pairs of lower energy photons in modes $a$ and $b$. This process satisfies energy and momentum conservation according to: $\omega_p = \omega_a + \omega_b$ and $k_p(\omega_p) = k_a(\omega_a) + k_b(\omega_b)$, where $\omega_j$ is the frequency in mode $j$, and $k_j(\omega) = n_j(\omega)\omega/c$ is the wave vector associated with the polarization of mode $j$, evaluated at frequency $\omega$.

Theoretically, the two-photon state generated via spontaneous parametric down conversion can be described by

$$|\psi\rangle = \int d\omega_a \int d\omega_b f(\omega_a, \omega_b)|\omega_a\rangle_a|\omega_b\rangle_b,$$  

where $|\omega_i\rangle$ is a one-photon Fock state of frequency $\omega_i$ prepared in mode $j$. The two photons are characterized by the bi-photon spectral function $f(\omega_a, \omega_b) = \alpha(\omega_a + \omega_b)\Phi(\omega_a, \omega_b)$, otherwise known as the joint spectral amplitude. The spectral properties of downconverted photons can be manipulated via the pump envelope function $\alpha(\omega_p) = \alpha(\omega_a + \omega_b)$ or the phase-matching function $\Phi(\omega_a, \omega_b)$. In general, the joint spectral amplitude cannot be factorized into a product of separable single-photon spectral amplitudes: $g(\omega_a)h(\omega_b)$. Factorization of the joint-spectral amplitude is desirable for the generation of pure heralded single photons, as discussed in Section IV-B.

In this paper, we focus on shaping of the phase-matching function. The phase-matching function is related to the nonlinearity profile of the crystal via the Fourier transform:

$$\Phi(\omega_a, \omega_b) = \int_{-\infty}^{\infty} \chi(z)e^{-i\Delta k(\omega_a, \omega_b)z}dz,$$  

where $\chi(z)$ represents the nonlinear coupling, dominated by the material nonlinearity; and where phase matching within the crystal can be described by the phase-mismatch $\Delta k(\omega_a, \omega_b) = k_p(\omega_a + \omega_b) - k_a(\omega_a) - k_b(\omega_b)$. We will use $\Delta k$ as shorthand for $\Delta k(\omega_a, \omega_b)$.

In principle, an arbitrary $\Phi(\omega_a, \omega_b)$ could be realized with appropriate design of the crystal’s nonlinearity profile such that it corresponds to the Fourier transform of the desired phase-matching function. Unfortunately, it is non-trivial to directly change the material properties of a nonlinear crystal, and different methods must be used.

A typical crystal has a uniform nonlinearity profile, and can be characterized by $\chi(z) = \chi_0\text{rect}(z/L)$. Here $\chi_0$ is the nonlinear coefficient of the material, $L$ is the length of the crystal, and $\text{rect}((z-x)/y)$ is a rectangular function of width $y$ centred at $x$. According to Eq. (2), the phase-matching function of such a crystal is $\Phi(\omega_a, \omega_b) = \chi_0L\text{sinc}(\Delta kL/2)$, where $\text{sinc}(x) = \sin(x)/x$.

The function $\Phi_{\text{rect}}(\omega_a, \omega_b)$ is peaked at $\Delta k(\omega_a, \omega_b) = 0$ meaning that photon-pairs will be generated within a range of frequencies that satisfy this condition. These photons will have a spectral shape characterized by the sinc function. One can control the width of the sinc function by cutting the crystal to a different length, however, the peak frequency and spectral shape remain fixed for a given material.

A common method for manipulating the central peak of the phase-matching function is a QPM technique called periodic poling. Here, the nonlinear medium is composed of $N$ domains of birefringent material with alternating orientation—either up or down. The width of each domain is given by the coherence length $l_c$, and the poled crystal is often characterized by the poling period $\Lambda = 2l_c$. A flip in the orientation of the domain introduces a phase shift of $\pi$.

In a poled crystal, the nonlinearity profile $\chi(z)$ is a discontinuous function that only takes on values of $\pm \chi_0$. To a good approximation, this technique produces a phase-matching function that, with increasing $N$, approaches

$$\Phi(\omega_a, \omega_b) \propto \text{sinc} \left( \frac{(\Delta k - \frac{2\pi}{\Lambda})L}{2} \right).$$

This shifts the center of the function by $2\pi/\Lambda$, allowing the generation of photons within a desired frequency range.

To understand this frequency shift, recall that the phase-matching function for the crystal arises from constructive interference between the phase-matching functions for each individual crystal domain:

$$\Phi(\omega_a, \omega_b) = \chi_0 \sum_{n=1}^{N} s_n \int_{-\infty}^{\infty} \text{rect} \left( \frac{z - z_n}{l_c} \right) e^{-i\Delta k z}dz,$$  

where $s_n$ accounts for the phase shift due to the orientation of the domain and $z_n$ specifies the origin of the $n$th domain. For first-order poling, $s_n = e^{in\pi}$ and $z_n = [(n - \frac{1}{2})l_c]$. The phase-matching function for each domain is centred about $\Delta k = 0$, however due to the phase factor associated with each domain, for first-order poling the functions constructively interfere at $\Delta k = 2\pi/\Lambda$, and approximate Eq. (3) up to some overall phase. Periodic poling provides control over the central peak of the function, however, the shape remains limited to a sinc function.

In Eq. (5), contributions from the $e^{i\Delta k z_n}$ factor are fixed by the relative position of the $n$th domain with respect to the rest of the crystal. However, contributions from $s_n = \pm 1$ are determined by the relative orientation of the domain—a controllable parameter. It is therefore natural to consider whether the phase-matching function can be tailored by manipulating the relative orientations of the individual domains in a non-trivial way. This was demonstrated by Braunyczyk et al. [21], where the authors designed and experimentally verified a Gaussian phase-matching function for a $1\text{ cm KTP}$ crystal with $\Lambda = 10.85\mu\text{m}$.

However, the design in [21]—and to a large extent the methodology—was specific to a particular set of parameters, and cannot easily be generalized. In this paper, we
introduce a general technique for optimizing the domain orientations in order to achieve a desired phase-matching function. In the next section, we describe the algorithm used for this optimization.

III. THE ALGORITHM

The task of finding an optimal domain configuration is formulated in terms of discrete optimization. The variables are the $N$ possible domain orientations $s_n$, constrained as $s_n \in \{-1, 1\}$. Thus, the solution space consists of $2^N$ possible crystal configurations. Neighbour configurations are defined as those which differ by exactly one domain orientation. Thus, any particular configuration has $N$ neighbours, since there are $N$ domains which could be flipped.

Each domain configuration $s = (s_1, \ldots, s_N)$ then has a corresponding non-linearity profile, with a phase-matching function $\Phi_s(\Delta k)$ given by the Fourier transform of this non-linearity profile. The task is to find the crystal alignment $s_0$ which yields a $\Phi_{s_0}(\Delta k)$ closest to some target function, $\Phi_{\text{target}}(\Delta k)$, on a specified range $[a, b]$. We define a cost function

$$d_s = \sum_{m=1}^{M} |\Phi_{s}(\Delta k_m) - \Phi_{\text{target}}(\Delta k_m)|$$

as a measure of the distance between $\Phi_{s}(\Delta k)$ and $\Phi_{\text{target}}(\Delta k)$. This distance is measured by selecting sufficiently many, say $M = 2000$, random points $\Delta k_m$ in the range $[a, b]$. The objective is to minimize $d_s$.

The solution space is simply too large to check each possible configuration. It is also not convex, meaning that there exist configurations which are locally optimal—that is, each neighbour is a worse configuration—but not globally optimal; one can not simply move from neighbour to neighbour, always selecting the better configuration. Thus, we use simulated annealing [23], a method which accepts worse configurations with a decaying probability $p$, and better configurations with some probability $q$ that is close to 1.

Starting with either a random or preselected domain configuration, the algorithm iterates through randomly-chosen neighbour configurations, deciding whether to flip a given domain or not. If $s$ is the current configuration, and $s'$ is a neighbour configuration being considered, then when $d_s \leq d_{s'}$, the algorithm moves to $s'$ with probability $p$; however, when $d_s > d_{s'}$, the algorithm moves to $s'$ with probability $q$.

The probability of accepting a worse configuration at the iteration $i$ is given by $p_i = h_i d_s / (A d_{s'})$, where $A$ is roughly the number of domains explored before the algorithm hits a local minimum, and $h_i$ is a decaying function known as the ‘heat function’ (in analogy with physical annealing). The algorithm is first run with constant probabilities $p_i = 0$ and $q = 1$, plotting the cost function at each iteration. The parameter $A$ is then given by the number of configurations explored at the point where the algorithm stops making significant progress. This procedure can be used to obtain a rough approximation to the parameter $A$. Best results are achieved by fine-tuning $A$, typically by trial and error approach.

The heat function is chosen to be $h_i = 2^{-2i/J}$, where $i$ is the iteration and $J$ is the total number of iterations. Finally, we use $q = 0.999$, determined heuristically. Any number close, but not equal to 1 tends to give good results.

Parameters $q$ and $A$ can be adjusted further to improve performance of the algorithm.

A. Specifying phase-matching function $\Phi_{\text{target}}(\Delta k)$

At the beginning of the algorithm, the target function $\Phi_{\text{target}}(\Delta k)$, normalized to a height $H$, is specified; as well as the range $[a, b]$. The target function will typically have an identifiable peak or center located at a $\Delta k$, which we will call $\Delta k_{\text{peak}}$. If there is no obvious peak, $\Delta k_{\text{peak}}$ can be chosen to be a point in the range $[a, b]$. The point $\Delta k_{\text{peak}}$ is used to determine the desired domain length, according to $l_c = \pi / \Delta k_{\text{peak}}$.

The number of domains $N$ and the height $H$ are estimated heuristically and can be modified to improve the performance of the algorithm. The number of domains $N$ is determined by the desired total length of the crystal $L$, according to $N = L / l_c$. An appropriate choice for $L$ is determined by first taking a Fourier transform of the target phase-matching function and taking $L$ to be the length over which the Fourier transform is appreciably non-zero. The algorithm assumes a bulk nonlinearity normalized to 1. For a bulk crystal with no poling (i.e. $s_n = +1$), we have a maximum nonlinearity of $L$ at $\Delta k = 0$ and negligible nonlinearity at $\Delta k = \Delta k_{\text{peak}}$. For first-order poling, we have a maximum nonlinearity of $2L / \pi$ at $\Delta k = \Delta k_{\text{peak}}$. The height of the target function should therefore be in the range $0 < H < 2L / \pi$.

The algorithm stops after a predetermined number of iterations, which should be large enough to allow the algorithm to find an optimal configuration. We note that the algorithm is probabilistic, and may therefore occasionally yield an unsatisfactory solution. If this occurs, the algorithm should be run again.

IV. RESULTS

In this section, we consider the task of engineering a Gaussian phase-matching function, and compare our method to existing methods. We then show how a Gaussian phase-matching function can be used to generate pure heralded single-photons—and compare the purity of photons generated using our method with that of other methods. We also show that the algorithm can be used to approximate other phase-matching functions of interest.
FIG. 1. Comparison of phase-matching functions for different apodization schemes (scaled target functions are shown in orange). a) Standard first-order periodically poled crystal (no apodization). We define the peak nonlinearity of this crystal to be unity and scale all other peak nonlinearities accordingly. b) Method proposed by Brańczyk et al. [21]. c) Method proposed by Dixon et al. [22]. In the Dixon scheme, the peak center is shifted due to the customized duty cycle. For comparison, we have shown the shifted target function in green. d) Domain configuration generated using the simulated annealing algorithm presented in this paper. We define Δk′ = Δk − 2π/Λ.

A. Gaussian phase-matching function

For comparison with the specific example given in Brańczyk et al. [21], we consider the generation of a Gaussian profile proportional to exp(−Δk²/2σ²) where σ = 568, using a ppKTP crystal with Λ = 10.85µm. This standard deviation corresponds to a Gaussian whose width matches the width of the phase-matching function generated using a first-order-poled crystal with N = 1045 domains, shown in Figure 1 a). We define the peak nonlinearity of this crystal to be unity and scale all other peak nonlinearities accordingly. Figure 1 b) shows the phase-matching function for the crystal from [21]. This design used N = 1846 domains.

The scheme proposed by Dixon et al. [22] has certain advantages over the design in [21]. The Dixon scheme can easily be adapted for any number of domains. Furthermore, for a given crystal length, the Dixon crystal outputs a higher peak nonlinearity than the Brańczyk crystal. However, for a given phase-matching-function bandwidth, the two designs have comparable peak nonlinearities, as can be seen by comparing Figure 1 c) with b). The Dixon scheme also suffers from the potential drawback that the generated phase-matching function is translated and slightly broadened from what one would expect of a first-order-poled crystal with the same N and Λ. However, one can compensate for this shift and broadening by modifying the parameters N and Λ.

The phase-matching function generated using our simulated annealing algorithm is shown in Figure 1 d). To generate this function, we used a first-order poled crystal with N = 1846 domains as the initial trial function. We then ran the algorithm with 10⁶ iterations, using probability q = 0.999 and A = 1000 in the definition of the acceptance probability p₁, see Section III. We used M = 2001 random samples to compute the objective function dₙ over the range [a, b] = [(1 − 0.02)²/Λ, (1 + 0.02)²/Λ]. The height of the target function was 0.45 × 2L/π where L = Nlₐ. The side lobes in Figure 1 d) are noticeably reduced compared with the other schemes—Figures 1 a)-c)—and the output function is a much closer approximation to the desired Gaussian function.

We next show how the phase-matching function generated with our algorithm can be used to increase the purity of heralded single photons.
as well as the purity of the heralded photon, we characterize by the joint spectral amplitude.

The Schmidt coefficients for the same material and poling period \(\Lambda\), with a duty cycle customized according to Dixon et al. [22]. In the Dixon scheme, the peak center is shifted due to the customized duty cycle. c) Domain configuration generated using the simulated annealing algorithm presented in this paper. All plots were generated using the Sellmeier equations given in [25, 26]. We define \(\Delta\omega_j = \omega_j - \bar{\omega}_j\) where \(\bar{\omega}_j = \omega_p / 2\).

### B. Pure heralded single photons

Recall that bi-photons generated via downconversion are characterized by the joint spectral amplitude \(f(\omega_a, \omega_b) = \alpha(\omega_a + \omega_b) \Phi(\omega_a, \omega_b)\), where \(\alpha(\omega_a + \omega_b)\) is the pump envelope function and \(\Phi(\omega_a, \omega_b)\) is the phase-matching function. The detection of a single photon in one mode of the downconverted state heralds the presence of another single photon in the other mode. Spectral correlations in \(f(\omega_a, \omega_b)\), i.e., entanglement between the downconverted modes, reduce the spectral purity of the heralded photon.

To characterize the degree of entanglement in \(|\psi\rangle\), as well as the purity of the heralded photon, we first consider the bi-photon’s Schmidt decomposition \(|\psi\rangle = \sum_k b_k |u_k\rangle_a |v_k\rangle_b\). The Schmidt modes \(|u_k\rangle_a = \int d\omega_a g_k(\omega_a) |\omega_a\rangle\) and \(|v_k\rangle_b = \int d\omega_b h_k(\omega_b) |\omega_b\rangle\) each form a discrete basis of complex orthonormal functions. The Schmidt coefficients \(b_k\) are real and satisfy \(\sum_k b_k^2 = 1\) if \(f(\omega_a, \omega_b)\) is normalized. For a completely decorrelated joint-spectral amplitude, \(f(\omega_a, \omega_b) = g(\omega_a)h(\omega_b)\) and the Schmidt decomposition contains only one term: \(|\psi\rangle = b_1 |u_1\rangle_a |v_1\rangle_b\), where \(b_1 = 1\).

The degree of entanglement of a pure bipartite state can then be characterized by the entropy of entanglement [24]. In terms of the Schmidt coefficients, this is given as \(E = -\sum_k b_k^2 \log_2(b_k^2)\).

Given detection of a single photon in mode \(b\) by a detector that does not provide any spectral information, the single-photon state in mode \(a\) can be written as \(\rho_a = \sum_k b_k^2 |u_k\rangle_a \langle u_k| \cdot a\). The purity of the reduced density matrix is given by \(P = Tr[\rho_a^2] = \sum_k b_k^2\).

A popular method for reducing spectral correlations in the joint spectral amplitude is to work in a regime where the group velocities satisfy \(k'_p = (k'_a + k'_b) / 2\), where \(k'_p = \partial k_j(\omega) / \partial \omega|_{\omega = \bar{\omega}_j}\) and \(\bar{\omega}_j\) are the central frequencies [13]. For maximal decoherence, the width of the phase-matching function should match the width of the pump function as closely as possible. For a standard type-II first-order-poled crystal, this is achieved when \(4/\sigma^2 = \gamma L^2(k'_a - k'_b)(k'_b - k'_a)\), where \(\sigma\) is the pump bandwidth, \(L\) is the crystal length and \(\gamma \approx 0.193\). Figure 2 a) shows the joint-spectral amplitude and Schmidt coefficients for a ppKTP crystal with \(\Lambda = 46.1\mu m\) and \(N = 570\) domains, pumped by a 791nm laser with a bandwidth of \(\sigma = 1.3\text{nm}\). Notice that the side lobes that arise from the sinc profile of the phase-matching function admit some correlations between the two photons, resulting in a heralded photon purity of \(P = 0.865\).

Figure 2 a) shows the joint-spectral amplitude and Schmidt coefficients for the same material and poling period \(\Lambda\), with a duty cycle customized according to Dixon et al. [22]. To compensate for the broadening induced by this method, we increased the number of domains to \(N = 660\) to achieve the most factorable joint-spectral amplitude, and therefore the highest possible heralded-photon purity. We didn’t compensate for the spectral shift as this does not affect the purity of the heralded photons. The side lobes are drastically reduced and the heralded photon purity is increased to \(P = 0.979\).
The joint spectral amplitude and Schmidt coefficients generated using our algorithm are shown in Figure 2 c). To generate this function, we used a crystal with \( N = 1000 \) randomly oriented domains as the initial trial function (in the previous section, the initial trial function was predetermined). We then ran the algorithm with \( 2 \times 10^5 \) iterations, using \( q = 0.999 \) and \( A = 1000 \). We used \( M = 2001 \) random samples to compute the objective function \( d_q \), over the range \([a, b] = [(1 - 0.005) \frac{2\pi}{L}, (1 + 0.005) \frac{2\pi}{L}]\). The height of the target function was \( 0.45 \times 2L/\pi \) where \( L = NL_c \). Using our algorithm, the heralded photon purity is increased to \( P = 0.999 \).

### C. Other functions

The simulated annealing algorithm can also be used to approximate other phase-matching functions of interest.

A triangular phase-matching function is shown in Figure 3 a). To generate this function, we used a crystal with \( N = 3500 \) randomly oriented domains as the initial trial function. We then ran the algorithm with \( 4 \times 10^4 \) iterations, using \( q = 0.999 \) and \( A = 1000 \). We used \( M = 2001 \) random samples to compute the objective function \( d_q \), over the range \([a, b] = [(1 - 0.005) \frac{2\pi}{L}, (1 + 0.005) \frac{2\pi}{L}]\). The height of the target function was \( 0.3 \times 2L/\pi \) where \( L = NL_c \). To generate the top-hat phase-matching function shown in Figure 3 b), we increased the number of domains to \( N = 5000 \). Due to the sharp edges, more domains were needed to produce a reasonable approximation. We then ran the algorithm with \( 10^5 \) iterations, using \( A = 1000 \). The height of the target function was \( 0.1 \times 2L/\pi \). All other parameters were the same as for the triangle example above.

### V. REMARKS AND FUTURE DIRECTIONS

We proposed a technique for shaping the phase-matching profile of a pair of downconverted photons. Our method can be used to approximate profiles of interest, such as Gaussian functions, triangles and top-hats. In particular, we demonstrated how a Gaussian phase-matching function generated using our technique can drastically increase the purity of heralded single photons. Compared to the alternative of spectral filtering, one can pump such a crystal at a much higher power without introducing loss of purity due to higher photon numbers. This allows the creation of purer multi-photon states for quantum information processing, e.g. Fock states with high photon number \([9]\).

Recent work by Quesada and Sipe \([27]\) shows that the joint spectrum for higher Fock states can differ from that of single-photon pairs. It would therefore be interesting to apply our technique in this regime. It might also be possible to apply our technique in the optimization of the entire joint spectral amplitude, rather than just the phase-matching function, as was shown by Phillips et al. \([28]\).

We also expect our technique to have applications in classical nonlinear optics, such as second harmonic generation, where similar spectral shaping techniques have been demonstrated \([19, 20]\).

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Appendix A: C++ code

A C++ implementation of the algorithm described in this paper is also available. The zipped package can be downloaded from [www.agatabranczyk.com/Dosseva2014code.zip](http://www.agatabranczyk.com/Dosseva2014code.zip). It contains a ‘readme.rtf’ file which details how to compile and run the code.

[1] A. Christ, A. Fedrizzi, H. Hubel, T. Jennewein, and C. Silberhorn, “Parametric down-conversion,” Experimental Methods in the Physical Sciences 45, 351–410 (2013).

[2] P. Kok, W. J. Munro, K. Nemoto, T. C. Ralph, J. P. Dowling, and G. J. Milburn, “Linear optical quantum computing with photonic qubits,” Rev. Mod. Phys. 79, 135 (2007).

[3] N. Gisin and R. Thew, “Quantum communication,” Nat Photon 1, 165–171 (2007).

[4] B. L. Higgins, D. W. Berry, S. D. Bartlett, H. M. Wiseman, and G. J. Pryde, “Entanglement-free Heisenberg-limited phase estimation,” Nature 450, 393–396 (2007).

[5] T. Nagata, R. Okamoto, J. L. O’Brien, K. Sasaki, and S. Takeuchi, “Beating the Standard Quantum Limit with Four-Entangled Photons,” Science 316, 726 (2007).

[6] G. Brida, M. Genovese, and R. Berchera, “Experimental realization of sub-shot-noise quantum imaging,” Nat Photon 4, 227–230 (2010).

[7] A. N. Boto, P. Kok, D. S. Abrams, S. L. Braunstein, C. P. Williams, and J. P. Dowling, “Quantum interferometric optical lithography: Exploiting entanglement to beat the diffraction limit,” Phys. Rev. Lett. 85, 2733–2736 (2000).

[8] M. Nasr, S. Carrasco, B. Saleh, A. Sergienko, M. Teich, J. Torres, L. Torner, D. Hum, and M. Fejer, “Ultra-broadband biphotons generated via chirped quasi-phase-matched optical parametric down-conversion,” Physical review letters 100, 183601 (2008).

[9] A. M. Brańczyk, T. C. Ralph, W. Helwig, and C. Silberhorn, “Optimized generation of heralded fock states using parametric down-conversion,” New Journal of Physics 12, 063001 (2010).

[10] A. Christ, C. Lupo, M. Reichelt, T. Meier, and C. Silberhorn, “Theory of filtered type-II parametric down-conversion: A method for single-mode EPR-entanglement generation,” ArXiv e-prints (2014).

[11] W. P. Grice, A. B. U’Ren, and I. A. Walmsley, “Eliminating frequency and space-time correlations in multiphoton states,” Phys. Rev. A 64, 063815 (2001).

[12] M. G. Raymer, J. Noh, K. Banaszek, and I. A. Walmsley, “Pure-state single-photon wave-packet generation by parametric down-conversion in a distributed microcavity,” Phys. Rev. A 72, 023825 (2005).

[13] A. B. U’Ren, C. Silberhorn, K. Banaszek, I. A. Walmsley, R. Erdmann, W. P. Grice, and M. G. Raymer, “Generation of pure-state single-photon wavepackets by conditional preparation based on spontaneous parametric downconversion,” Laser Physics 15, 146–161 (2005).

[14] K. Garay-Palmett, H. J. McGinness, O. Cohen, J. S. Lundeen, R. Rangel-Rojo, A. B. U’Ren, M. G. Raymer, C. J. McKinzie, S. Radic, and I. A. Walmsley, “Photon pair-state preparation with tailored spectral properties by spontaneous four-wave mixing in photonic-crystal fiber,” Opt. Express 15, 14870–14886 (2007).

[15] P. J. Mosley, J. S. Lundeen, B. J. Smith, P. Wasylczyk, A. B. U’Ren, C. Silberhorn, and I. A. Walmsley, “Heralded generation of ultrafast single photons in pure quantum states,” Phys. Rev. Lett. 100, 133601 (2008).

[16] B. J. Smith, P. Mahou, O. Cohen, J. S. Lundeen, and I. A. Walmsley, “Photon pair generation in birefringent optical fibers,” Opt. Express 17, 23589–23602 (2009).

[17] L. E. Vicent, A. B. U’Ren, R. Rangarajan, C. I. Osorio, J. P. Torres, L. Zhang, and I. A. Walmsley, “Design of bright, fiber-coupled and fully factorable photon pair sources,” New Journal of Physics 12, 093027 (2010).

[18] T. Gerrits, N. Thomas-Peter, J. C. Gates, A. E. Lita, B. J. Metcalf, B. Calkins, N. A. Tomlin, A. E. Fox, A. L. Liparoti, J. B. Spring, N. K. Langford, R. P. Mirin, P. G. R. Smith, I. A. Walmsley, and S. W. Nam, “On-chip, photon-number-resolving, telecommunication-band detectors for scalable photonic information processing,” Phys. Rev. A84, 060301 (2011).

[19] M. Fejer, G. Magel, D. H. Jundt, and R. Byer, “Quasi-phase-matched second harmonic generation: tuning and tolerances,” Quantum Electronics, IEEE Journal of 28, 2631–2654 (1992).

[20] G. Imeshev, M. Fejer, A. Galvanauskas, and D. Harter, “Pulse shaping by difference-frequency mixing with quasi-phase-matching gratings,” Journal of the Optical Society of America B 18, 534–539 (2001).

[21] A. M. Brańczyk, A. Fedrizzi, T. M. Stace, T. C. Ralph, and A. G. White, “Engineered optical nonlinearity for quantum light sources,” Opt. Express 19, 55–65 (2011).

[22] P. B. Dixon, J. H. Shapiro, and F. N. C. Wong, “Spectral engineering by gaussian phase-matching for quantum photonics,” Opt. Express 21, 5879–5890 (2013).

[23] P. J. Van Laarhoven and E. H. Aarts, Simulated annealing (Springer, 1987).

[24] C. H. Bennett, D. P. DiVincenzo, J. A. Smolin, and W. K. Wootters, “Mixed state entanglement and quantum error correction,” Phys. Rev. A 54, 3824 (1996). arXiv:quant-ph/9604024.

[25] K. Fradkin, A. Arie, A. Skliar, and G. Rosenman, “Tunable midinfrared source by difference frequency generation in bulk periodically poled ktp,” Applied physics letters 74, 914–916 (1999).

[26] F. König and F. N. Wong, “Extended phase matching of second-harmonic generation in periodically poled ktp with zero group-velocity mismatch,” Applied physics letters 84, 1644–1646 (2004).

[27] N. Quesada and J. E. Sipe, “Effects of time ordering in quantum nonlinear optics,” ArXiv e-prints (2014).
[28] C. Phillips, L. Gallmann, and M. Fejer, “Design of quasi-phasematching gratings via convex optimization,” Optics express 21, 10139–10159 (2013).