Engineering the frequency correlations of entangled two-photon states by achromatic phase matching

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We put forward a new method to control the spectra of photon pairs generated in parametric downconversion that allows their spectral properties to be tuned from correlation to anticorrelation, including uncorrelation. The method employs tilted pulses and can be implemented in materials and frequency bands for which conventional methods do not hold. © 2005 Optical Society of America

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Spontaneous parametric downconversion (SPDC) provides quantum frequency entanglement between the generated photons,[1] thus opening a wealth of opportunities in quantum information. Different potential applications need different specific degrees of frequency entanglement. For example, quantum-enhanced position and clock synchronization require frequency-correlated photon pairs.[2] The deleterious effects of chromatic dispersion in quantum crytography[3] and in quantum optical coherence tomography,[4] can be mitigated by use of frequency-anticorrelated photons. At another limit, the generation of frequency entanglement that involves more than two photons in simultaneous or cascaded SPDC requires removing the correlations between photons in each pair,[5] i.e., frequency uncorrelation.

In principle, any degree of frequency entanglement can be achieved by use of special phase-matching conditions. For example, the photons generated with a continuous-wave pump are frequency anticorrelated. However, many applications require ultrashort pump pulses, and under such conditions the frequency anticorrelation is lost.[6,7] On the other hand, frequency-correlated photons can be obtained in a special nonlinear crystal in which the group velocity of the pump beam happens to match the average group velocity of the downconverted photons.[8,9] Similarly, it is possible to obtain completely frequency-uncorrelated entangled photons by adjusting the properties of the downconversion process,[5] such as the crystal length or the pump beam width. However, a desired degree of frequency entanglement can be achieved only with special crystals and pump light conditions; thus elucidation of techniques to expand the available possibilities for obtaining frequency-uncorrelated photons is of paramount importance. A partial solution is the use of noncollinear geometries.[10–12] However, in this Letter we show that achromatic phase matching provides a powerful solution that may be applied to a wide variety of wavelengths and materials. The proposed technique passes the pump beam through a medium with angular dispersion such as a prism[13] or a diffraction grating[14,15] and henceforth employs Poynting vector walk-off to tailor the effective group velocity and group-velocity dispersion experienced by all involved photons.

Consider a second-order nonlinear optical crystal of length L, illuminated by a laser pump beam propagating in the z direction. The two-photon quantum state $|\Psi\rangle$ at the output of the nonlinear crystal, within first-order perturbation theory, is given by $|\Psi\rangle = |0, 0\rangle - (i/\hbar)\int_0^\tau \mathrm{d}t H_I(t)\,|0, 0\rangle$, where $|0, 0\rangle$ is the vacuum state, τ is the interaction time, and $H_I(t)$ is the effective Hamiltonian in the interaction picture, given by $H_I = \epsilon_0 \int_V \mathrm{d}V \chi^{(2)} \hat{E}_p^{+} \hat{E}_s^{-} \hat{E}_i^{-} + \mathrm{H.c.}$, where ϵ_0 is the permittivity of free space, $\chi^{(2)}$ is a second-order nonlinear susceptibility tensor, V is the volume of the crystal illuminated by the pump beam, $\hat{E}_p^{+}$ refers to the positive-frequency part of the pump electric-field operator, and $\hat{E}_{s,i}^{-}$ refer to the negative-frequency parts of the signal and idler electric-field operators, respectively.

To achieve achromatic phase matching, let the input pump beam with frequency and transverse momentum distribution $E_0(\omega_0 + \Omega, p_x, p_y)$ be diffracted by a grating located in front of the nonlinear crystal.[15,16] Here $\mathbf{p} = (p_x, p_y)$ is the transverse momentum, ω_0 is the central angular frequency of the pump beam, and Ω is the angular frequency deviation from the central frequency. In the grating, each spectral component is dispersed in a different direction. For a grating oriented in the x direction, the output signal of the grating is a tilted pulse that is written as $E_0[\omega_0 + \Omega, p_x/\alpha - \Omega \tan(\varphi)/(c\alpha), p_y]$, where φ is the tilt angle, c is the velocity of light, $\alpha = -\cos\theta_0/\cos\beta_0$, θ_0 is the input diffraction angle, and β_0 is the output diffraction angle. The resultant beam is a tilted pulse, so its peak intensity is located at a different time for each value of x. For normal incidence upon the nonlinear crystal, one has[14] $\tan\varphi = -md/(\lambda\cos\beta_0)$, where d is the groove spacing of the grating and m is the diffraction order. The pump beam that propagates inside the nonlinear crystal is given by $E_p^{+}(\mathbf{x}, z, t) = \int \mathrm{d}\omega_p \mathrm{d}\mathbf{p} E_0[\omega_p, p_x/\alpha - \Omega_p \tan(\varphi)/(c\alpha), p_y]\exp[ik_p z + i\mathbf{p}\cdot(\mathbf{x} + z\rho_p) - i\omega_p t]$, where $\omega_p = \omega_0 + \Omega_p$, $\mathbf{x} = (x, y)$ is the position in the transverse plane, $k_p(\omega_p, \mathbf{p}) = [(\omega_p n_p/c)^2 - |\mathbf{p}|^2]^{1/2}$ is

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the longitudinal wave number inside the crystal, n_p is the refractive index at the pump wavelength, and $\rho_p = (\rho_{0x}, \rho_{0y})$ is the Poynting vector walk-off.

Here we concentrate on phase matching for degenerate SPDC when the signal and idler photons propagate collinearly with the pump beam. Under such conditions the electric-field amplitude operator for the signal photon is given by $\hat{E}_s^{-}(\mathbf{x}, z, t) \propto \int \mathrm{d}\omega_s \mathrm{d}\mathbf{p} \exp[-ik_s z - i\mathbf{p}\cdot(\mathbf{x} + z\rho_s) + i\omega_s t]\hat{a}_s^{\dagger}(\omega_s, \mathbf{p})$, where $\omega_s = \omega_0/2 + \Omega_s$, $\mathbf{p} = (p_x, p_y)$ is the transverse momentum of the signal photon with momentum $\mathbf{p}$ and frequency ω_s, n_s is the refractive index inside the nonlinear crystal at the signal wavelength, and $\rho_s = (\rho_{1x}, \rho_{1y})$ are the walk-off parameters for the signal wave. The electric-field amplitude operator for the idler photon is written similarly, where $\mathbf{q} = (q_x, q_y)$ is the transverse momentum of the idler photon, $\rho_i = (\rho_{2x}, \rho_{2y})$ are the walk-off parameters, and $\omega_i = \omega_0/2 + \Omega_i$. We focus on type II (*eoe*) downconversion in a uniaxial crystal, such as β-barium borate (β-BaB_2O_4; BBO), for which the optical axis is contained in the XZ plane; then all walk-off parameters vanish, except for $\rho_{0x} \equiv \rho_0$ and $\rho_{1x} \equiv \rho_1$.

At the output face of the nonlinear crystal, a second grating is used to recollimate the beam by compensating for the angular dispersion introduced by the first grating.[15,16] We can describe the effect of the second grating by making the substitutions $p_x \rightarrow \alpha p_x + \Omega_s \tan\varphi/c$ and $q_x \rightarrow \alpha q_x + \Omega_i \tan\varphi/c$. The quantum state of the photon pair at the output face of the nonlinear crystal can be written as $|\Psi\rangle = \int \mathrm{d}\omega_s \times \mathrm{d}\omega_i \mathrm{d}\mathbf{p}\mathrm{d}\mathbf{q}\Phi(\omega_s, \omega_i, \mathbf{p}, \mathbf{q})\hat{a}_s^{\dagger}(\omega_i, \mathbf{p})\hat{a}_i^{\dagger}(\omega_i, \mathbf{q})|0, 0\rangle$, where the state function is

$$\Phi(\omega_s, \omega_i, \mathbf{p}, \mathbf{q}) = E_0(\omega_p, \mathbf{p} + \mathbf{q}) \times W(\Delta_k L/2)\mathcal{F}_s(\omega_s)\mathcal{F}_i(\omega_i), \quad (1)$$

with $\omega_p = \omega_s + \omega_i$. The joint spectral amplitude is multiplied by Gaussian spectral filter functions $\mathcal{F}_{s,i}$ to take into account the spectral filters collocated in front of the detectors. The function $W(\Delta_k L/2) = \mathrm{sinc}(\Delta_k L/2)\exp(i\Delta_k L/2)$ comes from the phase-matching condition along the longitudinal direction z, and

$$\Delta_k = k_p(\omega_p, \overline{p}_x + \overline{q}_x, p_y + q_y) + (\overline{p}_x + \overline{q}_x) \times \tan\rho_0 - k_s(\omega_s, \overline{p}_x, p_y) - \overline{p}_x \tan\rho_1 - k_i(\omega_i, \overline{q}_x, q_y), \quad (2)$$

where $\overline{p}_x = \alpha p_x + \Omega_s \tan\varphi/c$ and $\overline{q}_x = \alpha q_x + \Omega_i \tan\varphi/c$.

The central physical result of this Letter is contained in Eqs. (1) and (2). In general, Eq. (1) describes an entangled state in both transverse momentum ($\mathbf{p}$, $\mathbf{q}$) and frequency (Ω_s, Ω_i). However, in a specific detection scheme, e.g., projection into Gaussian modes with monomode optical fibers, Eq. (1) has to be integrated into the required spatial modes. Here we address the case when the downconverted photons are projected into spatial modes with large beam widths, which corresponds to monitoring the function $\Psi(\Omega_s, \Omega_i) = \Phi(\Omega_s, \Omega_i, \mathbf{p} = 0, \mathbf{q} = 0)$, a quantity that can be acquired experimentally with the signal and idler photons passing through an appropriate $2f$ optical system.

The joint spectral intensity $|\Psi(\Omega_s, \Omega_i)|^2$ is strongly affected by the slope in the (Ω_s, Ω_i) plane of the loci of perfect phase-matching frequencies,[7] and this slope is determined by the tilt angle of the angularly dispersed pump beam. To make this readily apparent let $k_s \approx k_s^0 + N_s\Omega_s$, $k_i \approx k_i^0 + N_i\Omega_i$, and $k_p \approx k_p^0 + N_p\Omega_p$.[6] Under this approximation, perfect phase matching is achieved for signal and idler frequencies given by

$$\frac{\Omega_i}{\Omega_s} = -\frac{N_p + \tan\varphi\,\tan\rho_0/c - N_s - \tan\varphi\,\tan\rho_1/c}{N_p + \tan\varphi\,\tan\rho_0/c - N_i}, \quad (3)$$

where $N_{s,i,p}$ are the inverse group velocities of the signal, idler, and pump, respectively. This expression can be understood as follows: The temporal evolution of the pump beam, for a tilted pulse with a large beam width, is governed by the effective inverse group velocity[17] $N_p' = N_p + \tan\varphi\,\tan\rho_0/c$, and the signal and idler photons acquire the same tilt as the pump photons. Therefore one can tailor the group velocities of all involved photons by acting on the angular dispersion of the pump photon, a feature that can be directly used to control their frequency correlations. Figures 1 and 2 illustrate the point.

In Fig. 1 we plot the phase-matching function $|W(\Omega_s, \Omega_i)|^2$ and the joint spectral intensity for highly correlated and anticorrelated photons. The case with no tilt is also shown for comparison. To draw this figure we considered a Gaussian pulse before the first grating, i.e., $E_0(\Omega_p) \propto \exp[-\Omega_p{}^2 T_0{}^2/(8\ln 2)]$, where T_0 is the pulse's temporal width (full width at half-maximum at intensity, i.e., FWHM), which pumps a degenerate type II SPDC in a BBO nonlinear crystal pumped at a frequency-doubled Ti:sapphire laser ($\lambda_p{}^0 \sim 400$ nm). We considered Gaussian spectral filters $\mathcal{F}_{s,i} \propto \exp[-(2\ln 2)\Omega_{s,i}{}^2/\sigma^2]$, where σ is the filter's FWHM. Notice that, according to the figure, for example, highly anticorrelated photons ($\Omega_s = -\Omega_i$) are obtained with tilt angle $\varphi \simeq 39.6°$, whereas highly correlated photons ($\Omega_s = \Omega_i$) are obtained with tilt angle $\varphi \simeq -53.2°$. It is worth stressing that the anticorrelated photons are obtained not with cw radiation but with a broadband pump pulse. The degree of anticorrelation can be increased by use of longer nonlinear crystals. Similar results, not shown here, were obtained with other crystals and wavelengths than those displayed in this Letter.

Next we consider the case when the signal and idler photons are to be completely uncorrelated. Their state function has to be separable, i.e., $\Psi(\Omega_s, \Omega_i) \propto f(\Omega_s)g(\Omega_i)$. One can perform a Schmidt decomposition, namely, $\Psi(\Omega_s, \Omega_i) = \sum_n \sqrt{\lambda_n} f_n(\Omega_s)g_n(\Omega_i)$, where λ_n, f_n, and g_n are solutions of the integral eigenvalue

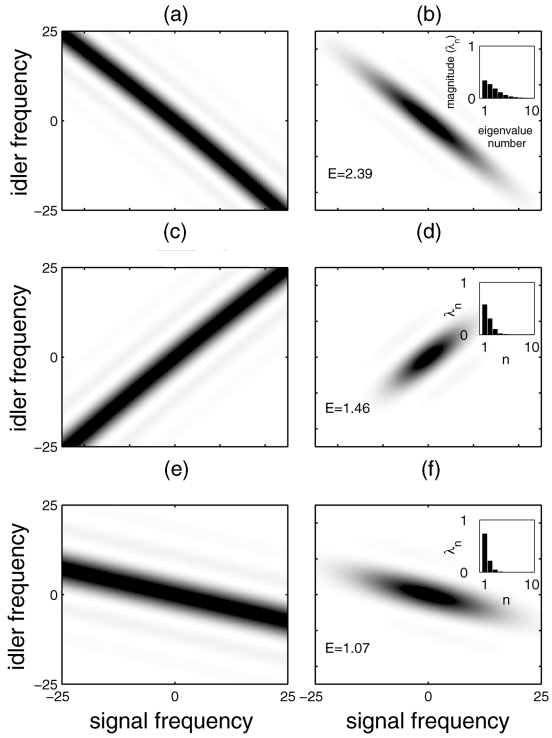

Fig. 1. Illustrative examples of the joint spectral intensity and the phase-matching function for degenerate type II downconversion under typical conditions. (a), (c), (e) Intensities of the phase-matching function; (b), (d), (f) joint spectral intensities. (a), (b) $\varphi = 39.6°$; (c), (d) $\varphi = -53.2°$; (e), (f) $\varphi = 0°$. Inset, numerical value of the entropy of entanglement as well as eigenvalues of the Schmidt decomposition of the corresponding state function. Angular frequency plotted in terahertz. Calculations were made for a BBO nonlinear crystal with length $L = 3$ mm. $T_0 = 100$ fs, ${\lambda_p}^0 = 400$ nm, ${\lambda_s}^0 = {\lambda_i}^0 = 800$ nm, $\Delta\lambda_0 = 10$ nm.

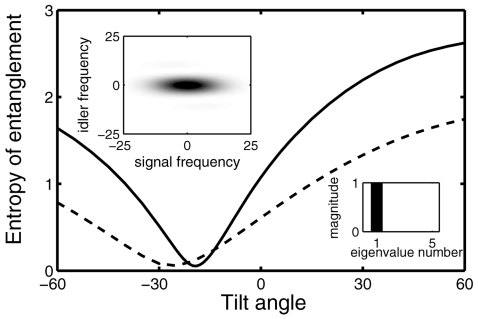

Fig. 2. Entropy of entanglement as a function of the tilt angle. Solid curve, $L = 3$ mm; dashed curve, $L = 1.5$ mm. Tilt angle is given in degrees. Inset, eigenvalues of the Schmidt decomposition and joint spectral intensity when the downconverted photons are nearly uncorrelated ($\varphi = -19.3°$). Calculations made with the same parameters as for Fig. 1.

equations for continuously variable systems[18]: $\int d\omega_2 K_1(\omega_1, \omega_2) f_n(\omega_2) = \lambda_n f_n(\omega_1)$, $\int d\omega_2 K_2 \times (\omega_1, \omega_2) g_n(\omega_2) = \lambda_n g_n(\omega_1)$, where $K_1(\omega_1, \omega_2) = \int d\omega_3 \Psi(\omega_1, \omega_3) \Psi^*(\omega_2, \omega_3)$ and $K_2(\omega_1, \omega_2) = \int d\omega_3 \times \Psi(\omega_3, \omega_1) \Psi^*(\omega_3, \omega_2)$. In Fig. 2 we show the behavior of the entropy of entanglement of the two-photon state, defined as $E = -\sum_n \lambda_n \log_2 \lambda_n$,[19] as a function of the tilt angle for two different crystal lengths. Notice that for $\varphi = -19.3°$ and $L = 3$ mm, the entropy of entanglement is $E \simeq 0$, which corresponds to a nearly uncorrelated two-photon state. The inset in Fig. 2 shows the joint spectral intensity and the Schmidt decomposition. The potential of the technique to engineer the spectral correlations of the entangled states is clearly apparent.

We thus stress the potential of the new technique to engineer the frequency correlations and frequency entanglement of two-photon states. The method expands the frequency bands and nonlinear materials at which quantum information protocols based on frequency entanglement can be implemented. Diffraction gratings with suitable groove spacing to produce the required tilted pulses are readily available.

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