A method to entangle photons in frequency is discussed. It uses short pump pulses and long crystals and allows the generation of maximally-entangled two-photon states that are correlated in frequency and anti-correlated in time of arrival. An experimental setup to test the entanglement of the created states is proposed.

Spontaneous parametric downconversion (SPDC) is the conventional source of entanglement used in most experiments. However, the state generated through SPDC using the customary phase-matching condition is maximally entangled only when a continuous-wave (cw) pump is used. In particular one sees that by decreasing the pulse length, the visibility of the interference experiments decreases. Advantages offered by pulsed SPDC have prompted interest in finding ways to retain or to restore the entanglement in this regime.

In this paper a method to achieve maximum entanglement in the pulsed regime is analyzed. It does not require any filtering or post-selection, and it utilizes long crystals resulting in high downconversion efficiencies. Although the customary phase-matching condition used in SPDC crystals is appropriate for cw pumping, the phase matching conditions we propose here are designed for pulsed pumps. The biphoton twin-beam state generated by conventional phase matching and cw pumping is given by

\[ |\text{TB}\rangle = \int \frac{d\omega}{2\pi} \phi(\omega)|\omega_p/2 - \omega\rangle_s|\omega_p/2 + \omega\rangle_i, \]

where \( |\omega\rangle \) is the state in which there is one photon at frequency \( \omega \) and no photons at other frequencies, \( \omega_p \) is the frequency of the cw pump, \( \phi(\omega) \) is the spectral function of the twin-beam state, and \( s \) and \( i \) refer to the signal and idler modes, respectively. On the other hand, the state that is generated from the method presented here will be shown to be

\[ |\text{DB}\rangle = \int \frac{d\omega}{2\pi} \phi(\omega)|\omega_p/2 + \omega\rangle_s|\omega_p/2 + \omega\rangle_i. \]

This state has been previously proposed by Erdmann et al. The state \( |\text{TB}\rangle \) is anti-correlated in frequency: the sum of the frequencies of the two downconverted photons is fixed and is equal to the cw pump frequency. In contrast, the state \( |\text{DB}\rangle \) is correlated in frequency: the difference in the frequencies
of the downconverted photons is a fixed quantity. Analogously, whereas the state $|TB\rangle$ has fixed photon time-of-arrival difference (as shown in the famous “Mandel dip” experiment\textsuperscript{7}), the state $|DB\rangle$ has a fixed photon time-of-arrival sum, as can be exhibited with a Mach-Zehnder interferometer. Moreover, whereas the photons of the state $|TB\rangle$ can be discriminated by a frequency measurement, the photons of $|DB\rangle$ can be discriminated by a time-of-arrival measurement. In this sense one may claim that the state $|DB\rangle$ proposed here is the dual of the state $|TB\rangle$ that is obtained under the customary phase-matching condition used in most entanglement experiments. Among other possible applications, the $|DB\rangle$ state can be employed in the localization protocols described in Ref. 8 to achieve enhancements and cryptographic advantages in position measurements and clock synchronization.

At the output of a compensated SPDC crystal of length $L$, pumped with a coherent classical pump with spectrum $|\alpha(\omega)|^2$ (peaked at the average pump frequency $\omega_p$), the biphoton state component is given by

$$|\Psi\rangle = \int \frac{d\omega_s}{2\pi} \frac{d\omega_i}{2\pi} \alpha(\omega_s + \omega_i) \Phi_L(\omega_s, \omega_i) |\omega_s\rangle_s |\omega_i\rangle_i ,$$  \hspace{1cm} (3)

where a normalization factor has been omitted and the phase-matching function is defined as

$$\Phi_L(\omega_s, \omega_i) = \frac{\sin (\Delta k(\omega_s, \omega_i)L/2)}{\Delta k(\omega_s, \omega_i)/2} ,$$  \hspace{1cm} (4)

with $\Delta k = k_p(\omega_s + \omega_i) - k_s(\omega_s) - k_i(\omega_i)$, for $k_p,s,i$ being the wave vectors of the pump, signal, and idler beams, respectively.

In order to generate the customary twin-beam state $|TB\rangle$ of Eq. (1), one must use a cw pump, which implies $\alpha(\omega_s + \omega_i) \propto \delta(\omega_p - \omega_s - \omega_i)$ in Eq. (3). The spectral function $\phi(\omega)$ of Eq. (1) can then be obtained from the phase-matching function as $\phi(\omega) \propto \Phi_L(\omega_p/2 + \omega, \omega_p/2 - \omega)$. This, however, is not the only way one can obtain a maximally frequency-entangled state starting from (3). One of the two frequency integrals may also be eliminated by forcing $\Phi_L$ to approach a Dirac delta function, instead of relying on $\alpha \rightarrow \delta$ as is done for the $|TB\rangle$ state. This can be accomplished by taking the limit $L \rightarrow \infty$, that transforms the spectral function $\Phi_L$ into a Dirac delta $\delta(\Delta(\omega_s, \omega_i)/2)$. Now we need to impose the additional condition that $\Delta k(\omega_s, \omega_i) = 0$ if and only if $\omega_s = \omega_i$. Using the first-order Taylor series approximations for the signal and idler wave vectors around the degeneracy point $\omega_p/2$, the desired $\Delta k$ condition is obtained if one chooses the crystal wave vectors to satisfy

$$n_p(\omega_p) = \frac{n_s(\omega_p/2) + n_i(\omega_p/2)}{2} ,$$  \hspace{1cm} (5)

$$k_p'(\omega_p) = \frac{k'_s(\omega_p/2) + k'_i(\omega_p/2)}{2} ,$$  \hspace{1cm} (6)

where $n_{p,s,i} = c k_{p,s,i}/\omega$ and the primes denote first derivatives with respect to $\omega$. Equation (5) is the customary phase-matching condition that is used in
the generation of the $|TB\rangle$ state. On the other hand, Eq. (6) is an “extended
phase-matching condition” that ensures that the signal and idler photons are
correlated in detuning from $\omega_p/2$ over the entire pump spectrum. Under these
conditions and taking the limit $L \to \infty$, the biphoton state $|\Psi\rangle$ of Eq. (3)
reduces to the $|DB\rangle$ state of Eq. (2) with $\phi(\omega) = \alpha(2\omega + \omega_p)$.

1 Experimental characterization

A simple way to characterize the two maximally frequency-entangled states
$|TB\rangle$ and $|DB\rangle$ is to feed these states into either a Hong-Ou-Mandel
interferometer\(^7\) or a Mach-Zehnder interferometer.\(^9\) It can be shown\(^6\) that
the output signatures of these interferometers are strongly dependent on the
type of frequency correlation between the two downconverted photons. An
example of these signatures is given in Fig. 1.

![Figure 1](image-url)

**Figure 1.** Left: Mandel dip in the coincidence rate $P(\tau)$ at the output of a Hong-Ou-Mandel
interferometer as a function of the delay $\tau$ between the signal and idler beams. The plotted
curve refers to a $|TB\rangle$ input state. The characteristic triangular shape derives from
the $\sin(x)/x$ form of the biphoton spectrum in the $|TB\rangle$ state. Had a $|DB\rangle$ state been fed
into such an interferometer, one would observe a constant $P(\tau)$. Right: Peak and fringes
in the coincidence rate $P(\tau)$ at the output of the Mach-Zehnder interferometer when the
$|DB\rangle$ state is at the input. The Gaussian envelope derives from the Gaussian spectral profile
of the pump beam, and the fringes modulate the envelope at the pump frequency. Had a
$|TB\rangle$ state been fed into the Mach-Zehnder interferometer, one would not see any Gaussian
envelope on the fringes. The parameters in the plots are $|k'_s(\omega_p) - k'_i(\omega_p/2)|L = 0.14$ ps
(for the left plot) and $\omega_p = 10^{15}$ s\(^{-1}\) with a pump bandwidth of 90 THz (for the right plot).

A crystal satisfying both conditions (5) and (6) is difficult to find. However, one can enforce (5) via quasi-phase-matching in a periodically-poled $\chi^{(2)}$ material.\(^10\) In particular, introducing a grating period $\Lambda$ in the crystal, the zeroth-order term in the $\Delta k(\omega_s, \omega_i)$ expansion (5) is replaced by

$$n_p(\omega_p) = |n_s(\omega_p/2) + n_i(\omega_p/2)|/2 - 2\pi c/\Lambda \omega_p.$$  

This condition, together with (6), are satisfied, for example, by periodically-poled potassium titanyl phos-
phate (PPKTP) at a pump wavelength of 790 nm with a grating period of 47.7 µm when propagation is along the crystal’s x axis, the pump and idler are y-polarized, and the signal is z-polarized. To what extent is the condition \( L \rightarrow \infty \) physically realizable? It can be shown,\(^6\) for example, that, for a 167 fs transform-limited pump pulse, a crystal length of the order 0.23 cm \( \ll L \ll 19.7 \) cm is sufficient to ensure a high degree of positive-correlated frequency entanglement.

In conclusion, we have shown a potentially realizable method to create photons that are frequency entangled with positive correlation and a procedure to test the generated entanglement to compare it with customary frequency entangled biphotons.

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References