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The role of two-photon entanglement in sum-frequency generation

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Abstract: The theory of sum-frequency generation (SFG) as a two-photon measurement process is used to infer the role of two-photon entanglement in this process, and an experimental setup and preliminary data are presented as a way towards quantifying the dependence of SFG on entanglement.

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A fundamental open question in quantum optics is to what extent does entanglement enhance two-photon processes such as two-photon absorption (TPA). This question remains difficult to investigate due in part to the inherently low cross section for two-photon processes at the flux levels typical for entangled photon-pair (EPP) generation. For example, even using the brightest sources of EPP based on parametric downconversion (PDC), it has been shown that TPA cross sections in most materials are too low to produce a usable signal [1]. Meanwhile, it has been shown that sum-frequency generation (SFG) using EPP, not only has a much more appreciable cross section at realistic flux levels, but also mimics TPA in its dependence on the input two-photon state [2].

In previous work [3], we have shown how to construct the positive operator-valued measure (POVM) associated with two-photon detection based on SFG followed by a mode-resolved single-photon detection of the SFG photon. Characterizing this POVM is useful for applications where two-photon detection plays a central role, such as in quantum teleportation [4] and entanglement swapping [5]. Here we extrapolate on those results, using quantum measurement theory, to infer what two-photon states are optimal for a given SFG process, what role spectral-temporal entanglement plays in this two-photon process.

Consider a two-photon state $|\Psi\rangle$ generated by PDC, which is described by

$$|\Psi\rangle = \int d\omega_s d\omega_i f(\omega_s,\omega_i)\,|\omega_s\rangle_s\,|\omega_i\rangle_i\,, \tag{1}$$

where $f(\omega_s,\omega_i)$ is the joint-spectral amplitude (JSA), which contains complete spectral-temporal information about the two-photon state, and $|\omega_{s(i)}\rangle_{s(i)}$ is a single-photon state at frequency $\omega_{s(i)}$ in the signal (idler) mode. This state contains spectral-temporal entanglement when the JSA is not separable into a product of functions of ω_s and ω_i. Consider this two-photon state undergoing an SFG process with a phase-matching function given by $\Phi(\omega_s,\omega_i)$, after which the upconverted single photon is projectively resolved onto a spectral mode described by $\alpha(\omega)$. Then it can be shown, from a modal-overlap argument, that the optimal two-photon JSA for such an SFG process is given by

$$f_{opt}(\omega_s,\omega_i) \propto \alpha(\omega_s+\omega_i)\Phi(\omega_s,\omega_i). \tag{2}$$

Furthermore, it follows that, if $\alpha(\omega_s+\omega_i)\Phi(\omega_s,\omega_i)$ is non-separable, then the optimal two-photon state to excite this process is necessarily non-classical, and is entangled.

To investigate the dependence of two-photon SFG on the JSA and the entanglement of the input state, we use two experimental configurations, one based on a continuous-wave (CW) laser source, and one based on a pulsed laser source. In both configurations, light from the laser source undergoes PDC in a non-linear crystal, creating a spectrally entangled photon pair (EPP). After dispersion compensation and manipulation in the spectral domain, the EPP undergo SFG at a second, identical non-linear crystal, and in each experiment, the SFG signal is analyzed as a function of various accessible parameters.

In the CW experiment [7], we employ a green (532 nm) laser source and pump a degenerate PDC process in a periodically-poled lithium niobate crystal (PPLN), generating EPP with a bandwidth of 60 nm centered at 1064 nm. After dispersion compensation, the EPP are incident on a second PPLN, where the resultant SFG signal is collected. Here we analyzed the dependence of the SFG signal on the attenuation of the green laser pump, which shows the characteristic linear dependence that is a signature of the quantum nature of the EPP [6], in comparison with the quadratic dependence on direct attenuation of the EPP. Furthermore, we compared the

SFG signal resulting from EPP against SFG resulting from a similar flux of classical CW 1064 nm laser light. This comparison shows that the EPP enjoy an efficiency up to two orders of magnitude greater than that of the classical light with respect to SFG, which is consistent with our predictions based on spectral entanglement. This preliminary data is shown in Fig. 1.

In order to further characterize the dependence of SFG on the JSA of the input two-photon state, we employ a pulsed pump derived from a titanium-sapphire laser frequency-doubled to generate blue pulses at 415 nm. This blue pump generates EPP at a periodically poled potassium titanyl phosphate (PPKTP) crystal, with a bandwidth of 120 nm centered at 830 nm, at a maximum flux of 20 nW. These EPP are routed through a spatial-light modulation based pulse shaper, where direct amplitude and phase manipulation of the JSA can be implemented and subsequently measured using time-of-flight spectrometry, before undergoing SFG at a second, identical PPKTP. This experiment is underway and is expected to allow not only quantifiable manipulation of the amount of entanglement of the two-photon state, but also direct comparison with SFG resulting from classical pulsed light at a similar flux.

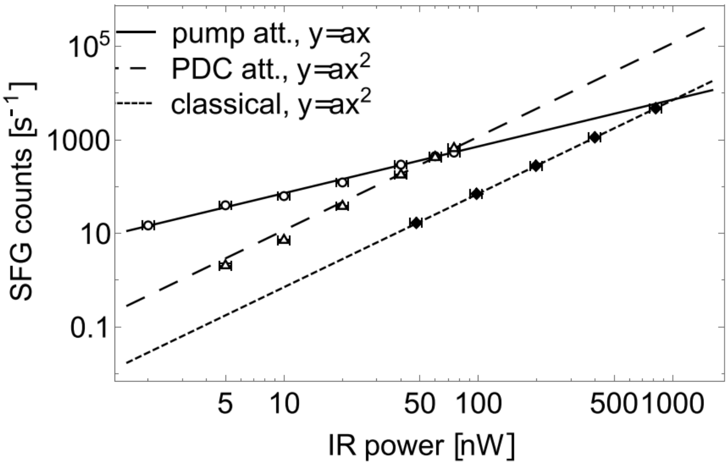

Fig. 1. Preliminary data showing the dependence of SFG excited by EPP on attenuation of the pump and on attenuation of the PDC, as well as the dependence of SFG induced with classical light on attenuation of the input light. It can be seen that SFG is enhanced by up to two orders of magnitude when induced by EPP over classical light in the low flux regime.

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