Single-Photon-Level Nonlinear Optics Through Quantum Interference

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Abstract: We demonstrate suppression of spontaneous parametric down-conversion via quantum interference with two weak coherent states. Pairs of photons upconvert with high efficiency exhibiting a nonlinearity enhanced by 10 orders of magnitude. This constitutes a two-photon switch.

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Interactions between optical fields are thought to be necessary for constructing critical two-photon quantum logic gates such as the controlled-not. Typical material nonlinearities are negligible except at high intensities, which makes them unsuitable for quantum computation. However, with respect to their ease and accuracy of manipulation and low decoherence rate, photons are ideal for quantum computation. Nonlinear effects that are significant at intensities corresponding to one photon would open the door to a field of quantum nonlinear optics and the possibility of all-optical quantum logic gates. In this experiment, we demonstrate an effective two-photon nonlinearity mediated by a strong classical field. In a typical setup for second-harmonic generation, a peak intensity on the order of 1 GW/cm^2 is required to provide an upconversion efficiency on the order of 10%. Instead, here beams with peak intensities on the order of 1mW/cm^2 undergo second harmonic generation with an efficiency of about 1% - roughly 11 orders of magnitude higher than would be expected without any enhancement. What is more, *conditional* measurements show effects with near-unit efficiencies, indicating strong dependence on the presence or absence of a single photon.

Our experiment relies on the process of spontaneous parametric down-conversion. If a strong pump laser beam with a frequency 2ω passes through a nonlinear crystal with a nonzero second-order susceptibility, χ^2 , then pairs of photons with nearly degenerate frequencies ω can be created, each in a distinct mode. Experiments have shown that the phase information of the pump is not lost during down-conversion, and that even though the phase of one of the down-converted beams is random, the phase of both together must sum to the phase of the pump[1-3]. This allows a well-defined phase difference to exist between a *pair* of laser beams and a *pair* of down-converted beams. We use this fact to set up quantum interference that enhances the effective nonlinearity of the crystal. A simplified schematic of our experiment is shown in fig. 1. Modes 1 and 2 lead to a nonlinear crystal and initially contain weak coherent states. Mode p contains a strong classical pump at half the wavelength of the coherent states. To lowest order in light intensity, there are two Feynman paths that can lead to a pair of photons, one in each of the two output modes of the crystal, 3 and 4. Each of the coherent states can contribute a photon to make up the pair (dashed lines), or the pair can come from down-conversion (solid lines). The challenge is to make these paths indistinguishable so that they interfere.

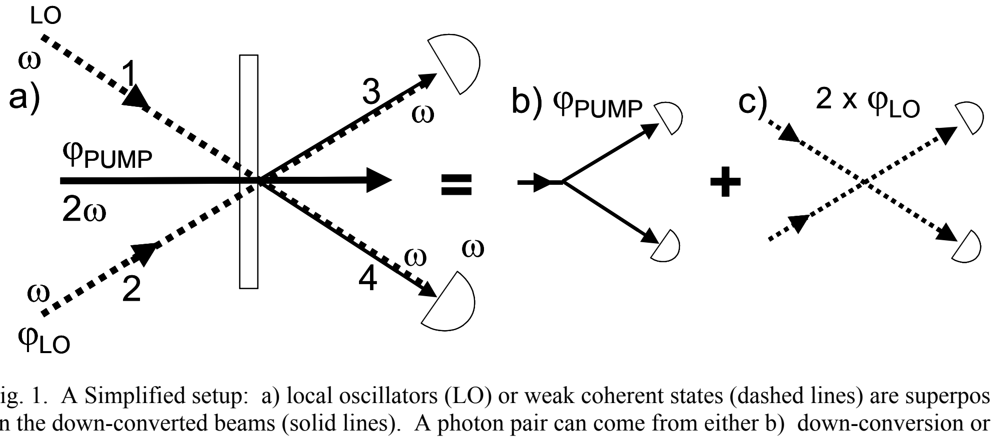

Fig. 1. A Simplified setup: a) local oscillators (LO) or weak coherent states (dashed lines) are superposed on the down-converted beams (solid lines). A photon pair can come from either b) down-conversion or c) the weak coherent beams.

To eliminate distinguishability we match the spectral, spatial and temporal distributions of the down-converted and weak coherent beams by using narrow interference filters, a pinhole spatial filter, and a femtosecond laser (centered at 810 nm). The latter is used as a source for the weak coherent beams and is also frequency-doubled to pump the down-conversion process, thus creating a phase relationship between the two Feynman paths. By using type-II phase-matching we can use modes which are collinear with the pump but have orthogonal polarizations. When we change the phase of the pump relative to the coherent states with an optical delay, we observe modulations in the photon pair production rate in the output modes. The rate of coincident detection events between single-photon detectors placed in modes 3 and 4 is shown in fig. 2. At a peak in the interference pattern the total pair production rate is enhanced, being greater than the sum of the rates from the independent paths. At a valley in the fringe pattern, the rate of photon pair production is similarly suppressed by destructive interference. Here, photon pairs are removed from the weak coherent beams at the crystal. If, however, one of the coherent beams is blocked, then those photons that would have been removed are now transmitted through the crystal. This constitutes an optical switch in which one weak coherent state controls the transmission of the other coherent state - even when there is on average less than one photon in either input pulse. An inherent noise source in the switch arises from the amplitude to get a photon pair even when both input states are blocked. Another limitation is that since the switch relies on phase, single-photon states cannot be switched. Instead, the amplitude to have a single photon is switched. While these features might limit the usefulness of this effect for a controlled-not gate, conditional-phase operations have also been demonstrated. This effect is closely related to the "railcross" experiment [4], in which suppression

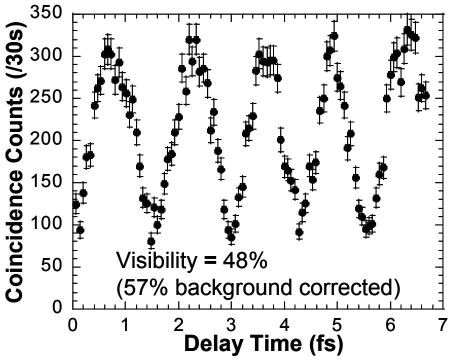

Fig. 2. The coincidence rate as a function of the delay time.

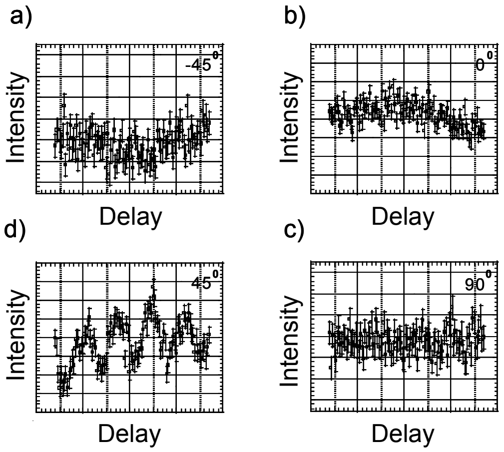

Fig. 3. The intensity at one detector (singles rate) versus delay time. a) Both weak coherent states or local oscillators (LO) blocked. b) Vertical LO blocked. c) Horizontal LO blocked. d) Both LO's unblocked.

and enhancement of down-conversion was observed. In this work, we use independent beams instead of a closed interferometer, so that it is possible to independently control the input beams.

In a typical two-photon interference experiment such as the Hong-Ou-Mandel interferometer [5], photon pair or coincidence oscillations occur but the intensity of the beams remains constant. Instead, photons bunch together in both the output beams. In contrast, in this experiment, photon pairs are actually removed from the input beams resulting in a change in intensity, which is shown in fig. 3. The nonlinear character of the effect is evident from the fact that it requires both weak coherent states to be present. Energy conservation requires that the photon pairs removed from the weak coherent beams are upconverted into the pump beam. This is similar to the time-reverse of the down-conversion process, in which 100% second harmonic generation is possible, even for weak beams. However, it difficult to measure the upconverted photons directly as they are dwarfed by the strong pump. Instead, we look for the removal of photon pairs from the weak coherent states, as shown in fig. 4. To magnify the absolute size of this removal, in light of our imperfect interference visibility, we increased the amplitude for a photon pair from the weak coherent states. We observe that 15.7% of the photon-pairs in the weak coherent beams are removed.

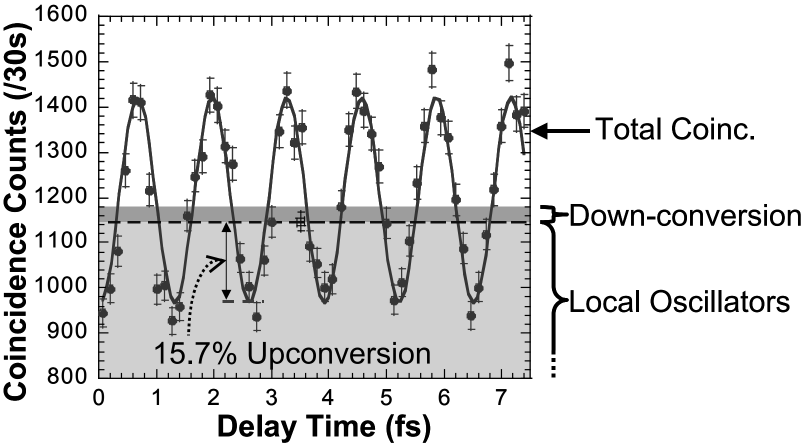

Fig. 4. The coincidence rate for unbalanced Feynman paths. For certain delay settings the number of photon pairs drops below the dashed line, which represents the number of photon pairs from the weak coherent states alone. This demonstrates that 15.7% ± 1.7% of the input photon pairs upconvert.

It is important to clarify which aspects of this experiment can be explained by classical nonlinear optics in place of the quantum description used in this summary. A formal theoretical quantum mechanical description of the action of the crystal on the optical fields predicts both the photon-pair and intensity modulations. It indicates that in the limit where the amplitudes for getting a photon-pair from down-conversion and the weak coherent beams are equal, 100% visible photon-pair oscillations are possible. Classically, these oscillations should be factorizable into the intensity oscillations in the two output beams. However, the predicted intensity modulations have low visibility and hence, cannot account for the photon-pair oscillations. Whereas the intensity modulations can always be described by either a quantum or classical description, the suppression and enhancement of photon-pair production reported by this experiment can only be described by quantum physics. Specifically, while the coherent states are transmitted nearly unmodified (to within 1%), all photon *pairs* in the coherent beams are removed through this quantum interference effect.

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