Nonlinear Optics with Less Than One Photon

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We demonstrate suppression and enhancement of spontaneous parametric down-conversion via quantum interference with two weak fields from a local oscillator (LO). Effectively, pairs of LO photons up-convert with high efficiency for appropriate phase settings, exhibiting an effective nonlinearity enhanced by at least 10 orders of magnitude. This constitutes a two-photon switch and promises to be applicable to a wide variety of quantum nonlinear optical phenomena.

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Nonlinear effects in optics are typically limited to the high-intensity regime, due to the weak nonlinear response of even the best materials. An important exception occurs for resonantly enhanced nonlinearities, but these are restricted to narrow bandwidths. Nonlinear effects which are significant in the low-photon-number regime would open the door to a field of *quantum* nonlinear optics. This could lead to optical switches effective at the two-photon level (i.e., all-optical quantum logic gates), quantum solitons (e.g., two-photon bound states [1]), and a host of other phenomena. In this experiment, we demonstrate an effective two-photon nonlinearity mediated by a strong classical field. Quantum logic operations have already been performed in certain systems including trapped ions [2], NMR [3], and cavity QED [4], but there may be advantages to performing such operations in an all-optical scheme—including scalability and relatively low decoherence. A few schemes have been proposed for producing the enormous nonlinear optical responses necessary to perform quantum logic at the single-photon level. Such schemes involve coherent atomic effects (slow light [5] and electromagnetically induced transparency [6]) or photon-exchange interactions [7]. We recently demonstrated that photodetection exhibits a strong two-photon nonlinearity [8], but this is not a coherent response, as it is connected to the amplification stage of measurement. While there has been considerable progress in these areas, coherent nonlinear optical effects have not yet been observed at the single-photon level for propagating beams. In a typical setup for the second-harmonic generation, for instance, a peak intensity on the order of 1 GW/cm^2 is required to provide an up-conversion efficiency on the order of 10%. In the experiment we describe here, beams with peak intensities on the order of 1 mW/cm^2 undergo a second-harmonic generation with an efficiency of about 1%—roughly 11 orders of magnitude higher than would be expected without any enhancement. While this 1% effect in the intensities of the outgoing modes can be described by a classical nonlinear optical theory, the underlying origin of the effect is observable in the correlations of the outgoing modes and requires a quantum mechanical explanation. Furthermore, the effect in the correlations was measured in this experiment to be about 70 times larger than in the intensities and, in theory, 100% of the photon pairs can be up-converted.

Our experiment relies on the process of spontaneous parametric down-conversion. If a strong laser beam with a frequency 2ω passes through a material with a nonzero second-order susceptibility, $\chi^{(2)}$, then pairs of photons with nearly degenerate frequencies, ω, can be created. In past experiments, interference phenomena have been observed between weak classical beams and down-converted photon pairs [9–11]. Although spontaneously down-converted beams have no well-defined phase (and therefore do not display first-order interference), the *sum* of the phases of the two beams is fixed by the phase of the pump. Koashi *et al.* [10] observed this phase relationship experimentally using a local oscillator (LO) harmonically related to the pump. More recently Kuzmich *et al.* [11] performed homodyne measurements to directly demonstrate the anti-correlation of the down-converted beams' phases. Some proposals for tests of nonlocality [12] have relied on the same sort of effect. Such experiments involve beating the down-converted light against a local oscillator at one or more beam splitters, and hence have multiple output ports. The interference causes the photon correlations to shift among the various output ports of the beam splitters.

In contrast, in this experiment the actual photon-pair production rate is modulated. A simplified cartoon schematic of our experiment is shown in Fig. 1. A nonlinear crystal is pumped by a strong classical field, creating pairs of down-converted photons in two distinct modes (solid lines). Local oscillator beams are superposed on top of the down-conversion modes through the nonlinear crystal and are shown as dashed lines. A single-photon counting module (SPCM) is placed in the path of each mode. To lowest order there are two Feynman paths that can lead to both detectors firing at the same time (a coincidence event). A coincidence count can occur either from a down-conversion event (Fig. 1b), or from a pair of LO photons (Fig. 1c). Interference occurs between these two possible paths provided they are indistinguishable. Depending on the phase difference between these two paths (φ), we observe enhancement or suppression of the coincidence

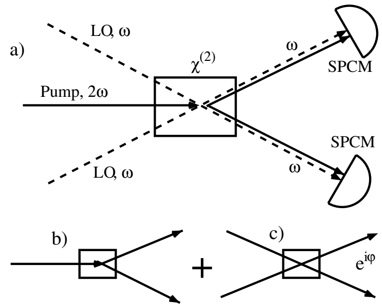

FIG. 1. A cartoon of our experiment. (a) Local oscillator (LO) beams (shown by dashed lines) are overlapped with the pair of down-converted beams. A coincidence count is registered either if (b) a down-conversion event occurs, or if (c) a pair of laser photons reaches the detectors (SPCMs).

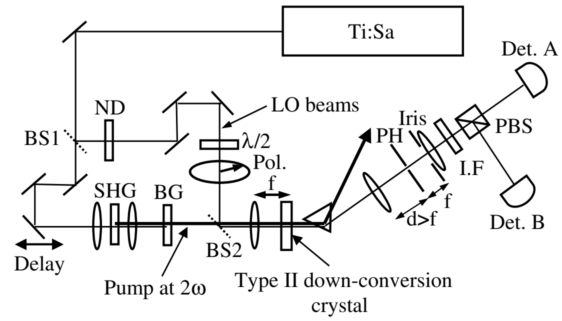

FIG. 2. Experimental setup: BS1 and BS2 are 90/10 (T/R) beam splitters; SHG consists of two lenses and a BBO nonlinear crystal for type-I second-harmonic generation; BG is a colored glass filter; ND is a set of neutral density filters; $\lambda/2$ is a zero-order half-wave plate; PH is a 25-μm diameter circular pinhole; I.F. is a 10-nm-bandwidth interference filter; PBS is a polarizing beam splitter; and Det. A and Det. B are single-photon counting modules. The thin solid line shows the beam path of the 810-nm light, and the heavy solid line the path of the 405-nm pump light.

rate. A phase-dependent rate of photon-pair production has been observed in a previous experiment using two pairs of down-converted beams from the same crystal [13]. By contrast, our experiment uses two independent LO fields which can be from classical or quantum sources and subject to external control. If the phase between the paths (Figs. 1b, 1c) is chosen such that coincidences are eliminated, then photon pairs are removed from the LO beams by up-conversion into the pump mode. If, however, one of the LO beams is blocked, then those photons that would have been up-converted are now transmitted through the crystal. This constitutes an optical switch in which the presence of one LO field controls the transmission of the other LO field—even when there is less than one photon in the crystal at a time. This switch does have certain limitations. First, it is inherently noisy because it relies on spontaneous down-conversion, which leads to coincidences even if one or both of the LO beams are blocked. Second, since the switch relies on interference, and hence phase, it does not occur between photon pairs but between the *amplitudes* to have a photon pair. While this may limit the usefulness of the effect as the basis of a "photon transistor," a simple extension should allow it to be used for conditional-phase operations.

In order for the down-conversion beams to interfere with the laser beams, they must be indistinguishable in all ways (including frequency, time, spatial mode, and polarization). Down-conversion is inherently broadband and exhibits strong temporal correlations; the LOs must therefore consist of broadband pulses as well. We use a mode locked Ti:sapphire laser operating with a central wavelength of 810 nm (Fig. 2). It produces 50-fs pulses at a rate of 80 MHz. This produces the LO beams, and its second harmonic serves as the pump for the down-conversion. Thus, the down-conversion is centered at the same frequency as the LO, and the LOs and the down-converted beams have similar bandwidths of around 30 nm. To further improve the frequency overlap, we frequency postselect the beams using a narrow bandpass (10-nm) interference filter [14]. As this is narrower than the bandwidth of the pump, it erases any frequency correlations between the down-conversion beams. In addition to spectral indistinguishability, the two light sources must possess spatial indistinguishability. The down-conversion beams contain strong spatial correlations between the correlated photon pairs; measurement of a photon in one beam yields some information about the photon in the other beam. Such information does not exist within a laser beam; since there is only a single transverse mode, the photons must effectively be in a product state and exhibit no correlations. We therefore select a single spatial mode of the down-converted light by employing a simple spatial filter. The beams are focused onto a 25-μm diameter circular pinhole. The light that passes through the pinhole and a 2-mm diameter iris placed 5 cm downstream is collimated using a 5-cm lens. In order to increase the flux of down-converted photons into this spatial mode, we used a pump focusing technique related to the one demonstrated by Monken *et al.* [15]. The pump laser was focused directly onto the down-conversion crystal. Since the coherence area of the down-converted beams is set by the phase-matching acceptance angle, the smaller pump area reduced the number of spatial modes being generated at the crystal, improving the efficiency of selection in a single mode. Imaging the small illuminated spot of our crystal onto the pinhole, we were able to improve the coincidence rate after the spatial filter by a factor of 30.

The final condition necessary to obtain interference is to have a well-defined phase relationship between the LO beams and the down-conversion beams. To achieve this, the same Ti:sapphire source laser is split into two different paths (Fig. 2). The majority of the laser power (90%) is transmitted through BS1 into path 1, where it is type-I frequency doubled to produce the strong (approximately 10-mW) classical pump beam with a central frequency of 405 nm. This beam is used to pump our down-conversion crystal after the 810-nm fundamental light is removed by colored glass filters. Instead of using down-conversion with spatially separate modes as shown in Fig. 1, we use type-II down-conversion from a 0.5-mm β-barium borate (BBO) nonlinear crystal. In this process, the photon pairs are emitted in the same direction but with distinct polarizations. The photon pairs are subsequently spatially filtered, spectrally filtered, and then split up by the polarizing beam splitter (PBS). The horizontally polarized photon is transmitted to detector A, and the vertically polarized photon is reflected to detector B. Detectors A and B are both single-photon counting modules (EG&G models SPCM-AQ-131 and SPCM-AQR-13). Path 1 also contains a trombone delay arm which can be displaced to change the relative phase between paths 1 and 2. To create the LO laser beams, we use the 10% reflection from BS1 into path 2. The vertically polarized laser light is attenuated to the single-photon level by a set of neutral-density (ND) filters, and its polarization is then rotated by 45° using a zero-order half-wave plate, so that it serves simultaneously as LO for the horizontal and vertical beams. After the wave plate, the light may pass through a polarizer, which can be used to block one or both of the polarizations from this path. This is equivalent to blocking one or both of the LO beams. Ten percent of the light from path 2 is superposed with the down-conversion pump from path 1 at BS2. The LO beams are thus subject to the same spatial and spectral filtering as the down-conversion and are separated by their polarizations at the PBS. This setup is similar to certain experiments investigating two-mode squeezed light [16]. Rather than investigate the noise characteristics of the output modes, we study the effect of a photon in one LO beam on the transmission of a photon in the other beam.

In order to maximize the interference visibility, we chose the ND filters so that the coincidence rate from the down-conversion path was equal to the coincidence rate from the laser path. The singles rates from the down-conversion path alone were 830 and 620 s^{-1} for detectors A and B, respectively, and the coincidence rate was $(11.0 \pm 0.3)\ s^{-1}$ (the ambient background rates of roughly 340 s^{-1} for detector A and 540 s^{-1} for detector B have been subtracted from the singles rates, but no background subtraction is performed for the coincidences). The singles rates from the LO paths were 34 560 and 31 350 s^{-1} for detectors A and B, respectively, and the coincidence rate from this path is $(11.6 \pm 0.4)\ s^{-1}$. The LO intensities need to be much higher than the down-conversion intensities to achieve the same rate of coincidences because the photons in the LO beams are uncorrelated. Nonetheless, the mean number of LO photons per pulse is on the order of 0.01 at the crystal and for this reason the process of stimulated emission is negligible. As the trombone arm was moved to change the optical delay, we observed a modulation in the coincidence rate (Fig. 3). We have explained that this interference effect leads to enhancement or suppression of photon-pair production; naturally, this should be accompanied by a modification of the total photon number, i.e., the *intensity* reaching the detectors. The visibility of the coincidence fringes is (56.0 ± 1.5)%, and the visibilities

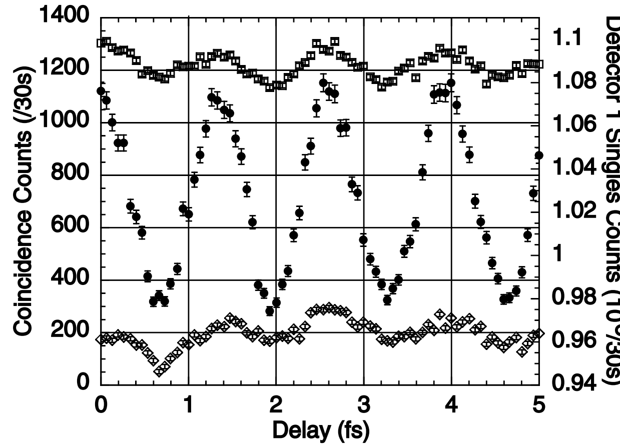

FIG. 3. The coincidence rate and singles rates as functions of the delay time. The coincidence counts (solid circles) demonstrate a phase-dependent enhancement or suppression of the photon pairs emitted from the crystal. The visibility of these fringes is (56.0 ± 1.5)%. The corresponding effects in the singles rates at detectors A (open squares) and detector B (open diamonds) are also shown; the visibilities are 0.83% and 0.78%.

in the singles rates are approximately 0.83% and 0.78% for detectors A and B, respectively. In theory, the visibility in coincidences asymptotically approaches 100% in the very weak beam limit for balanced coincidence rates. At the peak of this fringe pattern, the total rate of photon-pair production is greater than the sum of the rates from the independent paths. At the valley of the fringe pattern, the rate of the photon-pair production is similarly suppressed. With appropriate experimental parameters, we have observed coincidence rates drop 16% below the rate from the laser beams alone, an 8σ effect. The coincidence and singles fringes are all in phase and have a period corresponding to the 405-nm pump laser. To ensure that the observed oscillations in the coincidence rate were not due to a spurious classical interference effect, we verified that interference was destroyed by insertion of either a blue filter in the LO path or a red filter in the pump laser path, but unaffected by red filters in the LO path or blue filters in the pump path.

Figure 4 shows four sets of singles rate data for detector A, corresponding to four different polarizer settings. Recall that the light is incident upon the polarizer at 45°, so when the polarizer is set to 45°, both of the LO beams are free to pass. When the polarizer is set to 0° or 90°, one of the LO beams is blocked, and when the polarizer is set to −45° both of the LO beams are blocked. The left-hand side of Fig. 4 shows the data for the two orthogonal diagonal settings of the polarizer, −45° (top panel) and 45° (bottom panel); the right-hand side shows the data for the two orthogonal rectilinear settings, 0° (top panel) and 90° (bottom panel). When the polarizer is set to 0°, only the LO going to detector A is allowed to pass; on the other hand, when it is set to 90°, only the LO going to detector B is allowed to pass, so A measures only background plus

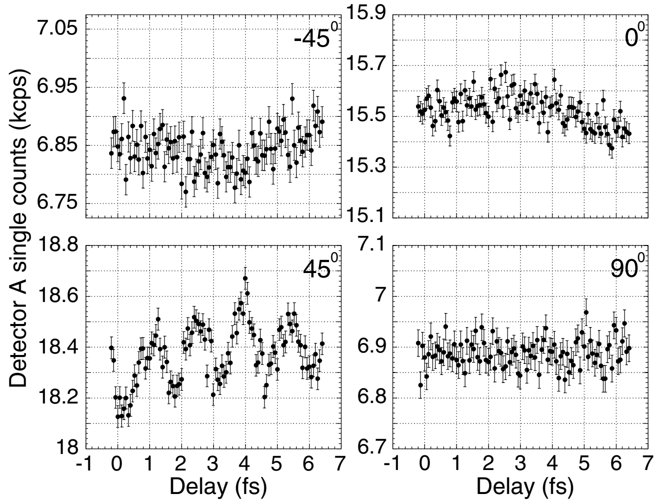

FIG. 4. The singles rate at detector A versus the delay for four different polarizer angle settings (labels in upper right corners). At $-45°$ no LOs can pass; at 45° both LOs can pass; at 0° the LO to detector A can pass; at 90° the LO to detector B can pass. The fringes are apparent only for the $+45°$ polarizer setting, and have a visibility of 0.7%. These four data sets show that both horizontally and vertically polarized photons must be present for the effect to occur.

down-conversion. For the 45° data, the singles rate at detector A shows fringes with a visibility of about 0.7%. This visibility is roughly 70 times smaller than the corresponding visibility in the coincidence rate because only about 1.4% of detected photons are members of a pair, due to the classical nature of our LO beams. The fringe spacing in the singles rate corresponds to that of the pump laser light at 405 nm even though it is the 810-nm intensity that is being monitored. By examining the other three polarizer settings ($-45°$, 0°, and 90°), it is apparent that in order to observe fringes in the singles rate, both LO paths must be open. This is evidence for a nonlinear effect of one polarization mode on another.

The intensity (singles rates) fringes can be explained by a classical nonlinear optical theory. Although the intensity of the difference-frequency light generated by one LO beam and the pump is negligibly small, its amplitude beats against the other LO to produce a measurable effect in analogy with optical homodyning. However, in a classical picture, the coincidence rate is just proportional to the product of the two singles rates [17]. Therefore, the maximum visibility in the coincidences in a classical theory is just the *sum* of the visibilities in the singles rates. In our case, that would correspond to a coincidence visibility of only 1.6%. Our 56% visibility can be explained only by a quantum mechanical picture in which the probability for one photon to reach a detector is strongly affected by the presence or absence of a photon in the other beam. A theoretical description of the intensity and coincidence effects has been performed [18].

We have demonstrated a quantum interference effect which is an effective nonlinearity at the single-photon level. We have shown that *pairs* of photons may be removed from two LO beams, although the system is transparent to *individual* photons. The phenomenon is closely analogous to second-harmonic generation in traditional nonlinear materials, but is enhanced by the simultaneous presence of a strong classical spectator beam with an appropriately chosen phase. For a different choice of phase, it should be possible to observe an effect analogous to cross-phase modulation between the two weak modes. Strong nonlinearities at the single-photon level should be widely applicable in quantum optics [19,20]. Overall, effects such as these hold great promise for extending the field of nonlinear optics into the quantum domain.

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