Direct Observation of the Second-Order Coherence of Parametrically Generated Light

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The second-harmonic spectrum of the output of an optical parametric emitter at degeneracy exhibits a narrow peak, much sharper than the spectrum of the parametric light. This peak is a manifestation of the second-order correlations in parametric light. Alternatively, it can be considered as resulting from the recombination of simultaneously generated (twin) wave packets. The twin–wave-packet correlation time is shown to be limited by the first-order coherence time of parametric light.

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Spontaneous parametric down-conversion arises from the splitting of an incident (pump) photon into two lower-frequency (signal and idler) photons which constitute a highly correlated photon pair.[1] Photon coincidence experiments[2,3] have shown that within instrumental resolution (100 ps in Ref. 3) the two photons in a pair are emitted simultaneously (twins), within a correlation time that should be limited only by their bandwidth.[1] At degeneracy, where the signal and idler photons are identical, the gain bandwidth of a free-running parametric generator is very large. In this case, the twin-correlation time is expected to be in the subpicosecond range, where experimental verification is out of the reach of photon-counting methods.

In this Letter, we give an alternative experimental answer to this problem by measuring the correlation time for degenerate parametric light through purely optical means. In addition, we show that the strong correlations between twin photons are preserved at high intensity, i.e., under conditions where the spontaneous parametric emission is enhanced by parametric amplification. This allows us to work with intensities high enough to perform nonlinear optical mixing experiments easily, and to observe directly the second-order coherence of parametric light.

At the heart of our method is the idea that the spectrum of light generated by nonlinear-optical mixing reveals the spectral and temporal correlations of the two beams being mixed. In the absence of correlations, this spectrum is given by the convolution of the spectra of the two incoming beams, since each frequency component of one beam can mix with all frequency components of the other. In the case of light produced by the parametric splitting of pump photons, however, photon wave packets are produced in pairs satisfying energy and wave-vector conservation. Optical mixing of such a wave packet *with its twin* is the exact inverse of parametric generation. It should thus give rise to a very sharp spectrum, identical to that of the pump, irrespective of the spectral widths of the two recombining wave packets. The sharp peak may be superimosed on a broad background due to the mixing of uncorrelated pairs. Clearly, the sharp peak should disappear and give rise to a broad spectrum if the two beams being mixed are delayed with respect to each other more than the correlation time of twin wave packets. By a performance of optical mixing in a noncollinear phase-matched time-resolved configuration,[4] and by detection of only the sharp spectral feature through a monochromator, it is therefore possible to measure the correlation time of twin wave packets.

Figure 1 shows a block diagram of our experimental apparatus. The pump source consists of a KH_2PO_4-doubled mode-locked pulsed (20-Hz repetition rate) yttrium aluminum garnet (YAlG) laser which delivers single pulses at 532 nm, with 50-MW peak power. The pulses are approximately Gaussian, have a duration of 35 ps (FWHM) and their spectrum is transform limited. The pump beam is injected into the parametric generator (PG) which consists of two 25-mm-long $LiIO_3$ crystals (type-I phase matching) in series, angle tuned to degen-

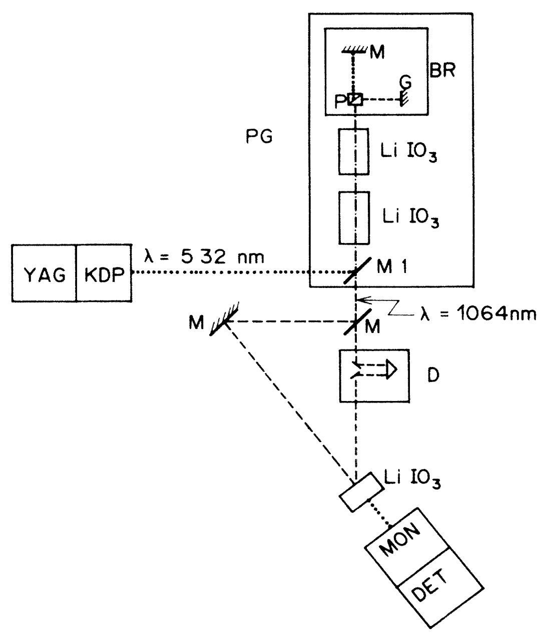

FIG. 1. Experimental setup (see text). PG, parametric generator; BR, backward reflector; P, Glan prism; G, grating; M, mirror; M1, dielectric mirror, R_{max} at 532 nm; D, variable optical delay; MON, monochromator; DET, detection.

eracy (1.06 μm). The PG is used in a double-pass mode by use of a backward-reflecting setup to be described later. The parametric light is thus spontaneously generated in the first few millimeters of $LiIO_3$ and gets amplified (gain of the order of 10^{10}) as it co-propagates collinearly with the pump through an effective length of 10 cm of crystal. At the exit we obtain 13-ps-long pulses of 1-MW peak power, having a divergence of 0.5 mrad. The parametric light is then studied in an optical mixing and spectral analysis setup. By means of a beam splitter each pulse is divided into two beams which are then recombined at an angle of 10°, focused in a 0.5-mm-long $LiIO_3$ crystal. The crystal is adjusted so that the superposition of the two beams is phase matched for frequency doubling along their bisectrix. The light produced by this frequency mixing is then analyzed by a 75-cm Jobin Yvon monochromator. In "spectral" experiments the entire spectrum is recorded and averaged over 32 or 100 pulses, by means of a B&M Spektronik GmbH optical multichannel analyzer. In "time-resolved" experiments, the exit slit is imaged on a photodiode. The intensity (averaged over 50 pulses) is plotted against the delay of the two beams which is stepped by a change of the pathlength of one beam after each 50-pulse-averaged measurement.

In the first series of experiments a mirror was used as the backward reflector of the PG, allowing the amplification of all wavelengths that satisfied phase matching near degeneracy. The spectrum of the parametric light (measured directly) fluctuated from shot to shot: Its width varied from 150 to 250 Å. Its average over 100 pulses was a Gaussian 200 Å wide (FWHM) centered at 10640 Å. The spectrum of the frequency-doubled parametric light contained a sharp peak at 5320 Å whose width was instrumentally limited (0.5 Å), as seen on Fig. 2(a). The sharp peak was superimposed on a broad background similar to the spectrum of the parametrically generated light. The intensity of the sharp peak as a function of the relative delay of the two beams (in the noncollinear setup) was approximately a Gaussian of width 0.20 ± 0.03 ps FWHM. The integrated intensity of the overall spectrum (measured by a removal of the monochromator) as a function of relative beam delay was a Gaussian of width 18 ± 1 ps FWHM.

These observations permit us to describe the output of the PG at degeneracy as pulses of an average duration of 13 ps (18-ps autocorrelation width) composed of twin wave packets with 0.20-ps twin-correlation time. This correlation time should equal the first-order coherence time of the parametric light.[1] The Gaussian width of 200 Å corresponds to a first-order coherence time of 0.08 ps, a factor of 2.5 shorter than the measured twin-correlation time. However, the average coherence time is given by the average of the inverse of the bandwidth, whereas in these experiments we have access only to the inverse of the average bandwidth. Given the spectral

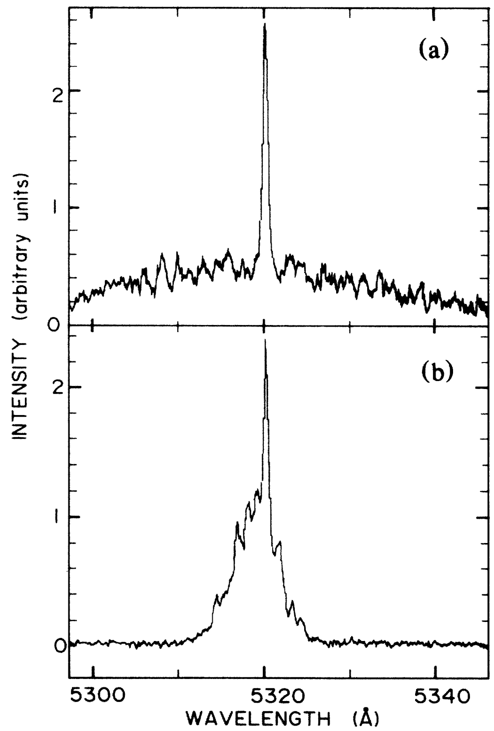

FIG. 2. Spectrum of frequency-doubled degenerate parametric light, averaged over 32 pulses: (a) Parametric generation with broad-band backward reflector, (b) parametric generation with frequency-dispersing backward reflector.

shot-to-shot fluctuations described above, this could account for the discrepancy of a factor of 2.5.

In order to confirm the equality of the twin-correlation and the first-order coherence times, a second series of experiments was undertaken in which the backward reflecting mirror of the PG was replaced by a frequency-dispersing setup, so that parametric light could be reinjected for amplification only within 10 Å around 10640 Å. This setup was composed of a Glan prism (to separate the pump from the parametric light), a mirror to backward reflect the pump and a 1200-g/mm grating in Littrow configuration tuned to back reflect the parametric light at 10640 Å. The average spectral width of the parametric light thus produced was about 22 Å, in correspondence to a coherence time of 0.75 ps. As before, the frequency-doubled spectrum contained a sharp peak at 5320 Å, superimposed on a fluctuating background whose average spectrum was only 8 Å wide [see Fig. 2(b)]. The monochromator slits were adjusted so that a significant fraction of the background could be transmitted along with the sharp line, so that we could measure the delay dependence of the intensities of the sharp peak and of the broad background simultaneously. The result is displayed in Fig. 3, which consists of a 18-ps (FWHM) Gaussian on top of which is a 1.3-ps-wide

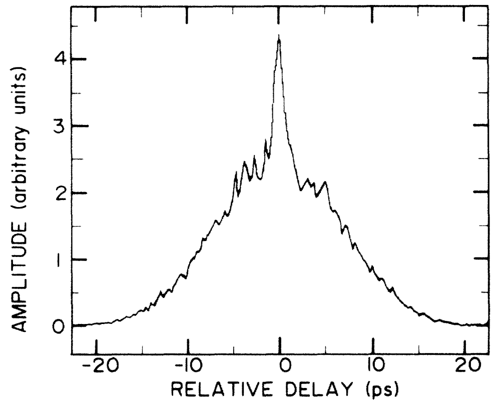

FIG. 3. Spectrally selected intensity autocorrelation of narrow-band degenerate parametric light. Narrow peak in center corresponds to twin-wave-packet correlation and follows the coherence profile of the pulses. Broad Gaussian corresponds to mixing uncorrelated wave packets and follows intensity autocorrelation profile of the pulses.

peak. The relative intensity of the two components depends on the amount of background rejection, and could be varied by an adjustment of the monochromator slits. The wide Gaussian follows the background intensity and corresponds roughly to the intensity autocorrelation of the parametric pulse, whereas the central peak follows the sharp line intensity and gives the twin-wave-packet correlation profile. The twin-correlation time (1.3 ps) thus followed the increase of 1 order of magnitude of the "average" coherence time (0.75 ps), while the discrepancy between them became smaller, consistent with the reduction of the shot-to-shot spectral fluctuations because of the grating. These measurements demonstrate that the twin correlation is limited by the first-order coherence time of the parametric light, as predicted in Ref. 1.

The appearance of a sharp peak in the intensity autocorrelation at $t=0$ is reminiscent of the Hanbury Brown–Twiss effect for chaotic light,[5] where the peak is due to the correlation of random intensity fluctuations. In parametric light, on the other hand, the sharp temporal peak can be observed only with appropriate spectral selection and is due to twin-wave-packet phase correlations. These correlations are most easily discussed away from degeneracy where the signal and idler modes are distinguishable. The signal and idler fluctuations are equal and opposite at all times to satisfy energy and momentum conservation. When the signal or idler is frequency doubled, the amplitude of the fluctuations doubles, and thus the corresponding spectra are broad. On the other hand, when the signal and idler are frequency mixed, their mutual fluctuations cancel each other because of their anticorrelation. The frequency sum thus does not fluctuate, and gives rise to a sharp spectrum.

At degeneracy, the same arguments can be applied. The frequency-doubled spectrum of degenerate parametric light contains both a sharp line and a broad background. The sharp line is due to the anticorrelated part of the phase fluctuations, and can be considered as the equivalent of signal-plus-idler mixing. It alone contributes to the sharp temporal feature in spectrally selected intensity autocorrelation measurements. The broad background arises from the mixing of wave packets undergoing identical or uncorrelated phase fluctuations, and can be considered as the equivalent of the signal- and the idler-frequency-doubling spectrum. The integrated intensity of the background should thus be twice that of the sharp line (signal-plus-signal and idler-plus-idler as compared with signal-plus-idler). Processes which reduce the phase anticorrelation of twin wave packets (such as momentum mismatch) would change the intensity ratio in favor of the background. In the experiments reported in this Letter, this intensity ratio could not be measured with sufficient accuracy.

Our experiments are designed with a classical viewpoint in mind, in that we deal with high-intensity pulses and measure only their mean intensity by conventional methods; we do not have direct access to their noise characteristics. Thus, the phenomena observed can be considered within a classical framework. However, degenerate parametric light is best described in terms of the quantum-mechanical two-photon coherent states,[6] which are superpositions only of even-numbered photon states of the corresponding mode of the electromagnetic field, and thus are the natural states for describing twin emission. These states have been the subject of a rich theoretical literature, especially regarding their properties of quantum uncertainty squeezing.[7] Very few experiments have been reported[8] as yet in which these purely quantum features of two-photon coherence have been studied. Spectrally resolved experiments on high-intensity parametric light, on the other hand, permit the investigation of some classical features of two-photon coherent states.

The description of our experiments can be formalized rigorously in terms of the three-time second-order correlation function of the electric field of the radiation. The spectrum of the frequency-doubled light is the Fourier transform of the two-time first-order correlation function of the second-harmonic field

$$S(\omega)=\int d\tau_1 e^{i\omega\tau_1}\int dt\, G^{(1)}(t,\tau_1), \qquad (1)$$

where

$$G^{(1)}(t,\tau_1)=\langle E_{2\omega_0}^{(-)}(t)E_{2\omega_0}^{(+)}(t+\tau_1)\rangle. \qquad (2)$$

Since the second-harmonic field is directly proportional to the product of the fields of the two beams being mixed, $G^{(1)}$ is proportional to

$$G^{(2)}(t,\tau_1,\tau_2)=\langle E_1^{(-)}(t)E_2^{(-)}(t+\tau_2)E_2^{(+)}(t+\tau_1+\tau_2)E_1^{(+)}(t+\tau_1)\rangle, \quad (3)$$

where the subscripts of the electric fields denote the two beams, while τ_2 is their relative delay.

Clearly, when no spectral selection is used in the detection, Eq. (1) has to be integrated over frequency giving a delta function in τ_1. With denotation of the intensity by $I=|E|^2$, Eq. (3) becomes

$$G^{(2)}(t,\tau_2)=\langle I_1(t)I_2(t,\tau_2)\rangle \quad (4)$$

giving the ordinary intensity autocorrelation, in which only the relative beam delay enters in the experiment. Spectral selection, on the other hand, gives access to a second-order correlation function (3) in which τ_1, the "spectral" time, is averaged over a wide range of values, while τ_2, the beam delay, is stepped experimentally. Our experiments are, to our knowledge, the first to measure an effect describable by a three-time second-order correlation function. The theoretical analysis of our experiments in terms of the three-time functions as well as experiments away from degeneracy will be published elsewhere.[9]

In conclusion, we note that the spectral and temporal analysis of frequency-doubled light gives access to several coherence properties of the fundamental. In particular, the spectrum of frequency-doubled degenerate parametric light contains a sharp peak centered at the pump wavelength. This peak can be considered as resulting from the recombination of twin wave packets. The temporal profile of the sharp peak gives the twin-wave-packet correlation-time profile. The twin-correlation time is thus found to coincide with the first-order coherence time of the parametric light. Alternatively, the sharp peak can be considered as due to phase correlations in parametric light manifesting its second-order coherence. The study of such correlations can be formalized in terms of the three-time second-order correlation function of the electric field.

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