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Spontaneous Parametric Processes in Modern Optics

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Spontaneous parametric processes have proved to be integral to
the advancement of quantum information science. Their incredible
versatility, in terms of the emission properties of entangled
photons, allows for nearly unimaginable applications in the
transmission and processing of quantum information.

spontaneous parametric processes can be used to generate entangled photon pairs. Quantum entanglement describes a physical system containing two or more subsystems that cannot be characterized solely by its individual components. Two entangled particles remain inextricably linked to each other, even if separated by an arbitrarily large distance. The possibility of such "spooky action at a distance" has provoked intense debate ever since Einstein, Podolsky and Rosen published a paper in 1935 showing that the predictions of quantum mechanics are at odds with both classical physics and with some of our most basic notions of everyday life. This debate has grown into an active and fascinating field of contemporary physics.

Since the late 1970s, a number of important experiments have eliminated doubts about the validity of quantum mechanics. Within the last two decades, research efforts have shifted towards harnessing quantum-mechanical effects such as entanglement for new technologies that would not be possible within the realm of classical physics.

In the 1980s researchers toyed with the idea of exploiting the properties of photonic quantum systems for the secure transmission of information. These systems would rely on the laws of physics to guarantee the security of such quantum key distribution protocols.

In the 1990s, two landmark quantum algorithms were discovered that could one day be run on a quantum computer. Peter Shor's algorithm showed that a quantum system can

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factorize a number N into its prime factors using exponentially fewer resources compared with a classical system. The second algorithm, published by Lov Grover, showed that a quantum system could outperform a classical counterpart in database searching. Much of the interest in quantum effects lies in their potential to revolutionize computation and communications in the near- and mid-term future.

Photons and quantum information

Photons have played a central role in advancing the field of quantum information. They are natural carriers of information, both classical and quantum, over distances ranging from very short to very long. Photons have been essential to developing technologies such as quantum key distribution and quantum computation. Among the available and proposed non-classical light sources, those relying on spontaneous parametric processes in nonlinear optical materials represent an established and flexible technology for generating photon pairs and heralded single photons.

Spontaneous parametric down conversion

The family of spontaneous parametric processes includes spontaneous parametric down conversion (SPDC), in which a pump laser beam illuminates a second-order nonlinear crystal; individual pump photons may then be annihilated, giving rise to the emission of photon pairs. SPDC may be categorized in terms of the polarization properties of the generated pairs, which also defines the resulting spatial intensity distribution. In type I SPDC, the signal and idler photons are co-polarized; this leads to a spatial light distribution in the form of a single cone. In type II SPDC, the signal and idler photons are cross-polarized, causing the emitted light to form two distinct cones. SPDC was first demonstrated more than 40 years ago, and it has led to a multitude of studies that have sharpened our understanding of the behavior of quantum systems.

Spontaneous four-wave mixing

Spontaneous four-wave mixing (SFWM) involves two photons, either from a single pump mode or from two distinct pump modes. The photons are annihilated, resulting in the emission of a photon pair, as mediated by a third-order optical nonlinearity—usually in an optical fiber.

SFWM was first demonstrated a decade ago and has become a viable fiber-based alternative to SPDC. This process has some important advantages over SPDC, including that it allows a much longer interaction length and hence a larger rate of emission. In principle, it also leads to the possibility of straightforward integration with existing fiber optical networks.

A limitation of SFWM is that the photon pairs it produces may be spectrally and spatially overlapped with single, uncorrelated photons generated by spontaneous Raman scattering.
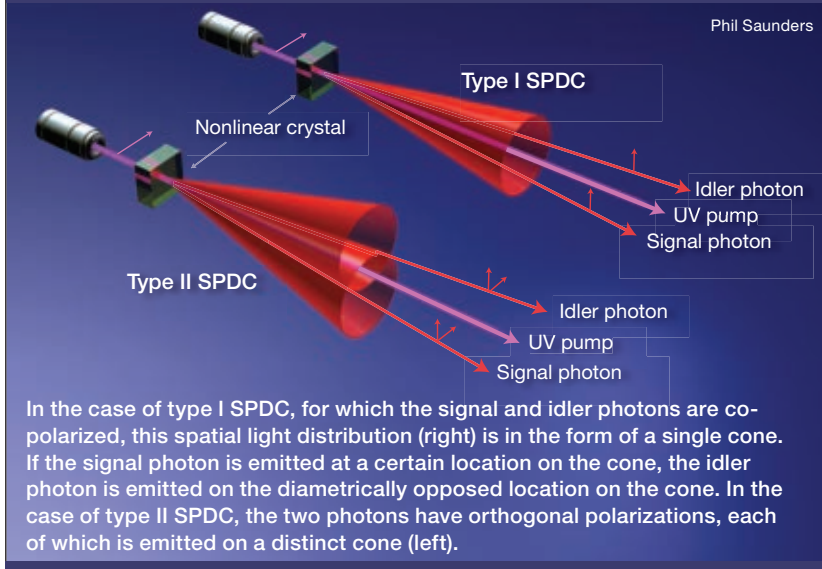
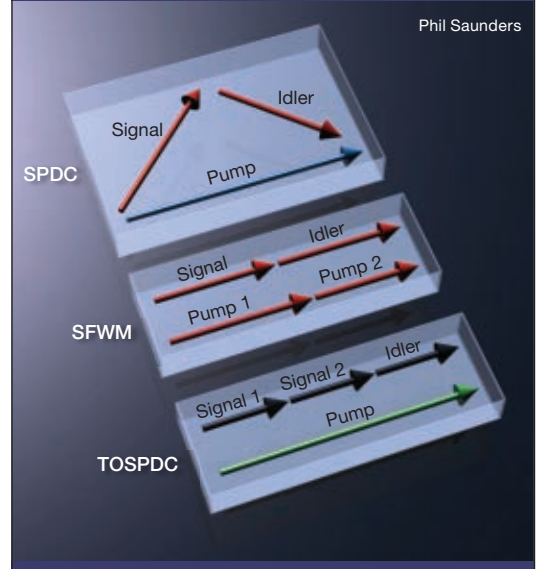[Spatial-spectral structure of photon pairs]
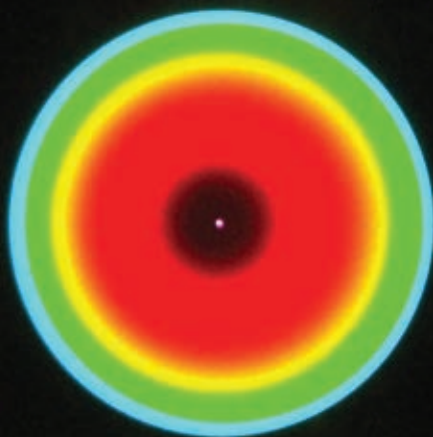
This photograph shows the structure emitted by the process of type I spontaneous parametric down conversion. A beam from a diode laser, centered around 405 nm, illuminates a beta-barium borate crystal. A color charge-coupled-device camera records the transverse-momentum intensity distribution.

U'Ren laboratory/Universidad Nacional Autónoma de México

Spatial light distribution of spontaneous parametric down conversion photon pairs

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Type I SPDC

Nonlinear crystal

Idler photon
UV pump
Signal photon

Type II SPDC

Idler photon
UV pump
Signal photon

In the case of type I SPDC, for which the signal and idler photons are co-polarized, this spatial light distribution (right) is in the form of a single cone. If the signal photon is emitted at a certain location on the cone, the idler photon is emitted on the diametrically opposed location on the cone. In the case of type II SPDC, the two photons have orthogonal polarizations, each of which is emitted on a distinct cone (left).

Phase matching diagrams for spontaneous parametric processes

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SPDC

Idler
Signal
Pump

SFWM

Idler
Signal
Pump 1
Pump 2

TOSPDC

Signal 1
Signal 2
Idler
Pump

This may lead to accidental coincidence events, thereby degrading the source quality. One solution to this problem is to rely on specific source designs for which the signal and idler photons are sufficiently spectrally removed from the pump so that the signal and idler modes remain uncontaminated. Other approaches—such as cryogenic cooling of the fiber—have been explored for the suppression of Raman scattering.

Third-order spontaneous parametric down conversion

The third member of the family of spontaneous parametric processes, which has been essentially unexplored from an experimental point of view, is third-order spontaneous parametric down conversion (TOSPDC). In the TOSPDC process, single pump photons are annihilated, giving rise to the emission of photon triplets. This process can be based on the same third-order nonlinearity in optical fibers that makes the SFWM process possible.
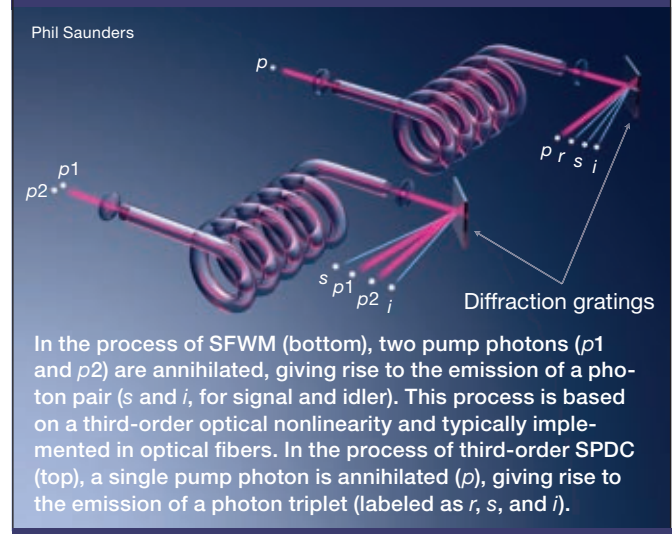
However, the very large spectral separation between the pump and the generated photons in TOSPDC makes phase matching more challenging than it is with SPDC or SFWM. We have recently proposed a specific technique that relies on thin optical fibers with a high dielectric contrast; in principle, it permits the practical implementation of TOSPDC. This would lead to the direct generation of photon triplets in which all three photons originate from a single quantum mechanical event. This is in contrast to existing schemes for generating entangled photon multiplets, based on multiple pair generation events or on pairs combined with highly attenuated coherent states.

A straightforward modification of a TOSPDC source could also be used to directly generate three-photon Greenberger-Horne-Zeilinger (GHZ) polarization-entangled states. Such a state involves two amplitudes: All three photons horizontally polarized and all three photons vertically polarized.

Generating GHZ states typically involves a two-step process of selecting three out of the four photons from two separate SPDC-generated photon pairs. Generating GHZ photon triplets can be reduced to a single step by using TOSPDC. This scheme requires a fiber Sagnac interferometer with the pump propagating through the fiber in both directions and with opposite polarizations.

GHZ states lead to correlations in the sense that measuring the polarization of two of the three photons determines the polarization of the third photon. Theories based on locality and realism (i.e., classic physics) cannot be used to predict the polarization of the third photon. Note that proofs of the validity of quantum mechanics based on photon pairs typically rely

Spontaneous four-wave mixing and parametric down conversion

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Diffraction gratings

In the process of SFWM (bottom), two pump photons (p1 and p2) are annihilated, giving rise to the emission of a photon pair (s and i, for signal and idler). This process is based on a third-order optical nonlinearity and typically implemented in optical fibers. In the process of third-order SPDC (top), a single pump photon is annihilated (p), giving rise to the emission of a photon triplet (labeled as r, s, and i).

on the violation of Bell's inequalities, which does not fully rule out hidden variable theories (designed to remain consistent with locality and realism) since certain loopholes remain. The contradictions between local realistic theories and quantum mechanics (made evident by three-photon GHZ states) lead to a stronger refutation of hidden variable theories than is possible with photon pairs.

The image at the bottom of p. 39 contains a representation of the fiber-based SFWM and TOSPDC processes. The image below shows an energy, or frequency, diagram indicating the participating modes for the SPDC, SFWM and TOSPDC processes.

Alternate processes for generating non-classical light

Spontaneous parametric processes are not the only mechanisms for generating non-classical light. Single photons may be created by the controlled, and ideally on-demand, de-excitation of a single quantum emitter. Researchers have demonstrated such single photon sources based on a variety of physical systems, including quantum dots, single molecules, single vacancy centers and single atoms in cavities.

The first photon-pair experiments, including work by Clauser et al. in the 1970s and Aspect et al. in the early 1980s, relied on atomic cascades. While atomic cascades led to the first convincing photon-based tests of the validity of quantum mechanics—i.e., the violation of Bell's inequalities—the count rates are low. More recently, photon pairs have been generated by the collective emission from an atomic ensemble, a promising approach that permits a programmable time delay between two emitted photons.

Strengths of the spontaneous approach

Spontaneous parametric processes remain widely used, and they exhibit some important advantages over other

While pure single photons are the most basic building blocks of light fields, their generation is challenging.

techniques for generating non-classical light. This family of processes is particularly versatile, permitting photon pair (and triplet) generation with a wide range of properties, depending on pump-mode choice and nonlinear media characteristics. Photon pairs may in principle be entangled in any photonic degree of freedom, or in any combination of degrees of freedom.

Researchers have demonstrated sources of photon pairs entangled in polarization, in transverse position/momentum (including the specific case of orbital angular momentum), and in frequency/time, as well as sources of hyper-entangled photon pairs with simultaneous entanglement in more than one degree of freedom.
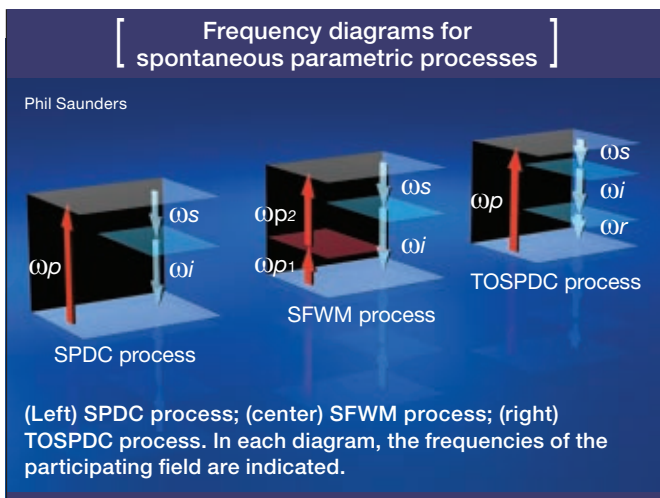
In the case of polarization, the source must be explicitly designed so that two amplitudes exist that cannot be resolved into factors, leading to quantum entanglement. A popular technique involves the spatial overlap of the emission cones of two separate but adjacent crystals, configured so that one of the crystals contributes an amplitude with both photons being horizontally polarized, and the other contributes an amplitude with both photons being vertically polarized.

For other photonic degrees of freedom, such as frequency/time and transverse position/momentum, phase matching and energy conservation constraints lead to the presence of entanglement for most source configurations, even if no explicit strategy is used to this end. The resulting photon pair sources may involve high-dimensional entanglement because the underlying Hilbert spaces for these degrees of freedom are of infinite dimensionality.

This is of interest because in principle the two parties sharing the two photons then have access to greater mutual information—which could be important for the design of a next-generation of quantum communication protocols. For example, this higher dimensionality could lead to quantum key distribution involving much larger alphabets.

The great flexibility of SPDC and SFWM processes, in terms of photon-pair properties, is apparent in the tremendous range of selectable emission bandwidth. At the low-bandwidth end of the scale, the nonlinear material can be placed inside an optical cavity so that emission occurs only at the resonant frequencies of the cavity. By making use of a high-finesse cavity, it becomes possible to obtain small emission bandwidths in the region of a few MHz. Photon pairs with such small bandwidths are crucial for atom-photon interfaces, so that single photons may be reliably absorbed by single atoms, leading to the possibility of efficient quantum memories.

Without the use of a cavity, the SPDC and SFWM photon-pair emission bandwidth tends to be considerably large—in the region of tens of THz. This happens because there are no well-determined intermediate energy levels for these processes, as shown in the image to the left. However, recently published

Frequency diagrams for spontaneous parametric processes

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ωp ωs ωi

SPDC process

ωp_2 ωs ωi ωp_1

SFWM process

ωp ωs ωi ωr

TOSPDC process

(Left) SPDC process; (center) SFWM process; (right) TOSPDC process. In each diagram, the frequencies of the participating field are indicated.

work shows an emission bandwidth that has been made considerably larger through careful source design.

For example, a chirped quasi-phase matched crystal has been used so that various portions of the crystal lead to different phase matched emission frequencies. In this case, the overall emission bandwidth can become very considerable. Alternatively, by selecting the frequencies so that they are centered at the zero dispersion frequency of the nonlinear medium, the natural phase matching bandwidth becomes extremely large.

The degree of photon-pair entanglement is determined by the ratio of the emission bandwidth to the pump bandwidth. Thus, photon pairs with a large emission bandwidth can be the basis for highly entangled photon pair sources, particularly if the pump bandwidth can be simultaneously made small. The SPDC and SFWM processes may then form the basis for photon pair sources ranging from factorable to highly entangled, and from nearly monochromatic to highly broadband.
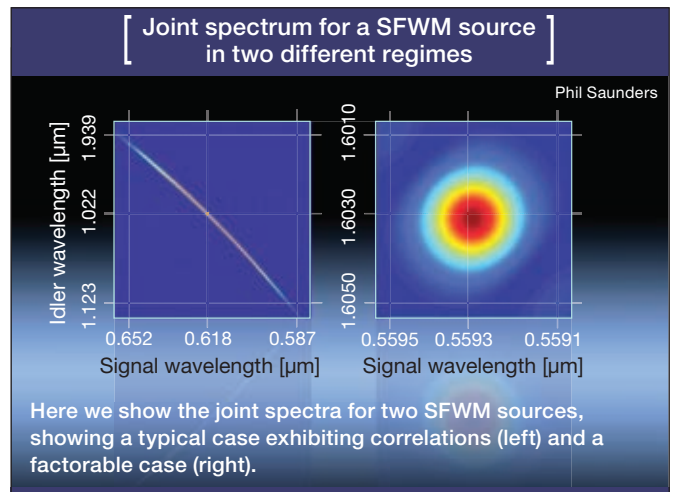
Generating pure single photons

Nevertheless, it is often the case that such spectral and spatial entanglement can become a limitation. Perhaps the clearest example of this is in relation to the design of heralded sources of pure single photons. While pure single photons are the most basic building blocks of light fields, their generation is challenging. Apart from being interesting from a fundamental physics point of view, they are crucial for implementing quantum information processing protocols. In the presence of photon-pair entanglement, each of the constituent single photons becomes a statistical mixture of the relevant spectral and spatial modes, so that the detection of one of the photons heralds (i.e., announces the presence of) a single photon described by a mixed state.

In order for the heralded photon to be in a pure state, the two-photon amplitude must be factorable: In other words, it must be the product of the single-photon amplitude functions of the heralding and the heralded photons. Thus, any protocol that relies on pure single photons must be based on factorable (unentangled) photon pairs. An approach that is often used to guarantee the purity of single photons is strong spatial and/or spectral filtering. However, this method results in a prohibitive reduction in the emitted flux, motivating a strong interest by a number of research groups (including our own) to devise reliable factorable photon-pair sources that do not rely on filtering.

It is interesting to note that a monochromatic pump in the SPDC process results in maximal spectral entanglement, and therefore also leads to highly impure heralded single photons. Therefore, a key requirement in strategies for generating factorable photon-pairs is a broadband pump, often in the form of a train of femtosecond pulses.

Research has shown that matching the group velocity of the pump photons to the group velocity of the generated photons in specific ways, referred to as group velocity matching, can lead to factorable photon pair sources. According to Mosley

[Joint spectrum for a SFWM source in two different regimes]

Phil Saunders

Here we show the joint spectra for two SFWM sources, showing a typical case exhibiting correlations (left) and a factorable case (right).

et al., an intuitive explanation for velocity matching is that traveling with equal group velocities forces one daughter photon to occupy the same narrow "time bin" as the pump pulse and therefore to be in a single broadband mode. Other relevant techniques that have been proposed involve using semiconductor Bragg wave guides, exploiting angular dispersion and using nonlinear photonic crystals.

Conclusion

We have explored the family of spontaneous parametric processes which occur in nonlinear optical materials, including SPDC, SFWM and TOSPDC. Of the three processes we have discussed, SPDC has been especially vital for the development of quantum information science, to a large degree because of the astonishing range of properties that can characterize SPDC photon pairs. We expect that these three processes will remain at the heart of future advances in experimental quantum information science. In time, these advances may translate into new computation and communication technologies that could transform our everyday lives. ◭

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