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PERSPECTIVE

A perspective on high photon flux nonclassical light and applications in nonlinear optics

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Abstract

Nonclassical light sources have a vital role in quantum optics as they offer a unique resource for studies in quantum technology. However, their applicability is restricted by their low intensity, while the development of new schemes producing intense nonclassical light is a challenging task. In this perspective article, we discuss potential schemes that could be used towards the development of high photon flux nonclassical light sources and their future prospects in nonlinear optics.

Keywords: quantum optics; nonlinear optics; high-power lasers

1. Introduction

The quantum description of a classically oscillating electromagnetic field[1,2] changed the course of the history on light technology and light–matter interaction, opening the way for the development of quantum optics which has led to countless applications in quantum technology[3,4]. The nonclassical light sources[5–11] have a vital role in this research domain, as they offer a unique resource for fundamental studies and applications in quantum technology. Despite the tremendous progress of this research domain, the majority of the achievements have been accomplished using relatively weak electromagnetic fields (low photon number light sources). Consequently, the applicability of the majority of the existing nonclassical sources is limited by their low intensity while the development of new schemes for the generation of high-intensity nonclassical light is considered as a challenging task. It is practically impossible to address in a single article the countless applications in basic research and technology that can be conducted using sources delivering high photon flux nonclassical light. For this reason, in this perspective article, after a brief introduction on the fundamentals of quantum optics and the generation of nonclassical light, we focus our discussion on the effect of the photon statistics of the light source in nonlinear optics emphasizing on multiphoton excitation processes.

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2. Scientific background: classical and nonclassical light

Quantum optics is founded on the quantization of the electromagnetic radiation and the quantum description of a classically oscillating current (coherent states). A key aspect of the studies in this research domain is the measurement and interpretation of light intensity fluctuations and the characterization of the quantum states of light. These are typically achieved by means of photon statistics measurements, phase sensitive homodyne detection schemes such as quantum tomography[7,8] and measurements of the Glauber correlation functions[12–16]. Nonclassical, or quantum, light states, are the light states where the electromagnetic field cannot be described by the classical wave mechanics. Such are, for example, the states of squeezed light, the photon number states, and the cat states. Some of the most useful criteria to distinguish the classical from the nonclassical light are based on the measurement of the (i) Glauber correlation functions $g^{(q)}(\tau)$, typically obtained by qth-order autocorrelation measurements, with q being the order of nonlinearity and τ the time delay between the signals, (ii) Wigner function (Q- or P-functions) in phase space[17], and (iii) photon number distribution, which can be obtained from the Wigner function or directly by photon statistics measurements. As it is nicely described in the book chapter of Strekalov and Lechs (see Ref. [18] and references therein), each of the criteria alone is sufficient but not necessary to distinguish classical from nonclassical light.

Regarding the statistical properties of light, there are several considerations to be taken into account. (i) A coherent state depicts a Poissonian photon number distribution with a normalized $g^{(2)} = 1$. This refers to the case where photons randomly reach a detector. (ii) A nonclassical light with super-Poissonian photon number distribution and $g^{(2)}(0) > 1$, is characterized as photon bunching. This refers to the case where the photons have the tendency to reach the detector in bunches, i.e., more close in space (time) than the photons of the coherent state. Chaotic, thermal, or stochastic light, although considered as classical, also corresponds to this case (with the photon number fluctuations to be determined by the coherence time of the light source). (iii) A nonclassical light with sub-Poissonian photon number distribution and $0 < g^{(2)}(0) < 1$, is characterized as photon antibunching. This is a purely quantum effect which refers to the case where the photons have the tendency to reach the detector more equally and further away in space (time) than those of a coherent state. However, in the present article, we consider as nonclassical the light states having a Wigner function that depicts negative values and/or non-Gaussian distributions, and a photon number distribution that deviates from the corresponding of a coherent state that is considered as the best quantum description of a classically oscillating field.

In particular, a coherent light state is a quantum state of the field that describes the classical behavior of the electromagnetic radiation typically produced by a conventional continuous wave (CW) or pulsed laser system. In this state, the quantum fluctuations of the quadrature components (which are equal to the fluctuations of the vacuum state and randomly distributed in the quadrature components) are equal and the uncertainty of their product is the minimum given by the Heisenberg relation. For a coherent light state, as is considered the light state of a laser field, the electric field variance ΔE remains constant within its cycle. An important measurable feature of a light source is its photon number distribution P_n, resulted by the projection of the light state to the photon number state $|\,n\rangle$. For coherent light states $|\,\alpha\rangle$ this distribution is Poissonian,

$$P_n = |\langle n|\alpha\rangle|^2 = \frac{<n>^n}{n!} e^{-<n>},$$

and for high mean photon numbers it can be approximated by a Gaussian,

$$P_n \approx \frac{1}{2\pi N_0} \exp\left[\frac{(n-N_0)^2}{2N_0}\right],$$

where N_0 is the mean photon number of the field. In addition, the Wigner function

$$W(p,q) \propto \exp\left[-\frac{(q_0-q)^2}{2\alpha^2} - \frac{(p_0-p)^2}{2(\alpha/\hbar)^2}\right]$$

depicts a Gaussian distribution in phase space (where q, p are the field quadratures and α the width of the distribution in q) and a second-order Glauber correlation function $g^{(2)}(0) = 1$.

A good example of nonclassical light states is the well-known and extensively studied squeezed light states. These are a special class of quantum states where the quantum noise is not randomly distributed between the field quadratures. It is reduced in one of the quadrature components and increased in the other. In this case, the variance of the field quadratures is modulated within the cycle of the field. The photon statistics of these nonclassical light sources significantly deviates from the Poissonian of the coherent states and the Wigner function depicts a distribution that significantly deviates from the Gaussian. Other well-known examples of nonclassical light are the photon number states (or Fock states) and the ‘cat’ states (see Ref. [18] and references therein). One of the main characteristics of these light states is that their Wigner function, despite its non-Gaussian form, depicts negative values.

3. Sources of nonclassical light

Nowadays, the nonclassical light states are usually produced by parametric down/up-conversion methods in solids, Kerr effects in optical fibers, semiconductor lasers, wave-mixing processes in atomic ensembles, etc.[18,19] and/or by implementing light engineering protocols having as recourses the squeezed, photon number states, and detection approaches[20–25]. Although, these sources typically deliver low photon number nonclassical light, recent developments have shown that high-gain parametric down conversion processes can be used for the generation of high photon number squeezed light states[26–28]. In addition, in the last few years the fully quantized description of the strong field laser–matter interaction, which takes into account the back action of the interaction on the coherent state of the driving field, has attracted a considerable interest from the theoretical[29–31] and experimental[32,33] point of view, with the very recent investigation of Ref. [34], to demonstrate in a rigorous way that strongly laser-driven materials can be used for the generation of unique nonclassical light states with

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controllable features. Taking into account that these sources are driven by intense laser pulses[35] capable of inducing interactions in the moderate and relativistic regime[36–38] makes them a very promising candidate for the generation of intense nonclassical light.

4. Photon statistics effects in nonlinear optics

Multiphoton processes are the essence of nonlinear optics with countless applications in basic research and technology. This cannot be better outlined than the way that is done using the sentence '*At this point, one may raise a question: are all media basically nonlinear? The answer is yes. Even in the case of vacuum, photons can interact through vacuum polarization. The nonlinearity is, however, so small that with currently available light sources...'* in the introduction of Shen's book *Principles of Nonlinear Optics*[39]. Harmonic generation[25,39,40], high harmonic generation in moderate[41–43] and relativistic intensity regimes[35–38], multiphoton processes in atoms[44], polymerization[25,39], vacuum polarization in super-relativistic intensities[35,45–47], visual science[25,48], etc. are some examples illustrating the importance of nonlinear optics in different research directions of basic research and technology. Eventually the observation of the nonlinear effects requires driving forces that can efficiently induce nonlinear processes up to the level of observation by the available detection systems. In case that the driving force is induced by an electromagnetic field, nonlinear processes can be observed by increasing the photon flux as well as the quantum fluctuations of the electromagnetic field.

For coherent light states, this is shown by the dependence of the transition rate W of a qth-order multiphoton process that is proportional to the qth power of the driving field intensity, i.e., $W \propto F^q$ (where F is the photon flux of the driving field). However, the complete relation which includes the photon fluctuations of a light source is $W \propto g^{(q)}F^q$, where $g^{(q)}$ is the qth-order Glauber functions with $\langle : n^q : \rangle \equiv \left\langle (a^\dagger)^q a^q \right\rangle$, n is the photon number incident reaching the detector, $\langle n \rangle$ is the average photon number (time integrated) reaching the detector, $F = \langle n \rangle$ is the mean photon number or photon flux, and $a^\dagger$, a the photon creation and annihilation operators, respectively. Evidently, the photon statistics of a light source, which appears in the $g^{(q)}$ functions, can dramatically influence the q-photon transition rates of a multiphoton process[25,48–61] simply because any nonlinear effect with ultrashort response time will experience the high/low and ultrafast/slow fluctuating photon numbers (photon bunching/antibunching). This remarkable effect, has tremendous advantages in nonlinear optics. For example, while for a coherent light state $g^{(q)} = 1$, the corresponding functions of a chaotic and vacuum squeezed state are $g^{(q)} = q!$ and $g^{(q)} = (2q-1)!!$, respectively (Figure 1).

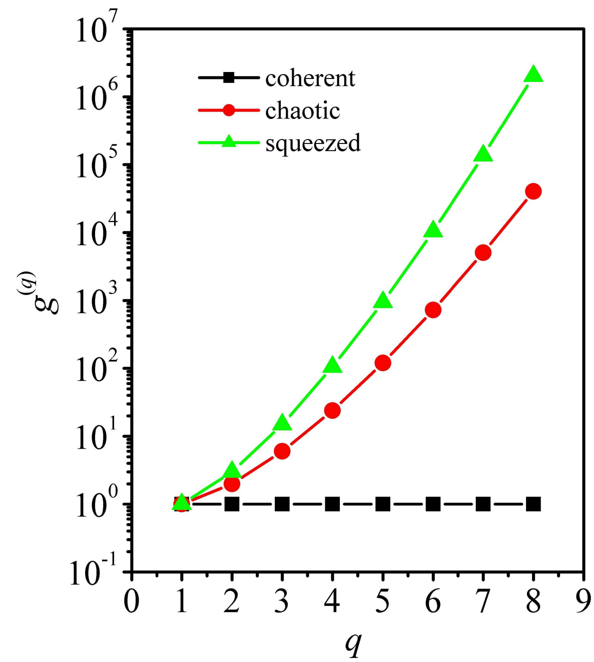

Figure 1. Dependence of g on the order of nonlinearity q, for coherent (black squares), chaotic (red circles), and squeezed (green triangles) light.

Hence, for the same F, chaotic and nonclassical light states with super-Poissonian photon number distribution can lead to a significant enhancement of the transition rates of highly nonlinear processes compared with those of the coherent light sources (Figure 2).

Evidently, such light sources can also be proved highly beneficial for observing nonlinear processes such as laser-induced pair production or vacuum polarization effects[35,46–48], which is one of the most challenging tasks in ultrarelativistic interactions. The enhancement of the transition rates was clearly shown using stochastic and vacuum squeezed states. This was achieved by measuring the ion yield produced by multiphoton ionization process of xenon (Figure 2(a))[57] and the harmonic yield produced by nonlinear process in a crystal (Figure 2(b))[28], respectively. It is noted that, owing to limitations of producing high-intensity thermal light from natural sources, the enhancement of the multiphoton transition rates shown in Figure 2(a), was achieved by mimicking a natural source using stochastic laser pulses generated by an multimode phase unlocked laser system.

The enhancement of the multiphoton transition rates was also studied theoretically and observed experimentally in the XUV spectral region, using FEL sources[62], whereas the differences compared with the laser-driven coherent XUV sources have been discussed in Ref. [63].

A direct consequence of this enhancement is the ability to study nonlinear processes in all states of matter using light intensities below the damage threshold of the materials. This makes the quantum light a unique resource for studies in

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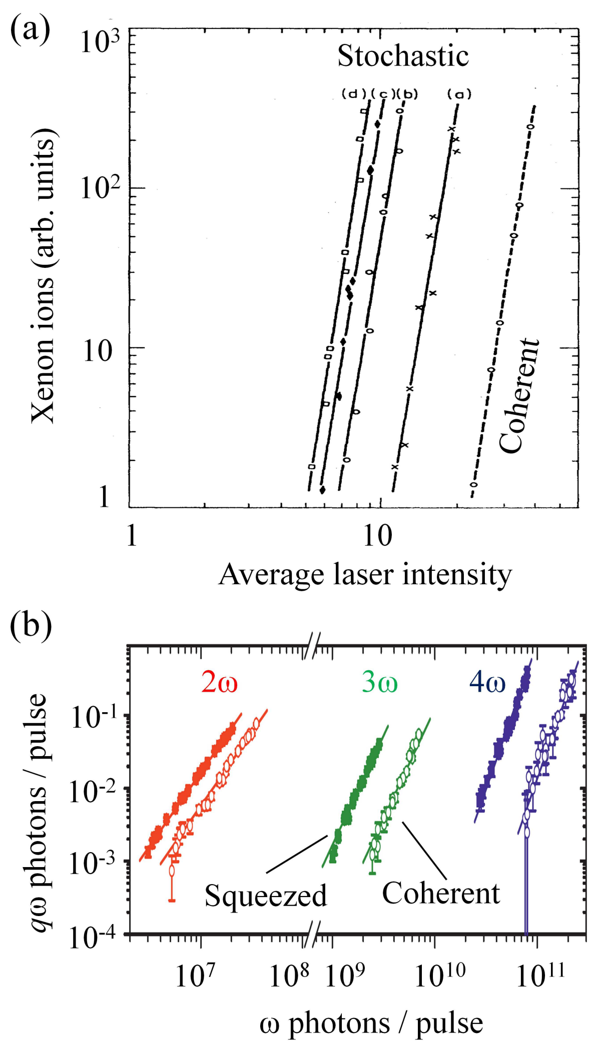

Figure 2. (a) Dependence of the 11-photon multiphoton ionization of Xe on the intensity of a coherent and stochastic light field operating with 10, 30, 70, and 100 phase-unlocked modes (shown with a, b, c, and d in the graph). (b) Dependence of the harmonic yield, produced by nonlinear processes in a crystal, on the intensity of a vacuum squeezed (solid squares) and a coherent (open circles) light states. Parts (a) and (b) are reproduced from Refs. [57] and [28], respectively.

visual science, ultrafast science and nonlinear spectroscopy providing the means to observe and control nonlinear processes on a fundamental quantum level. The advantages of using the quantum light towards these directions have been beautifully described in Ref. [64]. As is briefly discussed in Ref. [65]:

> Quantum light offers several advantages to spectroscopy – by enhancing signal strengths, by creating new ‘control knobs’ for the manipulation of optical signals, or by even allowing entirely new types of signals. The strong fluctuations of quantum light can enhance the nonlinear signal strength relative to linear absorption[66]. In addition, time-frequency entanglement of photons can be employed to control excitation pathways and excited state populations in aggregates[67]. Third, the quantum nature of light may be used to study collective effects in many-body systems by back and forth projection of entanglement from the field onto the matter. This allows to prepare and control higher excited states in molecular aggregates, and access dark multi-particle states. Finally, photon coincidence counting experiments can access useful material information imprinted on the quantum statistics of emitted light fields.

The most recent example which depicts the impact of the intense quantum light, is demonstrated in the theoretical work of Ref. [68] where the authors have shown the influence of the statistical properties of light in atomic spectroscopy and particularly the AC Stark splitting effect.

5. Conclusions

Over the last few decades, tremendous efforts in laser engineering have led to the development of laser systems delivering high-power laser pulses with duration down to 5 fs and power up to the tens of petawatts range. Such systems have been employed in groundbreaking investigations in strong laser field physics[35]. However, the development and the upgrade of these high-power lasers have mainly been focused on the energy enhancement of the coherent light states of the laser field, leaving unexploited the potential effect that the intense nonclassical or stochastic light sources can have for investigations in nonlinear optics. In this perspective article, we aimed at highlighting the important role that high-power laser systems may play towards the development of intense quantum or stochastic light and its novel applications in nonlinear optics including interactions in the ultrarelativistic regime (such as laser-induced pair production or vacuum polarization effects[35,46–48]) where the enhancement of the desired signal remains a challenging task. After a brief presentation of the potential schemes that can be used towards this direction, we have discussed the remarkable effects of photon statistics in nonlinear optics.

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