Photon correlations in both time and frequency

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While quantum mechanics precludes the perfect knowledge of so-called “conjugate” variables, such as time and frequency, we discuss the importance of compromising to retain a fair knowledge of their combined values. In the case of light, we show how time and frequency photon correlations allow us to identify a new type of photon emission, which can be used to design a new type of quantum source where we can choose the distribution in time and energy of the emitted photons.

In quantum theory, one cannot have full information over a system. This is one of the most important features of the theory, that caused much exasperation to Einstein who insisted that in order to be satisfactory, a physical model has to be complete, i.e., describes all that can be measured (what he called “elements of reality”). Let us take the case of light. There are many things that one can measure from a light field, such as its frequency (that corresponds to its color) or its intensity (its brightness). In fact, one is typically interested in the more complete information that provides the intensity at the various frequencies, the so-called “spectral distribution” or spectrum of light. There is also the polarisation (if light “points” in a direction), the coherence (its ability to produce fringes if superimposed to itself) and of course the position (in space) where the field is measured or how these various quantities change spatially.

With the advent of quantum mechanics, our understanding of light underwent considerable modifications, starting with a revival of one of the great scientific controversies: the particle versus wave nature of light. This had opposed Newton to Hooke, Huygens, Young and other prestigious names but also to most the evidence of the time. When light was later measured to travel slower in a dense medium, the particle interpretation was believed to be definitely buried. Two centuries later, Bose’s derivation of the blackbody radiation (spectrum of emission due to temperature) and the photoelectric effect (how light produces current from metals it shines on), related the energy, \( E \), to the frequency, \( \omega \) (through Planck’s relation \( E = h\omega \)), and it appeared that the light field is quantized, that is, light is made up of particles after all: the photons. This exacerbated the completeness problem as this makes particularly salient the impossibility to know simultaneously two basic and crucial quantities for a photon: its energy and position (or, equivalently, its time of emission or time of detection, etc.) Time and frequency are indeed conjugate, meaning that they refer to complementary features that cannot be defined together. This is illustrated in Fig. 1 for three possible cases of the light field: i) where the energy (frequency) is perfectly known, resulting in complete indeterminacy in the time of emission, iii) the opposite case where the time of emission is perfectly known, resulting in indeterminacy of frequency and ii) a compromise where both energy and time are known within some finite accuracy. Since all the evidence in favour of the wave interpretation could not be disposed of, but remained in startling contradiction, the view emerged of the wave-particle duality. This was one of the first uncanny quantum-mechanical concepts, arguing for the coexistence of mutually excluding aspects.

Light is even more bizarre when one considers its correlations. The concept of “correlation” describes the tendency of variables to exhibit some degree of relationship, which can be small or even zero (“uncorrelated”) or on the contrary large or even total (in which case one can equate them through a mathematical function). A shocking property of light was discovered by Hanbury Brown in the mid 1950s, with the observation that the photons arrival times on a detector are positively correlated—meaning that photons tend to arrive together, in “bunches”—and this even if they come from different, uncorrelated sources, such as two stars from separate galaxies. This always happens as long as de-
tectors are “blind” to the photonic properties (i.e., they do not resolve their frequency, polarisation, etc.) Such dramatic and counter-intuitive properties caused a commotion and were even initially rejected as absurd by the scientific community, but it was soon understood as a manifestation of indistinguishability of particles which can be accumulated in the same state (“bosons” or wave-like objects), as opposed to those which cannot co-exist (“fermions” or matter-like objects). This led to the development of quantum optics and brought another revolution in our understanding of a central theme of Physics: coherence. Rather than being linked to the monochromaticity of a field and the stability of its amplitude, coherence is more fundamentally a measure of the degree of correlations between photons. A coherent light source, like a laser, is one for which photons have no mutual correlations. This happens when the granularity of the field does not matter and removing a photon leaves the field essentially unperturbed, corresponding to the classical case where something can be observed without affecting it. As a consequence, photons arrive at the detector without any time relationship between them, as one would have expected in the first place. Blackbody radiation, where photons are in thermal equilibrium, comes on the contrary with strong positive correlations, namely, the bunching reported by Hanbury Brown. There is a third, negative type of correlations, which is of special interest as it describes quantum states of the light field which have no classical counterpart and can power quantum technology. A single photon is an example. Measuring the light field in this case removes the photon and one is left with the vacuum. Such states of the light-field are highly sought after because they can be used for so-called “quantum information processing”, where “qubits” replace the bits (0 or 1) to enhance considerably the computational power to the point that computation currently out of reach, such as factoring very large integers, would become feasible. The case of integer factorisation would become feasible. The case of integer factorisation would be out of reach, such as factoring very large integers, would be out of reach, such as factoring very large integers, would be out of reach, such as factoring very large integers, would be out of reach. Since it is impossible to have a complete characterization of photons in both time and frequency, photon correlations have largely been focused on their temporal aspect alone. It is, however, important for quantum applications that photons remain indistinguishable, or the quantum effects would be washed out and a stream of photons would reduce to a mere classical stream of bits. This means that if frequency is to be resolved in a measurement, photons must be closer in frequency than the detector resolution. To properly describe what happens when one compromises between time and frequency, one needs a theory of photon correlations not only in time alone, but that covers for these two variables. The formal aspects of such a theory have been developed in the late 80s by providing the mathematical expression for the joint probability distribution $g^{(n)}(t_1, \ldots, t_n)$ to detect $n$ photons such that the one detected at time $t_i$ has energy $\omega_i$. The uncertainty has been brought here in frequency through the detector resolution (the so-called “filter spectral linewidth”) $\Gamma_i$, that gives the accuracy one has on $\omega_i$. Physically, this corresponds to filtering the light and thus determining (or measuring) its frequency.

One fundamental source of light has been particularly fit for the study of correlations with combined frequency and time information: resonance fluorescence. This consists of a laser exciting a two-level system, which emits light by spontaneous emission (rather than by scattering or reflecting the laser; this gives the “fluorescence” part of the name) and when the energy of the laser matches the energy of the system’s transition (this gives the “resonance” part of the name). This is therefore a fundamental case of driving a system at the frequency at which it emits. In such a case, the spectrum of emission consists

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**FIG. 2.** The spectrum of emission (distribution of emitted frequencies) of a coherently driven two-level system, known as resonance fluorescence, features three peaks (the so-called “Mollow triplet”). This can be understood as photon transitions between neighbouring doublets of the level structure, shown to the right. As two of the four possible transitions have almost the same energy, one peak is twice as high as the others. A natural question that this structure raises is: how are the photons from the various peaks correlated?

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(a) (b)
of a triplet, shown in Fig. 2(a), that is called the Mollow triplet after the person who first provided its mathematical expression [9]. The physical reason for this peculiar lineshape was provided through the so-called dressed-atom picture [10], that shows how the combined two-level system plus resonant driving leads to a level structure that consists of an infinite ladder of doublets, shown in Fig. 2(b). The doublet comes from the two-level system and its infinite repetition comes from the photons of the laser. Now, photon transitions from one doublet to the next account for the spectral structure in much the same way that photon transitions between the states of the hydrogen atom account for its spectral lines (Lyman, Balmer, Paschen and other series). The exact computation of frequency and time-resolved correlations of this simple system, however, remained out of reach even for the case of two photons only, $g^{(2)}(\omega_1, t_1; \omega_2, t_2)$, until a technique was introduced by some of the Authors [11] that allows to compute this quantity without the approximations performed before. In particular, it lifted an important constrain of previous works to limit to photons whose energies are those of the emission peaks. This may not seem to be a serious limitation since the system emits mainly at these frequencies. However, computing the full two-photon correlation spectrum (2PS) of resonance fluorescence [12], it was found that, on the contrary, most of the interesting quantum emission does arise away from the peaks. The 2PS for resonance fluorescence is shown in Fig. 3(a), showing the joint probability distribution of detecting two photons at the same time for all the possible combinations of frequencies. This results in a two-dimensional landscape (as there are two frequencies). The color code is such that red corresponds to bunched photons (increased probability to detect two photons with the corresponding frequencies together), white is for no correlations (same probabilities as for two random sources) and blue is for antibunched photons (decreased probability to detect the photons simultaneously). The point at $1,0$, for instance, corresponds to correlations between photons coming from the central and high-energy satellite peaks. This is blueish, meaning that such photons tend to avoid being detected together.

The computation of this complete landscape of photon correlations let appear a clear feature: the three antidiagonal lines of strong bunching. These correspond to so-called “leapfrog transitions”, whereby transitions in the ladder of states of Fig. 2(b) are not between neighbouring doublets anymore, but jumping over one of these (hence the name) in a two-photon transition. The fact that the transition now occurs with two photons lifts the quantization of the spectrum, since only the sum $\omega_1 + \omega_2$ is quantized, so that $\omega_1$ can take any value, as long as the other photon fulfills energy conservation by being emitted at frequency $\omega_2 = (E/h) - \omega_1$. This is actually a known phenomenon from planetary nebulae that results in a rare situation where atoms are trapped in states which only have a two-photon channel escape, resulting in continuous spectra that have puzzled astrophysicists for some time [13]. In our case, there is no need of a lucky suppression of single-photon events, these can remain dominant as is the case in resonance fluorescence where most of the emission is indeed coming from the peaks. The filtering in frequency allows to literally unravel the two-photon emission, which one can show is maximally nonclassical [14]. This theoretical prediction for the structure of photon correlations has been confirmed experimentally with a spectacular agreement [15].

Computing photon correlations in both time and frequency thus led us to the discovery of a new type of photon emission which had remained unnoticed despite five decades of combined theoretical and experimental scrutiny on the fundamental problem of resonance fluorescence [16-18]. The importance of this finding lies in the obvious prospects it enables for the design of new types of photon sources. Quantum mechanics is notorious for making it possible for anything to happen, by providing a probability amplitude to any event whatsoever which, at the classical level, may be small or cancelled by others with which it interferes destructively. Resonance fluorescence is such a quantum system that, although it is globally a single-photon source that stems from a two-level system, actually embeds any type of photon emission, which one can distillate and harvest through frequency filtering [19]. One can focus on any desired event by coupling the system to a photonic resonator,
known as a “cavity”. This stimulates the emission at the frequencies of interest by a process known as “Purcell-enhancement”. For instance, placing the system in a single cavity with a frequency at $1/n$th of the distance (in frequency) between the central and satellite peak, turns the Mollow triplet into a pure source of $n$-photon emission \cite{20,21}. That is to say, this generalizes the case of a single-photon source to one that emits exactly and exclusively $n$ photons, for any integer $n$ (chosen by placing the cavity at the adequate frequency). But even this already remarkable extension only scratches the surface. By turning to more complex types of leapfrog transitions \cite{22}, one can realise other types of versatile and tunable quantum sources. For instance, one can design a configuration where a photon of a given energy “heralds” (announces) the subsequent emission of five photons equally split in frequency, that can be used as an input for a quantum gate. The number of photons and their distributions in energy are configurable, by placing cavities at the corresponding frequencies. This may be technically challenging, but the principle is simple and general enough to inspire actual implementations, in this or other quantum systems. The ability to tune and exploit photon correlations thus promises a wealth of applications, ranging from quantum spectroscopy \cite{23} to providing better quantum sources of the types already known, or of a completely new character \cite{24,25}.

In conclusion, although quantum mechanics does not allow us to know everything about a system, it is important to deal with the compromise on knowledge one can get from all the conjugate variables. In the case of photon correlations, we have shown that the time-information alone, which has dominated the field of quantum optics since its creation, gives an overly simplified picture of the structure of photon emission. This, in particular, has kept hidden important leapfrog processes that jump over the states by involving several photons at once. Such processes can power the quantum technology of tomorrow, for instance by turning such a simple and fundamental problem as resonance fluorescence into a configurable universal multiple-photon emitter.

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