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Abstract. The centrality of the photon concept in modern physics is strongly evident in wide spheres of photonics and nanophotonics. Despite the resilience and persistence of earlier classical representations, there are numerous optical features and phenomena that only truly photon-based descriptions of theory can properly address. It is crucial to cast theory in terms of observables, and in this respect the quantum theory of light engages most directly and pragmatically with experiment. No other theory adequately reconciles the discreteness in energy of optical quanta, with their characteristic quantum mechanical delocalization in space. Examples of the distinctiveness of a photonic representation are to be found in nonclassical optical correlations; intensity fluctuations and phase; polarization, spin, and information content; measures of optical chirality; near-field interactions; and plasmonics. Increasingly, links between these fundamental properties and features are proving significant in the context of nanoscale interactions. Yet, even as new technologies are being built on the framework of modern photonics, a number of difficult questions surrounding the nature of the photon still remain. Both in its flourishing applications and in matters of fundamental entity, the photon is still a subject very much at the heart of current research. © *The Authors. Published by SPIE under a Creative Commons Attribution 3.0 Unported License. Distribution or reproduction of this work in whole or in part requires full attribution of the original publication, including its DOI.* [DOI: 10.1117/1.JNP.8.081599]

Keywords: photon; quantum optics; spontaneous emission; nanophotonics; nano-optics; spin; optical angular momentum; twisted light; optical vortex; near-field; virtual photon; plasmonics; reflection; fluctuations; entangled states; quantum information; phase; multipoles; polarization.

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1 Introduction

The concept and physical connotations of the photon have undergone a lengthy period of accretion since Einstein introduced the simple notion of light being quantized in units of energy.[1] Additional attributes such as spin, subsequently bestowed upon the photon concept, have developed and enriched the subject, and greatly expanded its explanatory power. Although these and other, subsequent developments have not yet wrought a full elucidation of the underlying nature, our current understanding of photon behavior is sufficiently robust to have successfully built whole industries upon it. At heart, the science of the photon enables us to comprehend phenomena that underscore both the integrity of the quantum and the wave/particle duality of light; examples are readily to be found in fields as diverse as quantum optics and communication,[2] the sciences of color, spectroscopy, photophysics and photochemistry, and the technology of optical materials.

Distinctly photonic attributes are to be found in a variety of nanoscale applications, such as nanomechanics, nonlinear nano-optics, and intermolecular energy transfer. The term "nanophotonics" itself seems to have been coined as the promise of such applications first began to be recognized,[3] foreshadowing the subsequent development of single-photon sources. Prominent among the latter, quantum dots, nanocrystals, nanowires, and organic molecules now appear to offer scope for some of the greatest promise for advances in optical communications and

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quantum computing.[4–6] The arguments used 20 years ago by Willis Lamb and others, in heroic attempts to demolish the use of the photon concept, have since then become much less beguiling.[7]

Given the sweeping breadth of this domain, it might be considered astonishing that classical wave descriptions still pervade a great many reports, and even text books, dealing with nanoscale optical phenomena. To express the electric field of light with circular frequency ω as a classical wave, such as $E_0 \cos \omega t$ with E_0 the vector amplitude, is entirely inconsistent with the quantization of light. True enough, such representations do often provide a basis for acceptable accounts of numerous optical phenomena, delivering results that are in many cases immeasurably different from those of the photon-based theories. However, there are numerous exceptions, some of them especially notable. It is particularly striking that there is no form of spontaneous emission (whether by atoms, molecules, quantum dots, or any other form of nanoemitter) for which the classical representation gives satisfaction.[2] The reason is self-evident: the excited state of the emitter is an eigenstate of the system Hamiltonian—a stable stationary state solution of the field-free Schrödinger equation. In such a picture, there is simply no mechanism for the initially excited state to decay. In the quantum theory of light, however, the electric field becomes an operator on radiation states: it is no longer a variable to be simply equated to zero when no light is present. It is this, photon-based formulation of theory that correctly delivers the Einstein A-coefficient, the experimental determinant of a spontaneous emission rate. Even textbooks designed to inform their readers about light-matter interactions on the nanoscale can be misleading, commonly overlooking the need for a thoroughly photon-based account, and focusing instead on traditional thermodynamic arguments.[8]

It is freely acknowledged that there may be at least a perception of unusual difficulty, in the fully fledged quantum electrodynamical theory—perhaps compounded by the disconcerting sound of ancillary concepts such as "vacuum fluctuations," "entangled," or "virtual" photons. It may well be that this, to some extent, deters many scientists and engineers from fully engaging with the photon picture. The difficulty, at least, is exaggerated: the theory of quantized radiation[9,10] has much in common with the widely familiar quantum theory of simple harmonic motion,[11] applied throughout physics. Moreover, it is in the spirit of the modern formulation of quantum mechanics to focus on observables, and for the photon there is an obvious connection to be drawn with photon detection measurements, usually involving absorption. A concise, measurement-based interpretation, popularly ascribed to Roy Glauber, asserts: "A photon is what a photodetector detects." The photodetection process (signifying a resonant response to the electromagnetic field of impinging radiation) fundamentally comprises detection events, each of which registers the arrival of a discrete quantum of energy. In fact, the photoelectric effect provides an insufficiently conclusive basis for proving the existence of photons,[12] but the necessity of the concept has been proven in many other unequivocal connections.[13] It is important to notice that any experiment or measurement that can have a bearing on the subject is ultimately limited by the requirement for an observable, and this inevitably involves interactions with matter. The field of nanophotonics thus represents an obvious point of connection between the understanding of photons and the science of nanoscale optics.

Starting from a perspective on some of the broader issues of photon-based and classical representations, and developing a theme addressed in a recent conference paper,[14] this paper aims to set out some of the key issues, focusing on why the photon picture has such a central role in the realm of nanophotonics. Features more specifically associated with virtual photons are being addressed in a separate paper.[15]

2 Photonics, Nanophotonics, and Nano-Optics

The closeness of the link that exists (as will be shown) between nanophotonics or nano-optics, and photon attributes, invites close attention to the general properties of photons and their propagation characteristics. Accordingly it may be useful to begin with a workable definition of a photon: perhaps it is no surprise to report that, even at this level, there are immediately some difficult issues to address.

To begin, the word photon was coined in 1926 by Lewis,[16] who introduced it to describe "not light but [that which] plays an essential part in every process of radiation." Some of Lewis's views, at this early stage of development for quantum mechanics, will now appear odd. For example, he wrote that "[the photon] spends only a minute fraction of its existence as a carrier of radiant energy, while the rest of the time it remains as an important structural element within the atom." This was already a significant deviation from the original "light quantum" concept, of which Einstein had written: "When a light ray spreads out from a point source, the energy is not distributed continuously over an increasing volume … but [it] consists of a finite number of energy quanta that are localized at points in space, move without dividing, and can only be absorbed or generated as complete units."[1] Lewis also insisted that every photon was identical, but he recognized the key attributes of direction, frequency, and polarization determining differences in their observed properties. In this respect, the original concept is quite close in spirit to the accepted sense a century or more later, where definitions usually make reference to optical modes defined by wave-vector and polarization. Concisely, we can assert that an integer designating the quantum occupation number of such a mode signifies, in physical terms, the corresponding number of photons. Each photon signifies one quantum of a single normal mode of excitation of the microscopic electromagnetic field.[17]

In considering evidence for the quantization of light, it might at first seem significant that most of the historic "proofs"—such as the discovery of a wavelength threshold for photoelectron emission—relate to sources of light in which light-matter interactions could only occur one photon at a time. However, it should not be thought that such a situation is by any means unusual. A quick calculation based on the intensity of our most familiar source proves instructive; the mean intensity of sunlight on the Earth's surface, at around 1.4 $\mathrm{kW\,m^{-2}}$ in the spectral region 400 to 700 nm, translates into a photon flux of around 10 photons $\mathrm{Å^{-2}\ s^{-1}}$. In connection with the familiar process of photosynthesis in a leaf, for example, then after accounting for cloud cover, screening by tree canopies, etc., this level of photon delivery amounts to approximately one photon per chlorophyll molecule per second.[18] Needless to say, each photon will traverse even such a large molecule in a very short time, typically on the attosecond timescale, so that the chance of finding just one photon at a time in a particular molecular location is very small. In fact, the prospect of finding two or more photons simultaneously able to interact with any optical center of nanoscale dimensions is entirely negligible, under any ambient conditions. Photons usually travel alone.

In this connection, it is interesting to recall Dirac's famous axiom: "Each photon … interferes only with itself."[19] Although the context of that remark is obviously important (for example, there have been recent reports of interactions between photons whose electromagnetic fields are strongly engaged with atoms in highly excited Rydberg states)[20] this is a salutary consideration—especially given a current debate about whether the apparent coherences seen in ultrafast pulsed (and therefore ultraintense) laser studies of photosynthetic systems can meaningfully represent coherences that could arise under ambient sunlight conditions.[21] It appears unlikely—but the excited state lifetime clearly has a bearing on the question, and the experiments addressing such features are notoriously difficult to interpret.[22]

To broaden this perspective, we can further estimate the likelihood of finding, at any instant, a single photon within a substantially larger volume, such as might be spanned in each direction by the dimensions of the optical wavelength. Suppose we take a laser source: for a laser emitting 532-nm (green) wavelength radiation, the conditions necessary to ensure an average of one photon within this volume would require an intensity of around 7.0×10^5 $\mathrm{kW\,m^{-2}}$—which proves to be a level that is indeed only achievable with a laser source. The lesson is that, although the integrity of the photon is crucial in the optical sciences, most mechanisms involve one photon at a time. The only obvious exceptions are multiphoton absorption and other nonlinear optical processes that specifically engage more than one photon in each optical interaction. Certain techniques with "photon" nomenclature, such as photon echo spectroscopy,[22] specifically require high-intensity pulses and are, indeed, unworkable at the single-photon level.

More commonly in optics, experimental techniques and observations concern a collective response from discrete photon-particle interactions, and one needs to identify what is distinctive about the topics that fall under the umbrella heading "photonics." The early 1950s origin of this term is commonly traced back to the rocketry engineer Eugen Sänger, whose intent seems to

have been to characterize the emergence of technologies based on the photon, in analogy to the electron basis of electronics. Much of the research now reported under this heading pays scant heed to the quantum nature of light; it is not hard to find examples in which the word "photon" is entirely absent. Arguably, this is consistent with the absence of "electron" in most user-oriented material on electronics. There is, in fact, almost no common ground among the many definitions that can be found for "photonics." It is interesting to note that the derivative term "nanophotonics," coined around 50 years later, has already acquired a clearer focus, one that generally centers on optical interactions associated with submicron structures. This area, more obviously linked with the rapidly emerging field of nanotechnology, is now developing a much stronger and more obvious connectivity with the "photon" at its heart. The subject area of "nano-optics" is scarcely distinguishable from nanophotonics, save perhaps in suggesting a slightly greater degree of linkage with, or development from, traditional optics.[23]

3 Issues of Integrity, Indivisibility, and Localization

Photons are generally considered to be massless entities, notwithstanding their capacity to convey both energy E and linear momentum (the latter of magnitude $p = \hbar\omega/c$) in accordance with the relativistic energy-momentum equation $E^2 = p^2c^2 + m^2c^4$, with $m = 0$. Although possible manifestations of a finite mass remain a topic for discussion in a cosmological context, the likely upper bound of around 10^{-62} kg on any possible mass[24] is clearly way below any scale measurable in the realm of terrestrial optics. Despite their lack of mass, however, photons are not to be considered spatially dimensionless. As Michael Mishchenko has pointedly commented, "the lasting misinterpretation of photons as localized particles of light is kept flourishing by ignorant authors of many school and college textbooks on physics."[17] Photons do not behave or move like point particles; they are at every instant in time intrinsically delocalized in space, as a simple manifestation of quantum uncertainty.

Although the direct measurement of a photon may be localized by the physical extent of a detector, this cannot be interpreted as signifying localization of the photon itself.[25] As the process of light absorption involves photon capture, it brings into effect a collapse of the quantum state for the radiation field. Mandel and Wolf's classic text warns against attempts to build a picture of localized photons: "a photon has no precise position no matter what the state may be."[26] There have often been misleading attempts to directly interpret photon interactions in the old-fashioned sense of a conventional object with elementary quantum attributes—see, for example, a recent paper on coherent effects in nanostructured organic photovoltaic materials.[27] Some of the more obvious errors have already been noted and corrected, most notably by Mukamel.[28] Although it is tempting to think of the massless photon in point-like particle terms, it is salutary to recall that, as all know, even the electron in the simplest atom is an extensively delocalized entity.

Direct insights into the delocalized quantum nature of light are afforded by observations of processes based on spontaneous parametric down-conversion—the time-inverse of sum-frequency or second harmonic generation, in which single photons are converted into pairs that propagate away usually in slightly different directions. The result is an optical field exhibiting quantum entanglement,[29–31] signified by nonclassical correlations between the polarization states of the two emerging components. Many other experiments in quantum optics underscore the more general inseparability of such photon states. When a single photon is intercepted by a beam-splitter, the resulting state of the radiation field with one well-defined quantum of energy can be cast as a linear superposition of "reflected" and "transmitted" states—each having a distinct character, since different wave-vectors are involved. Such linear combinations of state are not to be conceived as linear combinations of particles: the radiation state comprises the energy of a single quantum, but not "a photon" in the sense we have been considering. Although beyond the scope of this paper, the possibilities of using such entangled photons for quantum information and communication have been the subject of much interest and extensive investigation.[32] Recombining light from the two paths out of the beam-splitter provides a definitive proof of quantum optical behavior.[33] It is interesting to recall that the difficulty of explaining instances of partial reflection was one of the main reasons Newton's original corpuscular theory of light eventually fell out of favor.

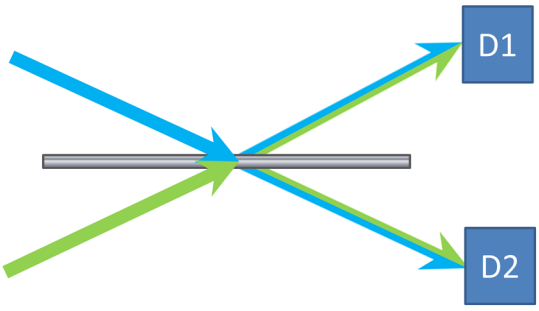

Fig. 1 Schematic optical geometry for study of the Hong-Ou-Mandel effect, detectors D1 and D2 receiving light that is transmitted and reflected from the two pulsed beams (input from the left-hand side) by the centrally placed beam-splitter (which, in the experiments, can be moved to allow or disallow simultaneity of the two input pulses). The two input beams are identical in wavelength (different colors on the figure only used for clarity).

A classic illustration is the Hong-Ou-Mandel effect, the name now given to an experiment illustrating the specifically nonseparable nature of a two-photon state.[34] The setup, shown in Fig. 1, is a beam-splitter in the form of a two-sided, partially transmissive mirror, fabricated and oriented in such a way that exactly half the light impinging on either side (from a set direction) is reflected, the rest passing through. Identical input beams of light are directed onto each side of this beam-splitter at an identical angle, impinging at the same point on either of its two faces. Two photodetectors are positioned to intercept the light coming off the mirror on each side: each might in principle equally detect either the reflection of one input beam or the transmission of the other. Both theory and experiment reveal remarkable quantum effects when the intensity of each input is reduced to a level that provides individual photons only sporadically. Whenever two photons arrive at the mirror together, one from each source, it transpires that there is no possibility at all that both detectors will simultaneously register a photon: it is impossible to have either both photons transmitted, or both photons reflected. The only possible outcome is for two photons to emerge in the same direction. This nonseparability is a principle that extends to states comprising any number of photons.

It is worth further emphasizing the distinction between the wavefunction for a simple radiation state with a single quantum of energy, and a potentially insidious "photon wavefunction" concept. Most mainstream theorists will agree that the photon does not have a wavefunction; the notion is essentially a misappropriation of terms. This assertion can alarm some experimentalists who routinely take the notion for granted. But in definitive textbooks of the quantum theory of light there is no mention of any such thing. In fact, those who use the term "photon wavefunction" usually mean something like the spatial distribution of the electric field—ignoring the equally important (but certainly different) form of the magnetic field, for example. Bialynicki-Birula et al., who use the Riemann–Silberstein vector—essentially a complex quantity whose real and imaginary parts represent the electric and magnetic fields, respectively—represent notable exceptions.[35–37] Overlooking the distinction between this and a true quantum mechanical wavefunction is just about defensible when single photons are involved, and the distinction from the state vector is less of a problem. For states with two or more identical photons, there is no sense in which one could consider each to have its own wavefunction.

Not surprisingly, attempts to cast the photon as a classical particle are particularly disastrous, a paper by Li providing a recent case in point.[38] The photon does not travel under the laws of Newtonian particle mechanics, along a track such as might trace out a helix in the case of circular polarizations, for example—nor should the oscillations of an electric field pointing in any particular spatial direction be regarded as a physical displacement in that spatial dimension. Correct representations can be found in standard literature such as the well-known texts by Craig and Thirunamachandran,[39] or the more recent one by Duarte.[40] Attempts to circumvent such well-founded theory can only exhibit disregard for the catalog of known photon behavior, especially interference phenomena. It is interesting to note that even from Roychoudhuri's very different perspective, in which the principle of wave noninteraction is used to support a notional bridge between classical and quantum behaviors, it is still rightly considered necessary for matter to be

engaged in explaining optical interference effects—whereas an entirely classical particle conception of the photon only leads to predictions that everyday observations would immediately prove demonstrably false.[41,42]

In the case of another recent experiment reporting direct measurement of a photon wavefunction,[43] the interpretation of results is itself based on flawed logic. The general argument, itself based on perfectly valid application of "weak measurement" theory,[44] concerns identifying positional information. Such an application would be valid if applied to a genuine material particle because such a particle continues to exist, beyond the measurement. The clever feature of weak quantum measurement is that the process of observation leaves the state of the observed particle essentially unchanged. But the photon is not a particle of matter, and almost any measurement of it involves its annihilation. Certainly, we can make observations that truly represent photon detection events, but what is it that is being observed? Let us look at the Hamiltonian operator for the measurement. Without interaction, the matter and radiation field both continue unchanged and no measurement is made; this is a simple consequence of the states with a specific number of photons being eigenstates of the Hamiltonian for radiation in a vacuum. Whether the interaction Hamiltonian is written in the -p.a (minimal coupling) or -μ.e (multipole coupling) formulation, it is clearly the field that is measured—a property of the photon, not the "presence of the photon."

4 Photon Number Distributions: Fluctuations and Phase

Every beam of light exhibits fluctuations of intensity: fundamentally, it is an unavoidable consequence of having light energy conveyed in discrete quanta. Furthermore, to produce any effectively constant beam would require unattainable levels of control over the individual photon emissions occurring within the source. Again, no source has the capacity to deliver photons that all have precisely the same wavelength: notwithstanding practical limitations, this would contravene the principle of quantum uncertainty in the emission events. In consequence, every beam has a characteristic coherence length, essentially determined by the inverse of the beam linewidth expressed in wavenumber terms. Although it is possible to ascribe the underlying fluctuations in beam intensity to stochastic patterns of interference between optical modes with differing wavelengths and frequencies, there are more fundamental aspects of phase that arise at the quantum level.

Each photon carries a phase $\exp[-(\mathrm{i}kr - \omega t)]$, referring the space coordinate r and time t to the position and time for photon creation. Yet this "absolute" phase, conveyed by the photon creation operator in the Heisenberg representation,[45] has little practical significance unless optical interference is somehow engaged. More significant still is the fact that the quantum operators for photon number and phase (the latter in any of several different representations) do not commute.[46] Loudon[10] has given one of the clearest descriptions of the implications: if the exact number of photons was to be precisely known, then precisely no phase information could be secured by any measurement. Again, this is an issue that appears relevant to some current arguments over the coherence of single photons.[21]

For radiation states with an exact phase, the photon number becomes infinitely uncertain. There are, however, quantum optical states that lie between the extremes represented by number and phase states. A familiar and important example is the "minimum uncertainty" state known as a coherent state,[10] generally considered closest in behavior to a classical wave, for which the associated intensity fluctuations take the form of a Poisson distribution. It is possible to construct such a state in which the mean (expectation value) number of photons is precisely unity—yet individual measurements might give other low integer values. Moreover, any act of measurement would destroy the latent phase information. The close link between issues of coherence and photon statistics is the subject of much of Mandel and Wolf's pioneering work.[26]

5 Polarization and Spin: Angular Momentum and Information

The integer spin associated with the photon marks it out as a boson—subject, as such, to Bose–Einstein distribution laws (as distinct from half-integer fermions, to which Fermi–Dirac statistics

apply). There is no limit to the number of bosons that can be accommodated in a single quantum state—in the case of photons this is indeed one of the key reasons that coherent laser light is feasible. The quantized spin is distinctive because, although the photon also conveys a quantum of energy and also of linear momentum, the latter both depend on optical frequency or wavelength: spin does not. Moreover, the states for which the component of photon spin along the direction of propagation is sharply defined (in the quantum mechanical sense) as ± 1 are states of left/right-handed circular polarization. As the propagating electromagnetic field vectors of circularly polarized light describe a helix, it is not surprising that this form of light affords the means to differentially interact with matter of opposite chiral form, such as the left- and right-handed enantiomers of optically active molecules. It has in fact been shown that every meaningful measure of chirality in light itself amounts to a simple difference in the numbers of left- and right-handed circularly polarized photons present.[47]

As a result of its spin, the capacity of the photon to convey information is not limited to its direction or frequency/wavelength: differentiation on the basis of polarization is also possible. Circular states of opposing handedness are perhaps the most obvious candidates (since they relate directly to eigenstates of the operator for spin)[26] although any two states corresponding to diametrically opposite positions on the Poincaré polarization sphere—see Fig. 2—would provide a suitable basis. Indeed, one may consider that the photon offers five degrees of freedom, for example three Cartesian components for the wavevector and two for the polarization, such as a latitude and longitude on the Poincaré sphere.

It is now recognized that there is a much wider scope in this whole field, afforded by the development of new structural forms of optical beam.[48] Typically such beams are produced by the passage of conventional Hermite–Gaussian beams through spatial light modulators,[49,50] q-plates,[51] or spiral phase plates,[52] with each of which it proves possible to engineer beams having intricately crafted wave-fronts, nonuniform polarization behaviors, singularities, and phase structures. Some of the most important examples are the "optical vortex" wave-front structures of Laguerre–Gaussian and Bessel beams.[53] The former represents the most widely studied type of optical vortex, illustrated in Fig. 3. Recent developments in theory suggest that such beams might in fact be produced directly, by exploiting the electronic relaxation of suitably engineered molecular complexes.[54,55] Such advances present new challenges for the nanostructural engineering of appropriate forms of emitter.

For such specifically directed and structured beams, which still offer the same options for polarization state, the three modal degrees of freedom usually associated with Cartesian components of the wave-vector now typically designate one (axial) wave-vector component and two other indices signifying an integer orbital angular momentum, and a specific Laguerre or Bessel polynomial determining the radial form of the beam cross section. The detailed vortex structure of such beams is primarily characterized by a topological charge, l, signifying the number of wavelengths over which each of l intertwined components of the wave-front complete one cycle about the beam axis. Each photon in such a beam conveys an orbital angular momentum of

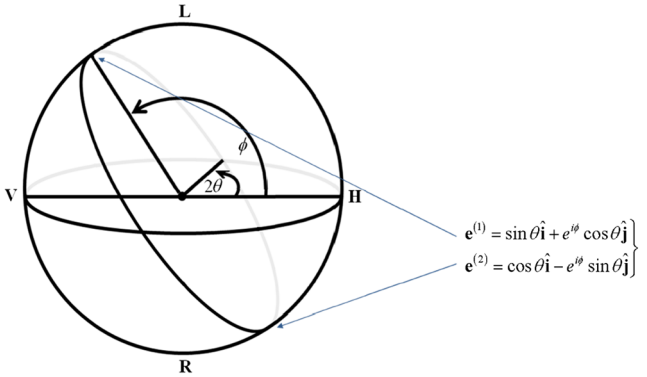

Fig. 2 The Poincaré sphere, upon which any diametrically opposed positions represent a suitable pair of polarization basis states: L, left-handed (circular); H, horizontal (plane); R, right-handed (circular); and V, vertical (plane).

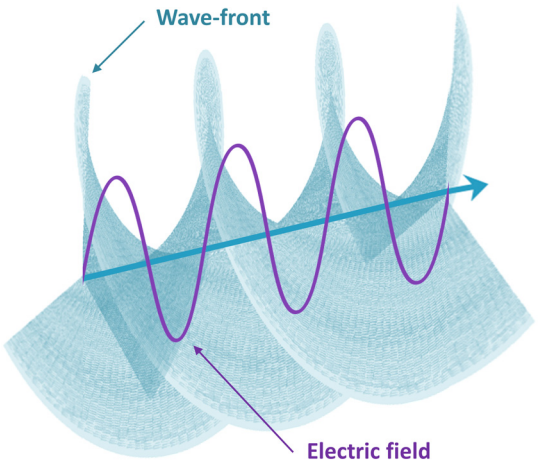

Fig. 3 Propagation of a plane polarized optical vortex. Here, the left-handed helical wave-front has topological charge $l = 3$, i.e., a wave-front with three intertwined helical surfaces, each completing one full rotation about the axis over a span of three wavelengths. The orbital angular momentum is $+3\hbar$ per photon. Circular polarization would add or subtract one unit of $\hbar$, according to whether the sense of rotation was the same, or opposite, to the sense of the helical wave-front.

magnitude $l\hbar$. The topological charge can take any real integer value, positive or negative, denoting either left- or right-handed twist, respectively. Although it might have been supposed that some structural attributes could only be supported in beams comprising a high density of photons, it transpires that most of these novel features can indeed be interpreted in terms of the properties of individual photons; experiments convincingly demonstrate that individual photons convey information on the structure of complex beams.[56,57] Recalling the intrinsically delocalized nature of the photon makes it a little easier to comprehend such properties.

Evidently, complex light beams with structured wave-fronts offer different degrees of freedom, and based on this premise it has been considered possible for individual photons to convey a far greater information content than was previously considered possible.[58,59] There are, in various quarters, reservations on how far this principle might be extended, although from some of the most ardent exponents there have even been suggestions that it might be possible to send the information content of a whole image, in a single photon. What has been proven is that modal information can be encoded in single-photon states, and subsequently resolved by beam tomography.[60] A holographic method has also demonstrated the possibility of discriminating between objects even when they are illuminated by no more than a single photon.[61] Most important of all, a variety of increasingly sophisticated means has been developed both to sort and to measure, with high fidelity, the topological charge content in vortex beams.[62–64] Such methods hold the promise of achieving the encoding and retrieval of a genuinely increased amount of information per photon—methods that hold an enticing promise for development in future forms of optical communication. At present, no upper bound on the potential information content per photon has been proposed: it is indeed curious to reflect that practical considerations alone might determine an upper limit, when it might be supposed that quantum principles would impose less arbitrary constraints.

Another noteworthy feature connects some of the earliest and some of the most recent research on optical angular momentum. A certain Darwin, an early pioneer in quantum mechanics, is widely accredited as conceiving of photons conveying multiple quanta of angular momentum. In an attempt to explain the apparent mismatch between the angular momentum carried away by photon spin in atomic decay,[65] Darwin drew attention to the case of "quadrupole emission where $2\hbar$ of momentum is lost in a single photon," concluding that "[f]or quadrupole emission the pure particle concept is a failure." Subsequent efforts based on the premise of an offset displacement for the point of emission were also unsuccessful.

In fact, the modern quantum electrodynamical theory of quadrupole (in general, multipole) emission has recently proven that the process of electronic decay in an emitter does not in general produce photons measurably imprinted with the corresponding angular momentum—the

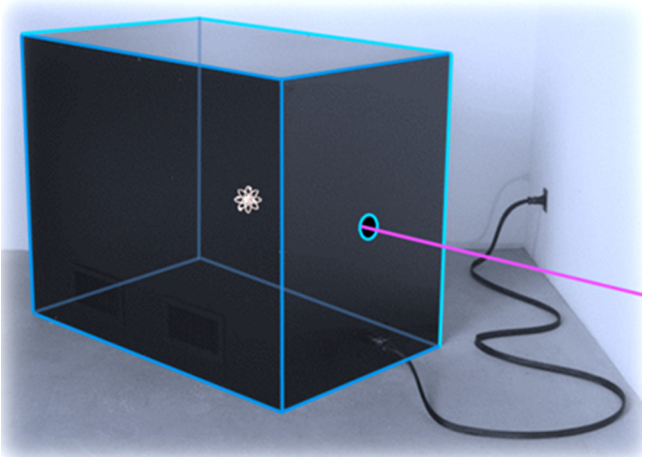

Fig. 4 Schematic illustration of the emission from an excited state atom inside a black box. It proves impossible to determine, from the emission in any specific direction, the multipole character of the transition responsible for the photon (Refs. 67 and 68).

character of any such emission can only be registered statistically, in the spatial distribution of the emerging radiation. In a sense, this ought not to be surprising. Consider, for example, the emission of a ruby laser, which results from a ^2E to $^4A_2$ transition of Cr^{3+} ions, each such ion being positioned in an octahedral field produced by six surrounding O^{2-} ions. The emission process, which is both spin and dipole forbidden, is only electric quadrupole allowed. If a strict rule of angular momentum transfer to the emitted photons was to apply, then only special "electric quadrupole" detectors would be able to detect the laser output. Of course this is not the case. The correct understanding is perfectly consistent with the angular quantum uncertainty principle.[66] In the case of any source with multipolar emission, it is now clear that removal of a detector from the immediate vicinity of the source produces a decreasing angular uncertainty in photon propagation direction, and this is manifest in an increasing range of possible integer values for the angular momentum of the detected light, as indicated by the thought-experiment depicted in Fig. 4.

6 Fields Near and Far: Real and Virtual Photons

In the detection of photons far from their source of emission (over distances, that is, much larger than the optical wavelength) the processes of emission and photodetection are generally considered separate. However, if the detector is moved closer to the source, to within a fraction of a wavelength, then the emission and absorption events become close enough in time for quantum uncertainty effects to intervene. Such effects arise as a result of source-detector coupling by virtual photons. The character of the electromagnetic coupling proves to exhibit subtle changes in this "near-field" regime, with implications that are especially significant for nanoscale interactions, as will be shown in Sec. 7.

A virtual photon might be described as a photon that passes between two particles of matter without intervening measurement. This provides a good working basis for describing interactions on the nanoscale, where the most distinctive features of "virtuality" are exhibited. Behind this definition is a concept first introduced in the quantum electrodynamical theory for noncontact couplings between charges, as for example, in electron-electron scattering.[69] Two key facets of the virtual photon representation are its intrinsic accommodation of causal constraints, in accordance with the precepts of special relativity; the other is the requirement for a summation to be effected over all wave-vectors and polarization states, for each virtual photon. The latter calculational strategy is consistent with the quantum mechanical principle of summing over every variable for unobserved intermediates. It emerges that, when virtual photons escape beyond the immediate vicinity of their birthplace, their characteristics nonetheless acquire an

increasingly real character, so that energy and momentum conservation become increasingly tight constraints.

To appreciate the progressive character of virtual photon propagation, it proves instructive to consider resonance energy transfer—a process widely prevalent in nature, and one that affords the mechanism for electronic excitation to pass between electrically neutral, independent particles such as molecules, chromophores, quantum dots, and the colored ions in many optical crystals.[70] The quantum electrodynamical representation entails the creation of a virtual photon at the particle which is acting as excitation donor, and subsequent annihilation of this virtual photon at another particle with the role of acceptor. Highlighting the paradoxical nature of virtual photons, calculation has to allow for virtual photons of all energies and all directions of propagation, not only those traveling directly toward the acceptor. Account must even be taken of virtual photons traveling outward from the acceptor.[71–73] The result reveals that the character of the energy transfer slowly changes between asymptotic forms, as the distance between the energy donor and acceptor increases. Close by the donor, in the near-field region where the distance is below ∼100 nm, the result is a rate that falls off with the inverse sixth power of distance. This is the result familiarly known as Förster (or fluorescence) energy transfer.[74] It is this form of energy transfer between quantum dots, for example, that has demonstrated utility as a single-photon emission source.[75] However, for distances well in excess of 100 nm, the distance dependence of the rate approaches the asymptotic form of an inverse square law, in accordance with the acceptor capturing a "real" photon released in spontaneous emission by the donor.

In fact, as distance increases there is a smooth transition in every respect from virtual to real photon behavior.[71] This can be understood as a manifestation of quantum uncertainty, corresponding to increasingly tight constraints on energy (and momentum) conservation as the photons travel for longer (and over larger distances). Such a perspective also lends new insights into the physical origin and form of the electromagnetic fields in the immediate vicinity of a photon emitter. An example of such features is the fact that the electric field has longitudinal, as well as the conventional transverse, character with respect to the displacement of a detector from the source. Related, and yet more striking, is the fact that there is a small but significant effective variation of wavelength close to the source,[76] as shown in Fig. 5. The wide range of virtual photon involvement in the field of nanophotonics, which has now emerged as an extremely important topical area in its own right, has been more comprehensively surveyed elsewhere.[15] Most recently, a role for the concept has even emerged in the ionization dynamics of helium nanodroplets.[77]

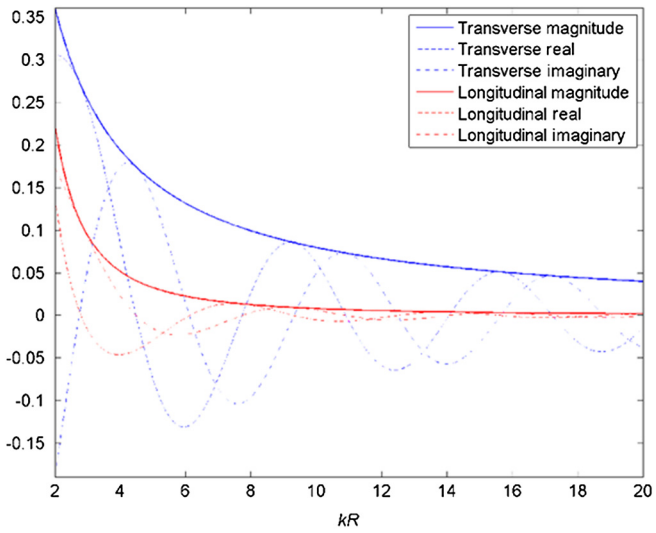

Fig. 5 Magnitude of the longitudinal and transverse components of a dipole-emitted electric field, plotted against kR (arbitrary vertical scale). The real and imaginary parts of each component are also included. Near the source, the interval between successive wavefronts is slightly larger than the wavelength; the interval between the indicated nodes corresponds to a distance $R \sim 1.022\lambda$, where λ is the wavelength.

7 Placing the Photon in Nanophotonics

The strength of a photon-based formulation of theory—and especially its deployment with the derivative concept of the virtual photon—is especially evident in the sphere of nanophotonics. Here, it is very evident that fundamental processes and properties associated with the nanoscale must properly account for the quantum nature of both the matter and the radiation field. A familiar example is the Casimir force that operates between electrically neutral, nonpolar nanoparticles, whose role and significance in nanoelectromechanical systems are now well recognized; their correct representation invokes the creation of short-lived virtual photons from the vacuum.[78,79] In another area, the laser-induced interparticle forces known as "optical binding,"[80] now a prominent focus of research in connection with optically induced nanoparticle self-assembly,[81] were also first predicted using the virtual photon formalism.[82] Further examples can be found in the field of light-harvesting materials[83] and the interparticle transfer of excitation associated with coupled nanoantenna emission.[84]

It is interesting to reflect on the changes in photon property that result from traveling through a medium with significant optical dispersion in the wavelength region of the photon. The variation in phase velocity associated with the frequency or wavelength dependence of the refractive index means that the velocity varies in a characteristic way across the absorption spectrum. In the familiar dispersion curves, which plot optical frequency against wave-number k, frequency regions well away from material absorption exhibit approximately linear sections of the plot with a slope approaching the vacuum speed of light—this line separates into asymptotes of zero slope in the vicinity of each absorption frequency. The quantum interpretation, for the regions of diminished slope above and below each resonant frequency, is that the photon seamlessly morphs into what is known as a polariton[85]—an electromagnetic quantum that is associated with strong interactions between propagating radiation and electronic excitations of the material. There is a continuum of such behavior, the photon acquiring a progressively modified character as its own electromagnetic fields are "dressed" by those of the material it encounters.

The strongest interactions arise in metals, where surface conduction electrons are most free to engage and respond to incident light by the formation of surface plasmon polaritons (sometimes known as dressed photons). Here, the strong coupling between the electromagnetic waves and charge density oscillations provide for the confined (subwavelength) transmission of optical frequency signals in nanostructures. Based on this principle, the emerging field of plasmonics[86] promises a wide scope for informatics and communications applications.[87] There is also a connection here with optical vortices. For example, a Laguerre–Gaussian beam impinging on a thin metallic layer will generate a surface plasmon optical vortex (SPOV).[88] If such a film is

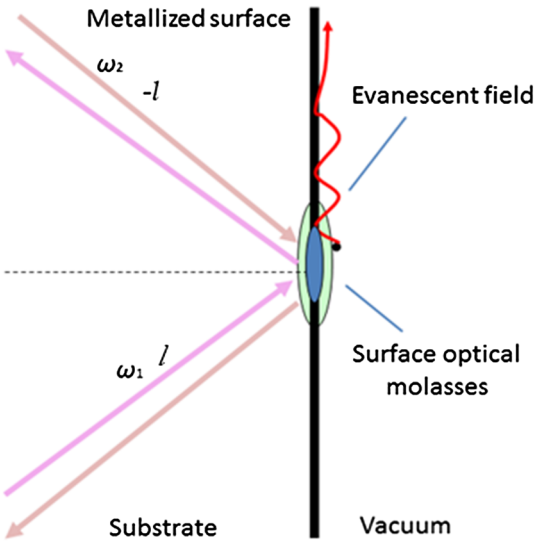

Fig. 6 Generation of a surface plasmon optical vortex at a metalized surface, using counter-propagating beams of opposite topological charge but similar optical frequency to produce a stable region of circulating charge.

surface-adsorbed on the surface of a transparent substrate, the beam can engage with the surface conduction electrons through total internal reflection, imparting a circulation of charge. When the SPOV is produced at the intersection of two counter-propagating beams with a slightly different optical frequency (Fig. 6), the pattern of interference between the surface optical vortices produces a rotation at the different frequency.[89] Shen et al. have demonstrated an effective method for measuring the orbital angular momentum and intensity distribution of an SPOV using near-field scanning optical microscopy.[90,91]

Other recent work has shown how it is possible to measure the orbital angular momentum of light using a circular plasmonic lens. This interference method owes its success to the partitioning of amplitudes between the surface plasmons and directly transmitted light, resulting in the formation of characteristic intensity distributions near the plasmonic lens.[92]

8 Conclusion

The photon description of light provides grounds for theory that successfully and comprehensively addresses not only the breadth of optics and photonics, but also into photobiology and photochemistry,[93] and still further beyond into the spheres of elementary particle physics and cosmology. Despite the continued presence of classical descriptions in optics, it is interesting to observe the present convergence of different approaches to light in its interactions with complex forms of matter.[94] In the condensed phase, where bulk refractive indices (or dielectric constants and magnetic permeabilities) are the most widely used means of quantifying optical response, it is now becoming obvious that for materials with nanoscale heterogeneity, correct theoretical representations require different kinds of treatments, generally requiring the light to be properly described in terms of photon propagation. Examples range from the need to adopt statistical methods to model the multiple scattering of photons on passage through media such as biological tissues,[95] through to the effects of surrounding media on energy transfer processes.[96]

Despite its successes, even within its most obviously appropriate province of photonics, numerous questions nonetheless remain for which current answers are either unsatisfactory, or in some cases entirely unknown. Top of the list right now is the issue of how much information you can get into a photon—and if there is a limit, what is it? Given the tantalizing prospect of directly emitting photons that can convey orbital angular momentum, with a correspondingly distinctive modal structure,[54,55,97]—and also the reporting of methods to sort and identify photons that differ in these respects[62,98,99]—what should now be considered the ultimate constraint on the capacity of optical communication channels? A more prosaic question is: how does a photon interact with the atoms of a gas to produce the effects manifest as refractive index? Simple calculations based on repeated forward scattering prove to yield results that are hugely in error compared to measured values: accordingly, there appear to be no literature reports properly addressing the issue. The issue of what one might adopt as a sensible scattering cross section is made significantly more complex not only by the fact that photons are not localizable, but that the process of elastic forward scattering by individual atoms or molecules has no directly practicable measurable.

Again, with a reminder of Dirac's assertion that a photon only interacts with itself, over what scale of length does a photon interact with itself? This issue also has a strong bearing on the following: Given that beams of light can form standing waves on normal reflection, what is the photon-based picture of mirror reflection? Here, the recent discovery of odd effects in the vicinity of a mirror—positions at which the superposition of input and output optical fields produces different patterns of interferences for the electric and magnetic fields[47,100]—invites pointed questions of how far away from the mirror those and other such effects might be registered at the single-photon level. Also, what happens to the angular momentum mismatch in a multipolar form of radiative decay? Calculations on hydrogen atoms seem to give conflicting results[101–103] while, as we have seen, the application of fundamental symmetry principles underscores a principle precluding the unambiguous transmission of information on the multipolar character of a source transition. On this, and each of the questions above, active research is ongoing to secure a clear answer.

It is reassuring to recognize that the perverse difficulty of these and many other such issues is a strong indicator of the activity and health of the subject area. Far from becoming fossilized, the photon concept, one hundred years and more after its birth, is very much alive.

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