# On the nanoscale transmission of quantum angular momentum

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## ABSTRACT

The electromagnetic propagation of angular momentum associated with photon spin has evolved into a subject of much broader remit, following the theoretical and experimental realization of optical beams that can convey quantized orbital angular momentum. The possibility of transmitting such information over nanoscale distances raises numerous issues. For example, it is known that electron spin can be relayed by near-field communication between exciton states in quantum dot assemblies; the question arises, can orbital angular momentum be conveyed in a similar way? There are fundamentally important technicalities surrounding such a prospect, representing potentially serious constraints on the viability of angular momentum transfer between electronically distinct components in structured nanomaterials. To resolve these issues it is necessary to interrogate the detailed form of near-field electromagnetic coupling of the relevant transition multipoles. The emerging results exhibit novel connections between angular momentum content in the near-field. The analysis leads to a conclusion that there are specific limitations on the nanoscale transmission of quantum angular momentum, with challenging implications for quantum optical data transmission.

**Keywords:** Metamaterials, angular momentum, spin, multipole, quantum dot, near-field, quantum information, quantum electrodynamics

## 1. INTRODUCTION

Over the past twenty years the understanding of optical angular momentum has undergone a huge development, producing nothing short of a paradigm shift in its conceptualization and opening up whole new areas of study [1,2]. This resurgence of interest was given a major stimulus by a realization [3] that it is possible to optically engineer structured beams of light that can convey azimuthal (orbital) angular momentum, as a result of phase singularities and associated vortex structures in the beam wavefront [4]. This form of angular momentum is quite distinct from the well-known connection of integer spin angular momentum with circularly polarized light. Indeed, with suitably engineered twisted beams it proves possible to convey significant integer multiples of the fundamental unit of angular momentum, $\hbar$, per photon [5]. As in the pioneering original study, most of the research in this area has focused on Laguerre-Gaussian modes of light, although the same principles operate for a variety of vortex mode structures.

The capacity of structured beams to carry angular momentum that is independent of any circularity of polarization has been thoroughly established by the development of studies to elicit such features, the experiments often based on interferometric interrogation with differently structured beams [6]. This led to a realization of the astonishing possibility of conveying more information per photon than was previously thought possible, prompting suggestions for a variety of quantum communication and information applications [7]. In connection with free space propagation, these possibilities are very much the subject of active investigation – but there are numerous questions concerning transmission over nanoscale distances. For example, although it is known that exciton spin can be communicated by near-field resonance energy transfer between neighboring quantum dots [8], is it conceivable that orbital angular momentum information might be conveyed by similar optical means? The answer is not obvious, and the question not very easy to address, since most of the existent theory on structured light addresses free-space propagation. There are, indeed, serious issues to resolve concerning angular momentum transfer by nanoscale transmission within structured materials, their significance amplified by the goals of much current research.

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A very important and influential study by Leach et al. [9] showed that it is possible to distinguish between even and odd values of the azimuthal quantum number at the single-quantum level. However, in this and similarly-directed research, it is notable that the status of the observed field as comprising a single photon is a retrospective inferral rather than measurement. Indeed one cannot expect to secure unequivocal information on both photon number and phase (the latter being intimately associated with orbital angular momentum) due to the number-phase uncertainty relationship that holds for optical states [10]. It is a little surprising that this principle, which is particularly relevant to low-number states, has seemingly received very little consideration in the literature to date. Certainly, there are numerous other practical issues to consider alongside this fundamental limitation; a very useful survey has recently been provided by Franke-Arnold and Jeffers [11].

As a particularly relevant context for such considerations, it is interesting to note that there have recently been theoretical advances in the tensor formulation of multipolar light, accommodating its near-field behavior and fully identifying the form of longer range retardation effects. In establishing a connectivity with short-range behavior, this form of analysis has also identified novel features in the angular momentum content of the electromagnetic near-field [12, 13]. Building on this work, the present study addresses specific technical constraints connected with the nanoscale transmission of quantum angular momentum. The results present challenging implications for quantum optical data transmission between electronically distinct components, over the scales of distance that are most actively sought for miniaturized devices and metamaterials.

## 2. PHOTON INTERACTIONS IN QUANTUM OPTICAL INFORMATION TRANSFER

In the development of quantum optical communication and informatics, attention generally focuses upon relatively low photon number Fock states of the radiation field. Whilst some of the classic effects that elicit the quantum nature of radiation, such as the Hong-Ou-Mandel effect [14], entertain fundamental interactions that can engage the combined effect of two input photons, these are not in general optically nonlinear processes. Rather, they are effects that occur only as a result of relatively rare correlations in the arrival of input photons: the intensities are far too low to significantly engage optically nonlinear properties of a material medium.

A variety of clever schemes have been envisaged for the transmission of quantum information using light endowed with orbital angular momentum [11]. Generally, such approaches focus upon states of the radiation field, rather than the electrodynamics of their interactions with matter. However, at the photon level, the only means of eliciting encoded information – whether of an intense beam of a single photon – is ultimately by engagement with matter, and it is this aspect that will concern us here. Even interferometric methods of beam interrogation require detector material for the formation or registration of images. Certainly, at the level where genuinely quantum effects might be manifest, the types of fundamental photonic process that can be involved in producing a signal are relatively few, being generally limited to the realm of linear optics: they are photon emission, absorption, and scattering. In connection with twisted optical beams, spontaneous photon emission is not relevant for consideration: these beams are in practice produced by the passage of conventional light through suitably designed optical elements – moreover, such types of light cannot be spontaneously generated from the vacuum [15]. Stimulated emission and photon absorption are of interest as they are likely candidates for coding and decoding processes.

As has been shown previously [16], orbital angular momentum exchange through the single-photon interaction of structured light with atoms or molecules occurs primarily in electric dipole coupling, and that angular momentum engages only local centre of mass motions. When higher order electric multipole interactions are entertained, it transpires that a limited degree of orbital angular momentum exchange with internal electronic transitions can occur – permitting the gain or loss of one unit in the case of electric quadrupole transitions, for example. Crucially, however, there is no possibility of a one-to-one mapping between the angular momentum content of the beam and the internal electronic motions; there are no selection rules on which to base atomic or molecular protocols for unequivocally decoding orbital optical angular momentum. Self-evidently, the same principles apply to both photon absorption and emission.

To proceed with a more general analysis, it is expedient to expand the focus of enquiry to other photon-level processes, entailed in the progress of structured radiation through an optical system. In connection with nanoscale transmission, a key phenomenon is scattering, in the quantum electrodynamical sense that couples the removal of a photon from one optical mode with the installation of a photon into another (or indeed the same) mode. In the course of passage through any optical element each photon generally experiences a sequence of scattering interactions (including forward elastic scattering); in the absence of absorption and emission there is nothing else that can change the state of the light at the level of low photon numbers. The following development of theory therefore focuses on angular momentum issues that arise first in the case of an isolated scattering event, and then a sequence of two such events. The implications that are thereby drawn are readily generalized to accommodate a succession of scattering events. It is the character of the electromagnetic propagation between these events, and the information that can thereby be conferred, that are of central interest.

For these initial studies, attention is focused upon cases where there is an unequivocal correlation between a specific input and output (or detected) photon. In principle this allows for the complete representation of photon transit through an optical system, in the same spirit as Jones calculus or ray transfer matrix analysis. Subsequent studies will entertain cases where there is a possibility of quantum interference between alternative photon histories. However, if the present analysis can identify any constraint or compromise of information fidelity emerging in the study of a single-thread photon history, it is certain that no lesser problems and frustrations are likely to beset more intricately designed systems.

## 3. THE ELECTRODYNAMICS OF PHOTON PROPAGATION AND SCATTERING EVENTS

The premise of the following analysis is that high-fidelity transmission over nanoscale distances can become meaningful only when information is retrievable from a single-photon input state necessarily within the optical near-field, *i.e.* for a distance $R$ where $kR \ll 1$, with $k = 2\pi/\lambda$ for an appropriate wavelength $\lambda$. This does not constrain the means of electromagnetic information transfer; even the so-called 'radiationless' (Förster) transfer, which occurs over much more severely restricted distances (typically tens of nanometers), operates on a basis of near-field coupling (which, in the context of a quantum electrodynamical description [17], is cast in terms of virtual photon propagation). The theoretical framework for addressing excitation propagation by such means can therefore also elicit the issues that arise in connection with angular momentum. Indeed, this principle has been demonstrated in connection with the local transmission of exciton spin, referred to earlier [8]. In that study, it was shown that the transfer of excitation along a column of quantum dots with a common orientation preserves spin information, absolutely.

Let us first consider a single photon scattering event. The theory has been well-rehearsed elsewhere [10]; suffice it to begin with the following expression for the matrix element;

$$M^{fi} = -\left(\frac{n^{\frac{1}{2}}\hbar ck}{2\varepsilon_o V}\right) e_i^{(\lambda)}(\mathbf{k})\bar{e}_j^{(\lambda')}(\mathbf{k}')\alpha_{ij}^{00}(-k;k). \tag{1}$$

Here, $\alpha_{ij}^{00}(-k;k)$ is the dynamic polarizability, a molecular counterpart to the conventional linear susceptibility;

$$\alpha_{ij}^{00}(-k;k) = \sum_r \left\{ \frac{\mu_i^{0r}\mu_j^{r0}}{E_{r0} - \hbar ck} + \frac{\mu_j^{0r}\mu_i^{r0}}{E_{r0} + \hbar ck} \right\}. \tag{2}$$

In the scattering process the input light has $n$ photons of wave-vector $\mathbf{k}$ and polarization vector $\mathbf{e}^{(\lambda)}(\mathbf{k})$; what emerges is differentiated by primes. Whereas coherent optical processes such as forward Rayleigh scattering, surface reflection and second harmonic generation (SHG) owe much of their high efficiency to the constructive interference of signals from different locations [18], this is a feature whose significance is lost at very low levels of intensity. Nonetheless, when the emergent light continues to propagate in the forward direction, and with unchanged polarization, then the factor of $n^{1/2}$ in (1) is squared. In this single-centre scattering case, it is trivially evident that an optical input with a given angular momentum – whether spin or orbital – can generate an onwardly propagating output with the same angular momentum character; the application of time-reversal symmetry [19] to equation (1) shows it by inspection. In particular it is instructive to observe that the polarizability tensor (2) is cast in terms of products of the time-inverse transition electric dipole moments for 'up' and corresponding 'down' transitions, $0 \leftrightarrow r$. Although (2) is specifically limited to the electric dipole approximation, it is reasonable to suppose that these transitions must be electric dipole allowed if forward-directed propagation is to be efficient. The restrictions of the electric dipole approximation will be removed in the ensuing development of theory, once the formalism of method is established.

For the special case of a topologically charge (optical vortex) input beam, the arriving photons have an orbital angular momentum that is quantized azimuthally. Forward propagation processes retain this orbital angular momentum absolutely – indeed this is true both for forward Rayleigh and forward harmonic generation [20]. As regards the spin angular momentum that is uniquely quantized in circular polarizations, the same rule applies for forward Rayleigh propagation in an isotropic environment. Such retention of spin is not possible in forward harmonic generation because the single emitted photon can convey away only one quantum [21], and so angular momentum could not be conserved as Noether's theorem [22] would require. However, if deflection occurs – where the emergent, detected light emerges in a direction that differs from the input; then, the one-to-one mapping of angular momentum content is entirely lost, both for Rayleigh and harmonic scattering. Again, this is readily shown by casting the emergent light in terms of the incident mode set, a procedure that scrambles the topological charge and also any spin angular momentum.

Next we consider the case of a double-scattering event. This represents the crux of this investigation, for it addresses electromagnetic propagation between two material interactions, neither of which is limited to the role of absorber or emitter. This is important for two reasons. First, this detaches propagation issues from others associated with photon creation or annihilation. Secondly, it elicits the distance-dependent characteristics of such propagation, supporting the present focus on nanoscale transmission. It might be considered that there is nothing further to analyze, given the simple outcome of the above consideration of single-center scattering. However a moment's reflection shows that this is far from the case. Over nanoscale dimensions, and certainly in the optical near-field, one cannot presume a specific causal sequence of these scattering events. Consistent with the basic tenets of quantum theory – and reflecting quantum uncertainty – one has to sum over all possible sequences of the fundamental photon events. Quantum electrodynamical calculation of the quantum amplitude for double-scattering (at sites to be labeled A and B) thus entails the consideration of an unobserved state of the radiation field, cast in terms of virtual photons. There is a total of four photonic interactions (input photon annihilation, virtual photon creation, virtual photon annihilation and output photon creation) constrained only by the coupling necessity that the sites of creation and annihilation of the virtual photon are different. It is also simplest to assume that the sites of real input and output differ; in the following it will be assumed that a real photon enters at A and one emerges from B. There are twenty-four time-orderings that contribute to the overall matrix element for the process – corresponding to twenty-four Feynman graphs [23] of the form illustrated in Fig. 1, or the same number of routes across a more comprehensive state-sequence construct, as in Fig. 2 [24].

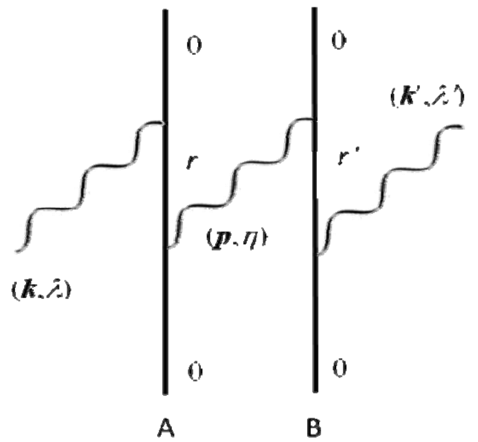


Fig. 1. One of 24 distinct time-ordered diagrams (time progressing upwards) for photon double-scattering, with an incident photon of mode ($\boldsymbol{k}$,$\lambda$), virtual photons ($\boldsymbol{p}$,$\eta$) and an emergent photon ($\boldsymbol{k}'$,$\lambda'$).

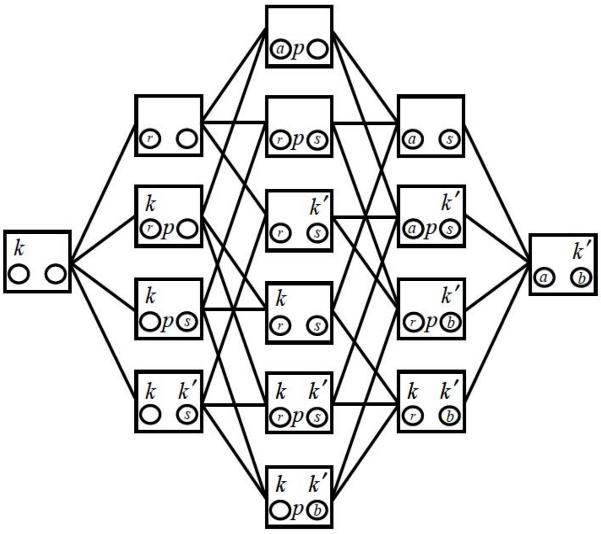


Fig. 2. State-sequence diagram for double scattering

In the ensuing calculation, with the requisite virtual photon summations necessarily to be effected over all possible states of the radiation field, any complete basis set will suffice. Thus, if the two scatterers are labeled *A* and *B*, on summing the quantum amplitudes the matrix element emerges as follows [24] (now dropping the superscript implied labels 00 on the polarizability tensors):

$$M^{fi}\left(\mathbf{k},\mathbf{k}',\mathbf{R}_{\mathrm{A}},\mathbf{R}_{\mathrm{B}}\right)=\left(\frac{n^{\frac{1}{2}}\hbar ck}{2\varepsilon_o V}\right)e_i^{(\lambda)}\left(\mathbf{k}\right)\bar{e}_l^{(\lambda')}\left(\mathbf{k}'\right)\alpha_{ij}^{\mathrm{A}}\left(-k,k\right)V_{jk}\left(k,\mathbf{R}_B-\mathbf{R}_A\right)\alpha_{kl}^{\mathrm{B}}\left(-k,k\right)\exp\left(i\left(\mathbf{k}\cdot\mathbf{R}_{\mathrm{A}}-\mathbf{k}'\cdot\mathbf{R}_{\mathrm{B}}\right)\right);\quad(3)$$

the same result is also deliverable by a more direct route that circumvents the lengthy virtual photon calculus [23]. Here, *V* is the retarded resonance electric dipole-electric dipole coupling interaction [17, 25] which accommodates both near-field and wave-zone limits. However, in the near-zone, that limit is identifiable with the usual formula for the coupling of static dipoles. In equation (3), each molecular polarizability again entertains a sum over states – for each such state *r* it is again reasonable to suppose that the transitions are electric dipole allowed. The matrix element (3) thus accommodates a sum of terms of the form:

$$\sum_{r_{\mathrm{A}}}\sum_{r_{\mathrm{B}}}\left(\frac{\mu_j^{r_{\mathrm{A}}0_{\mathrm{A}}}}{E_{r0}^{\mathrm{A}}\pm\hbar ck}\right)V_{jk}\left(k,\mathbf{R}_{\mathrm{B}}-\mathbf{R}_{\mathrm{A}}\right)\left(\frac{\mu_k^{r_{\mathrm{B}}0_{\mathrm{B}}}}{E_{r0}^{\mathrm{B}}\pm\hbar ck}\right),\quad(4)$$

which has been shown to allow the near-field transfer of spin angular momentum through the *V*, as shown in the work with Scholes [8].

A comparison of equations (1) and (3) shows that it is reasonable to interpret the electric near-field impinging upon B, associated with scattering at A, as follows;

$$e_i^{(\lambda)}\left(\mathbf{k}\right)\alpha_{ij}^{\mathrm{A}}\left(-k,k\right)V_{jk}\left(k,\mathbf{R}_{\mathrm{B}}-\mathbf{R}_{\mathrm{A}}\right),\quad(5)$$

subject to a phase factor. The physical interpretation of (5) is therefore the field associated with a collectivity of the virtual photon modes coupling *A* with *B*. Although, when the initially incident light is endowed with orbital angular momentum, there is no specific need to cast the virtual photon sums in terms of specifically structured modes, it is perfectly admissible to do so – and in this case it is expedient to do so, because of the angular momentum properties that can then be inferred. Essentially one is effecting a transformation over three degrees of freedom, between optical mode sets: in the conventional basis these degrees of freedom are the Cartesian components of the wave-vector, whilst in a Laguerre-Gaussian basis for example they are the axial magnitude of the wave-vector and the Laguerre polynomial indices *l* and *p*.

Attention now focuses on the symmetry of the coupling tensor *V* in equation (5). However, present purposes demand a result of wider applicability than the electric dipole approximation allows. It has recently been shown [12] that the simplest virtual photon transit engages a generalized *V* tensor expressible as:

$$V_{i_1 \ldots i_m j_1 \ldots j_n}(k, \mathbf{R}) = \frac{(-1)^m}{4\pi\varepsilon_0} \nabla_{i_2} \ldots \nabla_{i_m} \nabla_{j_2} \ldots \nabla_{j_n} \left( -\nabla^2 \delta_{i_1 j_1} + \nabla_{i_1} \nabla_{j_1} \right) \frac{e^{ikR}}{R} . \quad (6)$$

where *m* and *n* denote the electric multipolar order of the source and detector. It then transpires that the engagement and high fidelity transfer of *orbital* angular momentum, desirable because of the higher (in principle unlimited) number of quanta that each photon might convey, is indeed possible in the near-field – but it is frustrated by the necessity of engaging higher multipoles. Detailed analysis has, for example, demonstrated the dominance of a weight 3 character in electric quadrupole – electric dipole transfer in the near-zone, i.e. over nanoscale distances; the short-range contribution (decaying with the fourth power of distance) with three units of angular momentum engages two from the source with one at the detector. However, when scattering events are concerned then the associated reduction in efficiency with every additional order will still compromise the viability of usage for the propagation of quantum information. It may be recalled that for each unit increment in multipolar order, the associated quantum amplitude for a single photon event (creation or annihilation, whether of a real or a virtual photon) is diminished by a factor of typically $10^{-3}$; the quadratic involvement of such moments in the corresponding multipolar polarizabilities [12] means a 60dB loss per scatterer – 120 dB for the pair – even if only electric quadrupoles and no higher multipoles needed to be invoked. The only alternative that could preclude such a loss of efficiency is that the engaged $0 \leftrightarrow r$ transitions are additionally allowed by lower multipoles – which would critically undermine any fidelity in angular momentum information transfer. Thus it is in practice impossible to circumvent the key obstacle observed earlier – that it is not possible to secure fidelity of angular momentum transfer from light to matter in any of the $0 \leftrightarrow r$ transitions connected with the resonance coupling tensor *V*. Again, therefore, it emerges that there are considerable, perhaps insurmountable difficulties in attempting to achieve the unambiguous nanoscale transmission of orbital angular momentum information.

## 4. CONCLUSION

Issues associated with the fidelity of conveying photon orbital angular momentum information over nanoscale distances have been addressed by identifying relevant symmetry properties in the tensor for photon propagation between arbitrary interactions with matter, accommodating all orders of electric multipole coupling. The results of this analysis indicate that one cannot assume a one-to-one mapping between the input and output, a conclusion that has significant implications for some of the sought developments in quantum information technology.

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