

# Coherent delocalization: Views of entanglement in different scenarios

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**Abstract.** The concept of entanglement was originally introduced to explain correlations existing between two spatially separated systems, that cannot be described using classical ideas. Interestingly, in recent years, it has been shown that similar correlations can be observed when considering different degrees of freedom of a single system, even a *classical* one. Surprisingly, it has also been suggested that entanglement might be playing a relevant role in certain biological processes, such as the functioning of pigment-proteins that constitute light-harvesting complexes of photosynthetic bacteria. The aim of this work is to show that the presence of entanglement in all of these different scenarios should not be unexpected, once it is realized that the very same mathematical structure can describe all of them. We show this by considering three different, realistic cases in which the only condition for entanglement to exist is that a single excitation is coherently delocalized between the different subsystems that compose the system of interest.

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Entanglement is one of the main traits of quantum theory. For some, *the need to describe even systems that extend over macroscopic distances in ways that are inconsistent with classical ideas* [1] is a *troubling weirdness* of Quantum Mechanics. Since the publication of the seminal Gedanken EPR experiment by Einstein, Podolsky and Rosen (EPR) [2], the appearance of the first comments by Bohr about this subject [3] and the introduction of the entanglement concept by Schrödinger [4], innumerable theoretical discussions and experiments related to this topic have appeared. Arguably the most relevant contribution to this discussion has been the introduction, fifty years ago now, of the nowadays well-known Bell inequalities [5]. One of these Bell-like inequalities, the Clauser-Horne-Shimony-Holt (CHSH) inequality [6], is surely the most commonly used in experiments [7].

Originally, the application of the concept of entanglement was restricted to composite systems made up of two spatially separated subsystems. However, correlations of similar nature to the ones existing between physically separated subsystems may also exist when considering different degrees of freedom of a single system [8]. Indeed, entanglement can be measured in this kind of systems, provided that one is able to perform independent measurements in the degrees of freedom involved.

Along these lines, Gadway *et al.* [9] demonstrated the presence of entanglement by measuring correlations in two degrees of freedom (polarization and path) of a single photon; more recently, Vallés *et al.* [10] enlarged this analysis by considering other degrees of freedom (orbital angular momentum) and a more general class of quantum states (mixed states). Violation of Bell-like inequalities, a concept related to the presence of entanglement, has also been used to characterize properties of classical beams containing many photons, i.e., intense beams. Borges *et al.* [11] considered coherent beams whose total electric field writes  $\mathbf{E}(\mathbf{r}) = \Psi_H(\mathbf{r})\hat{\mathbf{e}}_H + \Psi_V(\mathbf{r})\hat{\mathbf{e}}_V$ , and used a CHSH inequality to characterize their coherence properties in one of the two degrees of freedom involved. Kagalwala *et al.* [12] added the consideration of partially coherent beams and also considered the relationship between the degree of Bell inequality violation and the degree of partial coherence in each degree of freedom. This so-called *non-quantum entanglement* has been considered as a fundamental tool for investigating important properties of classical fields [13, 14, 15, 16, 17, 18].

Here we intend to show that by looking at what is entanglement in specific, physically realistic scenarios, one can get a better understanding of what it means to be entangled. We will see that when considering systems in the single-excitation manifold, entanglement will always exist as long as the excitation is *coherently delocalized*. We will refer here to coherence as first-order coherence [19, 20], and delocalization as the fraction of parameter space where the single-excitation takes place. In general, coherence and entanglement do not imply each other, and might address different aspects of a particular physical system. However, in the single-excitation manifold [21], coherence and entanglement can become mathematically equivalent. In this particular regime the presence of entanglement entails coherence and vice versa, which means that any measure of entanglement is also a measure of coherence [22, 23, 24, 25].

The consideration of the single-excitation regime could be seen as overrestricting our analysis. However, the great majority of studies of entanglement, both theoretical and experimental, can be easily demonstrated to belong to this category. Surely, the implications of being entangled in different contexts might not be the same, especially when considering subsystems spatially far away from each other. Notwithstanding, a common conceptual understanding of entanglement in all of these different scenarios is still valid and illuminating. For the sake of simplicity, and keeping a common notation, we will use a quantum language to describe all scenarios, even when we refer to systems that might as well be described using classical concepts.

In what follows, we will be more specific about what *single-excitation* regime, *localization* and *coherence* mean. With these concepts at hand, we will explore different scenarios, some of them perfectly described classically, in which entanglement has been observed. Even though the appearance of entanglement in some of these cases might cause certain surprise, we will show that its presence should not be unexpected if one realizes that such systems can be described within the single-excitation manifold, and are therefore completely analogous to the systems where entanglement is usually considered.

## 1. Entanglement in light-harvesting complexes

Due to its importance and relevance for explaining and describing life on earth, photosynthetic light-harvesting complexes have been a topic of study for decades [26]. In recent years, they have attracted a renewed attention [27, 28, 29] mainly due to the experimental observation of long-lived electronic coherences in the energy transfer process of bacterial and algal light-harvesting complexes [30, 31, 32, 33]. Although the relevance of some quantum-born concepts, such as entanglement, for explaining the highly efficient energy transport observed in photosynthetic systems is still under discussion [34, 35, 36, 37, 38], we will show that the presence of entanglement should not be unexpected anyway. In the following, we will see that the appearance of entanglement is a direct consequence of considering a coherent nature of the photosynthetic complex, provided the state describing its dynamics is defined within the single-excitation manifold.

In general, a single excitation in a network of  $N$  chromophores (or sites) can be represented by a density matrix of the form

$$\rho = \epsilon |\Psi\rangle \langle\Psi| + (1 - \epsilon)I_D, \quad (1)$$

where

$$|\Psi\rangle = \sum_i^N \alpha_i |i\rangle, \quad (2)$$

$$I_D = \sum_i^N |\alpha_i|^2 |i\rangle \langle i|, \quad (3)$$

with  $|i\rangle$  indicating that the excitation is on site  $i$  with probability  $p_i = |\alpha_i|^2$ . The key consideration of *single-excitation* implies that only one site at any time can be in the

excited state. The parameter  $\epsilon$  determines the degree of coherence of the system.

In order to quantify coherence we make use of the degree of coherence, a function that corresponds to the absolute value of the normalized first-order coherence function [39]. We can then write the degree of coherence as

$$g_{ij}^{(1)} = \frac{\text{tr}(\rho\sigma_i^\dagger\sigma_j)}{[\text{tr}(\rho\sigma_i^\dagger\sigma_i)\text{tr}(\rho\sigma_j^\dagger\sigma_j)]^{1/2}}, \quad (4)$$

where  $\sigma_i^\dagger$  and  $\sigma_i$  are the raising and lowering operators for the  $i$ th site, respectively, and  $\text{tr}(\dots)$  stands for the trace. Making use of Eqs. (2) and (3), it is straightforward to find that for the state in Eq. (1), the degree of coherence writes

$$|g_{ij}^{(1)}| = \epsilon, \text{ for all } i \neq j. \quad (5)$$

Notice from Eq. (5) that, depending on the value  $\epsilon$ , the degree of coherence can take values from zero, when there is no coherence, to one, for a fully coherent system.

For the sake of simplicity, and to make more compelling the comparison with the other cases that will be discussed below, we restrict our attention to the case of two coupled sites or dimer. In this scenario, the density matrix in Eq. (3), in the basis  $\{|1\rangle, |2\rangle\}$ , reads

$$\rho = \begin{pmatrix} |\alpha_1|^2 & \epsilon\alpha_1^*\alpha_2 \\ \epsilon\alpha_1\alpha_2^* & |\alpha_2|^2 \end{pmatrix}. \quad (6)$$

Different measures—such as logarithmic negativity [40] and global entanglement [22]—have been used for quantifying entanglement in light-harvesting complexes. Here, we will quantify the amount of entanglement present in a two-site system by making use of the concurrence [41, 42], which for a density matrix of the form (6) is given by (see Supporting Material of Ref. [43])

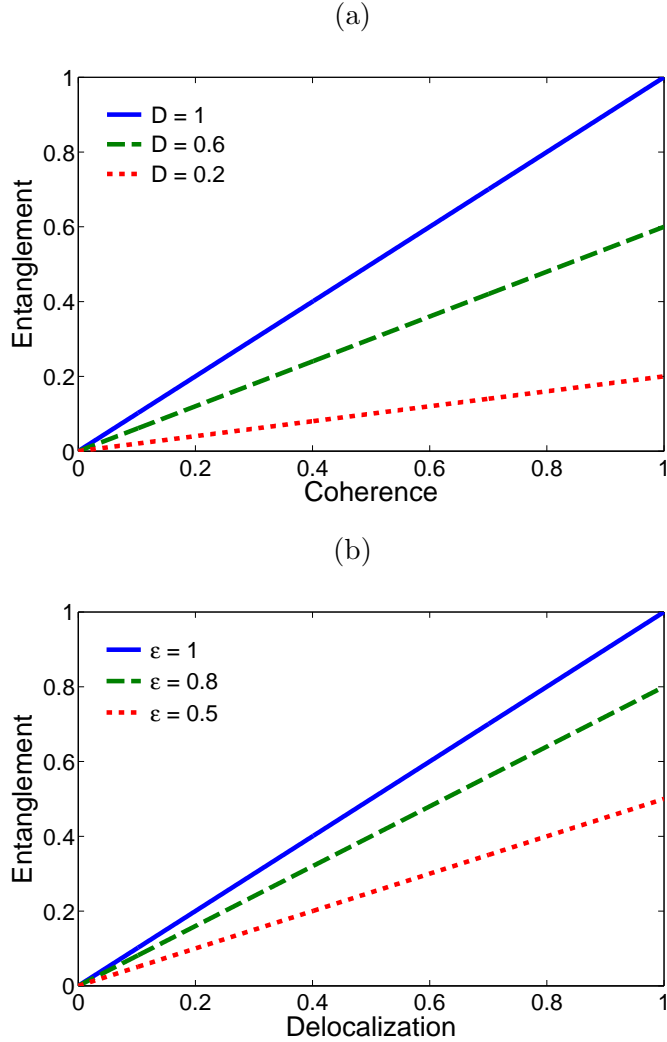
$$C = 2 \max\{0, \epsilon\sqrt{p_1p_2}\} = 2\epsilon\sqrt{p_1p_2}. \quad (7)$$

Finally, to quantify the degree of excitation's delocalization in the system given by Eq. (6), we introduce a measure of delocalization that can be defined as

$$D = 2\sqrt{p_1p_2}. \quad (8)$$

According to Eq. (8), if the excitation spreads equally over all sites (maximum delocalization), i.e.,  $p_1 = p_2 = 1/2$ , one obtains  $D = 1$ ; whereas if the excitation resides in a single site (maximum localization), i.e.,  $p_1 = 1$  and  $p_2 = 0$ , or  $p_1 = 0$  and  $p_2 = 1$ , we obtain  $D = 0$ . Notice that local unitary transformations that affect the excitation in sites 1 and 2 independently do not affect the value of  $D$ . Moreover, for a coherent state ( $\epsilon = 1$ ), Eq. (6) is equivalent to the Schmidt decomposition of the system [44], which justifies the validity of  $D$  as a good measure of the excitation's delocalization in the system.

Figure 1(a) shows the amount of entanglement (as quantified by the concurrence) as a function of the degree of coherence  $[g_{12}^{(1)}]$  for a fixed value of the degree of delocalization. For a given delocalization, the degree of entanglement increases for increasingly larger



**Figure 1.** Entanglement, as quantified by the concurrence, as a function of: (a) degree of coherence  $\epsilon$ ; and (b) delocalization  $D$  of the single excitation.

values of the coherence. Also, Fig. 1(b) shows the amount of entanglement as a function of the degree of delocalization for a fixed value of the coherence. Notice that also in this case, increasingly larger values of delocalization provide larger values of entanglement.

Using Eqs. (4), (7) and (8), and the results provided in Figs. 1 (a-b), one can find that

$$\text{Entanglement} = \text{Delocalization} \times \text{Coherence}.$$

From this relationship, we can conclude that maximum entanglement always requires a maximum delocalization of the excitation with maximum degree of coherence. This situation has been defined by previous authors as *coherent delocalization* [45]. In contrast, a maximally delocalized excitation ( $D = 1$ ) with no coherence, the so-called *incoherent delocalization*, produces no entanglement. Finally, as one can naturally expect, a fully coherent system with maximum localization ( $D = 0$ ) will not exhibit

entanglement.

In the following sections, we will show that similar results can explain the presence or lack of entanglement in different scenarios. Even though in the cases that we will describe below there is not an actual excitation being shared by the subsystems, we will borrow this term from the present discussion and use it to describe physical operations that modify certain properties of a photon, i.e., its polarization or its orbital angular momentum content. In this way, we will be able to define “ground” and “excited” states of each subsystem, thus allowing us to demonstrate the mathematical equivalence of all the cases considered in this work.

## 2. Polarization entanglement in a two-photon state

The most convenient way to generate entanglement between two parties is by making use of the nonlinear process of spontaneous parametric down-conversion (SPDC), where an intense pump beam interacts with the atoms of a non-centrosymmetric second-order nonlinear crystal and mediates the generation of paired photons (signal and idler) that can be entangled in any of the degrees of freedom that define the parameter space of the photons [46]. Polarization entanglement is the most common type of photonic entanglement, widely used in many quantum computing and quantum information applications [47], mainly because of the ease with which it can be generated and manipulated.

In general, the density matrix of the two-photon system can be written in the same form as Eq. (1), with [46]

$$|\Psi\rangle = \alpha_1 |H\rangle_s |V\rangle_i + \alpha_2 |V\rangle_s |H\rangle_i, \quad (9)$$

$$I_D = |\alpha_1|^2 |H\rangle_s |V\rangle_i \langle H|_s \langle V|_i + |\alpha_2|^2 |V\rangle_s |H\rangle_i \langle V|_s \langle H|_i, \quad (10)$$

where  $|H\rangle$  and  $|V\rangle$  stand for horizontal and vertical polarization states, respectively, and  $s, i$  are the commonly used labels for the signal and idler photons. Here, the values of  $\alpha_{1,2}$  depend on specific polarization-dependent characteristics of the photon-generation process [46].

One can easily realize that the two-photon state defined by Eq. (9) is equivalent to considering a two-site state in the *single-excitation* manifold by identifying the corresponding “ground” and “excited” states of each photon (or subsystem). For this, we can take  $|V\rangle_{s,i}$  as the ground states and  $|H\rangle_{s,i}$  as the excited states, so we find that the state given by Eq. (9) lives in a Hilbert subspace where only one of the two photons can be in the excited state, that is, the single-excitation subspace.

The degree of coherence of this system can be written as

$$g_{H_s V_i; V_s H_i}^{(1)} = \frac{\text{tr}(\rho A_{H_s V_i}^\dagger A_{V_s H_i})}{\left[ \text{tr}(\rho A_{H_s V_i}^\dagger A_{H_s V_i}) \text{tr}(\rho A_{V_s H_i}^\dagger A_{V_s H_i}) \right]^{1/2}}, \quad (11)$$

where  $A_{H_s V_i}^\dagger = (a_{H_s}^\dagger a_{V_i}^\dagger)$  and  $A_{V_s H_i}^\dagger = (a_{V_s}^\dagger a_{H_i}^\dagger)$ , with  $a_{H_s, V_s}^\dagger$  and  $a_{V_i, H_i}^\dagger$  being the operators that create signal ( $s$ ) and idler ( $i$ ) photons with horizontal ( $H$ ) and vertical ( $V$ )

polarizations. Using Eqs. (1), (9) and (10), one obtains that the degree of coherence reads

$$\left|g_{H_s V_i; V_s H_i}^{(1)}\right| = \epsilon, \quad (12)$$

which is a result that one can anticipate from Eq. (5). On the other hand, it is easy to see that  $D = 2|\alpha_1||\alpha_2|$ .

Concurrence is again used for quantifying entanglement in this system, as well as Eq. (8) for the excitation's degree of localization. Notice that, in the present scenario, maximum delocalization ( $D = 1$ ) designates the case where pairs of photons with polarization  $|V\rangle_s |H\rangle_i$  are as likely to be generated as photons with polarization  $|H\rangle_s |V\rangle_i$ . Indeed, the same results as those discussed in the previous section can be obtained for the two-photon case, which means that measuring entanglement in this system is fully equivalent to measuring coherence.

Experimentally, the quantum state described by Eqs. (9) and (10) may be generated by using two second-order nonlinear crystals, where degenerate and collinear type-II SPDC can take place. The input pump beam is divided with the help of a beam splitter and illuminates both crystals. The probability of generating two pairs of photons, one pair in each crystal, is assumed to be negligible for sufficiently low values of the pumping power. Then, down-converted photons of each crystal are redirected to a polarizing beam splitter (PBS), where they enter through different input ports. In this way, in each output port of the PBS, horizontally and vertically polarized photons can be detected. The probabilities  $p_{1,2}$  that the pair of photons originates in each of the two crystals may be engineered in several ways. For instance, one can control the phase-matching conditions, or the amount of pump power, independently in each crystal, effectively varying  $p_1$  and  $p_2$ , and so  $D$ . In the case where all pairs of photons come from a single crystal one would obtain  $D = 0$ ; whereas in the case when the pumping power and phase-matching conditions are equal in both crystals, one would have  $D = 1$ . The coherence  $\epsilon$  can be controlled by introducing/removing delays between paired photons originating from different crystals, which effectively introduces/erases distinguishability between them [46].

Finally, it is important to remark that a two-photon entangled state could also be described by a state of the form  $|\Psi\rangle = \alpha_1 |V\rangle_s |V\rangle_i + \alpha_2 |H\rangle_s |H\rangle_i$ . Note that, in this case, the signal photon's polarization is rotated, which means that, in order to remain in the single-excitation manifold, its corresponding "excited" and "ground" states should rotate as well. Using these new states one can obtain the same results as the ones discussed above. Finally, we highlight that the fact that the density matrix of the two-photon system lies in the single-excitation subspace allows one to implement experimental setups, such as the one described in Ref. [48], in which the degree of entanglement between the two photons is controlled by directly modifying the off-diagonal terms of the system's density matrix, that is, the degree of coherence.

### 3. Spin-orbit entanglement in single photons

The spatial shape of photons, or its orbital angular momentum (OAM) content, is a degree of freedom that has received increasing attention in the last few years, because it has opened a new window, easily accessible experimentally, to explore high-dimensional quantum spaces encoded in single- or two-photon systems [49, 50].

Let us consider the case of a single-photon state in which the OAM and polarization degrees of freedom are used. It has been shown that it is possible to generate single-photon states in which the spatial shape and polarization degrees of freedom are effectively entangled [51, 10]. In this scenario, the quantum state of the photon would be described by the so-called single-photon spin-orbit state, whose density matrix has the same form as Eq. (1), with [52, 53]

$$|\Psi\rangle = \alpha_1 |H, -1\rangle + \alpha_2 |V, +1\rangle, \quad (13)$$

$$I_D = |\alpha_1|^2 |H, -1\rangle\langle H, -1| + |\alpha_2|^2 |V, +1\rangle\langle V, +1|. \quad (14)$$

Here, the integer  $\pm 1$  corresponds to the value of the OAM index ( $m = \pm 1$ ) of the photon.

Again, we can see that the single-photon spin-orbit state lies within the single-excitation manifold by identifying the “ground” and “excited” states for each subsystem. If we define  $|V\rangle$  and  $|-1\rangle$  as the ground states, and  $|H\rangle$  and  $|+1\rangle$  as the excited states for the polarization and OAM degrees of freedom, we can readily find that Eqs. (13) and (14) describe a state that is equivalent to a Hilbert subspace where only one “excitation” in any of the two degrees of freedom can exist, i.e., the single-excitation subspace.

Following the same procedure as in previous sections, we can quantify coherence in the single-photon system by writing the first order correlation function as

$$g_{H-1;V+1}^{(1)} = \frac{\text{tr}(\rho a_{H-1}^\dagger a_{V+1})}{[\text{tr}(\rho a_{H-1}^\dagger a_{H-1}) \text{tr}(\rho a_{V+1}^\dagger a_{V+1})]^{1/2}}, \quad (15)$$

where  $a_{jm}^\dagger$  is the operator that creates a photon with the polarization state  $j = H, V$  and OAM index  $m = \pm 1$ .

Using Eqs. (1), (13) and (14) we thus find that the degree of coherence of the single-photon system is given by

$$|g_{H-1;V+1}^{(1)}| = \epsilon. \quad (16)$$

Finally, for quantifying entanglement in this system, we can make use of the basis  $\{|H, +1\rangle, |H, -1\rangle, |V, +1\rangle, |V, -1\rangle\}$  to write the density matrix of the single-photon system, and find that it has the exact same form as the one described in Eq. (6). It is then straightforward to obtain that the concurrence for this state is  $C = 2\epsilon\sqrt{p_1 p_2}$ .

In experiments, the quantum state described by Eqs. (13) and (14) may be generated by making use of a single-crystal collinear type-II SPDC configuration. In this configuration, one of the photons is projected into different polarization states while the remaining photon traverses an optical device that correlates polarization with OAM [10].



We can control the values of  $p_{1,2}$ , and therefore  $D$ , by defining a proper polarization-state projection. For instance, by projecting one photon into the polarization state  $|H\rangle$ , the remaining photon would be in the state  $|V, -1\rangle$ , and therefore  $D = 0$ . Similarly,  $D = 0$  if we project the photon into the polarization state  $|V\rangle$ . Interestingly, if we project one photon into the state  $|H\rangle + |V\rangle$ , the remaining photon will be in a quantum superposition of both states, thus giving us a maximum value of delocalization,  $D = 1$ . Also, as discussed in the previous case, coherence can be controlled by introducing/removing delays between the generated pairs of photons.

Finally, from the results discussed above, we can conclude that measuring entanglement in a single-photon spin-orbit system is the same as measuring coherence. By identifying that the single-photon spin-orbit state lies within the single-excitation manifold, we can anticipate the existence of entanglement between the spin and OAM degrees of freedom, provided that coherence between them is preserved. This conclusion has been recently verified by experiments in which the degree of entanglement between different degrees of freedom of a single photon is controlled by properly tuning the degree of coherence ( $\epsilon$ ) [10].

#### 4. Conclusions

Entanglement seems to be a ubiquitous concept that, even though it was introduced to explain a very specific phenomenon of quantum theory, it can apply as well to many different scenarios. Here we have shown that indeed this should not be unexpected, because when considering correlations between different parties—namely photons, degrees of freedom or sites—in the important case of the single-excitation manifold, entanglement is equivalent to coherence or, more specifically, to coherent delocalization.

We have investigated the conditions for the existence, or lack, of entanglement in three different systems: a) the process of exciton transport in photosynthetic light-harvesting complexes, which is generally modeled as a single excitation propagating in an  $N$ -site network, b) the two-photon state generated by means of spontaneous parametric down-conversion in nonlinear crystals and c) the coupling between different degrees of freedom of a single photon. Our results show that even though the physical scenarios of all the cases considered here are different, their mathematical equivalence is what allows one to expect and observe entanglement in each one of them. Furthermore, we have seen that within the single-excitation Hilbert subspace any measure of entanglement is equivalent to a measure of the degree of coherence and localization. This implies that any system that may be described in a similar manner to the single-excitation manifold will exhibit entanglement as long as coherence and delocalization between its subsystems are preserved.

Finally, we have explored the reason why entanglement can even be observed in classical coherent systems [9, 11, 12]. The analysis presented here demonstrates that the observation of entanglement, even if the system can be described classically, should

not be unexpected because the concept of entanglement in the single-excitation manifold is essentially the same as coherence.

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