Probing exciton/exciton interactions with entangled photons: theory

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Quantum entangled photons provide a sensitive probe of many-body interactions and offer an unique experimental portal for quantifying many-body correlations in a material system. In this paper, we present a theoretical demonstration of how photon-photon entanglement can be generated via interactions between coupled qubits. Here we develop a model for the scattering of an entangled pair of photons from a molecular dimer. We develop a diagrammatic theory for the scattering matrix and show that one can correlate the von Neumann entropy of the outgoing bi-photon wave function to exciton exchange and repulsion interactions. We conclude by discussing possible experimental scenarios for realizing these ideas.

I. INTRODUCTION

As evidenced by recent theoretical and experimental advances, quantum entangled photons provide a sensitive measure of collective and many-body dynamics. The sensitivity stems from the “spooky action at a distance” nature of entangled photons, whereby measurement of one photon gives information about its entangled partner photon. This information can be extracted through either coincidence detection, interference, or quantum state reconstruction.

We recently presented a theoretical basis for how entanglement can be produced in 2-photon scattering from a system of coupled excitonic sites. In our approach, we assume that the bi-photon scattering matrix can be decomposed into a product of two single-photon terms and an irreducible two-photon term of the form

$$S^{(2)}(\omega_1, \omega_2; \omega'_1, \omega'_2) = S^{(1)}(\omega_1, \omega'_1)S^{(1)}(\omega_2, \omega'_2) \times e^{g(\omega_1, \omega_2; \omega'_1, \omega'_2)} \quad (1)$$

where $S^{(1)}$ gives the single photon (Raman or Rayleigh) scattering and $g(\omega_1, \omega_2; \omega'_1, \omega'_2)$ is an irreducible term that can be related to exciton/exciton cross-correlations. We suggest that by measuring the photon entanglement entropy change, one can deduce a direct measure of exciton/exciton cross-correlations. Here, we perform a theoretical analysis of the two-photon scattering produced by a simple two-qubit system, coupled by exchange interactions which allow a single excitation to be transferred between qubits and a direct interaction which introduces an energetic cost for double excitation. We show that the bi-photon scattering can be related to the cascade emission from the double-excited system, can be “tuned” by changing the nature of the exchange term.

Significant amounts of theoretical and experimental efforts have been invested to achieve photon pair polarization entanglement using photon cascades in semiconductor quantum dots. In that case, special conditions making indistinguishable two alternative emission passes via split intermediate states need to be satisfied. In contrast, below we study the energy/time photon entanglement generation which turns out to have much less restrictions to be achieved.

II. THEORETICAL MODEL

Our theoretical approach is to use the Feynman diagram technique to compute the time-integrated two-photon correlation intensity following either 2-photon scattering or 2-photon radiative cascade from a J- or H-aggregate dimer system. We assume the bi-exciton...
system can be described by
\[
H_{xx} = E_e (\sigma^+_1 \sigma^-_2 + \sigma^+_2 \sigma^-_1) + U (\sigma^+_1 + 1/2)(\sigma^-_2 + 1/2)
\] (2)

where by the first term corresponds to the uncoupled qubits, the second promotes exchange between qubits and the third introduces two-body interactions corresponding to the energy cost to add a second excitation to the system. Writing this in a and the third introduces two-body interactions corresponding to the energy cost to add a second excitation to the system. Writing this in a

\[
E_o = 2E_x + U \quad \text{biexciton} \quad (3a)
\]
\[
E_d = E_x - J \quad \text{dark} \quad (3b)
\]
\[
E_b = E_x + J \quad \text{bright} \quad (3c)
\]
\[
E_a = 0 \quad \text{ground} \quad (3d)
\]

Figure 1b gives a sketch of relative placement of the energy levels in this system. In the uncoupled system, an excitation can be placed in either the |10⟩ or |01⟩ state. The exchange interaction J splits these states into a symmetric bright state (|ψ⟩ = (|10⟩ + |01⟩)/√2) and an anti-symmetric dark state (|ψ⟩ = (|10⟩ − |01⟩)/√2) and the anharmonic interaction shifts the energy of the doubly excited (|ψ⟩ = |11⟩) U. We add to this the radiation field and coupling so that the full Hamiltonian becomes
\[
H = H_{xx} + H_r + H_I
\] (4)

Under the rotating-wave approximation \( H_I \) would include an exchange term between photons and excitons, and hence |ψ⟩ becomes “dark” and all of the photo-physics occurs between the ground-state and states |ψ⟩ and |ψ⟩ = |11⟩. For a J-aggregate such as anthracene, J < 0 such that the dark-state |ψ⟩ lies above the lower |ψ⟩ state. For H-aggregates, the reverse occurs and the “bright” state is higher in energy than the “dark” state. The coupling term J is mediated by dipole-dipole interactions between transition densities, the coupling itself is depends upon both the distance of separation between qubits as well as their relative orientation.

Since the states are embedded in the continuum of photon states, they acquire an energy shift and decay and we shall denote such states that are dressed by the radiation field as |ψ⟩ with energy \( E_n = E_n + \Delta_n + i\hbar \Gamma_n / 2 \). While the ground-state is also dressed by the radiation field, it is in fact stable with \( \Gamma_n = 0 \). Of the dressed-states, only the ground state is an eigenstate of \( H \). Generally, the energy shifts \( \Delta_n \) are very small and can be ignored. However, the \( \Gamma_n^{-1} \) are the radiative lifetimes of the various excited states. For ππ transitions in organic materials, radiative lifetimes are on the order of 10-100 fs corresponding to homogeneous line-widths of 40-400 meV.

These assumptions allow us to write the resolvents for when the system is a given state as
\[
G_n(z) = (\psi_n) \frac{1}{z - H} |\psi_n⟩ = \frac{1}{z - E_n + i\hbar \Gamma_n / 2}. \]

Also, we write the matrix element for radiative transitions between states as \( ⟨i|j⟩ = [a^+(k), H_I] \) and \( ⟨i|j⟩ = [a^+(k), H_I] \) where \( \tilde{μ}^+(k) = [a^+(k), H_I] \).

Having established the system, we first consider the two photon cascade decay of the upper most, doubly excited state. We shall assume that this state can be prepared by simply pumping the system to create a population of bi-excitons and that the resulting photon cascade can be detected via single-photon counting and coincidence.

A. Entanglement by Radiative Cascade

Before computing the full 2-photon-in → 2-photon-out scattering process, we first consider the cascaded radiative decay from the upper bi-exciton state. Referring to the left Feynman diagram in Fig. 1b, we prepare the system in state c and disregard the bottom half of the diagram. In essence, the two photon process preparing state c can be considered as the time-reverse of the 2-photon cascade. Thus, once we have an expression for the cascade, it is trivial to obtain the 2-photon scattering term. Since both b and c are unstable, they acquire a line-shape and the two photon decay from \( ψ_c → ψ_0 \) only needs to pass through the density of states around state b. We start from state |ψ⟩; 0⟩, that is the upper state with no free photons, and decay to the ground-state to produce 2 free photons |ψ⟩; 0⟩ = a(k) \( a^+(k) |ψ_0⟩ \). The amplitude for the |ψ⟩; 0⟩ → |ψ⟩; 0⟩ → |ψ⟩; 0⟩ transition is then given by

\[
G(z) = \mu_{0,b}(k_2) \mu_{b,c}(k_1) G_0(z - \hbar \omega_1 - \hbar \omega_2) G_0(z - \hbar \omega_1) G_c(z)
\]

\[
= \frac{\mu_{0,b}(k_2) \mu_{b,c}(k_1)}{(z - (\hbar \omega_1 + \hbar \omega_2 + E_0))(z - \hbar \omega_1 - E_b)(z - E_c)}.
\]

Given this, we can calculate the time-dependent amplitude
\[
U(τ) = \lim_{τ→∞} \frac{1}{2πi} \int_{-∞}^{∞} e^{iEτ/\hbar} G(E + i\eta) dE.
\]

From this we get the integrated intensity \( I(\omega_1, \omega_2) = |U(τ)|^2 \) as a symmetric function of the two frequencies.
In this case, the integrated intensity can be written as a purely separable function of the two frequencies.

Taking τ ≫ Γ−1 \( b \) & Γ−1 \( c \), only the pole around the ground-state contributes to the integral.

\[
U(\tau) = e^{i(E_0 + \hbar \omega_1 + \hbar \omega_2)\tau / h} \frac{\mu_{0b}^+ \mu_{bc}^+}{(E_0 - E_b + \hbar \omega_1 + \hbar \omega_2 + i\hbar \Gamma_b / 2)(E_0 - E_c + \hbar \omega_1 + \hbar \omega_2 + i\hbar \Gamma_c / 2)}. \tag{8}
\]

Furthermore, we need to include the amplitude corresponding to the case where the \( \omega_1 \) photon is emitted before the \( \omega_2 \) photon. Consequently, the full amplitude is given by

\[
U_{k_1, k_2}(\tau) = \frac{\mu_{0b}^+ \mu_{bc}^+ e^{i(E_0 + \hbar \omega_1 + \hbar \omega_2)\tau / h}}{\hbar \omega_1 + \hbar \omega_2 - \hbar \omega_c + i\hbar \Gamma} \left( \frac{\hbar \omega_1 + \hbar \omega_2 - 2\hbar \omega_b + i\hbar \Gamma}{(\hbar \omega_1 - \hbar \omega_b + i\hbar \Gamma / 2)(\hbar \omega_2 - \hbar \omega_b + i\hbar \Gamma / 2)} \right). \tag{9}
\]

Hence, one can write the entangled intensity as a normalized probability distribution

\[
I(\omega_1, \omega_2) = \lim_{\tau \to \infty} |U(\tau)|^2 \tag{11}
\]

In the special case where \( \omega_c = 2\omega_b \) which corresponds to the case when the intermediate level is exactly half way between level c and the ground state. For our model system, this occurs when \( J < 0 \) and \( U > 0 \) the condition that \( 2\omega_b = \omega_c \) can not be satisfied. For an H-aggregate, however, this condition can be satisfied over a range of both \( J \) and \( U \). Under this special case condition Eq. \( 10 \) can be further factored to

\[
U_{k_1, k_2}(\tau) = e^{i(E_0 + \hbar \omega_1 + \hbar \omega_2)\tau / h} \left( \frac{\mu_{0b}^+ \mu_{bc}^+}{(\hbar \omega_1 - \hbar \omega_b + i\hbar \Gamma / 2)(\hbar \omega_2 - \hbar \omega_b + i\hbar \Gamma / 2)} \right). \tag{12}
\]

In this case, the integrated intensity can be written as a purely separable function of the two frequencies.

\[
I(\omega_1, \omega_2) = \frac{2|\mu_{0b}|^2 |\mu_{bc}|^2}{\hbar^4} \frac{1}{(\omega_1 - \omega_b)^2 + \Gamma^2 / 4} \frac{1}{(\omega_2 - \omega_b)^2 + \Gamma^2 / 4}. \tag{13}
\]

In Figure 2(a-c) we show the integrated intensity \( I(\omega_1, \omega_2) \) for two-photon emission from a bi-exciton state.
in a model J-aggregate dimer system with $\hbar \omega_x = 1$ setting the energy scale and with $U$ and $J$ indicated on the plots. In each, we assume $\Gamma = 0.6$ which in consistent with a lifetime of about 6 fs for an exciton with $\hbar \omega_x = 1$ eV. The line shape is symmetric about the line $\omega_1 = \omega_2$ reflecting the fact that we summed over both photon paths. In the uncoupled case, $U = 0$ and $J = 0$, the distribution is clearly separable into two terms. Increasing the either the hopping term $J$ or the repulsion term $U$ leads to intensity distributions that are no longer separable into single photon terms.

B. Double photon scattering

We now consider the case depicted in the left-most Feynman diagram in Fig. 2. In this case, the two input photons place the system into the doubly-excited $|c\rangle = |11\rangle$ state and are re-emitted leaving the system back in its ground state: $|a\rangle \rightarrow |b\rangle \rightarrow |c\rangle \rightarrow |b'\rangle \rightarrow |a'\rangle$.

For this, we shall write the amplitude in terms of the Møller operators to propagate the initial state $|a, n_1, n_2\rangle$ from $t \rightarrow -\infty$ forward to some intermediate time $t$, where the system is in the $|c, n_1 - 1, n_2 - 1\rangle$ state then from $t \rightarrow +\infty$ to $|a, n_1', n_2'\rangle$. The Møller operator interwines the asymptotic (i.e. free) observables to those in the fully interacting theory. These are especially important considering the scattering of quantum photons since the atomic/material target is never fully free of the radiation field. The operators are defined by writing the interaction picture ket as

$$|\psi(t)\rangle_f = e^{iH_o t/\hbar} e^{-i(H_o + V)t/\hbar} |\psi\rangle$$

where $|\psi\rangle$ is the asymptotic state. Inverting this,

$$|\psi\rangle = e^{i(H_o + V)t/\hbar} e^{-iH_o t/\hbar} |\psi(t)\rangle_f.$$

Upon taking the limits of $t \rightarrow \pm \infty$, one defines the Møller operator

$$\Omega^{(\pm)} = \lim_{t \rightarrow \pm \infty} e^{i(H_o + V)t/\hbar} e^{-iH_o t/\hbar}. \tag{16}$$

Assuming we have two photons in the asymptotic states, the relevant states are

$$|\psi^- (k_1, k_2)\rangle = a^\dagger_{k_1} a^\dagger_{k_2} |a; 0\rangle \tag{17a}$$

$$|\psi^+ (k_1', k_2')\rangle = a^\dagger_{k_1'} a^\dagger_{k_2'} |a'; 0\rangle. \tag{17b}$$

Thus we transform the input state $|\psi^- (k_1, k_2)\rangle$ to the output state

$$|\psi^+ (k_1', k_2')\rangle = \Omega^{(-)}\Omega^{(+) |\psi^- (k_1, k_2)\rangle} \tag{18}$$

$$= S^{(2)} |\psi^- (k_1, k_2)\rangle \tag{19}$$

where $S$ is the scattering matrix. Since the initial and final atomic states will be the same ground state, energy and momentum conservation will require that $\hbar \omega_1 + \hbar \omega_2 = \hbar \omega_1' + \hbar \omega_2'$ and $k_1 + k_2 = k_1' + k_2'$. The final transition amplitude can now be deduced from Eq. 10 by forward propagating the input state and reverse propagating the final state to some intermediate time $\tau$ where the system is in $|c\rangle$.

$$S^{(2)} = \int_{-\infty}^{+\infty} d\tau U_{k_1'k_2'}^{\dagger} (\tau) U_{k_1k_2} (\tau) \tag{20}$$

Integrating over all intermediate times the energy conservation $\omega_1 + \omega_2 = \omega_1' + \omega_2'$, and finally one finds that

FIG. 2. Two-photon cascade emission intensity distributions as varied by exciton/exciton interaction terms, $U$, and $J$. (a) Non-interacting qubits. (b) H-aggregate ($U > 0$ & $J > 0$). (c)J-aggregate ($U > 0$ & $J < 0$).
\[ S^{(2)}(\omega_1, \omega_2; \omega_1', \omega_2') = \frac{\mu_\text{ab} \mu_\text{bc} \mu_\text{hc}^{\dagger}}{\hbar^2} \left( (\omega_1 + \omega_2 - 2\omega_b)^2 + \Gamma^2 \right) \left( (\omega_1 + \omega_2 - \omega_e)^2 + \Gamma^2 \right) \times \left( \frac{1}{(\omega_1 - \omega_b + i\Gamma/2)(\omega_2 - \omega_b + i\Gamma/2)(\omega_1' - \omega_b - i\Gamma/2)(\omega_2' - \omega_b - i\Gamma/2)} \right). \] (21)

Since the general form of the two-photon scattering matrix is identical in form to what we arrived at for the cascade (aside from a constant term), the resulting entanglement change reflects the cascade dynamics from state \( |c\rangle \).

In Fig. 3 we show the results for scattering an initial input bi-photon Fock state \( |\omega_1, \omega_2\rangle \) under various parametric conditions. In (A) and (B) we consider the case where only 1 of the input photons is resonant with the single-exciton transition \( (E_x / \hbar = 1) \) with the other photon being very much off-resonant \( \omega_2 = 2\omega_1 \). The value of the exciton transfer term \( J \) is deliberately chosen to be large to highlight the effect of resonant coupling between the two qubits. Clearly, choice of input states has a profound effect upon the outgoing state. However, even when one of the photons is off-resonance, the outgoing state is entangled due to photon-photon coupling introduced by interactions with the medium.

C. Entanglement Entropy Generation

The entropy \( S \) provides a useful metric for the entanglement carried by the outgoing photons. This can be determined by singular value decomposition of \( I(\omega_1, \omega_2) \) in which we write the 2-photon intensity

\[ I(\omega_1, \omega_2) = \sum_n r_n f_n(\omega_1) g_n(\omega_2) \] (22)

as a weighted sum over single-component terms determined by Singular Value Decomposition (SVD). Taking \( r_n \) to be normalized to unity,

\[ S = -\sum_n r_n \ln r_n. \] (23)

The functions \( f_n(z) \) and \( g_n(z) \) are orthogonal polynomials forming a complete basis.

In Fig. 4 we computed the entropy as function of both \( J \) and \( U \) over a wide parametric range. The upper half \( (J > 0; U > 0) \) corresponds to the situation for most \( H \)-aggregate systems. Here, the positions of the two middle energy levels are swapped and the (now) upper state is what carries the coupling to the radiation field. In this regime, we have the possibility for satisfying the \( 2\omega_b = \omega_c \) criterion for a fully separable two-photon emission spectrum. The lower half \( (J < 0; U > 0) \) is corresponds to the parametric range for \( J \)-aggregate systems. Here, because the \( 2\omega_b = \omega_c \) cannot be satisfied, the entanglements are higher. For comparison, we consider two systems with identical entanglement entropy indicated on Fig. 4 by the letters "B" and "C". corresponding to an \( H \)-aggregate (Fig. 2b) and a \( J \)-aggregate (Fig. 2c). Since the coupling terms are sensitive to packing and aggregation, it is should be possible to control and select the entanglement in the emitted photon state.

The right-most Feynman diagram in Fig. 1d corresponds to two successive Raman scattering processes. In this case, the two processes will be independent in the limit that the line-shape of state \( b \) is sufficiently broad. (cite our last paper) It is important to distinguish this process from the double-excitation process discussed above. Assuming the two \( a \rightarrow b \rightarrow a' \) process are uncorrelated, the integrated intensity is the product of individual Raman intensities. As we showed in our previous work, even if the second excitation occurs within the homogeneous lifetime \( \Gamma^{-1}_b \) of state \( b \) the two events will not produce entangled photons. However, in the limit of slow-modulation the two transition moments can be correlated giving rise to entanglement in the outgoing photon state.

III. DISCUSSION

In this paper we have show how to construct the transition matrix for 2-photon resonant scattering which produces entanglements within the out-going photon state. We show that such an entanglement can be connected to exciton/exciton interactions occurring via exchange coupling between sites and exciton/exciton repulsion within a binary qubit system that corresponds to a molecular dimer. Since these parameters are exquisitely linked to the local structure of the system and relative orientation of the transition dipoles on each monomer, it should be possible to manipulate the outgoing entanglement via external means.

While we present results for a simple excitonic dimer, the model and methods are easily extendable to systems with multiple excitonic sites and internal vibronic degrees of freedom. Experiments based upon these ideas may offer valuable insights into the correlated dynamics occurring within complex excitonic systems. Our current results are valid only in the limit of low temperature and where vibronic coupling can be ignored. The inclusion of finite temperature and vibronic dynamics will certainly muddle the waters by limiting the time-frame over which entanglements can be established. As discussed in our recent paper, entanglement between the outgoing photons is contingent upon the both strength of the interaction and the magnitude of environmental fluctuations. Our current efforts are to include both implicit and explicit
quantized vibronic modes into the bi-photon scattering model.

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FIG. 3. Two-photon scattering distributions: (A,B) $J$ and $H$ aggregate system with only 1 photon on-resonance ($\omega_2 = 2\omega_1$) and (C) $J$ aggregate with both input photons being on resonance.

FIG. 4. Entanglement entropy for model interacting binary qubit system as varied by the interaction parameters $U$ and $J$. Units are such that $E_x = 1$. The white line corresponds to the condition where $2J = U$ whereby the single excitation energy is exactly half the double-excitation energy.

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