Resonant Transfer of Excitons and Quantum Computation

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Resonant energy transfer mechanisms have been observed in the sensitized luminescence of solids, in quantum dots and in molecular nanostructures, and they also play a central role in light harvesting processes in photosynthetic organisms. We demonstrate that such mechanisms, together with the exciton-exciton binding energy shift typical of these nanostructures, can be used to perform universal quantum logic. In particular, we show how to generate controlled exciton entanglement and identify two different regimes of quantum behaviour.

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The Förster energy transfer was first studied in the context of the sensitized luminescence of solids, in which an excited sensitizer atom can transfer its excitation to a neighbouring acceptor atom, via an intermediate virtual photon. This mechanism is also responsible for photosynthetic energy processes in antenna complexes, biosystems (BSs) that harvest sunlight. More recently, interest has focussed on such a transfer in quantum dot (QD) nanostructures and within molecular systems (MSs). In this Letter we give a general scheme for quantum computation that can be implemented in different nanostructures (NSs) by exploiting the Förster and exciton-exciton interactions. Thus, methods for controlled generation of exciton entanglement that use both diagonal and off-diagonal interactions are given.

Consider two coupled generic NSs (scalability will be addressed later). We assume that the excitations of each NS are charge neutral (i.e., of an excitonic nature) and that they can be produced by optical means. We also assume that tunnelling processes between them may be neglected, but that there is a strong exciton-exciton electromagnetic coupling. Our two-level—qubit system is represented in each NS by a single low-lying exciton (qubit state |1⟩) and the ground state (qubit state |0⟩). Then the interaction Hamiltonian can be written in the computational basis { |00⟩, |01⟩, |10⟩, |11⟩}, with the first digit referring to NS I and the second to NS II (as follows):

\[
\hat{H} = \begin{pmatrix}
\omega_0 & 0 & 0 & 0 \\
0 & \omega_0 + \omega_2 & V_F & 0 \\
0 & V_F & \omega_0 + \omega_1 & 0 \\
0 & 0 & 0 & \omega_0 + \omega_1 + \omega_2 + V_{XX}
\end{pmatrix}
\]

(1)

where the diagonal interaction \(V_{XX}\) is the direct Coulomb binding energy between the two excitons, one located on each NS, and the off-diagonal \(V_F\) denotes the Coulomb exchange (Förster) interaction which induces the transfer of an exciton from one NS to the other. These are the only Coulomb interaction terms which act between the qubits. \(\omega_0\) denotes the ground state energy and we define \(\Delta_0 \equiv \omega_1 - \omega_2\) as the difference between the exciton creation energy for NS I (\(\omega_1\)) and that for NS II (\(\omega_2\)).

Energy contributions due to spin singlet-triplet splittings do not significantly affect the present gating scheme and such effects are dealt with elsewhere.

The eigenenergies and eigenstates of the interacting qubit system are \(E_{00} = \omega_0\), \(E_{01} = \omega_0 + \omega_1 - \frac{\Delta_0}{2}(1 - A)\), \(E_{10} = \omega_0 + \omega_1 - \frac{\Delta_0}{2}(1 + A)\), \(E_{11} = \omega_0 + \omega_1 + \omega_2 + V_{XX}\); and \(|\Psi_{00}\rangle = |00\rangle\), \(|\Psi_{01}\rangle = c_1 |01\rangle + c_2 |01\rangle\), \(|\Psi_{10}\rangle = c_2 |10\rangle - c_1 |01\rangle\), \(|\Psi_{11}\rangle = |11\rangle\), where \(A = \sqrt{1+4(V_F/\Delta_0)^2}\), \(c_1 = V_F/\Delta_0 \approx 1\) and \(c_2 = (A-1)/2A\). The eigenenergies in the absence and presence of interqubit interactions are displayed in Figs. (a) and (b) respectively. Fig. (c) shows \(c_1\) and \(c_2\) as a function of the ratio \(V_F/\Delta_0\).

Single qubit operations can be achieved by inducing Rabi oscillations in the excitonic system (e.g., see Ref. [8]). The \(V_{XX}\) and \(V_F\) interactions lead to two regimes for achieving quantum entanglement. First, if the ratio \(V_F/\Delta_0 \approx 1\), then after selectively exciting NS I and creating \(|10\rangle\), the system will naturally evolve into one of the maximally entangled Bell states \(\frac{1}{\sqrt{2}}(|00\rangle \pm |11\rangle)\) and this evolution could then be stopped by suppressing the Förster coupling (see later). Second, if \(V_F/\Delta_0 \ll 1\), the computational basis states are essentially the eigenstates of the system, and we can exploit the \(V_{XX}\) term, together with appropriately tuned laser pulses, to perform two-qubit logic gates and hence to generate controlled entanglement. From Fig. (b) we can see how to implement the cnot gate: e.g., \(\text{CNOT}_{12}(|1\rangle |0\rangle \rightarrow |1\rangle |1\rangle\) can be achieved by illuminating the qubit system in the state \(|10\rangle\) with a \(\pi\)-pulse of energy \(\epsilon_{12}\). If we start in the ground state and first apply a \(\pi/2\) or \(3\pi/2\) pulse at energy \(\omega_1\), we create the states \(\frac{1}{\sqrt{2}}(|00\rangle \pm |11\rangle)\); if we now apply a \(\pi/12\) pulse, we generate the maximally entangled states \(\frac{1}{\sqrt{2}}(|00\rangle \pm |11\rangle)\).

In order to illustrate our ideas, we shall first calculate the strength of the interactions in a coupled QD molecule (in a previous study of QD excitons for quantum computing [9], the off-diagonal interaction terms were neglected). We start with the direct Coulomb interaction, \(V_{XX}\). Different dot geometries (e.g. spherical, pyramidal,
cuboidal) can be used to implement the scheme. In this Letter we assume that the dots are square-based cuboids and that the potential energy \( V \) of both electrons and holes increases abruptly at the cube boundaries where the semiconductor bandgap changes, and \( V = 0 \) inside the cubes (see Fig. 2(a)). This type of potential has the advantage of both a well defined dot size in all three dimensions, and of bound and unbound solutions in each direction (this is in contrast with the parabolic potential considered in Ref. [9]). The wavefunctions for single particles may be expressed in the envelope function approximation as \( \psi_r(r) = \phi_p(r) U_p(r) \) \( | \rangle \), where \( \phi_p(r) \) is an envelope function describing the changing wavefunction amplitude of confined states for particle type \( p \) over the dot region, and \( U_p(r) \) is the Bloch function which has the periodicity of the atomic lattice. In the effective mass approximation, the envelope functions are solutions of the single particle Schrödinger equation for the potential \( V_p \), and for the effective mass \( m^*_p \) of particle \( p \) (see Fig. 2(a)). These solutions may be obtained by expanding the Hamiltonian in a set of envelope basis functions of the form \( \Omega(r) = \xi_x(x) \xi_y(y) \xi_z(z) \), (the \( \xi_i(i) \) are the solutions of a one-dimensional square well potential) and then diagonalizing \( | \rangle \).

We now combine the ground states of electrons and holes into a Slater determinant type wavefunction which represents one exciton on each dot:

\[
\Psi_{XX} = A \{ \psi^{\dagger}_e(r_1) \psi^{\dagger}_e(r_2) \psi^{\dagger}_h(r_3 - R) \psi^{\dagger}_h(r_4 - R) \} ,
\]

where \( A \) indicates that the wavefunction has overall anti-symmetry, \( R \) is the vector connecting the two dot centres, \( r_1 \) and \( r_3 \) represent the position vectors of electrons relative to the centres of dot I and dot II respectively and \( r_2 \) and \( r_4 \) are the equivalent vectors for holes. The expansion of the associated Coulomb operator \( V_{XX} \) in a Taylor series around \( R \) to lowest non-zero order yields

\[
\hat{V}_{XX} = \frac{k}{\epsilon_r R^3} \left\{ \mathbf{p}_I \cdot \mathbf{p}_II - \frac{3}{R^2} (\mathbf{p}_I \cdot \mathbf{R}) (\mathbf{p}_II \cdot \mathbf{R}) \right\} ,
\]

where \( k \equiv \frac{1}{4 \pi \epsilon_0} \), \( \epsilon_r \) denotes the dielectric constant of the medium (\( \epsilon_r = 10 \) throughout our discussion), \( \mathbf{p}_I = e (r_1 - r_2) \), and \( \mathbf{p}_II = e (r_3 - r_4) \) are the overall dipole moments on dot I and II respectively. To evaluate the matrix element \( \langle \Psi_{XX} | \hat{V}_{XX} | \Psi_{XX} \rangle \), \( \mathbf{p}_I \) and \( \mathbf{p}_II \) in Eq. 3 are replaced by their expectation values for the wavefunction, Eq. 2. This procedure gives rise to a direct term and exchange terms. The exchange terms are zero in the absence of wavefunction overlap between dots. The direct term is obtained through the use of the envelope function approximation which leads to the expectation value \( \langle r_1 \rangle = \int \phi^*_e(r_1) \phi_e(r_1) \mathbf{R} dr_1 \), and similar expressions hold for the other position expectation values. For a cuboidal dot, where the electron and hole wavefunctions have a definite parity about the dot centre, Eq. 3 implies that the exciton-exciton coupling is zero. However, when this symmetry is broken, the electron and hole localize in different parts of the dot and the dipole moment is non zero. This occurs, for instance, in pyramidal dots or when an electric field is present.

We have simulated the effect of applying an electric field (along \( x \)) in our model and the results are displayed in Fig. 2(b), where the exciton dipole moment \( p_i \) is plotted as a function of the dot size and the field strength \( E \). Two cases are considered: the first is that of a cubic dot (\( a = h/2 \)) and the second is that of a flat cuboidal dot (\( a = 5h \)). \( V_{XX} \) is obtained by using the size of \( p_i \) for each dot, and Fig. 2(c) shows \( V_{XX} \) (normalized by \( R^{-3} \)) calculated for both geometries, with \( a = b \). It is interesting that the size of the induced dipole depends strongly only on the length, \( a \), of the dot in the direction of the applied field, and at large field is given approximately by \( p_i \approx ea; \) this limit should be valid for any NS (such as the BSs and MSs described later) with a well defined length-scale, and not just for QDs. The cuboidal structure is more typical of Stranski-Krstanow self assembled dots, where typically \( R = 5 \) nm, \( a = 10 \) nm, and \( b = 8 \) nm, and \( h_1 = h_2 = 2 \) nm. In a field of 100 kV/cm, these parameters give \( V_{XX} = 120 \) meV, which would result in a lower time limit for the gate operation of around 10 fs. This is relatively short; decoherence times on the order of nanoseconds have been observed for uncoupled dots.

Let us now consider the Förster (off-diagonal) coupling \( V_F \) in QDs, which may be expressed as a matrix element of the Coulomb operator between excitons located on each of the two dots. By Taylor-expanding this expression around \( R \) to lowest non-zero order, we obtain

\[
V_F = \frac{k e^2}{\epsilon_r R^3} | (r_I) \cdot (r_{II}) - \frac{1}{R^3} ( (r_I) \cdot (R) ( (r_{II}) \cdot (R) ) ) |
\]

Here \( e(r_i) \) represents the matrix element of the position operator between an electron and a hole state on dot \( i \). The expression is therefore equivalent to the interaction of two point dipoles, one situated on each dot. However, its magnitude is quite different from \( V_{XX} \). By again employing the envelope function approximation, we can rewrite the above equation as

\[
V_F = \frac{k e^2}{\epsilon_r R^3} O_i O_{II} \left( \langle r_I^e \rangle \cdot \langle r_{II}^h \rangle - \frac{3}{R^2} ( \langle r_I^e \rangle \cdot R ) ( \langle r_{II}^h \rangle \cdot R ) \right)
\]

where \( \langle r_i^p \rangle = \int \phi_i^p(r) \mathbf{r} U_i^p(r) dr \) is the interband position matrix element for the atomic part of the wavefunction for dot \( i \), and \( O_i = \int \phi_i^e(r) \phi_i^h(r) dr \) is the overlap of electron and hole envelope functions on dot \( i \). Thus, the effects of the QD size and shape (which determine the overlap integrals) are separated from the effects of the material composition of the dot (which determine the interband position operator). For other, smaller, NSs, the envelope function approximation does not apply. However, in these cases the above analysis is still valid if the overlap integrals are set to unity, and if the NS’s entire wavefunction is represented by the atomic basis part of the general wavefunctions described above.

We now introduce a simple Kronig-Penney model to
describe the atomic basis, and we assume that the infinite Kronig-Penney quantum boxes have a width of 2\(r\). This gives \((r^2) = 32x/9\pi^2\), or \(V_F \propto x^2\), and so we plot \(V_F/x^2\) as a function of \(R\) in Fig. 3(a). The simple Kronig-Penney model shows how the size of the Förster transfer depends upon the physical size of the atomic part of the wavefunction. \((r^1)\) is a widely measured quantity since it determines the strength of dipole allowed transitions in optical spectra. In CdSe QDs it can be in the range of 0.9 to 5.2 e\(\AA\), in atomic systems it can also be several e\(\AA\) \(\frac{1}{\pi}\) and in BSs and MSs has recently been observed to be about 1.7 e\(\AA\). The solid line in Fig. 3(a) represents the case when the overlap integrals are set to unity; the symbols represent simulations which take higher order terms into account and have \(a=b=2\) nm. Fig. 3(b) shows how \(V_F\) varies as a function of dot size and confinement potential through the \(O_i\)'s of Eq. 1. The envelope functions are as described in the previous section, except that no electric field term has been included in the envelope Hamiltonian. Thus the values of \(V_F\) should be regarded as an upper limit for the cuboid dot model: applying a field will serve to reduce \(V_F\) since it decreases the \(O_i\)'s; hence it could be used to tune \(V_F\). The overlap is enhanced when there is a larger confinement potential and for larger dots, since in these cases the shape of the wavefunction is less sensitive to the effective mass of the particle. As an illustration of the use of these curves, let us assume that we have a dot system in which, as before, \(R = 5\) nm, \(a = 10\) nm, \(b = 8\) nm, and \(h_1 = h_2 = 2\) nm. Furthermore, let us take the measured dipole value for CdSe dots of 0.9 to 5.2 e\(\AA\). In this case, the Förster strength is between 0.02 and 0.6 meV, which if \(\Delta_0 = 0\) would correspond to an on resonance energy transfer rate of between 206.8 and 6.9 ps. This is short enough to be useful for QIP: decoherence times as long as a few ns \(\frac{1}{\pi}\) have been observed in QDs. In MSs or BSs, the interacting units can be as close together as 1 nm: using this and taking a typical molecular or biomolecular dipole value of about 1.7 e\(\AA\), we obtain an interaction strength of 8.3 meV (or a transfer time of 497 fs). Furthermore, \(V_F\) must certainly be controlled if the alternative scheme using \(V_{XX}\) is to be implemented (and therefore cannot be neglected as in Ref. 6).

By also using the model outlined above to calculate \(\Delta_0\) in QDs (shown in Fig. 4(a) as a function of dot size ratio for quantum cubes) we can compute the size of the \(c_1\) component of the \(|\Psi_{10}\rangle\) and \(|\Psi_{01}\rangle\) states. This is shown in Fig. 4(b), where the dependence on atomic dipole and dot separation are incorporated by multiplying \(c_1\) by \(R^2x^{-2}\). It can be seen there that a range of \(c_1\) values can be obtained by choosing dots with appropriate values of \(x\), \(R\) and \(a/b\). Dots with large \(x\) (> 1 nm say), small \(R\) (< 3 nm say) and \(a/b \sim 1\) give a larger \(c_1\), and it is then more appropriate to use the Förster interaction itself to create entangled states. On the other hand, dots with smaller \(x\), larger \(R\) or a large mismatch in dot size would be more suited to the scheme which uses the \(V_{XX}\) for QC and entanglement generation. The fidelity of a typical operation (e.g. \(|11\rangle \rightarrow |10\rangle\)) in this case is equal to \(1 - \epsilon_1^2\)—and so one must be careful when using the biexciton scheme to use the available parameter space and make sure that the Förster transfer is suppressed to the desired accuracy. There are other sources of decoherence in this case (e.g. the interaction with optical and acoustic phonons \(\frac{1}{\pi}\) which will reduce the value of the fidelity to below \(1 - \epsilon_1^2\). To minimize the effects of such decoherence channels, it is important to maximize \(V_{XX}\), since this leads to an improved transition discrimination and so to a faster gating time. This can be done by applying an electric field and choosing an appropriate dot shape, size and separation (as described earlier). It is then necessary to minimize the basis state mixing for the chosen parameters by selecting a suitable dot size ratio and material composition.

Single shot qubit state measurement in QDs could be performed by using resonant fluorescent shelving techniques \(\frac{1}{\pi}\). The QD state measurement can also be achieved by means of projecting onto the computational basis and measuring the final register state by exploiting ultrafast near-field optical spectroscopy and microscopy \(\frac{1}{\pi}\): these allow one to address, to excite and to probe the QD excitonic states with spectral and spatial selectivity. In addition, the qubit register density matrix can be reconstructed by measuring the QD photon correlations via standard quantum state tomography techniques \(\frac{1}{\pi}\). Scalability of the scheme given here could also be possible by adopting a globally addressed qubit strategy \(\frac{1}{\pi}\) on a stack of self-organized QDs \(\frac{1}{\pi}\).

Light-harvesting antenna complexes \(\frac{1}{\pi}\) or arrays of strongly interacting individual molecules \(\frac{1}{\pi}\) could provide an excellent system in which the Förster interaction could be used for QIP tasks. They are generally very uniform structures, and we may compare them to QDs by setting \(a/b \sim 1\), or \(V_F/\Delta_0 \gg 1\). Then the one-exciton eigenstates of a two qubit system with a Förster coupling naturally allows the generation of the states \(|\Psi\rangle = (|10\rangle \pm |01\rangle)/\sqrt{2}\), which, apart from their applications to quantum protocols, can be particularly useful in the fight against decoherence. Spectroscopic, line-narrowing techniques (e.g., hole burning and site-selective fluorescence), infrared and Raman experimental studies reveal that the main decoherence mechanisms in the antenna complexes arise from energetic disorder, electron-phonon coupling, and temperature effects \(\frac{1}{\pi}\). In this scenario, the excitations couple to an environment that typically possesses a much larger coherence length than the biomolecular units (BChl’s) spacing. For example, the BChl’s in the antenna complex LH2, which we consider as potential qubits, are spaced by as little as 1 nm, and hence so-called collective decoherence is expected to apply. In this case, provided that the logical qubit encoding \(|j\rangle_i \equiv |01\rangle_{jk}\) and \(|\bar{j}\rangle_i \equiv |10\rangle_{jk}\) that uses two physical (exciton) qubits can be realized
in the BChl’s system, arbitrary superpositions of logical qubits such as \( (\alpha_i |\downarrow\rangle_i + \beta_i |\uparrow\rangle_i)^\otimes N \), \( i = 1, \ldots, N \), \( \alpha_i, \beta_i \in \mathbb{C} \), are immune to dephasing noise (described by a \( \sigma_z \) operator \([19]\)), and single qubit manipulations can be carried out on the timescale of the Förster coupling (which as we have seen can be as short as 497 fs). Two-qubit logic gates can also be implemented within a decoherence-free subspace by using the above encoding, thus completing a universal set of gates \([19]\). Initialization of the system requires the pairing of the physical qubits to the logical ‘ground’ state \( |\downarrow\rangle^\otimes N \), and readout is to be accomplished by identifying on which of the two structures the exciton is. Furthermore, rings of BChl’s appear side by side in naturally occurring antenna complexes and also display energy selectivity—smaller rings tend to have higher energy transitions \([3]\). Thus, following a scheme as above, it may be possible to scale up such biological qubits in a natural way and construct a robust energy selective scheme for quantum computation.

In conclusion, we have provided a general scheme for quantum computation and quantum entanglement generation that can be implemented in different NSs by exploiting the Förster and exciton-exciton interactions. In particular, we have shown how such interactions can be manipulated in molecular, biomolecular and QD nanostructures in order to produce an accurate degree of control for quantum logic.

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FIG. 1: A schematic diagram of the proposed quantum logic gate scenario: energy levels in (a) the absence and (b) the presence of the interactions $V_{XX}$ and $V_F$ for nanostructures of different excitation frequency. $\epsilon_{12} \equiv \omega_2 + V_{XX} - \delta$, $\epsilon_{21} \equiv \omega_1 + V_{XX} + \delta$, $\delta \equiv V_F^2/\Delta_0$; (c) dependence of the eigenstate coefficients $c_i$ as a function of $V_F/\Delta_0$. The inset shows the eigenenergies $E_{01}$ and $E_{10}$ of plot (b) for $\omega_1/\Delta_0 \equiv 20$.

FIG. 2: (a) Schematic diagram of the cuboidal dot model. The potential inside the cuboids is set to zero, that outside is determined by the band offsets of the conduction and valence bands within the heterostructure. (b) Exciton dipole moment as a function of the dot size and applied electric field for two dot shapes. The dot parameters $m_e = 0.06$, $m_h = 0.6$, $V_e = V_h = 500$ meV. (c) Exciton-exciton binding energy for $a = b$ and sequence of dot shapes, size and field strength as in (b).

FIG. 3: (a) Dependence of the Förster interaction strength on the interdot separation. The solid line represents the case where $O_i = 1$ in the dipole-dipole approximation and the atomic dipole operator is given by $32x/9\pi^2$. The symbols result from a full numerical simulation for $a = b = 2$ nm, and the dotted lines are the dipole-dipole predictions in these cases. (b) The $O_i$ factor appearing in Eq. 4 as a function of dot size and confinement potential. $m_e = 0.06 m_0$, $m_h = 0.6 m_0$. A lower cut-off occurs when the ground state of the dot is no longer a bound state.

FIG. 4: (a) Energy splitting $\Delta_0$ of the qubit exciton states $|\Psi_{01}\rangle$, and $|\Psi_{10}\rangle$ in the absence of the Förster interaction as a function of the different dots sizes $a$ and $b$. The splitting is independent of interdot distance. (b) The size of the mixing component of the wavefunction, $c_1$, as a function of the dot size ratio. $c_1$ has been scaled by its dependence on the interdot distance, $R$, and typical atomic spacing, $x$. 
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