Tunneling and optical control in quantum ring molecules

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The ability to manipulate the coupling between quantum states in a controllable way has become a major goal in the research of semiconductor-based quantum computation devices. We explore theoretically the use of orbital degrees of freedom to control quantum tunneling in a coupled quantum ring system of two vertically stacked rings with different radii. An external magnetic field can tune excitonic states to have distinct angular momentum ground states, thus blocking recombination processes. Controlled couplings between Stark-shifted direct and indirect exciton states appear as anticrossings in the optical absorption spectrum, allowing the control of the optical activity of the system and possible exciton entanglement.

I. INTRODUCTION

Since the pioneering proposals of using semiconductor quantum dots as solid state realizations of qubits,1 coherent control of excitons in individual quantum dots has been reported by several groups,2 while coherent oscillations of electron qubits have been beautifully demonstrated in recent experiments.3 Moreover, as basic quantum gate operations require controllable 2 qubit setups, coupled quantum dots have been proposed as scalable implementations. Several ideas for coupled electron spin operations4 as well as excitons5 and biexcitons6 have been put forward. Optical control of tunneling in multiple dot devices has also been proposed,7 enhancing the flexibility of dot structures.

Great attention has been given to the detection of quantum tunneling coupling between quantum dots in optical systems. Recent experiments report manifestation of this coupling using different techniques, such as signatures in the optical emission8–10 and photon correlation measurements.11 Charge tunneling provides strong coupling between Stark-shifted exciton states, which leads to anticrossings between direct and indirect exciton photoluminescence lines as the applied electric field is varied.9,10 However, the strength of the tunneling coupling is not easily controllable in situ in typical quantum dots, as it is defined by the overlap of the carrier wave functions frozen in the structure at growth.

In this work, we consider a mechanism to achieve tunable coupling control by considering excitons confined in coupled quantum ring structures. The electron tunneling rate between direct and indirect exciton states can be controlled in quantum ring molecules (QRMs) by tuning an external electric and/or magnetic field. This control arises from the suppression of electron tunneling between states with distinct angular momentum. Moreover, quantum rings have a unique magnetic field level dispersion: unlike quantum dots, the ground-state total angular momentum L in quantum rings changes from L=0 to L≠0 by the application of a moderate external magnetic field B.16–21 The values of B for which these transitions take place depend on the flux threading each ring, resulting in a different energy dispersion for excitons in QRMs with different ring radii. Thus, charge tunneling between states with distinct angular momentum is strongly suppressed by orbital selection rules. We emphasize that this feature makes QRMs unique, as they offer the interesting ability to control the effective coupling of direct-indirect excitons by changing experimentally controlled external parameters.

Our theoretical ideas are explored on realistic structures. The formation and optical characterization of self-assembled double quantum semiconductor rings has been reported using Stranski-Krastanow growth12 and droplet-epitaxial techniques.13 Theoretical studies of the electronic properties for concentric planar14 and vertically coupled15 double ring systems have also been published. Our results indicate that the system can be tuned as to allow the desired coupling of direct-indirect exciton states and that this can be attained in current ring samples,12 bringing an experimental verification of our results within reach.

II. MODEL

A quantum ring molecule consisting of two vertically coupled quantum rings of different radii, in general, is shown in Fig. 1. For completeness, we consider different radial confinements for holes and electrons, which allows for the creation of excitons with a net charge polarization in the radial direction within each ring. In single rings, this radial polarization couples to an external magnetic field, producing distinct Aharonov-Bohm (AB) phases for electrons and holes with observable consequences in their optical properties, the “optical AB effect.”17–21

The essential physics of these orbital effects can be captured by a model in which electrons and holes are restricted to rings with only one transverse channel (strong radial confinement) of radii R_e and R_h, respectively.19–21 In this geometry, electron (hole) single-particle states are labeled by the respective angular momentum number l_e (l_h). For a single ring, the exciton Hamiltonian can be written in this basis as

$$H_{\text{ring}} = \sum_{l} \varepsilon_{l} a_{l}^\dagger a_{l} + \varepsilon_{\beta} b_{\beta}^\dagger b_{\beta} - \sum_{l,j} \Delta_{l,j} a_{l}^\dagger b_{j}^\dagger a_{j},$$

where $a_{l}$ ($b_{\beta}$) annihilates an electron (hole) with integer-valued angular momentum $l$, $\varepsilon_{l} = [\hbar^{2}/(2m_{e}R_{e}^{2})]l(l+1)$, $\Delta_{l,j} = q_{l,j}e^{-Fz_{\beta}} (q_{\mu} = \pm 1, \mu = e, h)$ is the quantization energy for each ring, $q_{\mu} = \pi R_{\mu}^{2}B/\phi_{0}$ is the magnetic flux for the electron
rated by a distance quantum ring molecule produces a Stark shift in the z profile in the growth direction. Hole is localized on the upper particle energy. Diagonalization of Eq. states with total angular momentum L is classified as either spatially states obtained from the procedure outlined above can be localized in ring 1 or ring 2, while electron tunnels between rings. (d) Different radial confinement potentials for electrons and holes on each ring induce a radial polarization of the exciton, allowing for a net Aharonov-Bohm phase in the neutral exciton.

(hole) in units of \( \phi_0 = \hbar |\epsilon| / c, B \) is the magnetic field, and \( \nu_\phi \) is the Coulomb interaction term. An electric field \( F \) applied in the z direction (perpendicular to the plane of the rings) produces a Stark shift \( \pm eF \alpha \) in the electron (hole) single-particle energy. Diagonalization of Eq. (1) yields exciton states with total angular momentum \( L = l_\text{e} + l_\text{h} \) and energies \( E_i(B,F) \).

In a system of two uncoupled quantum rings, exciton states obtained from the procedure outlined above can be classified as either spatially direct [both electron and hole are localized in ring 1 (\( D_1 \)) or ring 2 (\( D_2 \))] or indirect [electron in ring 1 and hole in ring 2 (\( I_{12} \)) or electron in ring 2 and hole in ring 1 (\( I_{21} \))]. In the quantum ring molecule, however, there is a finite overlap of the electron wave functions on the top and bottom rings, giving rise to electronic tunneling between the rings, thus coupling direct and indirect excitons (we neglect the much weaker hole tunneling processes).

In this approximation, the exciton Hamiltonian reads \( H = H_{\text{ext}} + H_{\text{tunn}} + H_{\text{light}} \), where \( H_{\text{ext}} \) is the double ring Hamiltonian, \( H_{\text{tunn}} \) accounts for the electron tunneling between rings, and \( H_{\text{light}} \) describes the dipole coupling to the radiation:

\[
H_{\text{ext}} = \sum_{i,C} \epsilon_i^C(B,F) |i^C\rangle \langle i^C|, \]

\[
H_{\text{tunn}} = t_e \sum_{i,j} \delta_i j |D_1\rangle \langle J_2^i| + |D_2^j\rangle \langle J_2^j| + \text{H.c.}, \]

\[
H_{\text{light}} = - \sum_i \mu_{i,0} \delta_{i,0} |D_1\rangle \langle 0| + |D_2\rangle \langle 0| + \text{H.c.}, \]

where \( C (=D_1,D_2,I_{12},I_{21}) \) labels direct and indirect neutral exciton states \( |i\rangle \) (with total angular momentum \( L_i \)) as described above, \( |0\rangle \) is the vacuum state (no excitation in the system), \( t_e \) is the tunneling coupling, and \( \mu_{i,0} = d_{i,0} E(t) \) are the interband matrix elements, with \( E(t) = E_0 \cos \omega t \) the electric field of incident light and \( d_{i,0} (=d) \) the dipole matrix element in the field direction. We neglect the much weaker dipole matrix elements for indirect states as the electron-hole overlap is small. Note also that \( H_{\text{tunn}} \) couples excitons of type \( D_\alpha \) with indirect ones of type \( I_{\alpha'\alpha} (\alpha, \alpha' = 1, 2) \) with the same total angular momentum \( L_i \).

The linear optical absorption for the double ring system is a function of the incident light frequency \( \omega \), the magnetic field \( B \), and the electric field \( F \), being proportional to the imaginary part of the optical polarization \( P(\omega) = \Sigma d_{i,0} (\epsilon_i^C |\vec{p}| \langle 0 | = \Sigma d_{i,0} \rho_i (\vec{p} \) is the density operator). Here, \( \epsilon_i \) is the background dielectric constant. Note that resonances in \( \alpha(\omega,B,F) \) occur at frequency values corresponding to direct exciton eigenstates directly coupled to the radiation field (dipole active). Since the photon angular momentum is taken up by the interband transition matrix elements, angular momentum selection rules ensure that the dipole-active exciton states have zero orbital angular momentum \( (L_i = 0) \).

In the rotating wave approximation, the equation of motion for the density operator yields a set of coupled equations for the frequency-dependent \( \rho_i(\omega) \). Keeping linear terms only, one gets

\[
[\Omega + i \gamma - \epsilon_i^C] \rho_i(\omega) = t_e \sum_j (\Delta_{ij} + \Delta_{ji}) \rho_j(\omega)
\]

\[
= - \mu \delta_{i,0} (\delta_{i,D_1} \delta_{j,J_2^1} + \delta_{i,D_2} \delta_{j,J_2^2}),
\]

where \( \Omega = \omega - E_{\text{light}} \) is the detuning of the incident light from the band edge, \( E_{\text{light}} \) is the optical band gap, \( \gamma \) is the homogeneous broadening, and \( \Delta_{ij} = \delta_{i,D_1} \delta_{j,J_2^1} + \delta_{i,D_2} \delta_{j,J_2^2} \) enforces that only exciton states of classes \( D_i \) and \( I_{ji} \) are coupled by the tunneling term.

### III. RESULTS

The system of coupled equations (4) determines the absorption coefficient. We are interested in the low-lying optically active states which show as resonances in the optical absorption. For concreteness, we consider quantum rings of radii \( R_{12} = 16 \text{ nm} \) and \( R_{13} = 20 \text{ nm} \) (large ring) and \( R_{12} = 10 \text{ nm} \) and \( R_{13} = 12 \text{ nm} \) (smaller ring). These radii are within the range in typical samples. A determination of the tunneling constant \( t_e \) depends crucially on the inter-ring distance \( d \) and on the details of the structure. Such a calculation is beyond the scope of the present work. Instead, we chose a phenomenological approach: \( t_e \) and \( d \) are chosen so that \( 2t_e = |e| d \Delta F \), where \( \Delta F \) is the electric field variation necessary to resolve an antiercrossing between direct and indirect states, in agreement with the observed values in coupled quantum dot samples.

We calculate the absorption coefficient \( \alpha(\omega,B,F) \) for \( \gamma = 0.05 \text{ meV} \) and \( E_{\text{light}} = 1.5 \text{ eV} \) and take as electron effective
When electron tunneling is included, direct and indirect states are mixed when in near resonance. In quantum dot molecules, this produces that direct-indirect crossings become **anticrossings**, arising from the formation of bonding and antibonding states around the energy degeneracy point.9 In quantum ring molecules, however, not all crossings become anticrossings, as shown in the energy levels plotted in Fig. 2(b). Angular momentum selection rules ensure that only states with the same total angular momentum are coupled by electron tunneling.22

Most importantly, not all states are optically active, as shown in the color contour plot in Fig. 2(b). The lowest energy optically active state is the $L=0$ direct exciton in dot 2, $D_{2}^{L=0}$. At $B=2.3$ T, near the anticrossing region, the state with indirect character ($=I_{12}$) becomes active due to the coupling with $D_{2}^{L=0}$, similar to the case of quantum dots.9 The ability to control the character of the ground state of the quantum ring molecules and their optical response, expressed in Fig. 2(b), is the main result of this paper. We emphasize that this arises from a distinctive property of the ring geometry: the possibility of tuning the orbital degrees of freedom (namely, the ground-state angular momentum) by varying an external magnetic field.

Notice, moreover, that the system can be tuned into a different regime at $B=2.8$ T: a degeneracy point between uncoupled direct and indirect states. Around this point, the ground state of the system is doubly degenerate, and one would typically create a linear combination of states, rather than an entangled set, if creation of the excitons is slightly above resonance.

Experimentally, the coupling between direct and indirect states is easier to observe by varying the electric field $F$ which produces a linear Stark shift in the indirect exciton lines, while the peaks from direct exciton states are essentially “flat.” This is shown in Fig. 3 in the range $-12.5 < eFd < -11.5$ meV. Indirect exciton states $I_{12}$ appear as diagonal lines with positive slope.

At zero magnetic field, there are two indirect states (with $L=0$ and $L=1$) close in energy to the $D_{2}^{L=0}$ low-lying state. Tunnel coupling is forbidden for the $L=1$ indirect state, resulting in only one anticrossing in the optical spectrum (at...
For even larger tendency to study entanglement of more than two exciton states. Interestingly, the two anticrossings merge into one strong anticrossing for higher magnetic field ($B \approx 3$ T). This can be understood from the spectrum shown in Fig. 2(a). As $B$ increases, two $L=0$ indirect states approach in energy and are degenerate at $B \approx 3$ T. In the $\alpha(F)$ map, this corresponds to a merging of the anticrossings with the $D_{2}^{L=0}$ state, as shown in Fig. 3. At that point, the direct state is equally coupled to two distinct indirect states, providing a very interesting opportunity to study entanglement of more than two exciton states. For even larger $B$, the anticrossings become separated again and the general situation seen at $B=0$ is recovered.

Notice that the direction (sign) of the electric field is crucial in the level mixing: direct and indirect states in the same energy window are essentially decoupled for positive values of $F$. This is shown in Fig. 4. Due to the much smaller tunneling amplitude for the holes, anticrossings between $D_{2}$ and $I_{2}$ states appearing for negative $F$ [Fig. 4(a)] are replaced by crossings between $D_{2}$ and $I_{2}$ states [Fig. 4(b)]. Notice of course that increasing hole tunneling in a structure decreases, two $L=0$ indirect states approach in energy and are degenerate at $B=0$ indirectlike state can be seen close to $\Omega=0$, so that now two anticrossings appear. In addition, the two $L=1$ indirect states split in magnetic field and crossings with these levels are also seen.

In conclusion, we demonstrate the possibility of using double quantum ring molecules as a tool to control tunneling in semiconductor devices. In the proposed setup, direct and indirect exciton states can be Stark shifted in and out of resonance by an electric field along the ring axis. As states with different $L$ have distinct dispersion with magnetic field, the setup allows the use of orbital angular momentum selection rules as a “selector” of which states can be coupled.

The proposed model of a larger ring coupled to a smaller one naturally arises in realistic self-assembled double-layer quantum ring samples due to induced strain. Experimental verification of the effects discussed here could be achieved in different optical setups involving stacked quantum ring samples. For instance, an anticrossing in the photoluminescence spectrum when an electric field is varied while the magnetic field is small (so that no changes in the ground-state angular momentum occur) should be evident, as observed in self-assembled quantum dot molecules. As the magnetic field increases, the angular momentum of the exciton in the larger ring changes and the anticrossing becomes a single line crossing. This would result in a continuous line in the photoluminescence spectra since the direct exciton recombination dominates over the indirect one.

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In fact, the angular momentum conservation for tunneling processes holds for a general exciton wave function $\Psi_i(p, \theta) = R(p)e^{i\ell \theta}$. In this case, $t_{ij}^{\ell \ell'}(\ell) = \delta_{\ell, \ell'}$. This vanishes for $L_i \neq L_j$, independent of the radial term. Here, $V(p)$ is the inter-ring tunneling Hamiltonian for the electron.