Opportunities for single hole-spin control using delocalized states of quantum dot molecules

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Abstract. We present three unique spin properties of holes in the delocalized states of coupled pairs of InAs quantum dots: resonant changes in the hole g factor, antisymmetric hole molecular ground states, and hole spin mixing. These three properties suggest new opportunities for spin control that are unique to holes and further support holes as a competitive candidate for the storage and manipulation of quantum information.

1. Introduction
Holes confined in QDs have spin projections that could be used as bits in spin-based logic or quantum information processing devices. Hole spins were initially discounted as viable qubit candidates because the complex nature of valence band states was expected to drastically reduce the spin lifetimes and decoherence times. Recent work, however, has shown that hole spins in QDs can be initialized and stored with lifetimes comparable to those of electrons.\textsuperscript{[1, 2]} Moreover, hole states have Bloch wavefunctions with p-like symmetry, which dramatically suppresses the hyperfine interaction with nuclear spins that is a primary source of decoherence for electron spins. Consequently, hole spins have emerged as a competitive choice for the fundamental unit of information in spin-based logic devices. In this work we present studies of coupled pairs of InAs QDs that illustrate how the formation of molecular states for holes leads to new opportunities for controlling single hole spin projections. These discoveries suggest that coupled pairs of quantum dots may provide an architecture that simplifies implementation of spin control protocols and further support holes as a competitive candidate for the storage and manipulation of spin-based quantum information.

2. Delocalized Molecular States
The Quantum Dot Molecules (QDMs) we describe here consist of two InAs QDs separated by a GaAs barrier and are grown by molecular beam epitaxy. The energy levels of the two dots are slightly different such that the electron and hole energy levels of the two dots are not degenerate, but the hole levels can be tuned into resonance with an applied electric field. On resonance, coherent hole tunneling between the QDs leads to the formation of molecular orbitals. The formation of molecular orbitals can be understood by considering only the lowest energy levels of each QD and temporarily ignoring spin. When there is no interaction between the states of...
Figure 1. Measured PL energies of the neutral exciton in a QDM as a function of applied electric field. The diagrams schematically depict the wavefunction of the atomic-like basis states.

Figure 2. Zeeman splitting at 6 T of the molecular ground and excited states as a function of applied electric field for QDMs with (a) 2 nm and (b) 4 nm barrier. (c) Schematic depiction of the symmetric and antisymmetric molecular wavefunctions.

the individual QDs, there are two possible states for holes: the energetic ground states of the top and bottom dot. The QD asymmetry and applied electric field cause electrons in the top dot to rapidly relax into the bottom dot, so we only need to consider one possible state for the electron (the energetic ground state of the bottom QD). The two possible locations for holes create two atomic-like basis states with wavefunctions schematically depicted at the right side of Fig. 1.

Fig. 1 plots the energies of photoluminescence (PL) lines emitted from a QDM with 2 nm barrier as a function of the applied electric field. The two atomic-like basis states have PL energies that depend differently on the applied electric field, leading to the asymptotically horizontal and diagonal lines in Fig. 1. When the hole levels of the two dots come into resonance, coherent tunneling of the hole between the two dots leads to the formation of delocalized molecular states. The signature of the formation of molecular orbitals is the anticrossing gap (Δ) that appears in the spectra. On resonance, the molecular state can be described as the bonding (symmetric) or antibonding (antisymmetric) combination of the two basis states. The data have been plotted in black and red to illustrate that one line (black) is the molecular ground state on resonance and the other (red) is the molecular excited state.

3. g factor Resonance

Fig. 2a shows the measured Zeeman splitting for the sample presented in Fig. 1 when we apply a magnetic field in the Faraday configuration. The data in black show that the molecular ground state has a resonant decrease in Zeeman splitting at the electric field of resonance. In contrast, the data in red show that the molecular excited state has a resonant increase in Zeeman splitting at the same electric field. These changes in Zeeman splitting originate in a resonant change in the g factor for the hole due to the formation of hole states with molecular orbital character, as schematically depicted in Fig. 2c.[3, 4] The molecular ground state has a symmetric (bonding) molecular wavefunction with non-zero amplitude in the GaAs barrier between the two dots, which increases the contribution from the GaAs and leads to suppression of the hole g factor. The node in the wavefunction for the antisymmetric (antibonding) molecular excited state, on the other hand, suppresses the contribution of the GaAs and leads to the resonant increase in Zeeman splitting for the molecular excited state.

Electrically tunable g factors have previously been used to coherently modulate electron spin projections in static magnetic fields using a technique known as g Tensor Magnetic Resonance.
However, the heterostructure used to demonstrate gTMR does not localize single spins and is inherently unable to provide the basis for a scalable spin-based logic device with individual localized, addressable spins. The QDM geometry, on the other hand, can localize a single hole spin in the QDM. Moreover, the magnitude of the resonant change in hole g factor observed in QDMs is more than an order of magnitude bigger than that observed in coupled quantum wells. Hole states in QDMs are thus ideally suited for the application of gTMR to coherently control single spins.

4. Reversal of the molecular orbital character
The theory of molecular orbitals derived for natural diatomic molecules predicts that molecular ground states have symmetric (bonding) orbital character. The suppression of the Zeeman splitting for the molecular ground state, as shown by the black trace in Fig. 2a, is the experimental signature of the state’s symmetric molecular orbital character. In Fig. 2b we present data on the Zeeman splitting of the molecular orbitals for a QDM in which the GaAs barrier has been increased from 2 nm to 4 nm. The data clearly demonstrate that the molecular ground state (black) now has the resonant increase in Zeeman splitting that is indicative of an antisymmetric (antibonding) molecular orbital. In contrast, the molecular excited state (red) now shows the resonant decrease in Zeeman splitting indicative of the symmetric (bonding) molecular orbital. This reversal of the molecular orbital character can not be explained by analogy with natural diatomic molecules and has surprising consequences.

The reversal of the molecular orbital character originates in spin orbit interactions between light and heavy hole spin components and has two important consequences for the control of hole spins and the use of QDMs for information storage and manipulation. First, it allows one to choose an energetic ground state with the desired resonant increase or resonant decrease in Zeeman splitting that is best suited to the overall device architecture and spin manipulation protocol. Second, if both electron and hole levels can be brought into resonance at the same value of the applied electric field the tunneling of the electron will form a symmetric molecular orbital state while the barrier thickness could be chosen to create an antisymmetric orbital state for holes. The opposite symmetry of the two states means that there is a zero matrix element for the dipole transition between these two states - i.e. there is no radiative recombination. The creation of an energetic excitonic ground state not subject to radiative recombination would be revolutionary for optoelectronic devices and lead to new opportunities for optically-driven information storage and manipulation. The creation of such a state is a unique opportunity provided by the molecular states of holes in QDMs.

5. Hole spin mixing
When the symmetry of the QDM is broken by lateral offset of one dot relative to the other, additional spin-orbit induced interactions become possible. These additional mixing mechanisms couple states with orthogonal heavy-hole spin projections and opposite spatial locations, equivalent to a spin-flip tunneling processes. This novel hole spin mixing mechanism is mediated by states with significant light hole character and leads to new anticrossings in the experimental spectra, as indicated by the arrows in Fig. 3a.

The existence of a novel spin mixing mechanism suggest new opportunities for hole spin control. One example of a novel spin control mechanism using adiabatic fast passage techniques is depicted by the arrows in Fig. 3b. The hole is initialized in the spin down state within the bottom dot at an electric field well away from resonance so that the heavy-hole spin projections remain well isolated and viable states for the storage of quantum information. The electric field is swept through the first anticrossing (arrow 1) at a rate that causes the hole state to ‘jump’ across the anticrossing and follow the asymptotic states. The electric field is then returned to the starting value at a rate that causes the hole state to follow the adiabatic line (arrow 2),
ending as a spin up hole spin projection. This technique is similar to one recently used to control electron spins in lithographically defined QDs.[10] In the InAs QDMs proposed here, however, both optical and electrical spin manipulation methods can be utilized.

6. Summary
Coherent tunneling between neighboring InAs QDs leads to the formation of molecular states for holes. These molecular states have unique properties that are determined by the structure and symmetry of the QDM. The formation of molecular orbitals leads to resonant changes in g factors that could be the basis of gTMR control of single localized hole spins. Changing the spacing between the dots leads to the formation of antisymmetric molecular ground states for the holes that can control both the sign of the g factor resonance and the optical matrix elements for exciton states. Breaking the symmetry of the QDM by offsetting one QDM laterally leads to a novel spin mixing mechanism unique to holes that could enable electrically-driven coherent control of the hole spin projection. The combined results demonstrate that the molecular orbital states of holes provide a variety of novel spin control opportunities.

7. Acknowledgments
Acknowledgments
MFD acknowledges support from the University of Delaware Research Foundation and the NSF (0844747). Work at the Naval Research Lab was supported by NSA/ARO and ARO/MURI(W911NF0910406). JIC acknowledges support from the Ramon y Cajal program.

References
[1] Heiss D, Schaeck S, Huebl H, Bichler M, Abstreiter G, Finley J J, Bulaev D V and Loss D 2007 Physical Review B 76 241306
[2] Gerardot B D, Brunner D, Dalgaro P A, Ohberg P, Seidl S, Kroner M, Karrai K, Stoltz N G, Petroff P M and Warburton R J 2008 Nature 451 441–444
[3] Doty M F, Scheibner M, Ponomarev I V, Stinaff E A, Bracker A S, Korenev V L, Reinecke T L and Gammon D 2006 Physical Review Letters 97 197202
[4] Doty M F, Scheibner M, Bracker A S and Gammon D 2008 Physical Review B 78 115316
[5] Kato Y, Myers R C, Driscoll D C, Gossard A C, Levy J and Awschalom D D 2003 Science 299 1201–1204
[6] Poggio M, Steeves G M, Myers R C, Stern N P, Gossard A C and Awschalom D D 2004 Physical Review B 70 121305(R)
[7] Climente J I, Korkusinski M, Goldoni G and Hawrylak P 2008 Phys. Rev. B 78 115323
[8] Doty M F, Climente J I, Korkusinski M, Scheibner M, Bracker A S, Hawrylak P and Gammon D 2009 Physical Review Letters 102 047401
[9] Doty M F, Climente J I, Greilich A, Yakes M, Bracker A S and Gammon D 2010 Physical Review B 81 035308
[10] Petta J R, Lu H and Gossard A C 2010 Science 327 669–672 10.1126/science.1183628