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Invited Paper

Molecular Engineering with Artificial Atoms: Designing a Material Platform for Scalable Quantum Spintronics and Photonics

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ABSTRACT

Self-assembled InAs Quantum Dots (QDs) are often called "artificial atoms" and have long been of interest as components of quantum photonic and spintronic devices. Although there has been substantial progress in demonstrating optical control of both single spins confined to a single QD and entanglement between two separated QDs, the path toward scalable quantum photonic devices based on spins remains challenging. Quantum Dot Molecules, which consist of two closely-spaced InAs QDs, have unique properties that can be engineered with the solid state analog of molecular engineering in which the composition, size, and location of both the QDs and the intervening barrier are controlled during growth. Moreover, applied electric, magnetic, and optical fields can be used to modulate, *in situ*, both the spin and optical properties of the molecular states. We describe how the unique photonic properties of engineered Quantum Dot Molecules can be leveraged to overcome long-standing challenges to the creation of scalable quantum devices that manipulate single spins via photonics.

Keywords: Spintronics, Quantum Information, Photonics, Quantum Dots, Quantum Dot Molecules

1. INTRODUCTION

Optoelectronic devices that operate at the quantum limit of single photons, charges, and spins have long been viewed as a promising platform for quantum technologies[1–5] because the weak interaction of photons with their environment provides a near-ideal mechanism for the transmission of quantum information. The ideal device would leverage wafer-scale semiconductor processing methods to create on-chip photonic devices that emit, route, and absorb photons. The fundamental components of such a device would have to include optically-active nanostructures with quantized energy states capable of a) absorbing and emitting single photons and b) storing information. To preserve quantum superpositions, information would most likely be stored in the spin projections of confined electrons or holes or in nuclear spin baths whose projections are controlled via interactions with the confined electrons or holes.[6] These single photon sources, detectors, or memories could then be connected to each other and communications networks using well-established photonic device architectures.

Semiconductor quantum dots (QDs), often called artificial atoms, have long been considered as promising components of scalable quantum photonic devices because they locally confine single charges with well-defined spin projections in discrete energy states analogous to the orbital energy levels of natural atoms. InAs QDs, in particular, have enabled many impressive proof-of-concept quantum spintronic device experiments.[6–13] However, efforts to create chip-scalable platforms for single-photon technologies have been hampered by the challenge of fabricating QDs with both spatial control of their position and spectral control over their emission energy.

The best optical performance for InAs QDs is obtained when the QDs are permitted to self-assemble on a GaAs surface. Although the optical quality is high, the self-assembly process results in random spatial locations for the QDs, which is incompatible with scalable production of devices that require spatial overlap between the

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QDs and a photonic mode used to control the propagation of light within a device. Site-controlled growth has been demonstrated, but it remains challenging to achieve high optical quality with site-controlled QDs. Moreover, the discrete energy levels of single QDs within an ensemble, even when grown by random self-assembly, typically vary by approximately 50 meV. As a result, it is not possible to deterministically create spectral overlap between the QD emission and the target photonic cavity or waveguide mode.

Here we discuss complexes of quantum dots that can be thought of as artificial molecules because the spatial arrangement of QD artificial atoms mimics that of natural molecules. Our investigation of both lateral and vertical diatomic quantum dot pairs reveals that coherent tunneling between QDs leads to the formation of delocalized molecular states with surprisingly complex properties that could be tailored during growth, via molecular structure and composition, or modulated *in situ* by applied electric, magnetic or optical fields. The number of artificial molecular parameters that can be tuned, both during and after growth, dramatically exceeds the opportunities with natural molecules. As a result, these quantum dot molecules (QDMs) could be the foundation of a solid-state version of molecular engineering in which the properties of single charges, spins, and photons are tailored for new device applications. We will show how engineering diatomic QDMs provides paths to overcoming long standing obstacles to the creation of scalable devices that manipulate spins with photons. We will focus on the progress and prospects for controlling single hole spins, which have been shown to have substantially longer decoherence times due to reduced interaction with nuclear spins.[6]

2. SPATIAL AND SPECTRAL CONTROL OVER SINGLE QDMS

InAs QDs form when InAs deposited on a GaAs surface aggregates to minimize strain and surface energy that originates in the lattice mismatch between the InAs and the GaAs substrate.[14–16] When these small regions of InAs are capped by GaAs, three-dimensional confinement leads to a ladder of discrete energy levels for confined electrons and holes analogous to the s, p, d, etc. levels of natural atoms. The center emission wavelength of an ensemble of InAs QDs can be controlled using a "cap and flush" technique in which GaAs is deposited to partially cover the InAs QD. The exposed InAs above the GaAs coverage is removed and the remaining InAs is covered with GaAs to create a pancake-shaped InAs QD of defined thickness. This control does not lead to identical QDs; size and composition inhomogeneities originating in diffusion processes always lead to a distribution of energy levels in a QD ensemble, typically of order 50 meV. This spectral inhomogeneity severely hampers progress toward deterministic fabrication of photonic devices, which would rely on consistent spectral overlap between QD emission and photonic cavity or waveguide modes. The sensitivity to spectral alignment in quantum devices is exacerbated by the need for strong interactions between emitters and optical cavities. Strong interactions require cavities with high quality factor (Q) and consequently narrow bandwidth. As a result, precise overlap between the cavity and emitter wavelengths is required. The wavelength inhomogeneity of current single QD growth methods prevents the reliable production of an ensemble of QDs each at the target wavelength required for device integration. Because QD growth relies on diffusion, which is inherently random, it is unlikely that this inhomogeneity can be directly overcome. Instead, we must consider paths toward *in situ* tunability that allows us to overcome this limitation.

After a single layer of QDs has been grown and capped, the strain induced by the buried QDs propagates through the capping layer and creates a preferential nucleation site above each buried QD.[16] Consequently, deposition of sequential layers of InAs QDs leads to aligned vertical stacks of QDs. This strain-driven alignment has two important consequences for the design of scalable quantum spintronic devices. First, it proveds a mechanism for transferring spatial templates to future growth surfaces. Second, it allows for the controlled creation of Quantum Dot Molecules (QDMs) that allow us to both overcome the spectral inohomogeneity problem described above and leverage emergent spin functionality.

2.1 Site Control

Methods for templating the spatial locations of QDs include: Focused Ion Beam formation of preferential growth sites,[18] Ga droplet epitaxy,[19–22] growth on cleaved-edge or faceted substrates,[23,24] controlled hydrogenation of quantum wells,[25] nanoindentation,[26,27] and growth through nanochannel templates.[28] A thorough review of progress on controlled QD growth was published by Kiravittaya, et al. in 2009,[29] though of course there have been significant advances since that report was published. The state-of-the-art in MBE growth of InAs QDs is

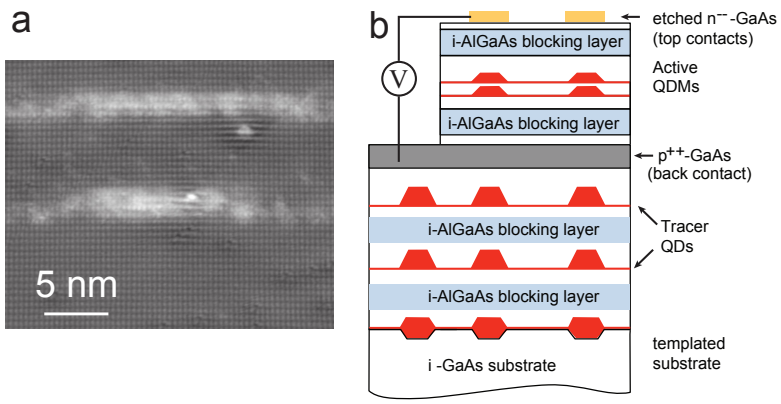

Figure 1. a) Cross-sectional scanning tunneling microscopy image of vertically-stacked InAs QDs grown by Allan Bracker of the Naval Research Laboratory.[17] b) Schematic depiction of device template. Nanofabricated features in the substrate guide nucleation of the initial QD layer. Strain propagation creates favorable vertically-aligned nucleation sites for subsequent tracer QDs whose optical quality is unimportant. QDMs, the optical element, are embedded in a p-i-n diode structure and spatially templated by the underlying tracer QDs.

to use GaAs surfaces pre-patterned with shallow holes or trenches that provide favorable QD nucleation sites. This approach has resulted in QDs with good optical properties,[30–32] but contaminants or native crystal defects arising from the fabrication process can have a severe impact on the optical quality of the resulting QDs. It is therefore desireable to separate the optically-active elements from the fabricated surface while preserving the spatial control over the QD location.

The strain-driven alignment described above provides a mechanism for overcoming this challenge: stressor QDs can propagate spatial registry through multiple layers of QD growth.[33] This approach can be fully implemented using InAs or InGaAs QDs, and typically results in vertical "columns" of QDs with poor optical quality close to the intially-fabricated surface and increasing optical quality in subsequent layers. A variety of other stressor QD compositions can be used to spectrally isolate stressor QD optical emission or absorption from the absorption and emission of the optically-active elements.[34–36] This approach is schematically depicted in Fig. 1b. The AlAs or AlGaAs layers grown in the spacer between QD layers suppress electronic transport from optically-active layers to optically-poor stressor QDs that provide nonradiative recombination sites.

2.2 Spectral Tuning

QDMs consist of two QDs aligned along the growth axis due to strain and separated by a barrier that is sufficiently thin to allow the formation of delocalized molecular orbitals for electrons or holes via coherent tunnel coupling between the two QDs. There is an extensive literature on the properties of QDMs.[17,37–54] In this section we focus on the presence of both direct and indirect optical transitions, as schematically depicted in Fig. 2a for a QDM embedded in a p-i-n device. Direct transitions are the "normal" optical transition involving an electron and hole in the same QD. Indirect transitions involve an electron and hole in separate QDs and are allowed because the wavefunctions of electrons and holes have non-negligible penetration into the barrier separating the QDs.

Fig. 2b presents the energies of the observed photoluminescence (PL) emitted by a neutral exciton (one electron, one hole) in a single vertical QDM with a 4 nm GaAs barrier between the QDs.[46] Because of the inhomogeneous distribution in QD energies, the electron and hole normally localize in individual dots. If the electron and hole are located in the same dot (lower inset to Fig. 2b), a direct optical transition occurs, with optical properties nearly identical to those of a single QD. For a direct transition, the emitted PL has a very weak dependence on applied electric field (nearly horizontal line at about 1319 meV). This weak dependence of the direct PL emission energy on applied electric field originates in the Stark Shift and the ~ 1 meV tuning over a moderate range of electric fields is typical of that observed in a variety of single QD structures. In contrast,

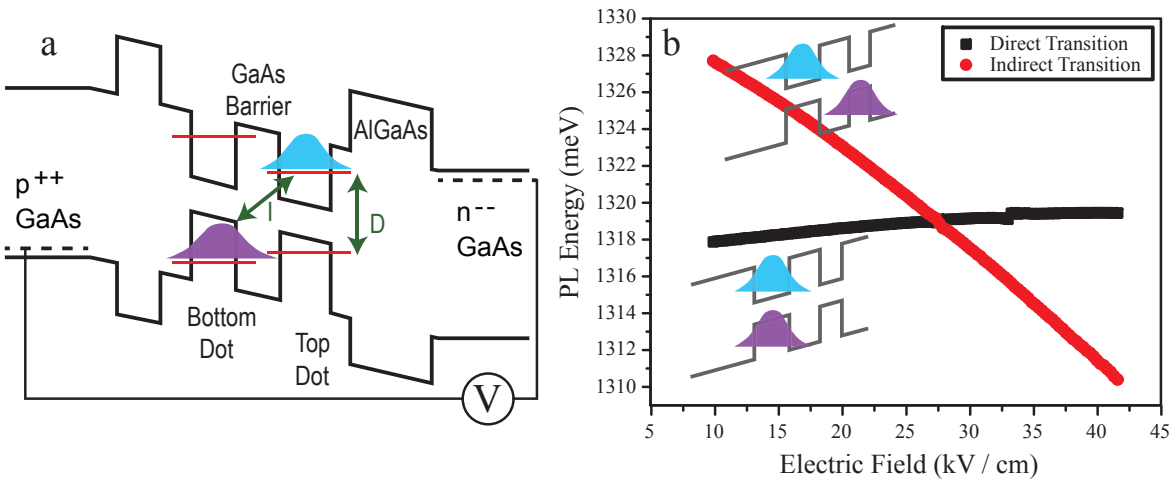

Figure 2. a) Schematic band diagram of the optically-active QDM, indicating Direct (D) and Indirect (I) optical transitions. Insets depict the wavefunctions for an electron and hole participating in an indirect transition. b) Experimental measurement of photoluminescence energy as a function of applied electric field in a similar QDM.[46]

when the electron and hole are located in separate QDs (e.g. as depicted in Fig. 2a and the upper inset of Fig. 2b) the applied electric field strongly affects the relative energies of the electrons and holes. As a result, the energy of PL emitted by an indirect transition, shown by the red symbols in Fig. 2b, tunes strongly with applied electric field. Fig. 2b demonstrates that the energy of an indirect transition can be tuned by at least 18 meV as the applied electric field is varied from 5 to 45 kV/cm. It is this strong tuning of indirect QDM emission with applied electric fields that allows individual QDMs to be tuned into resonance with the design wavelength of photonic devices, overcoming the spectral inhomogeneity problem inherent to individual QDs.

The wavelength tunability of indirect transition energies to applied electric field can be enhanced by placing a thicker barrier between the QDs. However, increasing the thickness of the barrier also decreases the optical dipole matrix element of the indirect transition by reducing the overlap between the wavefunctions of the electron and hole. For relatively closely spaced QDs, the indirect transitions can have relatively strong optical transition dipoles, allowing for strong interactions with light while retaining reasonable wavelength tunability.[46]

3. HOLE SPIN MIXING IN QDMS

An important capability that allows the deterministic control over wavelength tunability in QDMs comes from the partial cap technique described above, which removes the InAs above a controlled height to create a pancake-shaped dot of defined thickness. This technique allows the heights of the two QDs that comprise the QDM to be *independently* controlled. By selecting the relative size of each QD and embedding the QDM within a diode structure, as depicted in Fig. 2a, applied electric fields can be used not only to tune the energy of indirect optical transitions, but also to tune discrete energy levels into resonance to create delocalized molecular states in a controllable and reversible manner.[39] This electric field control over the formation of delocalized molecular states provides a powerful tool for manipulating molecular spin and charge properties *in situ*. These unique properties of QDMs provide many opportunities for quantum device technologies.[45,46,55,56] In this section we focus on one emergent spin property of QDMs that illustrates both the device functionality and engineering opportunities.

Figure 3 presents experimental photoluminescence spectra from a QDM in which the hole tunnels back and forth between the two dots while the electron remains confined in a single state with maximum wavefunction amplitude in the bottom dot. The right insets to Fig. 3 show the symmetric and antisymmetric orbitals formed by coherent tunneling of the hole. The magnitude of the anticrossing gap, Δ, depends on the overlap of the wavefunctions and thus on the thickness of the barrier separating the dots. The symmetry of molecular wavefunctions

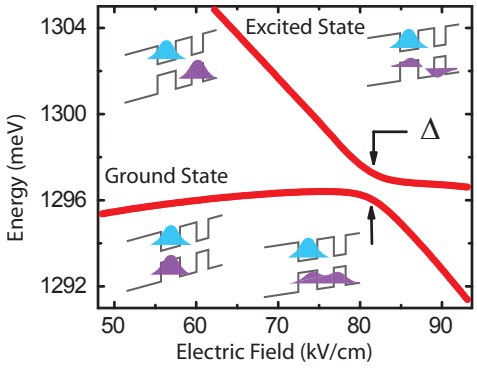

Figure 3. Discrete atomic-like states of an InAs QD molecule anti-cross (Δ) due to the formation of molecular orbitals, as depicted in the insets.[44]

depicted in the insets of Fig. 3 follows the energy ordering expected by analogy with diatomic molecules, but the symmetry reverses for thicker GaAs barriers.[44] This surprising and counter-intuitive results arises from spin-orbit interactions in the valence band that mix heavy- and light-hole states and may have significant implications for tailoring the optical properties of individual QDMs.[49]

New spin properties emerge when we apply a magnetic field along the optical axis, which is parallel to the stacking axis of the QDM. Each of the molecular states shown in Fig. 3 is comprised of two degenerate bright exciton spin configurations and the applied magnetic field Zeeman splits these two spin configurations. Surprisingly, we have found that the Zeeman splitting, which is proportional to the g factor, is not constant, but is a strong function of the applied electric field.[40] These resonant changes in g factor are understood to arise from the orbital character of the molecular states, which controls the magnitude of the barrier contribution to the g factor of a single confined carrier.[40,57,58] We have used heterostructure engineering to tailor the QDM composition and achieve similar g factor tuning for a single electron.[47] The resonant changes in g factor are functionally equivalent to a spin-dependent tunnel barrier (i.e. the magnitude of the anticrossing gap is spin dependent), suggesting that the controlled formation of these molecular states may provide a path toward spin filtering or manipulation.

In Fig. 4 we present a comparison of experimental and computational results for a different QDM in which the hole tunnels back and forth between the two QDs. Here we focus on the emergence of a family of additional

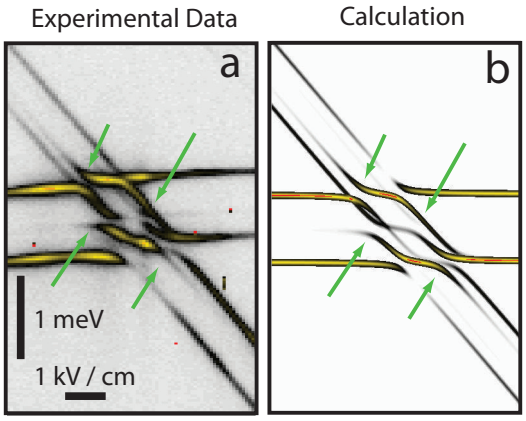

Figure 4. Experimental (a) and computed (b) spectra of spin mixing anticrossings of the neutral exciton in a single QDM with 4 nm barrier at 6 T.[17]

anticrossings, highlighted by the green arrows in Fig. 4. These anticrossings are the manifestation of hole spin mixing for tunneling holes, which results in a coherent coupling of bright and dark exciton states.[17,59] These anticrossings cannot be explained by Hamiltonians with no spin mixing. but, as shown in Fig. 4b, inclusion of a hole spin mixing term in the matrix Hamiltonians reproduces all of the observed anticrossings.[17]

The hole spin mixing originates in a lateral offset of the two QDs that breaks the molecular symmetry and enables a coupling between the light hole components of the nominally orthogonal Luttinger spinors that properly describe holes in QDs. This light hole mixing, which leads to a spinor mixing, results in the formation of molecular states that are an admixture of the two Luttinger spinors dominated by orthogonal heavy-hole spin projections —hole spin mixing.[17,49,59] This mixing can simplistically be understood as a spin-flip tunneling process, though the actual states include a mixture of both spin-conserving and spin-flip coherent tunneling. The discovery of hole spin mixing is exciting because it suggests that changes to the geometric symmetry of the QDM, along with the composition and structure, can result in new spin properties. We have developed detailed approaches that take advantage of these emergent spin properties to enable quantum information processing protocols with improved scalability.[46]

4. CONTROLLING SPIN PROPERTIES WITH ELECTRIC FIELDS

From a scalable device point of view there are two problems with attempting to use hole spin mixing as observed in Fig. 4. First, the magnitude of the hole spin mixing is not as large as we would like for optical control of coherent spin rotation. Second, the hole spin mixing arises due to lateral offsets of the two QDs along the stacking axis —offsets that cannot be controlled during growth. In this section we describe how application of in-plane electric field gradients can be used to turn on and off hole spin mixing.

To explore the possibility of *in situ* control over hole spin mixing, we computed the energy states for holes in QDMs under a variety of electric and magnetic field condidtions. Details can be found in Ref.[60] We first use atomistic tight binding simulations to generate exact eigenstates. We then use a finite basis matrix Hamiltonian extrapolation to compute the state energies as a function of varying electric field conditions. We verified that this approach achieves at least 97% precision relative to atomistic tight binding calculations when at least 16 eigenstates are included.

Our computational results reveal that hole spin mixing can emerge when lateral electric fields are applied to QDMs - i.e. when the electric field is orthogonal to the stacking axis of the QDM. For QDMs with no lateral offset along the stacking axis, the effect is relatively small. However, when the QDMs have a small lateral offset along the stacking axis, which is quite common in as-grown QDMs, the effect gets larger. Similarly, for lateral electric field magnitudes that are constant over the entire QDM, the magnitude of the resulting hole spin mixing is relatively small. When electric field gradients are applied instead, the magnitude can be significantly larger. This result can intuitively be understood by analogy with the structural lateral offset that is a necessary condition for hole spin mixing in the absence of lateral electric fields. A lateral electric field displaces electron and hole wavefunctions from the center to the side of the QDs. A lateral electric field gradient creates an asymmetric displacement of the hole wave functions, creating the asymmetry in the QDM required for hole spin mixing.

In Fig. 5 we present computational results for the energies of the hole states in a QDM with a 1 nm lateral structural offset when the applied lateral electric field has a gradient along the growth direction. Fig. 5a includes an outline of the boundaries of the two cylindrically-shaped QDs we use in the computations along with a color map depicting the electric field in the plane of each QD. Fig. 5b presents the hole spin mixing extracted from the computed hole energy levels as a function of the electric field gradient. The range of electric field gradients considered is chosen to be within the range achievable in real devices.[61] The key result is that the magnitude of hole spin mixing (Δ_{sm}) increases with increasing gradient magnitude, which means that the existence and magnitude of hole spin mixing can be modulated *in situ* by applied electric fields. This result establishes that it is technically feasible to electrically manipulate spin interactions for a single hole confined in a QDM and to thereby turn on and off coherent optically-driven spin initialization, rotation, or readout operation. Efforts to engineer devices that employ this spin manipulation protocol to create scalable spin-based quantum photonic devices with independent control over each hole spin-based qubit are underway.

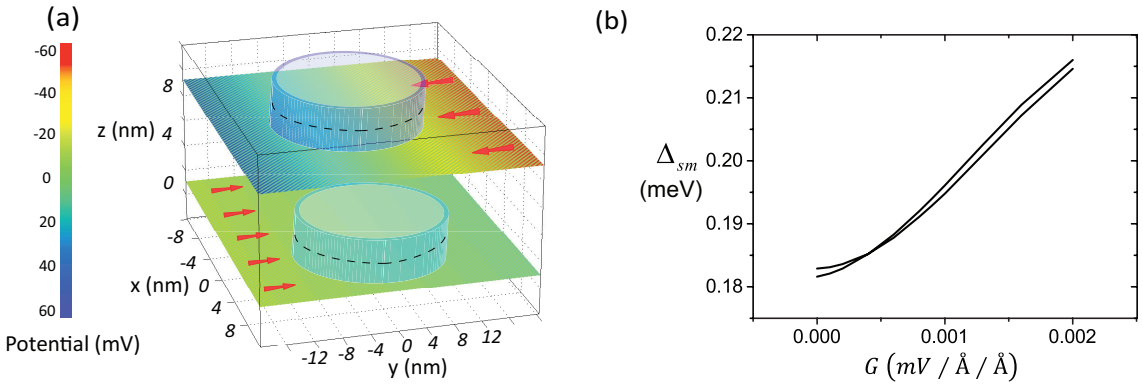

Figure 5. Effect of lateral electric field with a gradient in the growth direction on asymmetric QDMs. (a) Electric potential for gradient $G_{yz} = 0.002$ (mV/Å)/Å. (b) Hole spin mixing magnitude (Δ_{sm}) as a function of lateral electric field gradient G for two distinct spin mixing anticrossings.[60]

5. CONCLUSION

Quantum devices in which information is encoded in spin projections and manipulated by light have long been of interest, but numerous challenges to the design and production of scalable devices remain. InAs QDs remain one of the most promising materials for confining the spins and enabling the optical manipulation, but the random nucleation locations of InAs with good quality and the inhomogeneous distribution of QD optical transition energies remain major challenges. We have shown that the strain-driven preferential nucleation that underlies QDMs provides a path to overcoming the site control problem. Moreover, the indirect optical transitions of QDMs provide an opportunity for overcoming the spectral inhomogeneity problem by using growth-direction electric fields to tune the optical transition energies into resonance with design or photonic circuit component wavelengths. The existence of hole spin mixing in QDMs enables protocols for all-optical coherent control over hole spin initialization, manipulation, and readout. The dependence of hole spin mixing on lateral electric field gradients will allow the controlled creation and modulation of hole spin mixing, providing a path toward scalable devices in which each hole-spin qubit can be be independently manipulated by photons.

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