Electrically tunable effective $g$-factor of a single hole in a lateral GaAs/AlGaAs quantum dot

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Electrical tunability of the $g$-factor of a confined spin is a long-time goal of the spin qubit field. Here we utilize the electric dipole spin resonance (EDSR) to demonstrate it in a gated GaAs double-dot device confining a hole. This tunability is a consequence of the strong spin-orbit interaction (SOI) in the GaAs valence band. The SOI enables a spin-flip interdot tunneling, which, in combination with the simple spin-conserving charge transport leads to the formation of tunable hybrid spin-orbit molecular states. EDSR is used to demonstrate that the gap separating the two lowest energy states changes its character from a charge-like to a spin-like excitation as a function of interdot detuning or magnetic field. In the spin-like regime, the gap can be characterized by the effective $g$-factor, which differs from the bulk value owing to spin-charge hybridization, and can be tuned smoothly and sensitively by gate voltages.
In semiconductor spintronics, spin rotation is an essential element of coherent control. The most direct method of achieving spin rotation involves a pulsed magnetic field. This technique, however, produces slow rotations and leads to qubit addressability difficulties. Therefore, the preferred approach is electric dipole spin resonance (EDSR), involving an electrical microwave modulation of the system. The EDSR frequency is defined by the Zeeman energy $E_Z$ (spin splitting), which is set by the static external magnetic field. $E_Z$ depends on the effective $g$-factor of the host material, and therefore addressing individual spins in a multi-qubit device would benefit from the ability to tune the $g$-factor in a spatially selective manner.

In EDSR, the spin-flip results from electrical modulation of the particle’s position, and thus a mechanism coupling these two degrees of freedom is required. Such a mechanism can be engineered by creating position-dependent magnetic interactions with micromagnets or nuclei of the host material. In gated GaAs or Si devices confining electrons, the spin-orbit interaction (SOI) inherent in the semiconductor material has also been exploited. However, the resulting coupling is weak so coherent control of the spin is possible only on timescales comparable to the spin decoherence time. The SOI strength can be increased by confining electrons in InAs or InSb dots, however, switching the carrier type to holes that can deliver strong SOI holds the greatest promise. EDSR in hole systems has been more weakly with nuclear spins than electrons. This hyperfine interaction severely limits the control of the spin is possible only on timescales comparable to the spin decoherence time.

The system is further characterized by transport measurements in the high source-drain bias transport regime. The SOI is relatively weak owing to the inversion symmetry of the crystal lattice. However, a side effect of the increased SOI may result in a shorter hole spin relaxation time as this process is brought about by phonon-assisted SOI. Recently, $T_1$ in the GaAs gated hole device was measured and shown to be substantially shorter than that in electron-based GaAs devices. Nonetheless, the advantage of hole-based devices lies in the fact that holes interact more weakly with nuclear spins than electrons. This hyperfine interaction severely limits the $T_1$ of the electron spin at low magnetic fields, while the hole spin $T_1$ remains limited by the phonon-assisted SOI and therefore may become much longer. Moreover, the hole spin $T_1$ could be further extended by engineering the phonon density of states to eliminate the phonon modes mediating the spin-flip transitions. Preliminary work toward that end has already been demonstrated for electronic dots in nanowires.

Being a perturbative effect, the SOI in devices confining electrons is too weak to renormalize the Zeeman gap noticeably. This is why local electrostatic tuning of the $g$-factor for electrons has been attempted by other methods, e.g., by engineering of materials with composition gradients, exploiting interactions with magnetic impurities or nuclear spins, or utilizing a combination of SOI and the Stark shift. In silicon and germanium devices confining holes, weak SOI permits only a small degree of electrical control of the $g$-factor via engineering of valence subband mixing.

Here we demonstrate an electrically tunable effective $g$-factor, $g_{\text{eff}}$. In a laterally gated GaAs double quantum dot (DQD) confining one hole, in the strong interdot coupling regime. In this system, the spin-flip tunneling due to strong SOI introduces orbital (charge-transfer) effects. Consequences of strong level hybridization have been investigated previously in electron devices, where spin relaxation hotspots were detected in Si-based single dots and GaAs double dots, and valley orbit mixing effects were explored in Si dots. Here we demonstrate that the SOI-induced level hybridization alters the energy gap separating the levels involved in the microwave absorption (the EDSR gap), making $g_{\text{eff}}$ voltage dependent. Moreover, the SOI is sufficiently strong that one can utilize EDSR as a spectroscopic tool to map out the level structure of the DQD over a broad range of magnetic fields and gate voltages, which we carry out by electrical transport measurements. We engineer both the spectral content of the states and the effective $g$-factor by controlling the amount of mixing between different orbitals of the DQD. This is accomplished by adjusting the coupling and detuning between dots and the external magnetic field. The voltage dependence of $g_{\text{eff}}$ paves the way toward protocols for coherent control of single-hole spins involving fast voltage tuning of the EDSR gap character: from purely spin-like, conducive for long spin relaxation times, to a spin-charge hybrid, enabling fast coherent rotations in a constant magnetic field and under continuous microwave modulation.
Further model details, including the dependence of the energies of the four states on the interdot detuning and the magnetic field are presented in Supplementary Notes 1 and 2.

Resonant microwave absorption. Figure 2a shows a derivative of the single-hole high-bias transport triangle in the presence of monochromatic microwave modulation with frequency $f = 37.12$ GHz applied to gate $R$ at $B = 2.5$ T. Compared with Fig. 1e, we detect two new resonance lines (labeled I and II) resulting from the EDSR process, whereby microwaves are resonantly absorbed and the hole is promoted from GS to ES1. The arrangement of levels occurring at point b on the resonance line I is shown in Fig. 2b. Here the GS lies below the Fermi energy of the drain, i.e., the system is in the Coulomb blockade regime. Indeed, point b lies outside the transport triangle, where the current in the absence of microwaves is blocked. However, as ES1 lies above the Fermi energy of the drain, resonant microwave absorption lifts the Coulomb blockade resulting in current flow. This alignment of levels is analogous to that in the spin readout technique of Elzerman et al.\cite{Elzerman2004}. Without SOI, the...
microwave frequency \( f \) at which the EDSR signal occurs would correspond to the Zeeman energy \( E_x = g^* \mu_B B \), with \( g^* \) being the bulk effective \( g \)-factor and \( \mu_B \) being the Bohr magneton. However, the presence of strong SOI leads to noticeable repulsion of levels ES1 and ES2. As a result, the spin gap is renormalized and can be characterized by an effective \( g \)-factor defined as \( g_{\text{eff}} = \hbar f / \mu_B B \), with \( \hbar \) being Planck’s constant. The degree of renormalization clearly depends on the energy separation between ES1 and ES2, which can be controlled by adjusting the detuning, hence \( g_{\text{eff}} \) becomes voltage tunable. Figure 2a also reveals the second resonance line (II) parallel to I and equidistant from the charge transfer line \( \varepsilon = 0 \) but located at opposite (negative) detuning. The alignment of DQD levels at point c on that line is shown in Fig. 2c. At this detuning, without strong hybridization the energies of the left-dot orbitals of each spin would be higher than their right-dot counterparts, making it an energy blockade condition, and no current could flow. It should be noted that in our previous study we found that the rate of tunneling from GS into the drain (dashed arrow) is smaller than that from ES1 (solid arrow). This is because the tunneling barrier is not rectangular but is wider (narrower) at lower (higher) energies. The absorption of microwaves results in the hole being promoted from GS to ES1 and therefore in an enhancement of the tunneling current. This difference in the tunneling rates is necessary to observe current enhancement along resonance line II at the EDSR condition.

**Charge-like and spin-like regimes.** The central observation in Fig. 2 is that the EDSR signal occurs for specific detuning conditions set by the gate voltages. If the gap probed by microwaves were purely related to spin, i.e., defined only by a constant bulk \( g \)-factor, the resonance condition would depend only on the magnetic field and be voltage independent. To explore the voltage tuning of the EDSR, we follow the position of resonance line in the Coulomb blockade region, along the dashed yellow line in Fig. 3a, where we plot \( I_{\text{DOT}} \) at \( B = 2.5 \) T and microwave frequency \( f = 40.62 \) GHz. This line is equivalent to the line I in Fig. 2a. Figure 3b shows the magnetic field dependence of the detuning corresponding to the EDSR signal observed on scanning the gate voltages along the yellow dashed line in Fig. 3a, moving toward the \( \varepsilon = 0 \) charge transfer line denoted by the white dashed line. This procedure changes the detuning between the dots, keeping GS below the Fermi energy of the drain and ES1 above it. For a voltage-independent EDSR signal, we would expect the peak to occur only for one value of \( B \). However, we recover the EDSR peak at other magnetic fields, and the corresponding detuning position is magnetic field dependent. We analyze this dependence in detail in Supplementary Note 3. As examples, here we plot for \( B = 2.4 \) T and \( B = 3.0 \) T the calculated eigenenergies of the system with solid lines as a function of the detuning (Fig. 3c, d, respectively) with system parameters obtained from the experimental data (see “Methods”). The Hamiltonian parameters in Fig. 3c are \( t_N = 62.7 \) \( \mu \)eV, \( t_F = 42.0 \) \( \mu \)eV, and \( g^* = 1.24 \). While in Fig. 3d we use \( t_N = 54.5 \) \( \mu \)eV, \( t_F = 12.9 \) \( \mu \)eV, and \( g^* = 1.18 \). These two cases correspond to the two points, c and d, identified in Fig. 3b. In each diagram, for positive detuning, we find a unique match of the microwave energy (green bar) to the GS–ES1 gap, at the detuning 425 and 130 \( \mu \)eV, respectively, which accurately corresponds to the experimental resonance position in Fig. 3b. Here the crucial feature is the anticrossing of energies of ES1 and ES2 caused by the
spin-flip tunneling process. If this tunneling is switched off, the energy levels correspond to bonding and antibonding quantum molecular states with definite spin: down (blue dashed lines) and up (red dashed lines). However, in the presence of the spin-flip tunneling the ES1 and ES2 anticross with the corresponding mixing of spin states. The anticrossing defines two clear regimes for EDSR. For sufficiently high detuning, as in Fig. 3c, the resonance is spin-like, as it connects GS and ES1 characterized predominantly by opposite spins. For small detuning, as in Fig. 3d, the excitation is charge-like, as the resonance connects GS and ES1 mostly with the same spin but differing by hole charge distribution. Interestingly, in the small detuning limit, at large magnetic field, the signal observed is essentially driven by photon-assisted tunneling rather than EDSR. At intermediate detuning, the excitation is of a hybrid (spin-charge) nature. The effective factor $g_{\text{eff}}$ is therefore a three-dimensional parameter being both voltage and magnetic field dependent. The top axis in Fig. 3b shows the value of $g_{\text{eff}}$ corresponding to the observed EDSR signal at different magnetic fields. We find that in this experiment we are able to tune $g_{\text{eff}}$ by 30% by simultaneous adjustment of the gate voltages and the magnetic field.

**Voltage tuning of the effective $g$-factor.** In Fig. 4, we present examples of the voltage tunability of the EDSR signal by tracking the peak position in $V_I$, as a function of microwave frequency at $B = 1.5$ T (Fig. 4a), and $B = 1.75$ T (Fig. 4b). The smallest gap corresponds to zero detuning (see black double arrows in Fig. 3c, d). In Fig. 4c, we show the microwave frequency corresponding to this minimal gap (see green symbols in Fig. 4a, b) as a function of magnetic field. With the EDSR bridging the pure Zeeman gap, we would expect this dependence to be purely linear, with the slope corresponding to the material bulk $g$-factor, $g^*$. This linearity is clear for small magnetic field (region (1)). However, from its slope we extract $g_{\text{eff}} = 0.85$, which is 30% lower than the bulk value $g^* = 1.35$ measured by us previously, reflecting the strong spin-gap renormalization. As the magnetic field grows, the relationship becomes nonlinear, and at point (2) it reaches a maximum, corresponding to the onset of the anticrossing of ES1 and ES2 levels. Here the interpretation in terms of a renormalized spin gap reaches its limit even though $g_{\text{eff}}$ is finite. From Fig. 3d, one might expect that at even higher magnetic field the EDSR bridges the gap between states of the same spin, i.e., it is a charge-like excitation, and that the dependence in Fig. 4c should become magnetic field independent. However, in region (3) we find that the GS–ES1 gap decreases with increase of magnetic field. This is due to the decrease of both tunneling matrix elements resulting from diamagnetic effects as described in our previous work for much weaker interdot coupling.

**Fig. 3 Tuning effective $g$-factor with gate voltages.** a The high-bias transport triangle ($V_{SD} = +1$ mV) for a single hole with the electric dipole spin resonance (EDSR) feature at $B = 2.5$ T and microwave frequency $f = 40.62$ GHz. The detuning $\epsilon$ of the dots is swept along the yellow dashed line toward the charge transfer line $\epsilon = 0$ (white dashed line). b The detuning position of the EDSR peak and the smooth transition from a spin-like to a charge-like character as a function of the magnetic field (note smaller detuning is toward the top of the plot). Since the frequency is kept constant, the presence of an EDSR signal for different magnetic fields indicates an effective $g$-factor $g_{\text{eff}} = hf/\mu_B B$ (see top axis) that is voltage tunable. c, d The energies of the four levels GS, ES1, ES2, and ES3 as a function of detuning at $B = 2.4$ and $B = 3$ T, respectively. The energy gap of 168.0 $\mu$eV, corresponding to $f = 40.62$ GHz, is marked in each panel with a green bar. The EDSR signal at the two green dots marked in b occurs for the cases depicted in c, d. In the two lower panels, the minimal gap corresponding to zero detuning is marked by a black double arrow. The dashed lines show energies of spin-down (blue) and spin-up (red) states calculated for the case when spin-flip tunneling is absent. For each spin projection, the low-energy (high-energy) state can be identified as the bonding (antibonding) quantum molecular orbital. The black solid lines denote energies in the presence of spin-flip tunneling.
dependence of the EDSR frequency on magnetic field, however, now at large detuning, i.e., in spin-like regime. Here the dependence appears to be linear: at large detuning the ES1–ES2 anticrossing is expected only at much higher magnetic field. Owing to a much weaker renormalization by ES2, the extracted value of $g_{\text{eff}} = 1.25 \pm 0.03$ is much closer to the bulk value of $g^*$. We stress that $g_{\text{eff}}$ here is not determined from the slope of the dependence but by calculating it as $g_{\text{eff}} = hf / \mu_B B$ at a number of points. We note that in Fig. 4a, b we track the EDSR peaks on both sides of the charge transfer line in Fig. 2a, i.e., we observe a response at both positive and negative detuning (resonance lines I and II). This allows us to fit the experimental data (symbols) to the four-level model (Fig. 4c, solid lines) as a function of the microwave frequency and magnetic field and extract the system parameters (see “Methods” for details). The dependence of the tunneling matrix elements $t_N$ (spin conserving) and $t_F$ (spin flipping) on the magnetic field shown in Fig. 4f indicates a clear decrease in their magnitude due to the diamagnetic squeezing of the single-dot orbitals.

**Discussion**

Our work demonstrates that the single-hole GS–ES1 gap is tunable on adjusting the detuning between the dots and, in addition, is sensitive to the magnetic field through the Zeeman energy. Importantly, this gap can be probed by EDSR across the regimes...
of charge, spin, and hybrid excitations at a constant magnetic field by tuning the microwave frequency. The physics can be described in terms of a voltage-tunable g-factor, \( g_{\text{eff}} \). The identification of different EDSR regimes also enables electrical control of the spin relaxation time \( T_1 \), which may be useful, for example, for spin state initialization. Indeed, in the spin-like regime (region (1) in Fig. 4c) relaxation requires a higher-order process involving the SOI and phonons, and thus relaxation here has a longer timescale compared to that in the charge-like regime (region (3)) in which it is only mediated by the phonons. Spin qubit manipulation during the coherent control phase of a quantum algorithm should be undertaken with the longest possible \( T_1 \). Moreover, we expect shorter values of the decoherence time \( T_2^* \) in the charge-like regime owing to a strong dependence of the gap on the detuning, while in the spin-like regime the gap becomes voltage independent and is therefore less susceptible to the charge noise. This expectation follows from the gap behavior in the energy diagrams in Fig. 3c, d and is reflected by the EDSR signal position approaching a vertical asymptote in Fig. 4a, b, e.

The voltage dependence of the g-factor paves the way for local addressability of spin qubits in multi-qubit systems required for fast single-qubit addressability. This property removes the need of strongly focused and pulsed magnetic fields for single-qubit rotations or local nonuniform magnetic fields created by micromagnets. These magnetic techniques for implementing single-qubit operations are typically slow, with characteristic times comparable to the spin relaxation time \( T_1 \), and require complex material engineering. A system with a voltage-tunable g-factor may be manipulated at a constant magnetic field and with small, monochromatic, global microwave modulation by tuning the transition in and out of resonance through adjustment of the detuning, which can be fast. An optimal qubit design combining the advantages of the different regimes identified in our work and with optimized decoherence times would be a two-level system demonstrated for the charge qubit in Si\( _4 \), where the cavity-coupling between the qubit and the cavity photon was recently optimized\[50\]. The two-qubit operations in the time domain. The hole-based device makes it an ideal candidate for the long-range qubit coupling design, in which the two-qubit operations are

\[ T_1 = \frac{\alpha_L V_L}{2} + \frac{\alpha_R V_R}{2}, \]

\[ g_{\text{eff}} = \frac{a_L V_L}{a_R V_R}. \]

with the lever-arms parameters \( a_L = 63.4 \, \text{meV/V}, a_R = 33.9 \, \text{meV/V}, \)

\[ a_L = 46.7 \, \text{meV/V}, \quad a_R = 62.2 \, \text{meV/V}. \]

The goal is to determine the tunneling matrix elements \( t_{\text{F}, \text{F}'} \) and the bulk g-factor \( g' \) (which is the limit of experimentally measured \( g_{\text{eff}} \) at large detuning) by fitting experimental results to the theoretical model. Bearing in mind that \( t_{\text{F}} \) and \( t_{\text{F}'} \) may depend on the magnetic field, we focus on the datasets showing the position of the EDSR peak as a function of the microwave frequency (see Fig. 4a, b) as they are recorded at constant \( R \). For each dataset, we (i) extract the voltages \( V_L, V_R \) and frequency \( f \) at which we observe EDSR peaks, (ii) translate these values into energy units as described above, and (iii) fit all the data points with the calculated gap between the GS and ES\( _1 \) in our model parametrized with a single set of parameters \( t_{\text{F}}, t_{\text{F}'} \) and \( g' \). We adjust the values of these parameters using a genetic algorithm to optimize the quality of the fit. We repeat this procedure for several magnetic fields and allow the parameters to vary from one magnetic field to another. The results of the fitting procedure are presented in Fig. 4c of the main text and show extracted values of \( t_{\text{F}} \) and \( t_{\text{F}'} \) in Fig. 4f of the main text. We observe a clear decrease of both tunneling matrix elements as a function of magnetic field. This decrease is due to the diamagnetic effects as described in our previous work but for much weaker interdot coupling\[53\] (\( t_{\text{F}}, t_{\text{F}'} \approx 0.2 \, \text{eV} \)). As for the bulk g-factor \( g' \), the fitted values are found to lie within \( \pm 5\% \) of \( g' = 1.2 \). This value is somewhat smaller than the value of \( g' = 1.35 \) obtained by a magneto-transport tunneling measurement in our earlier study\[54\]. This difference may be due to details of sample tuning as well as the model not accounting for higher-order effects such as a small difference in the \( g' \) of the two dots or dependence of the tunneling matrix elements \( t_{\text{F}} \) and \( t_{\text{F}'} \) on the detuning.

Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Received: 19 June 2019; Accepted: 25 November 2019;
Published online: 13 December 2019

Methods

Device details. The experiments were performed on a DQD device fabricated from an undoped Ga\( _{1-x}\)Al\( _x \)As/Al\( _x \)Ga\( _{1-x} \)As (\( x = 90\% \)) heterostructure employing lateral split-gate technology\[50\]. A suitable DQD potential profile was defined by lateral surface Ti/Au gates. The two-dimensional hole gas (2DHG) was created by a global gate deposited above the structure separated by a 110-nm-thick Al\( _2 \)O\( _3 \) dielectric layer grown by atomic layer deposition. Left and right plunger gates, labeled in Fig. 1a as \( L \) and \( R \), respectively, were used to tune the hole energy levels in each dot, while the gate \( C \) was used to adjust the interdot tunneling barrier. The device was cooled down in a dilution refrigerator. The nominal lattice temperature was 60 mK. The charge state of the DQD was probed using the quantum point contact charge sensor with a constant source-drain bias voltage of 100 mV. The magnetic field was applied in the direction normal to the plane of the 2DHG. An Anritsu microwave signal generator Model 69377B was used and the nominal microwave power at source was typically between \(-20\) and \(-10\) dBm. In addition, the microwave signal was attenuated by \(-20\) dB at the 1K stage\[55\].

Fitting to experimental data. The EDSR data presented in the main text allow us to map out the gap between the two lowest-energy spinor states as a function of the magnetic field \( B \), the microwave frequency \( f \), and the detuning defined by voltages \( V_L \) and \( V_R \) (yellow dashed line in Fig. 3a). We convert the microwave frequency into energy \( \hbar f = hB \), where \( h \) is Planck’s constant. However, to establish a link between our theoretical model and the experimental data we need to express the onsite energies \( E \) and \( g' \) of the dots in terms of the gate voltages. We do this by utilizing lever arm parameters extracted from Coulomb diamond measurements (data not shown). \( t_{\text{F}} \) and \( t_{\text{F}'} \) can be expressed as

\[ t_{\text{F}} = \frac{\alpha_L V_L}{2} + \frac{\alpha_R V_R}{2}, \]

\[ t_{\text{F}'} = \frac{\alpha_L V_L}{2} + \frac{\alpha_R V_R}{2}, \]

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Acknowledgements
S.S. and A.B. thank the Natural Sciences and Engineering Research Council of Canada for financial support. M.T and Y.H thank Tohoku University GP-Spin program for support. This work was performed in part at the Center for Integrated Nanotecnologies, a U.S. DOE, Office of Basic Energy Sciences user facility, and Sandia National Laboratories, a multi-mission laboratory managed and operated by National Technology and Engineering Solutions of Sandia, LLC, a wholly owned subsidiary of Honeywell International, Inc. under the U.S. Department of Energy’s National Nuclear Security Administration under contract DE-NA-0003525. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

Author contributions
M.T. and S.S. performed the measurements; M.T., S.S., M.K., D.G.A., L.G., J.D., A.P.-B., A.B., A.S., and Y.H. analyzed and discussed the data; S.S., M.K., D.G.A., and A.S. wrote the paper; P.Z. provided maintenance of the cryogenic and measurement equipment; L.T., J.R., and T.H. fabricated and characterized the device.

Competing interests
The authors declare no competing interests.

Additional information
Supplementary information is available for this paper at https://doi.org/10.1038/s42005-019-0262-1.

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