Zero field splitting of heavy-hole states in quantum dots

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Using inelastic cotunneling spectroscopy, we observe a 55μeV zero field splitting in the spin triplet manifold of Ge hut wire quantum dots. The degeneracy of the heavy hole triplet state is lifted since the interplay of strong spin orbit coupling and strong confinement leads to a preferred direction of the heavy-hole pseudospin. The reported effect should be observable in a broad class of strongly confined hole quantum-dot systems and needs to be considered when operating hole spin qubits.

Hole states in semiconductor quantum dots have gained increasing interest in the past few years as promising candidates for spin qubits due to their strong spin orbit coupling (SOC) [1–3]. The SOC allows a full-fledged electrical control of the hole spins [4–7] either via the electric-dipole spin resonance [8], g-tensor modulation [9] or both of them [10]. Further, Rabi frequencies exceeding 100MHz have been reported [5, 6] and single-shot reflectometry measurements have revealed spin relaxation times approaching 100μs at 500mT [11], underlying the potential of hole spins as viable qubits.

Despite the fact that a hole is simply a missing electron, their spins behave strikingly different than their electron counterparts [12]. While the electron spin does not correlate with the direction of motion in typical semiconductors given their weak SOC [Fig. 1(a)], the Luttinger-Kohn Hamiltonian [13, 14], which describes holes near the Γ point of the valence band, imposes a coupling between the momentum and the hole pseudospin already for bulk materials: the pseudospin points in the same direction as the momentum [Fig. 1(b)]. By introducing a strong confinement potential creating a quantum well, the heavy-hole (HH) light-hole (LH) degeneracy is lifted and the pseudospin changes its direction. For the HH states, which become energetically favorable, the pseudospin points perpendicular to the momentum, i.e. in the direction of strong confinement [Fig. 1(c)] [12]. This implies that HHS confined in quasi two dimensional quantum dots (QDs), i.e. artificial atoms with strong confinement in one dimension, show spin anisotropy and could thus manifest similar effects as atoms show in the presence of a magnetocrystalline anisotropy, i.e. a magnetic anisotropy leading to a zero field splitting (ZFS). However, to the best of our knowledge, hitherto no work provided evidence of a ZFS for confined HH states in quantum dots.

For adatoms, on the other hand, ZFS studies have been at the focus of intense research as the magnetic anisotropy provides directionality and stability to the spin, which is the key for realizing nanoscale magnets. Scanning tunneling microscopy measurements have been used to reveal the magnetic anisotropy for several adatoms on surfaces and to understand how the local environment can influence it [15–19]. ZFS as high as 58meV, originating from the atomic spin orbit interaction, have been reported [15].

Here, we use inelastic co-tunneling (CT) as a tool to extract information about confined HH states. A hole-hole interaction strength of 275μeV, similar to that of GaAs is reported. We have furthermore investigated the magnetic anisotropy of HH states confined in quasi two dimensional QDs. A ZFS of up to 55μeV has been measured for the excited triplet states confined in a QD with an even hole occupation. The evolution of the triplet states both for perpendicular and parallel magnetic fields is in very good agreement with the anisotropic spin Hamiltonian.

The QDs used for this study are fabricated in Ge hut wires (HWs) grown by molecular beam epitaxy [20, 21]. These HWs are site-controlled as they are grown on Si wafers with predefined trenches [Fig. 2(a)]. The detailed description of the growth conditions can be found in Ref. [20]. They have a height of about 3.8nm and a width of approximately 38nm. Due to the strong confinement and compressive strain, the degeneracy between the HH and LH is lifted, leading to confined HH states [22, 23]. The HWs are contacted by two 25-nm-thick Pt electrodes, acting as source and drain, with a 50-nm spac-
ing between them. The gate electrode consists of 3-nm Ti plus 25-nm Pt, and is separated from the source and drain contacts by hafnium oxide, deposited in 80 cycles of atomic layer deposition [Fig. 2(b)]. Two nominally identical devices have been investigated in this study.

At low temperatures, transport through QDs is dominated by Coulomb blockade (CB), which leads to single electron transport. The stability diagram of a QD device with the characteristic Coulomb diamonds can be seen in Fig. 2(c). However, due to second-order elastic CT processes the conductance within the coulomb diamonds does not drop to zero [24, 25]. At zero magnetic field, once the energy due to the bias voltage across the QD exceeds the orbital level separation, \(|eV_{SD}| > E_{ORB}\), the inelastic CT process leaves the QD in the excited orbital state (\(e > 0\) denotes the elementary charge). The onset of inelastic CT is observed as a step in the differential conductance, \(dI/dV_{SD}\), at \(eV_{SD} = \pm E_{ORB}\) [24, 25], indicated by black arrows in Fig. 2(c).

Inelastic CT is an excellent tool for magnetotransport spectroscopy measurements as the step width is not lifetime limited but depends only on the effective temperature [25]. We first use it to extract information related to the strength of hole-hole interactions within a QD. When a QD confines an odd number of holes, the ground state is a (doubly degenerate) spin-doublet. On the other hand, with an even number of holes the ground state of the QD is a singlet state (assuming that the exchange coupling is weaker than the level splitting). Here, the two holes occupy the same (lowest in energy) orbital state with their pseudospins being antiparallel. The first excited states are the triplet states for which one hole occupies a (doubly degenerate) spin-doublet. On the other hand, with an even number of holes the ground state of the QD is a singlet state (assuming that the exchange coupling is weaker than the level splitting). Here, the two holes occupy the same (lowest in energy) orbital state with their pseudospins being antiparallel. The first excited states are the triplet states for which one hole occupies a higher orbital. This costs a higher energy for the orbital occupation but also gains some Coulomb repulsion and exchange energy compared to the singlet state [28, 29]. By comparing the difference between the singlet-triplet energy \(E_{ST}\) and the orbital level separation \(E_{ORB}\), one can thus obtain useful information about the strength of hole-hole interactions.

In order to conclude about the even/odd occupancy of the QD we investigate the evolution of the CT steps. For an odd number of holes, a magnetic field \(B\) lifts the spin
degeneracy of the doublet state by the Zeeman energy $E_Z = g\mu_BB$, where $g$ and $\mu_B$ are the hole $g$-factor and Bohr magneton, respectively. Once the energy due to the bias voltage across the QD exceeds the Zeeman energy, $|eV_{SD}| > E_Z$, the inelastic CT processes can flip the QD spin, leaving the QD in the excited spin state. This is visible as a step in Fig. 3(a). For zero magnetic field the observed feature corresponds to the first orbital excited state from which an orbital level separation $E_{ORB}$ of $690\mu$eV can be extracted. For an even number of holes, the magnetic field should split the three triplet states, and three inelastic cotunneling steps should be observed (note that the other state involved, the ground state singlet, does not split in a magnetic field). Indeed this behavior can be observed in Figs. 3(b) and (d). In this case the feature at zero field corresponds to the energy of the triplet states. The measured singlet triplet energy separation $E_{ST}$ is $415\mu$eV. The difference $E_{ORB} - E_{ST} = 275\mu$eV corresponds to the Coulomb interaction energy and is similar to what has been reported for GaAs QDs [28].

By inspecting carefully the behavior of the triplet states in Fig. 3(b), it can be seen that the triplets are not equally spaced (Fig. 3(d)); it actually seems that the three triplet states (marked by three dashed lines) are not degenerate at $B = 0\ T$ (this will become even more explicit when plotting the second derivative later). As has been recently shown, self-organized Ge HWs have a rather strong SOC similar to III-V materials [20]. As a consequence, it is preferable for the spin to point in the confinement direction as depicted in Fig. 1(c). In the spin Hamiltonian for the triplet state

$$H = -J/2\ SS + g_\perp\mu_B S_\perp B_\perp + g_\parallel\mu_B S_\parallel B_\parallel - DS^2_\perp,$$  

(1)

this is taken into account by an uniaxial anisotropy $D$. Note that this Hamiltonian only describes the effective triplet spin $S$, with $S_\perp$ and $S_\parallel$ being the projections in the $\perp$- and $\parallel$-direction of Fig. 2(b); $B_\perp$ and $B_\parallel$ denote the magnetic field strength in these directions which couple through different (anisotropic) $g_\perp$ and $g_\parallel$-factors; finally $J$ is the isotropic exchange.

The magnetic anisotropy term, $DS^2_\perp$, makes it preferably by an energy $D$ to align the spin-1 in the $\perp$-direction. At this stage of an effective Hamiltonian for the triplet state, we cannot distinguish the origin of this magnetic anisotropy. It might be due to (i) shape anisotropy caused by dipole interactions [30], (ii) single ion (single quantum dot) anisotropy caused by SOC-induced tran-
sitions to excited (virtual) states \( S_{1} \) or (iii) a SOC-induced anisotropic exchange \( J_A \) between two effective spin-1/2 holes on two different orbitals (1 and 2), i.e., \( H = -J_S S_1 S_2 - J_A S_{1\perp} S_{2\perp} \), which for \( S = S_1 + S_2 \) reduces to Eq. (4) with \( D = J A / 2 \), up to a constant.

Let us also note that the triplet state gains not only the Coulomb exchange \( J \) compared to the singlet state, but also a reduced Coulomb repulsion energy [which is beyond the effective spin Hamiltonian (1)]: the two holes of the triplet are in two different orbitals and hence further apart than the holes in the singlet state on the same orbital. On the other hand, we have to occupy an orbital at a higher energy, so that altogether the triplet state is \( E_{ST} \) higher in energy than the singlet state.

The eigenstates of Hamiltonian (1) can be easily calculated and are shown in Fig. 3 for a magnetic field applied once in the \( \perp \)- and once in the \( \parallel \)-direction. For \( B = 0 \) the two states with \( S_{\perp} = \pm 1 \) have a by \( -D \) smaller energy than the third triplet state with \( S_{\parallel} = 0 \). Hence, the lowest triplet state is doubly degenerate and the remaining one singly degenerate in Fig. 3. Applying now a magnetic field in the anisotropy direction \( B_\parallel \) Zeeman-splits the doublet and leaves the singly degenerate \( S_{\perp} = 0 \) state untouched (Fig. 3(a)).

The situation with the magnetic field \( B_\parallel \) orthogonal to the anisotropy direction is somewhat more complicated. Here, for small \( B_\parallel \), the eigenstates are still predominantly \( S_{\perp} = \pm 1,0 \) with only a small, perturbative readmixture \( \sim g_\parallel \mu B_\parallel / D \) as the magnetic field tries to align the spins in the \( \parallel \)-direction. This linear readmixture of the eigenstates, leads to a quadratic change of the energy eigenvalues in Fig. 3(b) for \( g_\parallel \mu B_\parallel \ll D \). For large \( g_\parallel \mu B_\parallel \gg D \), the usual Zeeman splitting of the triplet states into \( S_{\parallel} = \pm 1,0 \) is recovered as the HH pseudo spins now reorient along \( B_\parallel \).

These considerations clearly show that there is a ZFS and that the magnetic field dependence shows a quite different behavior for \( B_\parallel \) and \( B_\perp \). If we have an odd number of electrons, the doublet could also be described with Hamiltonian (1). But in this case, both \( S_{\perp} = \pm 1/2 \) states have the same anisotropy energy. Hence there is a Zeeman splitting but no ZFS as observed in Fig. 3(a).

In order to further elucidate this behavior of the triplet state, we study in Figures 3(c), (d) the dependence on a magnetic field \( B_\perp \). Using a second derivative to sharpen the features, it can be seen even more clearly that the HH triplet states are not degenerate at \( B = 0 \). Even more, the magnetic field evolution perfectly fits with that of Fig. 3(a), which is also indicated as dashed (white) lines. Figures 3(e)-(f) show the same split degeneracy also for a second device. In this case orbital effects also lead to a slight bending of the states for \( B_\perp \) and the ZFS is extracted to be 30\( \mu \)eV. Except for this extra bending Fig. 4(e) resembles Fig. 4(a) and Fig. 4(f) resembles Fig. 4(b) for the two different magnetic field directions. From the observed splitting it is obvious that the ZFS needs to be taken into account when considering the energy band diagram of double QDs, for which it has been assumed so far, that triplet HH states are all degenerate at \( B = 0 \).

In conclusion, we have demonstrated the zero field splitting for heavy hole states confined in a two dimensional quantum dot. Specifically, the triplet states are split into a double and a single degenerate level because of the strong spin orbit coupling and the strong confinement. This is both fundamentally interesting and important when operating heavy hole qubits [5-6], which are gaining significant interest in the past few years.

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[1] R. Winkler, ed., *Spin-Orbit Coupling Effects in Two-Dimensional Electron and Hole Systems* (Springer New York, 2003).

[2] C. Kloeckel, M. Trif, and D. Loss, Phys. Rev. B 84, 195314 (2011).

[3] L. Hong, E. Marcellina, A. Hamilton, and D. Culcer, Phys. Rev. Lett. 121, 087701 (2018).

[4] R. Maurand, X. Jehl, D. Kotekar-Patil, A. Corna, H. Bouhuslavsiky, R. Laviéville, L. Hutin, S. Barraud, M. Vinet, M. Sanquer, and S. De Franceschi, Nature Com. 7, 13575 (2016).

[5] H. Watzinger, J. Kukučka, L. Vukušić, F. Gao, T. Wang, F. Schäffler, J. J. Zhang, and G. Katsaros, Nature Com. 9, 3902 (2018).

[6] N. W. Hendrickz, D. P. Franke, A. Sambak, G. Scappucci, and M. Veldhorst, (2019), arXiv:1904.11443 (2019).

[7] A. Crippa, R. Ezzouch, A. Aprá, A. Amisse, R. Laviéville, L. Hutin, B. Bertrand, M. Vinet, M. Ur-dampilleta, T. Meunier, M. Sanquer, X. Jehl, R. Maurand, and S. De Franceschi, Nature Com. 10, 2776 (2019).

[8] V. N. Golovach, M. Borhani, and D. Loss, Phys. Rev. B 74, 165319 (2006).

[9] Y. Kato, R. C. Myers, D. C. Driscoll, A. C. Gossard, J. Levy, and D. D. Awschalom, Science 299, 1201 (2003).

[10] A. Crippa, R. Maurand, L. Bourdet, D. Kotekar-Patil, A. Amisse, X. Jehl, M. Sanquer, R. Laviéville, H. Bouhuslavsiky, L. Hutin, S. Barraud, M. Vinet, Y.-M. Niquet, and S. De Franceschi, Phys. Rev. Lett. 120, 137702 (2018).

[11] L. Vukušić, J. Kukučka, H. Watzinger, J. M. Milem, F. Schäffler, and G. Katsaros, Nano letters 18, 7141 (2018).
[12] C. Kloeffel, M. J. Rančić, and D. Loss, Phys. Rev. B 97, 235422 (2018).
[13] J. M. Luttinger and W. Kohn, Phys. Rev. 97, 869 (1955).
[14] J. M. Luttinger, Phys. Rev. 102, 1030 (1956).
[15] I. G. Rau, S. Baumann, S. Rusponi, F. Donati, S. Stepanow, L. Gragnaniello, J. Dreiser, C. Piamonteze, F. Nolting, S. Gangopadhyay, O. R. Albertini, R. M. Macfarlane, C. P. Lutz, B. A. Jones, P. Gambardella, A. J. Heinrich, and H. Brune, Science 988, 344 (2014).
[16] P. Jacobson, T. Herden, M. Muenks, G. Laskin, O. Brovko, V. Stepanyuk, M. Ternes, and K. Kern, Nature Com. 6, 8536 (2015).
[17] T. Miyamachi, T. Schuh, T. Mrkl, C. Bresch, T. Balashov, A. Sthr, C. Karlewski, S. André, M. Marthaler, M. Hoffmann, M. Geilhufe, S. Ostanin, W. Hergert, I. Mertig, G. Sch´on, A. Ernst, and W. Wulfhekel, Nature 503, 242 (2013).
[18] P. Gambardella, S. Rusponi, M. Veronese, S. S. Dhesi, C. Grazioli, A. Dallmeyer, I. Cabria, R. Zeller, P. H. Dederichs, K. Kern, C. Carbone, and H. Brune, Science 300, 1130 (2003).
[19] C. F. Hirjibehedin, C. Y. Lin, A. F. Otte, M. Ternes, C. P. Lutz, B. A. Jones, and A. J. Heinrich, Science 317, 1199 (2007).
[20] F. Gao, unpublished.
[21] J. J. Zhang, G. Katsaros, F. Montalenti, D. Scopece, R. O. Rezaev, C. Michel, B. Rellinghaus, L. Miglio, S. De Franceschi, A. Rastelli, and O. G. Schmidt, Phys. Rev. Lett. 109, 085502 (2012).
[22] G. Katsaros, V. N. Golovach, P. Spethis, N. Ares, M. Stoffel, F. Fournel, O. G. Schmidt, L. I. Glazman, and S. De Franceschi, Phys. Rev. Lett. 107, 246601 (2011).
[23] H. Watzinger, C. Kloeffel, L. Vukašić, M. D. Rossell, V. Sessi, J. Kukučka, R. Kirchschlager, E. Lausecker, A. Truhlár, M. Glaser, A. Rastelli, A. Fuhrer, D. Loss, and G. Katsaros, Nano letters 16, 6879 (2016).
[24] A. Kogan, S. Amasha, D. Goldhaber-Gordon, G. Granger, M. A. Kastner, and H. Shtrikman, Phys. Rev. Lett. 93, 166602 (2004).
[25] S. De Franceschi, S. Sasaki, J. M. Elzerman, W. G. van der Wiel, S. Tarucha, and L. P. Kouwenhoven, Phys. Rev. Lett. 86, 878 (2001).
[26] G. Katsaros, P. Spethis, M. Stoffel, F. Fournel, M. Mongillo, V. Bouchiat, F. Leffloch, A. Rastelli, O. G. Schmidt, and S. De Franceschi, Nature Nanotechnology 5, 458 (2010).
[27] A. Nenashev, A. Dvurechenskii, and A. Zinovieva, Phys. Rev. B 67, 205301 (2003).
[28] R. Hanson, L. P. Kouwenhoven, J. R. Petta, S. Tarucha, and L. M. K. Vandersypen, Rev. Mod. Phys. 79, 1217 (2007).
[29] L. P. Kouwenhoven, D. G. Austing, and S. Tarucha, Rep. Prog. Phys. 64, 701 (2001).
[30] For a pedagogical discussion of various magnetic anisotropy terms, see P. Bruno, “Physical Origins and Theoretical Models of Magnetic Anisotropy” in 24. IFF-Ferienkurs Forschungszentrum Jülich (Forschungszentrum Jülich, 3893361103.
[31] See e.g. M. Misiorny, M. Hell, and M. R. Wegewijs Nature Physics 9, 801 (2013).
[32] G. Burkard and D. Loss, Phys. Rev. Lett. 88, 047903 (2002).
[33] Y.-P. Shim, S. Oh, X. Hu, and M. Friesen, Phys. Rev. Lett. 106, 180503 (2011).