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1 Coherent spin-exchange via a quantum mediator

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6 **Coherent interactions at a distance provide a powerful tool for quantum simulation and com-
7 putation. The most common approach to realize an effective long-distance coupling ‘on-chip’
8 is to use a quantum mediator, as has been demonstrated for superconducting qubits ^{1,2} and
9 trapped ions ³. For quantum dot arrays, which combine a high degree of tunability ⁴ with
10 extremely long coherence times ⁵, the experimental demonstration of the time evolution of
11 coherent spin-spin coupling via an intermediary system remains an important outstanding
12 goal ^{6–25}. Here, we use a linear triple-quantum-dot array to demonstrate for the first time
13 a coherent time evolution of two interacting distant spins via a quantum mediator. The two
14 outer dots are occupied with a single electron spin each and the spins experience a superex-
15 change interaction through the empty middle dot which acts as mediator. Using single-shot
16 spin read-out ²⁶ we measure the coherent time evolution of the spin states on the outer dots
17 and observe a characteristic dependence of the exchange frequency as a function of the de-
18 tuning between the middle and outer dots. This approach may provide a new route for
19 scaling up spin qubit circuits using quantum dots and aid in the simulation of materials and**

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20 molecules with non-nearest neighbour couplings such as MnO ²⁷, high-temperature super-
21 conductors ²⁸ and DNA ²⁹. The same superexchange concept can also be applied in cold atom
22 experiments ³⁰.

23 Nanofabricated quantum dot circuits provide an excellent platform for performing both quan-
24 tum computation and simulation using single spins ^{4,31,32}. Many approaches to implementing co-
25 herent spin coupling between distant quantum dots have been proposed using a variety of coupling
26 mechanisms. These include superconducting resonators ⁶⁻⁸, surface-acoustic wave resonators ⁹,
27 floating metallic ¹⁰ or ferromagnetic couplers ¹¹, collective modes of spin chains ¹², supercon-
28 ductors ^{13,14}, Klein tunneling through the valence or conduction band ¹⁵ and superexchange or
29 sequential operations via intermediate quantum dots ¹⁷⁻²¹. A common theme among many of these
30 proposals is to create a coupling between distant spins by virtual occupation of a mediator quantum
31 system. So far, the use of these schemes to show the coherent time evolution of interacting distant
32 spins is lacking. More broadly, there are no experimental realizations so far of direct quantum
33 gates between any type of solid-state spins at a distance.

34 In this Letter we focus on the superexchange interaction to induce spin-spin coupling at a
35 distance. Superexchange is the (usually) antiferromagnetic coupling between two next-to-nearest
36 neighbour spins through virtual occupation of a non-magnetic intermediate state ²⁷. Given that
37 superexchange involves a fourth order process in the hopping amplitude, it is challenging to use
38 it for achieving coherent coupling. This is also the case for several related schemes relying on
39 quantum mediators.

40 We use a linear triple-quantum-dot array with one electron on each of the outer dots, and in-
 41 duce a superexchange interaction through the empty middle dot, which acts as a quantum mediator.
 42 This induces spin exchange of the two distant electron spins. Using repeated single-shot spin mea-
 43 surements we record the coherent time evolution of the spin states on the outer dots. We control
 44 the superexchange amplitude via the detuning of the middle dot electrochemical potential rela-
 45 tive to those of the outer dots, and study the cross-over between superexchange and conventional
 46 nearest-neighbour spin exchange.

47 The dot array is formed electrostatically in a two-dimensional electron gas (2DEG) 85 nm
 48 below the surface of a GaAs/AlGaAs heterostructure, see Fig. 1a. Gate electrodes fabricated on
 49 the surface (see Methods) are biased with appropriate voltages to selectively deplete regions of
 50 the 2DEG and define the linear array of three quantum dots. The left and right dot are each
 51 occupied with one electron, and each of the two electrons constitutes a single spin- $\frac{1}{2}$ particle. The
 52 interdot tunnel couplings are set to ≈ 8.5 GHz (left-middle) and ≈ 11.8 GHz (middle-right).
 53 The sensing dot (SD) next to the quantum dot array is used for non-invasive charge sensing using
 54 radiofrequency (RF) reflectometry to monitor the number of electrons in each dot³³. An in-plane
 55 magnetic field $B_{ext} = 3.2$ T is applied to split the spin-up (\uparrow) and spin-down (\downarrow) states of each
 56 electron by the Zeeman energy ($E_Z \approx 80 \mu\text{eV}$), defining a qubit. The electron temperature of the
 57 right reservoir is ≈ 75 mK.

58 In this system, superexchange can be seen as the result of the effective tunnel coupling t_{SE}
 59 between the outer dots. The amplitude of superexchange, J_{SE} , is approximated by $-\frac{t_{SE}^2}{\epsilon}$, with ϵ

60 the detuning between the electrochemical potentials of the outer dots ³¹, and $\epsilon = 0$ when (1,0,1)
61 and (2,0,0) are degenerate. Here t_{SE} can be described as $t_{SE} = (t_{m,l}t_{m,r})/\delta$, with $t_{m,l}$ ($t_{m,r}$)
62 the tunnel coupling between the middle and the left (right) site and δ the detuning between the
63 electrochemical potential of (1,1,0) and the average of the electrochemical potentials of (1,0,1)
64 and (2,0,0) ³⁴. The superexchange amplitude can thus be approximated as (see Supplementary
65 Information V for the range of validity)

$$J_{SE} = -\frac{t_{m,l}^2 t_{m,r}^2}{\delta^2 \epsilon}, \quad (1)$$

66 which illustrates the characteristic fourth-order hopping process underlying superexchange.

67 To provide direct evidence of coherent superexchange, we will probe the resulting time evo-
68 lution of the two spins via repeated single-shot measurements using spin-to-charge conversion ²⁶.
69 To achieve high read-out fidelities, we work at large magnetic field and perform the spin-to-charge
70 conversion as close as possible to the charge sensor (SD). In previous work, we therefore shuttled
71 electrons consecutively from left to middle to right with no detectable sign of spin flips upon shut-
72 ting ³⁵. Here, we explore a different approach, transferring the spin from left to right with only
73 virtual occupation of the middle dot, using the same long-range tunnel coupling that underlies co-
74 herent superexchange ²⁵. We test the two-spin read-out and long-range spin transfer as described
75 by the schematic diagrams of Fig. 1b and implemented by the pulse sequence depicted by the
76 blue and red arrows in Fig. 1c. Starting from an empty array, we load a random electron from
77 the reservoir into the right dot by pulsing into the charge state (0,0,1). Next we pulse into (1,0,0),
78 whereby the electron is transferred from the rightmost dot to the leftmost dot via a second-order
79 tunnel process across the middle dot. For this transfer we temporarily pulse δ closer to 0 to in-

80 crease the long-range shuttling rate (see Supplementary Information I). Finally, we once more load
81 a random electron in the right dot by pulsing to (1,0,1). We vary the waiting time in (1,0,1) during
82 which spins relax to the spin ground state $|\uparrow 0 \uparrow\rangle$. Then we reverse the pulse sequence and add
83 two calibrated read-out stages denoted by the green circles where spin-to-charge conversion takes
84 place. Fig. 1d shows the measured decays to the ground state spin-up for each of the two spins. We
85 report read-out fidelities of on average 95.9% and 98.0% for spin-down and spin-up respectively,
86 assuming no spin flips during the spin transfer³⁵ (see Supplementary Information III).

87 A key signature of superexchange driven spin oscillations is their dependence on the detun-
88 ing of the intermediate level (δ), see Eq. (1). We have therefore created linear combinations of
89 the gates P_1 , P_2 and P_3 in such a way that we can independently vary δ and ϵ as can be seen in
90 Fig. 2b. Superexchange occurs in the (1,0,1) charge configuration, and the superexchange am-
91 plitude, J_{SE} , increases for less negative ϵ , which translates to an operating point closer to the
92 (2,0,0)-configuration, see Fig. 2a. Similarly, J_{SE} increases with less negative δ , up to the point
93 where we cross the (1,0,1)-(1,1,0) transition indicated by the black dashed line in Fig. 2b and spin
94 exchange between nearest-neighbour dots will dominate (see Fig. 2c). To capture the expected
95 time evolution, we must take into account a difference in Zeeman energies between the two dots,
96 $\Delta E_z = E_{z,3} - E_{z,1}$, arising from slight differences in the g -factor for each dot³⁵. Spin exchange
97 defines one rotation axis, the Zeeman energy difference an orthogonal axis, as shown in the Bloch
98 sphere in Fig. 2d. In the experiment, ΔE_z is fixed, and J_{SE} can be controlled by gate voltage
99 pulses, as we discussed. By adjusting J_{SE} , we can thus define the net rotation axis and rate³⁶. A
100 similar Bloch sphere can be made for the nearest-neighbour regime.

101 The protocol for probing the time evolution is as follows. Starting with an empty array, we
 102 create a mixture of $|\uparrow 0 \downarrow\rangle$ and $|\uparrow 0 \uparrow\rangle$ and move to the position of the red star in Fig. 2b, where J_{SE}
 103 is small compared to ΔE_Z . This is achieved by sequentially loading the two spins as in Fig. 1c, in
 104 this case loading a \uparrow in the left dot and a random spin in the right dot. This procedure allows us
 105 to conveniently create an anti-parallel spin state without using more involved techniques such as
 106 electron spin resonance. Next, following the black dashed arrows in Fig. 1c, we pulse towards the
 107 $(2,0,0)$ regime and wait for several ns. The exact location in detuning space is marked in Fig. 2b
 108 by a red diamond. At this point J_{SE} is sizable, $|\uparrow 0 \downarrow\rangle$ is not an eigenstate of the Hamiltonian and
 109 is thus expected to evolve in time, periodically developing a $|\downarrow 0 \uparrow\rangle$ component ($|\uparrow 0 \uparrow\rangle$ will only
 110 acquire an overall phase). The larger $J_{SE}/\Delta E_Z$, the larger the $|\downarrow 0 \uparrow\rangle$ component. We pulse back
 111 to the position of the red star in $(1,0,1)$ and follow the same spin read-out procedure as was done
 112 for the T_1 -measurement in Fig. 1d. Fig. 2e shows the $|\uparrow 0 \downarrow\rangle$ and $|\downarrow 0 \uparrow\rangle$ probability as a function
 113 of the length of the detuning pulse. We see a sinusoidal dependence, with the $|\uparrow 0 \downarrow\rangle$ and $|\downarrow 0 \uparrow\rangle$
 114 populations evolving in anti-phase, as expected.

115 Returning to the key signature of superexchange, we fix the value of ϵ and vary δ along
 116 the vertical dashed line shown in Fig. 2b. For each choice of δ , we record the four two-spin
 117 probabilities as a function of the length of the detuning pulse (Fig. 3a). Starting from large negative
 118 δ , we first observe no oscillations at all: the superexchange mechanism is suppressed and the
 119 $|\uparrow 0 \downarrow\rangle$ -state remains fixed along the x -axis of the Bloch sphere. As we bring the electrochemical
 120 potential of the intermediate level closer to that of the outer dots, J_{SE} increases in magnitude and
 121 slow oscillations ~ 150 MHz start appearing that are still dominated by $\Delta E_z \approx 130$ MHz between

122 the outer dots, hence the low contrast of the oscillations. The oscillations become faster up to
123 ~ 900 MHz as δ is increased at which point J_{SE} is stronger than ΔE_z and the contrast increases.
124 When δ is further increased, the (1,1,0)-state becomes energetically favourable and the nearest-
125 neighbour exchange between the left and middle dot dominates. Here $\epsilon = -170 \mu\text{eV}$ and this
126 transition occurs around $\delta = 120 \mu\text{eV}$, which is where the black-dashed line in Fig. 2b is crossed.
127 Increasing δ even more enlarges the detuning between the left and middle dot and thereby slows
128 down the nearest-neighbour oscillations, as seen in the data.

129 For a quantitative comparison with the theoretical predictions, we show in Fig. 3b the ex-
130 pected time evolution of the system modeled using the measured nearest-neighbour tunnel cou-
131 plings, detunings δ and ϵ , and the difference in Zeeman energy probed through electric-dipole spin
132 resonance measurements³⁷. We include the effect of dephasing by charge noise³⁶ to match the
133 decay of the oscillations and account for the known read-out fidelities and hyperfine-induced de-
134 phasing⁴ (see Supplementary Information IV). We do not expect hyperfine-mediated electron spin
135 flips in the present operating regime, and hence no dynamical nuclear polarization. Fig. 2e shows
136 that it takes more than 1 ns for the superexchange to be turned on. This is caused by the finite rise-
137 time of the pulses produced by the arbitrary waveform generator and finite bandwidth of the coax
138 lines. The simulation includes this gradual turn on and off of J_{SE} . Comparing Fig. 3a and Fig. 3b
139 we report good agreement between theory and experiment, which supports our interpretation of
140 the data in terms of superexchange, including the transition to nearest-neighbour exchange.

141 In summary, we have demonstrated a first working example of a direct quantum gate between

142 solid-state spins at a distance via virtual occupation of a quantum mediator. This result underlines
143 the utility of arrays of quantum dots for the investigation and application of fundamental physical
144 processes driven by small-amplitude terms and higher-order tunneling. It is possible to extend
145 the distance between the coupled spins using elongated intermediate quantum dots or via different
146 (quantum) mediators altogether. Another interesting direction is to create non-nearest neighbour
147 spin-spin interactions with the centre dot occupied^{20,21,24}, which opens up further new possibilities
148 for quantum computation and modeling of complex materials.

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230 **Author contributions** T.A.B and T.F. performed the experiment and analyzed the data, C.R. and
231 W.W. grew the heterostructure, T.A.B., T.F. and L.M.K.V. contributed to the interpretation of the
232 data, and T.A.B. and L.M.K.V. wrote the manuscript, with comments from T.F.

233 **Additional information** Supplementary information is available in the online version of the paper.
234 Reprints and permission information is available online at www.nature.com/reprints. Correspon-
235 dence and requests for materials should be addressed to L.M.K.V.

236 **Competing financial interests** The authors declare no competing financial interests.

237 **Figure 1 Linear array of three quantum dots and long-range spin transfer**

238 **a** Scanning electron microscopy image of a sample nominally identical to the one used for the
239 measurements. Dotted circles indicate quantum dots and squares indicate Fermi reservoirs in the
240 2DEG, which are connected to ohmic contacts. The RF reflectance of the SD is monitored in order
241 to determine the occupancies of the three dots labeled numbers 1 to 3 from left to right respectively.

242 **b** Read from left to right and top to bottom. The array is initialized by loading two electrons from
243 the right reservoir. The spin that is loaded first is transferred to the left dot via a second-order
244 tunnel process across the middle dot. We load \uparrow -spins by tuning the loading position such that
245 only the \uparrow -spin level is accessible (as in the top left diagram). Random spins are loaded by making
246 both spin levels energetically available (top right). Spin read-out occurs using energy-selective
247 tunneling combined with charge detection via the SD. **c** Charge stability diagram of the triple dot
248 for $M = -412$ mV. Along the L and R axis, we linearly vary the voltages applied to gates P_1 , P_2 and
249 P_3 in such a way that we affect mostly the left and right dots, compensating for cross-capacitances.
250 Similarly, M controls mostly the middle dot (see Supplementary Information II). Labels (n, m, p)
251 indicate the number of electrons in the left, middle and right dot respectively. The middle dot
252 cannot be loaded directly from a reservoir and the left dot is only weakly tunnel coupled to the left
253 reservoir, leading to faintly visible charge transitions (black dotted lines indicate their positions).
254 The pulse sequence for loading and read-out is indicated in the charge stability diagrams via blue
255 and red arrows, see also panel b. The two black dashed arrows denote additional stages to probe
256 superexchange (see Fig. 2). **d** Measured single-spin populations averaged over 8000 cycles per
257 datapoint as a function of waiting time in $(1,0,1)$ for dot 1 (top) and dot 3 (bottom).

258 **Figure 2 Superexchange-driven spin oscillations**

259 **a** Energy diagram as a function of ϵ for $\delta < 0$. The long-range tunnel coupling induces an anti-
260 crossing between the (1,0,1) and (2,0,0) singlet states. The energy difference between T_0 and the
261 hybridized S is denoted J_{SE} . The T_- and T_+ states are split off by B_{ext} . **b** Charge stability diagram
262 in detuning space, allowing individual control of the detuning of the middle dot (δ) and between
263 the outer dots (ϵ), see panel c. **c** Cartoon depicting the transition from superexchange to nearest-
264 neighbour exchange as δ is made more positive. **d** Bloch sphere representation of $S - T_0$ subspace
265 in the superexchange regime with control axes J_{SE} and ΔE_Z . **e** Observation of superexchange-
266 driven spin oscillations. Starting with a mixture of $|\uparrow 0 \downarrow\rangle$ and $|\uparrow 0 \uparrow\rangle$ at the position of the red
267 star in b, we pulse ϵ for a varying amount of time to the position indicated by the red diamond.
268 Afterwards the four two-spin probabilities are measured by averaging over 999 single-shot cycles
269 per datapoint, two of which are shown.

270 **Figure 3 Transition from superexchange to nearest-neighbour exchange**

271 **a** Starting with a mixture of $|\uparrow 0 \downarrow\rangle$ and $|\uparrow 0 \uparrow\rangle$ at the position of the red star in Fig. 2b, we pulse ϵ
272 and δ for a varying amount of time to the position indicated by the vertical dashed line in Fig. 2b.
273 Afterwards the four two-spin probabilities are measured by averaging over 999 single-shot cycles
274 per datapoint. We clearly note the transition of oscillations dominated by ΔE_z ($\delta < -50 \mu\text{eV}$)
275 to increasingly faster superexchange dominated spin evolution and finally ($\delta > 200 \mu\text{eV}$) nearest-
276 neighbour exchange dominated evolution, which slows down as δ is further increased. Acquiring
277 this set of data took ~ 20 hours. **b** Simulation of the data shown in a. The independently determined
278 input parameters are: $t_{m,l} = 8.5 \text{ GHz}$, $t_{m,r} = 11.8 \text{ GHz}$, $E_{z,1} = 19.380 \text{ GHz}$, $E_{z,2} = 19.528 \text{ GHz}$,
279 $E_{z,3} = 19.510 \text{ GHz}$ and the risetime of the detuning pulse is 0.8 ns (see Supplementary Informa-
280 tion IV).

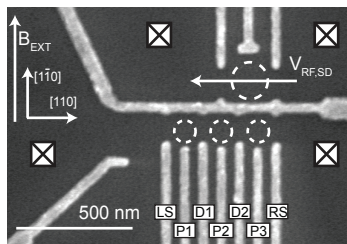
281 **Methods**

282 The experiment was performed on a GaAs/Al_{0.25}Ga_{0.75}As heterostructure grown by molecular-
283 beam epitaxy, with a 85-nm-deep 2DEG with an electron density of $2.0 \cdot 10^{11} \text{ cm}^{-2}$ and mobility
284 of $5.6 \cdot 10^6 \text{ cm}^2\text{V}^{-1}\text{s}^{-1}$ at 4 K. The metallic (Ti-Au) surface gates were fabricated using electron-
285 beam lithography. The device was cooled inside an Oxford Instruments Kelvinox 400HA dilution
286 refrigerator to a base temperature of 45 mK. To reduce charge noise, the sample was cooled while
287 applying a positive voltage on all gates (ranging between 250 and 350 mV)³⁸. The main function
288 of gates *LS* and *RS* is to set the tunnel coupling with the left and right reservoir, respectively. *D*₁
289 and *D*₂ control the interdot tunnel coupling and *P*₁, *P*₂ and *P*₃ are used to set the electron number in
290 each dot. Gates *P*₁, *P*₂, *P*₃ and *D*₂ were connected to homebuilt bias-tees (*RC*= 470 ms), enabling
291 application of d.c. voltage bias as well as high-frequency voltage excitation to these gates. The
292 microwaves were generated using a HP83650A source connected to *P*₂ via a homemade bias-tee at
293 room temperature. Voltage pulses to the gates were applied using a Tektronix AWG5014 arbitrary
294 waveform generator. RF reflectometry of the SD was performed using an LC circuit matching a
295 carrier wave of frequency 111.11 MHz. The inductor is formed from a microfabricated NbTiN
296 superconducting spiral inductor with an inductance of 3.0 μH . The power of the carrier wave
297 arriving at the sample was estimated to be -103 dBm. The carrier signal is only unblanked during
298 read-out. The reflected signal was amplified using a cryogenic Weinreb CITLF2 amplifier and
299 subsequently demodulated using homebuilt electronics. Real time data acquisition was performed
300 using a FPGA (field-programmable gate array DE0-Nano Terasic) programmed to detect tunnel
301 events using a Schmitt trigger.

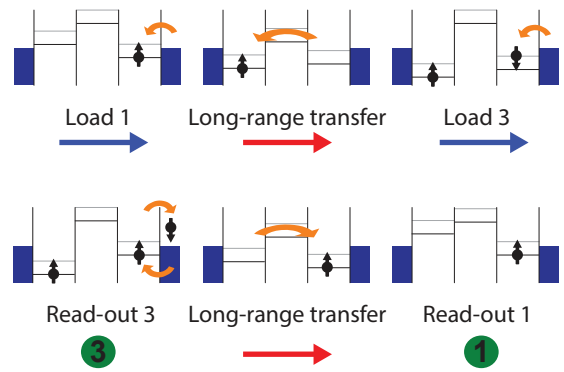
- 302 38. Long, A. R. *et al.* The origin of switching noise in GaAs/AlGaAs lateral gated devices. *Physica*
304 *E* **34**, 553–556 (2006).

Figure 1

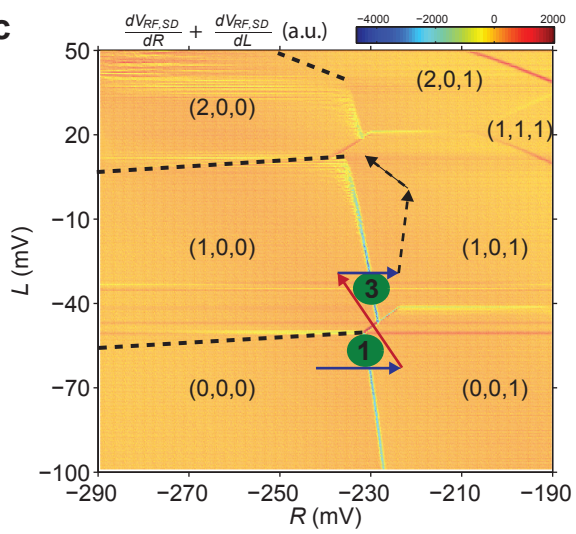
a



b



c



d

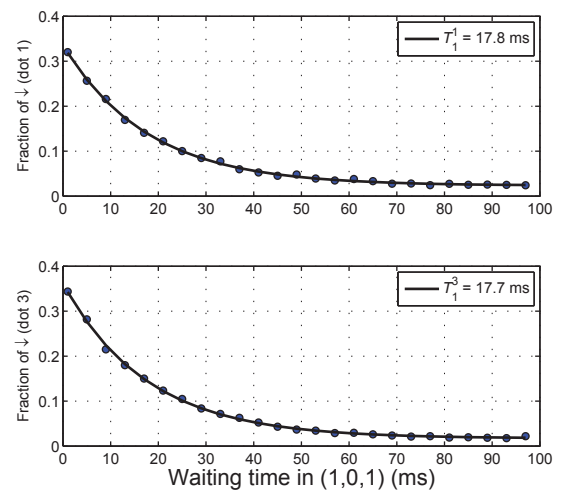


Figure 2

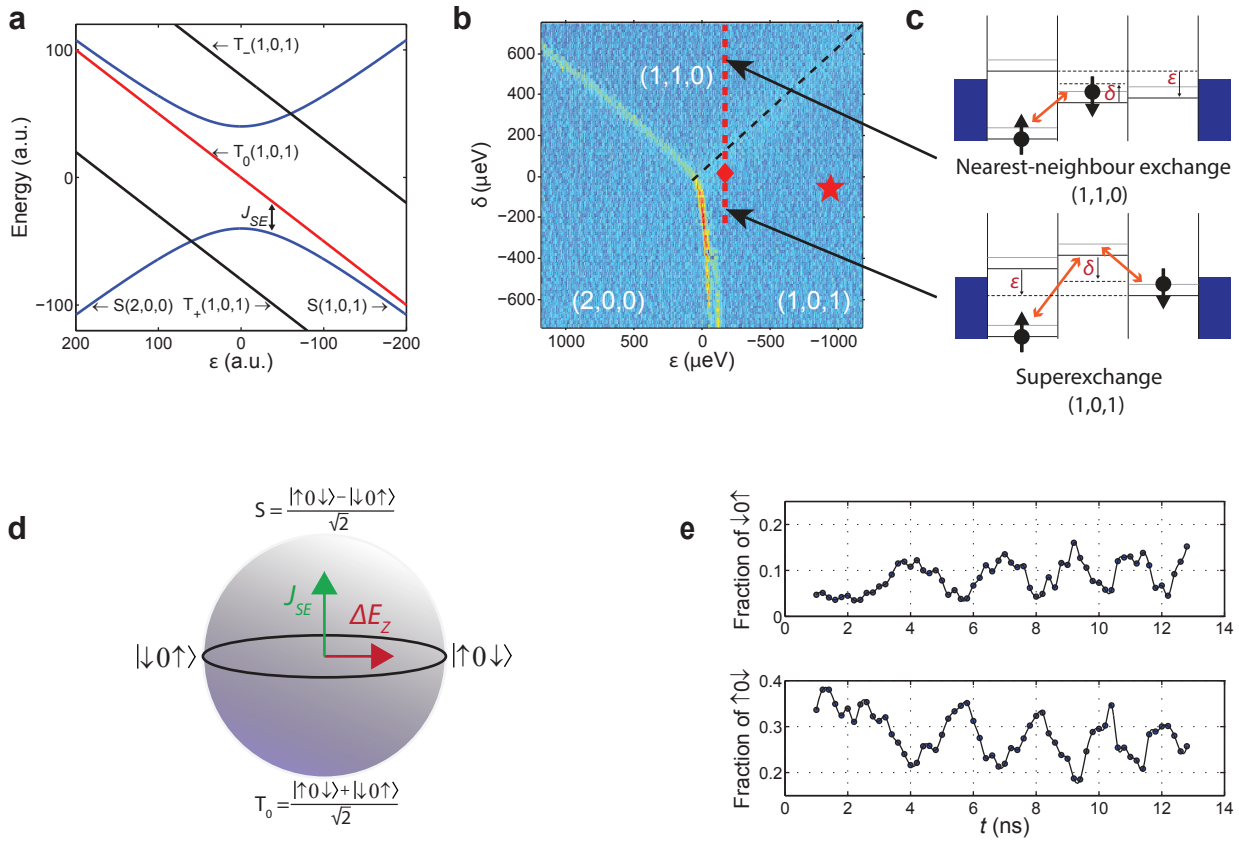


Figure 3

