Quantum photonics with spins in semiconductor nanostructures

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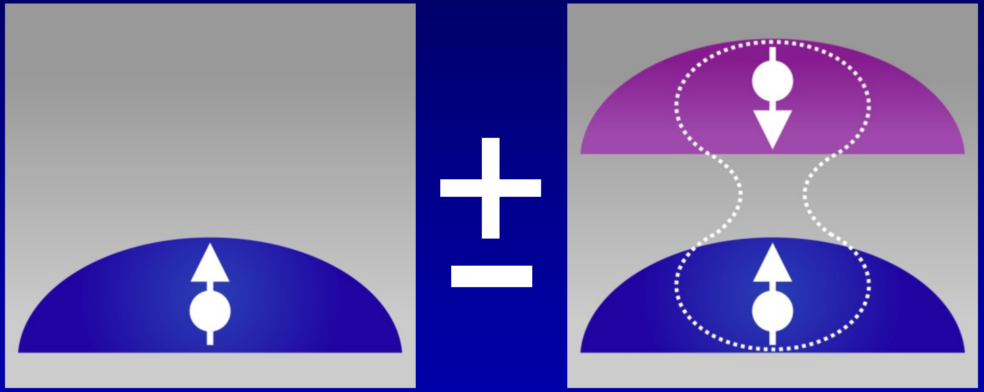
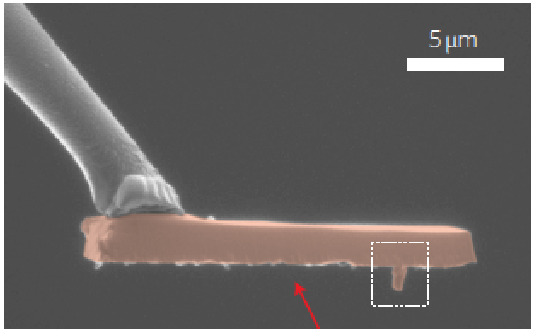


Quantum information

- ***Information*** is at the core of society:
    - ***generating*** information (using sensors to measure various things: *Big Data*)
    - ***processing*** information (computers: *Moore's Law*)
    - ***sharing*** information (via networks spanning large distances: *Internet*)
- Conventional devices – based on classical physics – are always becoming more powerful
- But they handle information in inherently inefficient ways for many (not all) important tasks
- "Information is physical" (Rolf Landauer, IBM) so must ultimately obey quantum mechanics
- ***Challenge:*** build devices that can harness resources provided by quantum mechanics (superposition and entanglement), to make more efficient use of full power of information

Quantum sensing



spin in diamond cantilever
as quantum sensor

Quantum computing

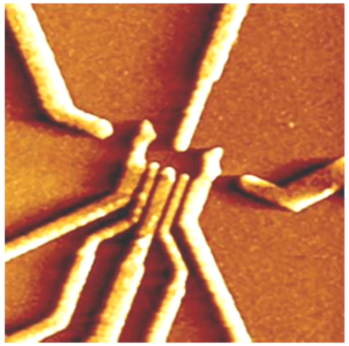
512-bit superconducting
quantum (?) processor

Quantum communication

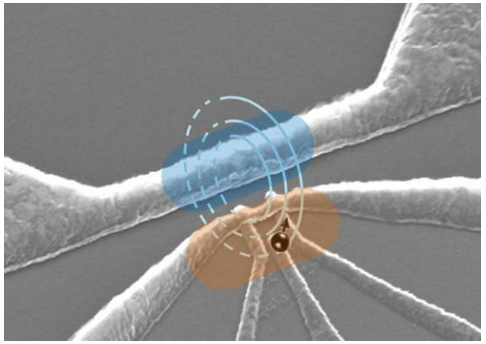
commercial fibre-based quantum
key distribution system

Quantum technology

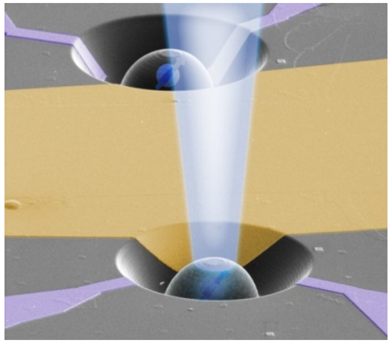
- Quantum states easily destroyed by unwanted interaction with environment: ***decoherence***
- Various approaches to reconcile conflicting demands of long coherence times and fast operations
- Why solid-state devices: more easily scalable? Not necessarily true... But solid state provides coherent spins and extensive tunability of electronic and optical properties
- Tempting to see quantum technologies as a race to build “the best qubit”
- But as in classical electronics, many materials serve different purposes:
  - silicon (ideal for electronics)
  - III-V (ideal for photonics)
  - organics (slower but very cheap and flexible)
- Same will likely be true for different qubit implementations



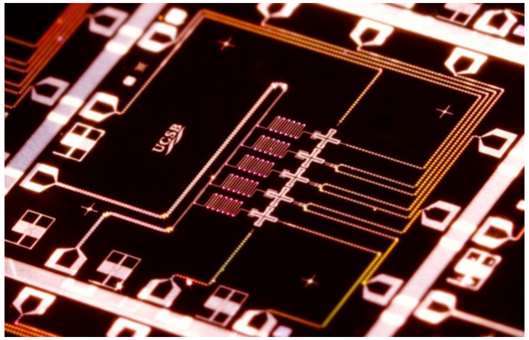
GaAs spin qubit



P:Si spin qubit



Diamond NV spin qubit



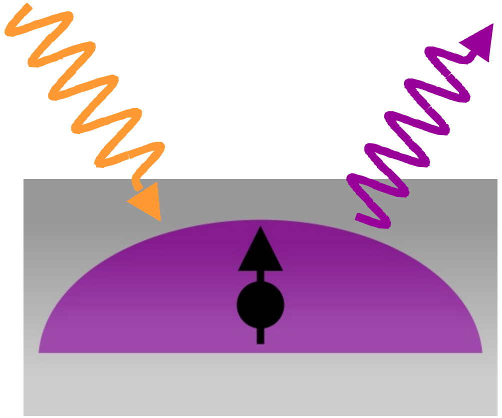
Superconducting quantum circuit

Quantum photonics

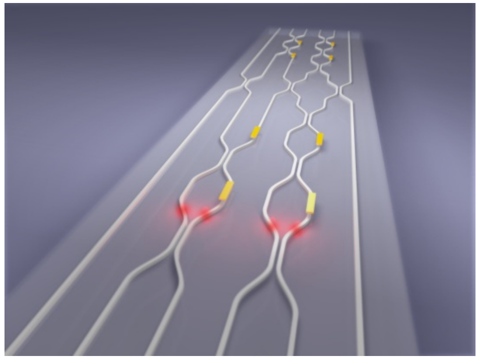
There will *certainly* be a need to interface with photons, which are the ideal long-distance carriers of quantum information: this is where optically active quantum emitters could fit in

Opportunities for optically active quantum emitters:

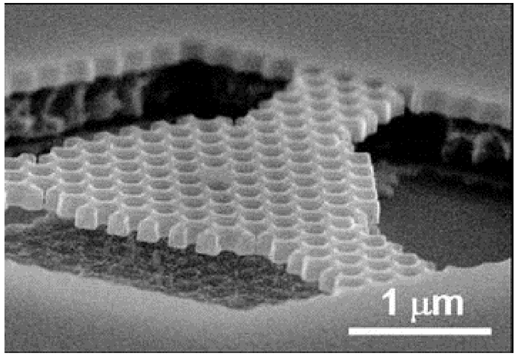
- Use light to control matter qubits (spins are the qubits)
- Use matter qubits to generate interesting states of light (photons are the qubits)
- Light-matter interface
  (quantum transducer, quantum memory, quantum repeater, quantum network, ...)



spin in optically active QD



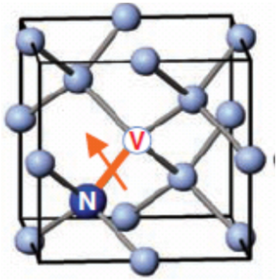
photonic quantum circuit



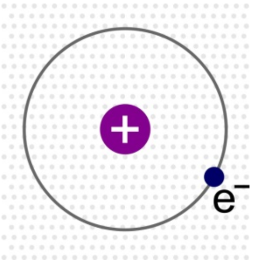
photonic-crystal membrane

Confining electron spins in solid state

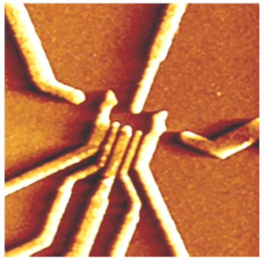
deep defects



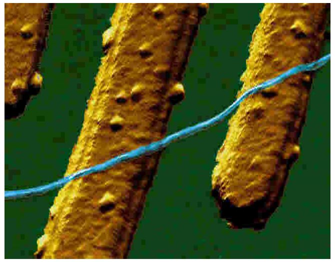
shallow donors



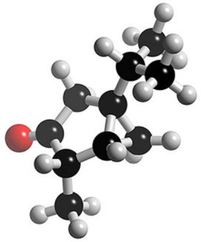
gate-defined QDs



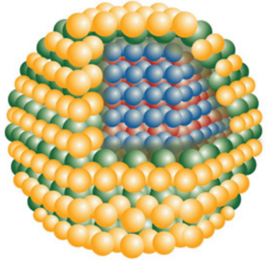
nanotubes



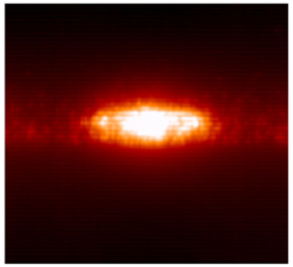
1 nm 10 nm 100 nm 1 μm



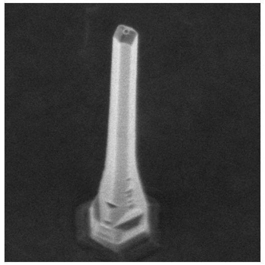
molecules



colloidal QDs



self-assembled QDs



nanowires

Why use self-assembled quantum dots?

- Provide electron (or hole) spins with modest coherence times: $T_1 \sim 10\ \mathrm{ms}$ and $T_2 \sim 3\ \mu\mathrm{s}$
- Fast source of near-lifetime limited photons: radiative lifetime $\sim 0.5\ \mathrm{ns}$, $\mathrm{FWHM} \sim 1\ \mu\mathrm{eV}$
- Excitonic transitions allow ultrafast ($\sim 10\ \mathrm{ps}$) control of electron spin
- Spin-photon coupling can be tailored using optical cavities, photonic crystals, waveguides, plasmonic nano-antennas, …
- Two self-assembled QDs can be controllably coupled for added flexibility: “*artificial molecule*”

Outline

- Introduction to self-assembled quantum dots as spin qubits

- Two-electron spin states in coupled quantum dots

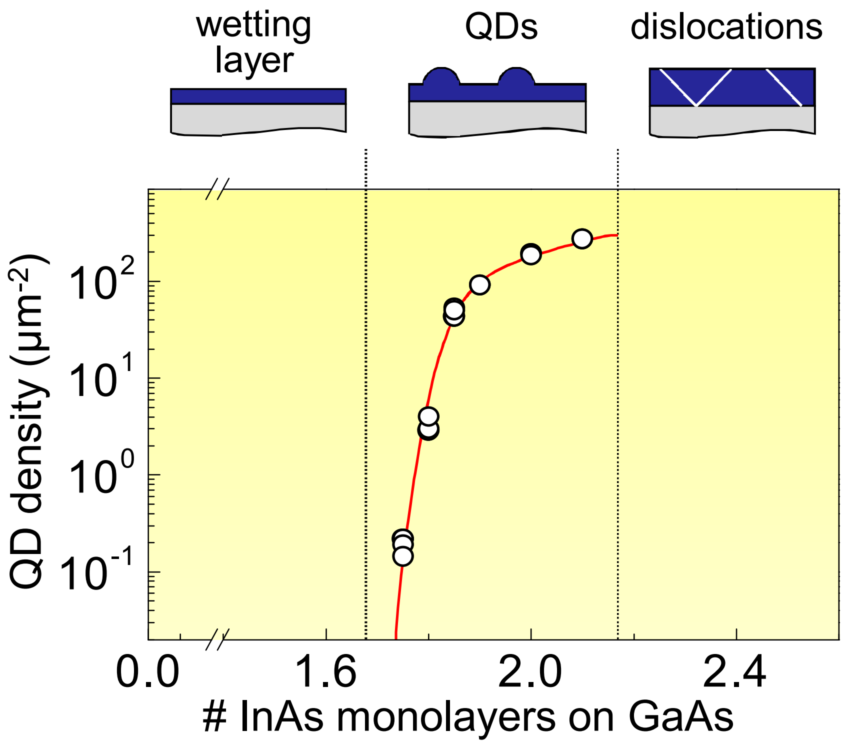
- Two-electron singlet-triplet states in a coupled QD form a lambda scheme
  JME, K. Weiss, J. Miguel-Sanchez & A. Imamoglu,
  Physical Review Letters **107**, 017401 (2011)

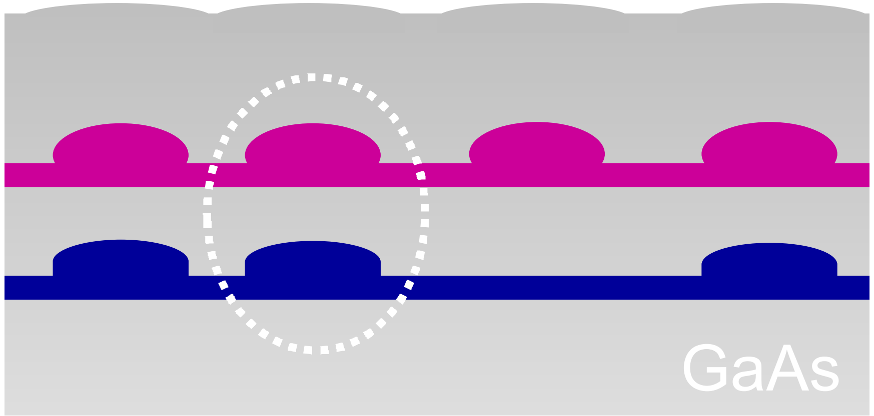
- Improved coherence time at "atomic clock transitions" of two coupled electron spins
  K. Weiss*, JME*, Y.L. Delley, J. Miguel-Sanchez & A. Imamoglu,
  Physical Review Letters **109**, 107401 (2012)

- Four coupled electrons spins form a magnetically tunable singlet-triplet qubit
  K. Weiss, J. Miguel-Sanchez & JME,
  Scientific Reports **3**, 3121 (2013)

- Conclusions and outlook

Growth of InAs/GaAs self-assembled QDs



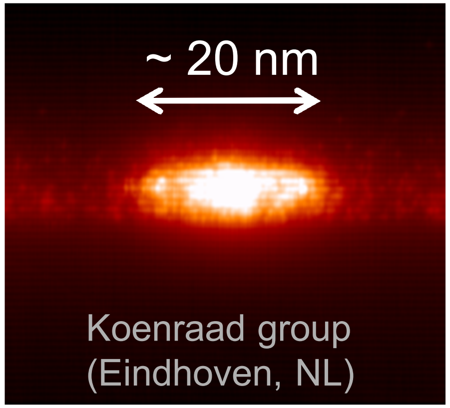


Self-assembled quantum dots:

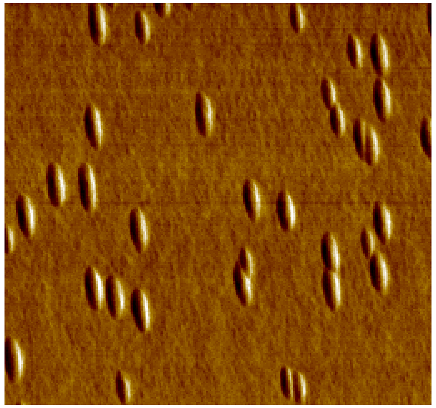
- Small InAs regions (~20 nm laterally and ~5 nm thick) form inside GaAs crystal
- Dots form spontaneously due to ~7% lattice mismatch between GaAs and InAs
- Vertical thickness can be controlled using partial overgrowth and annealing
- QDs maintain crystal structure of GaAs surroundings (i.e. no defects), but is highly strained
- QDs are connected via “wetting layer” (acts as a very thin quantum well)
- QDs in bottom layer (which is grown first) nucleate at uncontrolled positions
- QDs in subsequent layers can be stacked due to strain field emanating from each dot

Self-assembled In(Ga)As quantum dots

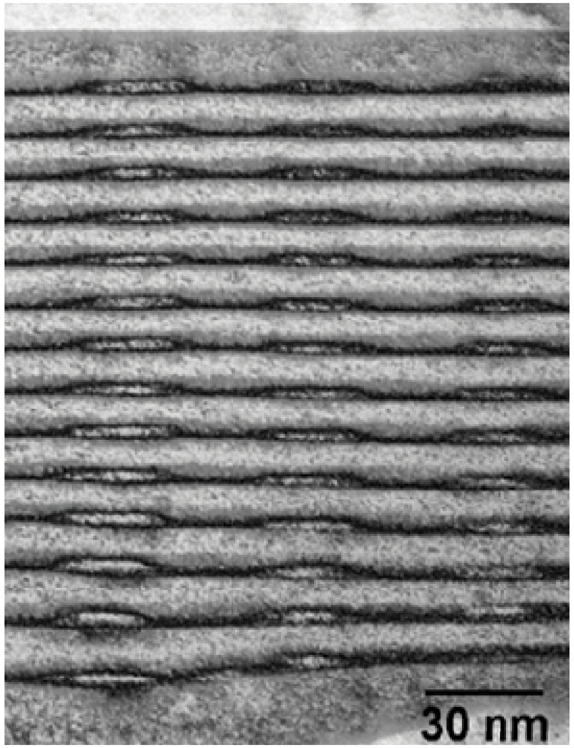
X-STM



AFM (10 x 10 μm^2)



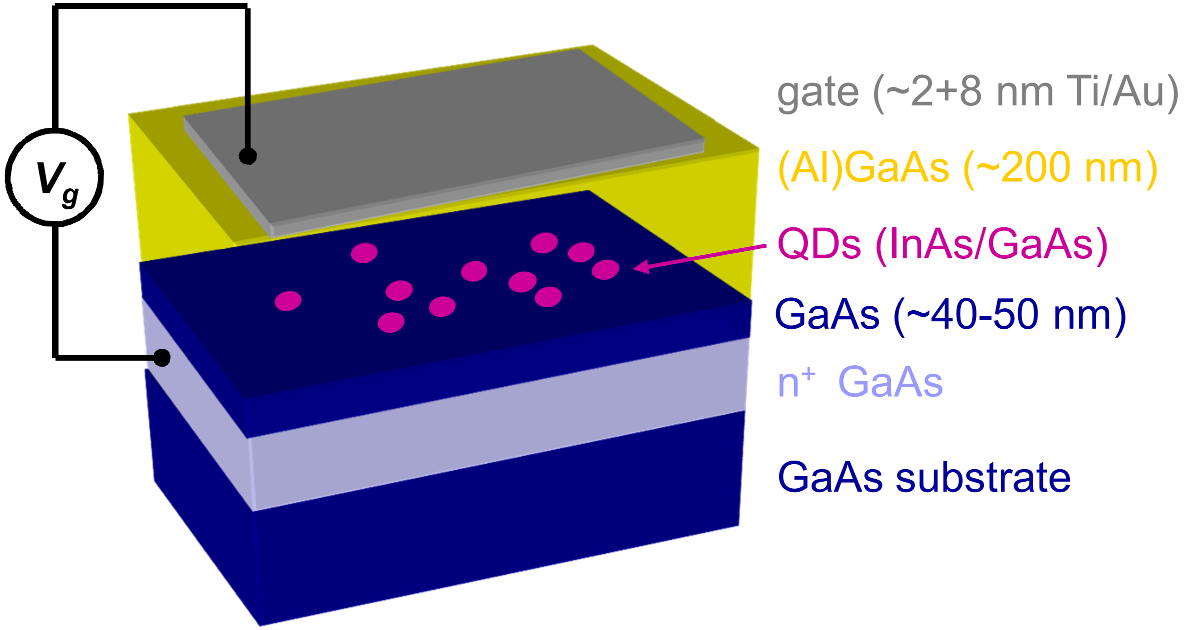
TEM

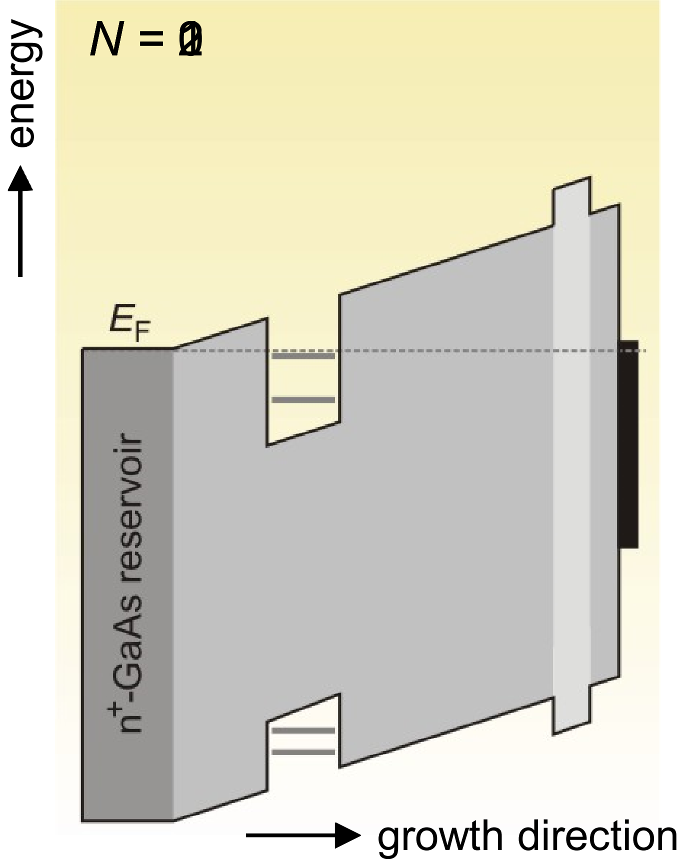


Optical properties of self-assembled quantum dots:

- Well-behaved photon emission (no blinking; linewidth $\sim\Gamma_{rad} \sim 1\ \mu\text{eV}$, lifetime $\sim 0.5\ \text{ns}$)
- Some control over wavelength via partial overgrowth ($\lambda \sim 900 - 1300\ \text{nm},\ \Delta\lambda \sim 10\ \text{nm}$)
- Control over average QD density (we choose $< 0.1\ \mu\text{m}^{-2}$) but no control over actual position
- But QDs can be vertically stacked to form "*artificial molecules*" (stacking probability > 80 %)
- Precise control over inter-dot tunnel coupling ($d \sim 10 - 30\ \text{nm} \Rightarrow 2t \sim 1\ - 0.01\ \text{meV}$)

QDs in Schottky diode structure

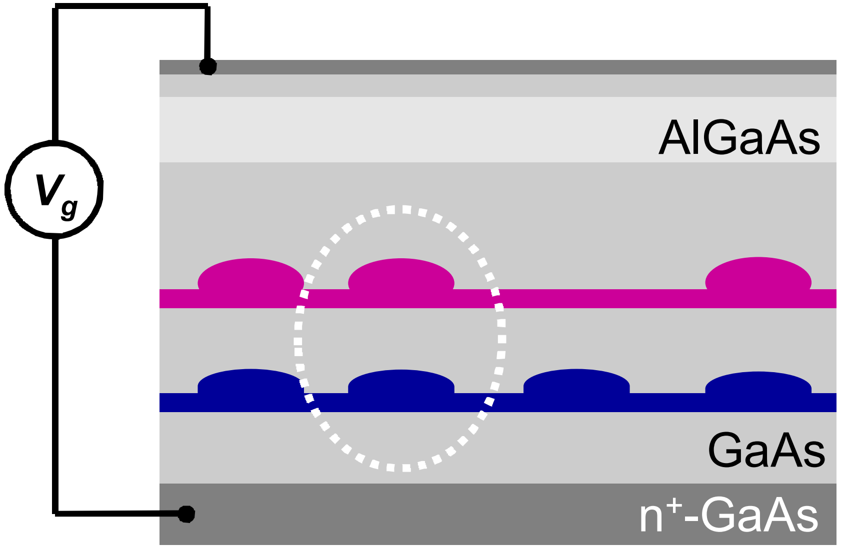


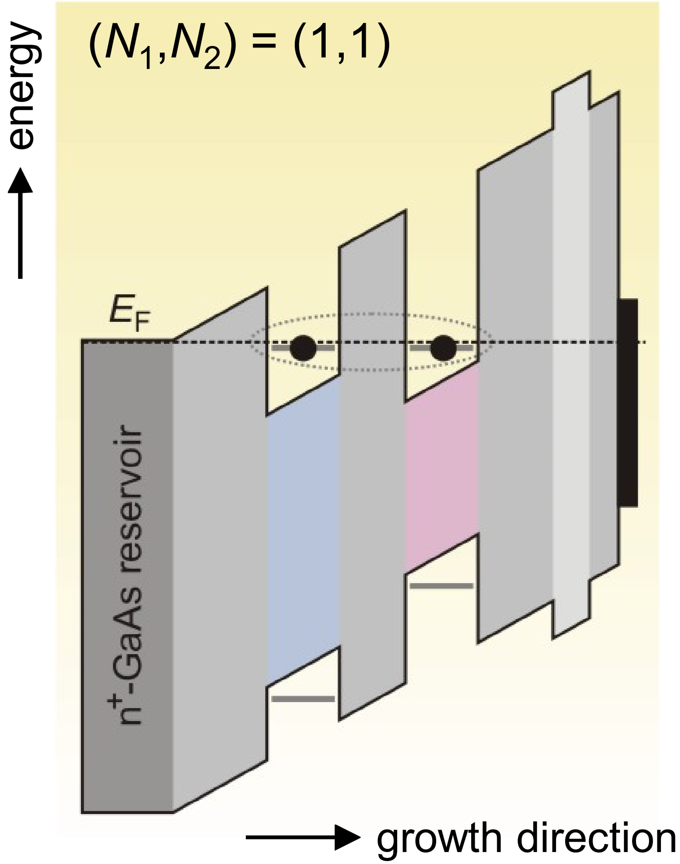


- QD layer embedded into Schottky diode
- Heavily doped back contact forms electron reservoir
- Make Ohmic contact from top surface to back contact
- Deposit semi-transparent top gate (~ 2+8 nm Ti/Au)
- Gate voltage V_g between top gate and back contact tilts CB and VB and thereby tuning QD energy levels with respect to Fermi energy E_F of back contact
- V_g allows control over QD charge ($N = 0,1,2, \ldots$)

Fill quantum dot with one electron or hole spin
⇒ ***spin qubit!***

CQDs in Schottky diode structure

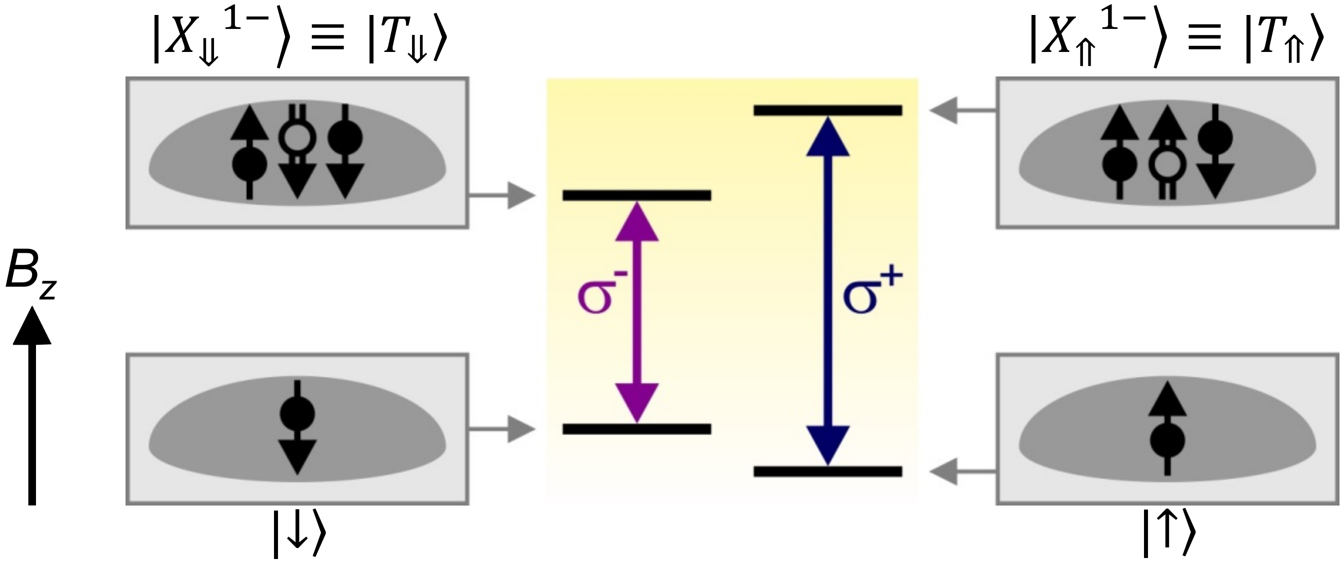




- Two QD layers embedded into Schottky diode
- Complete control over total charge in CQD via Coulomb blockade ($N_{total} = 0,1,2\ \ldots$)
- But limited control over internal charge distribution in CQD:$(N_1, N_2) = (2,0)/\ (1,1)/\ (0,2)$
- Requires careful search for particular CQD pair with correct level detuning (i.e. relative emission energy)
- Electrons are delocalized at anticrossings: $E(2,0) = E(1,1)$

Fill CQD with one electron spin in each dot ⇒ singlet (S) and triplet (T) spin states ⇒ ***entanglement!***

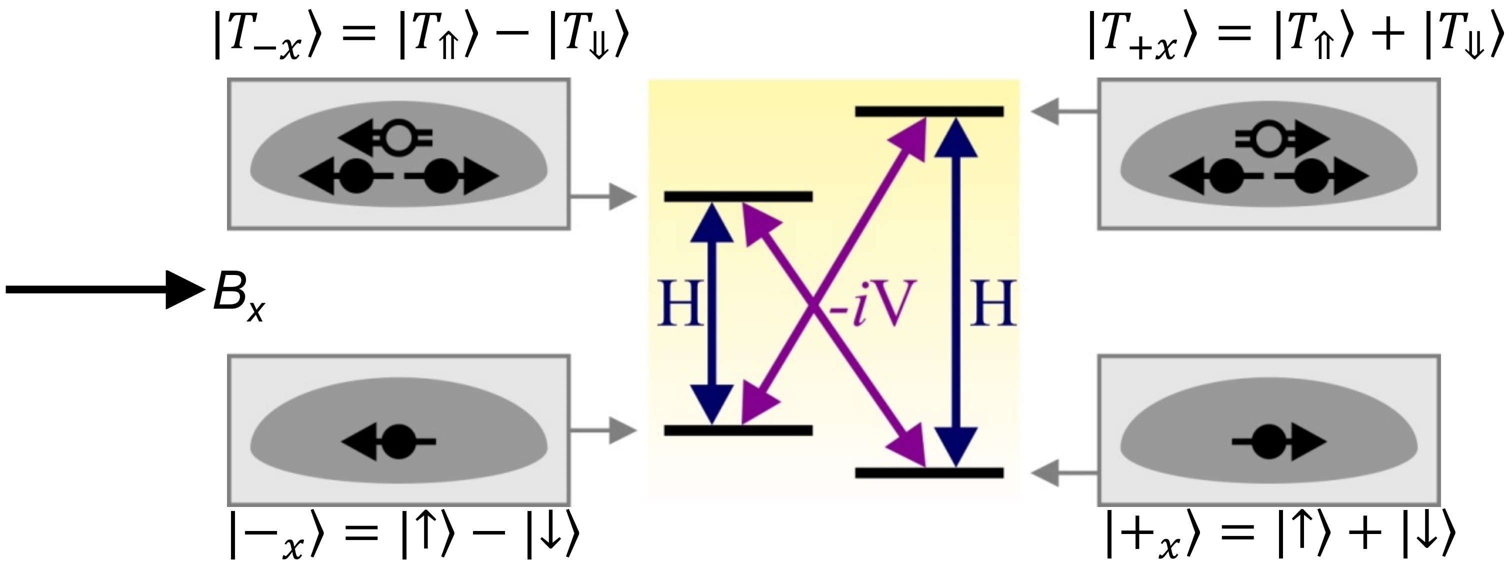
One-electron QD as spin qubit



Single electron (or hole) spin qubit:

- Qubit states: $|\uparrow\rangle$ (spin-up) and $|\downarrow\rangle$ (spin-down) along quantization axis defined by externally applied *B*-field
- Strain introduces built-in vertical quantisation axis
- Optical transitions involve heavy hole ($J_z = 3/2\hbar$)
- Faraday configuration (external *B*-field along vertical direction): quasi-recycling transitions with circularly polarised transitions
- Voigt geometry (in-plane external *B*-field): two lambda schemes with linearly polarised transitions

One-electron QD as spin qubit



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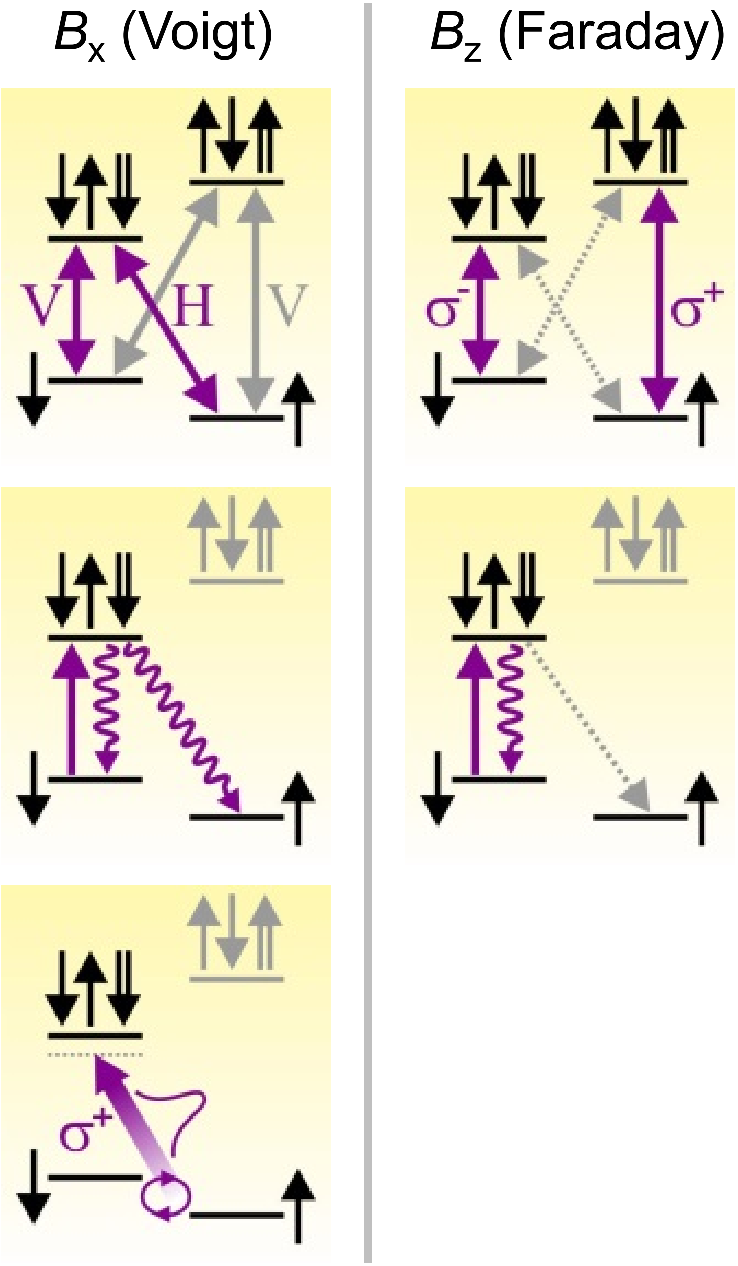
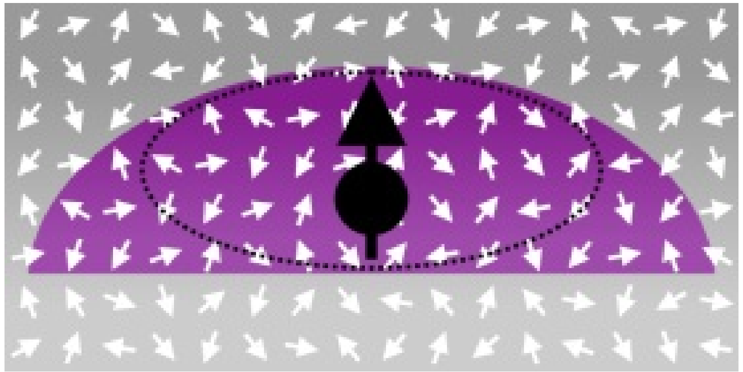
- Voigt geometry (in-plane external *B*-field): two lambda schemes with linearly polarised transitions

Self-assembled QDs for quantum information

- Single electron or hole spin qubit
  - charging: Warburton *et al.*, Nature 2000
- Spin initialization via spin pumping
  - Faraday ~ μs: Atatüre *et al.*, Science 2006
  - Voigt ~ ns: Xu *et al.*, PRL 2007
- Average spin readout via quasi-cycling transitions (or single-shot using additional QD)
  - CQD proposal: Kim *et al.*, PRL 2008
  - experiment: Vamivakas *et al.*, Nature 2010
- Spin manipulation (~10 ps) via off-resonant stimulated Raman transitions + Larmor
  - ensemble: Greilich *et al.*, PRL 2006
  - single electron: Press *et al.*, Nature 2008
  - single hole: Greilich *et al.*, Nature Phot. 2011
- Spin-photon entanglement via spont. emission
  - polarisation: DeGreve *et al.*, Nature 2012
  - energy: Gao *et al.*, Nature 2012
- Coupling to photonic-crystal nanocavity
  - *Q* ~ 4000: Carter *et al.*, Nature Phot. 2013



Single-spin coherence properties



Single electron spin coherence

$T_2^* \sim \mathrm{ns}$ for single electron spin (limited by fluctuating bath of nuclear spins)
Merkulov Efros & Rosen, PRB 2002
Press *et al.*, Nature Phot. 2010

1. Reduce bath fluctuations

- (Choose different material, e.g. Si, C, …)
- Freeze nuclear spin configuration via electron-nuclear feedback controlled by laser
  Faraday: Latta *et al.*, Nature Phys. 2009
  Voigt: Xu *et al.*, Nature 2009
  Sun *et al.*, PRL 2012

2.Dynamically decouple electron spin from bath

- $T_2 \sim \mathrm{\mu s}$ revealed by optical spin echo
  single electron: Press *et al.*, Nature Phot. 2010
  single hole: DeGreve *et al.*, Nature Phys. 2011

3. Engineer robust qubit states

- Use single hole spin as qubit
  Brunner *et al.*, Science 2009
  Greilich *et al.*, Nature Phot. 2011
- Use second QD to make qubit states robust against both magnetic and electric fluctuations
  $T_2^* < 1\ \mathrm{ns}$: Kim *et al.*, Nature Phys. 2010
  $T_2^* > 200\ \mathrm{ns}$: Weiss *et al.*, PRL 2012

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- Two-electron singlet-triplet states in a coupled QD form a lambda scheme
  JME, K. Weiss, J. Miguel-Sanchez & A. Imamoglu,
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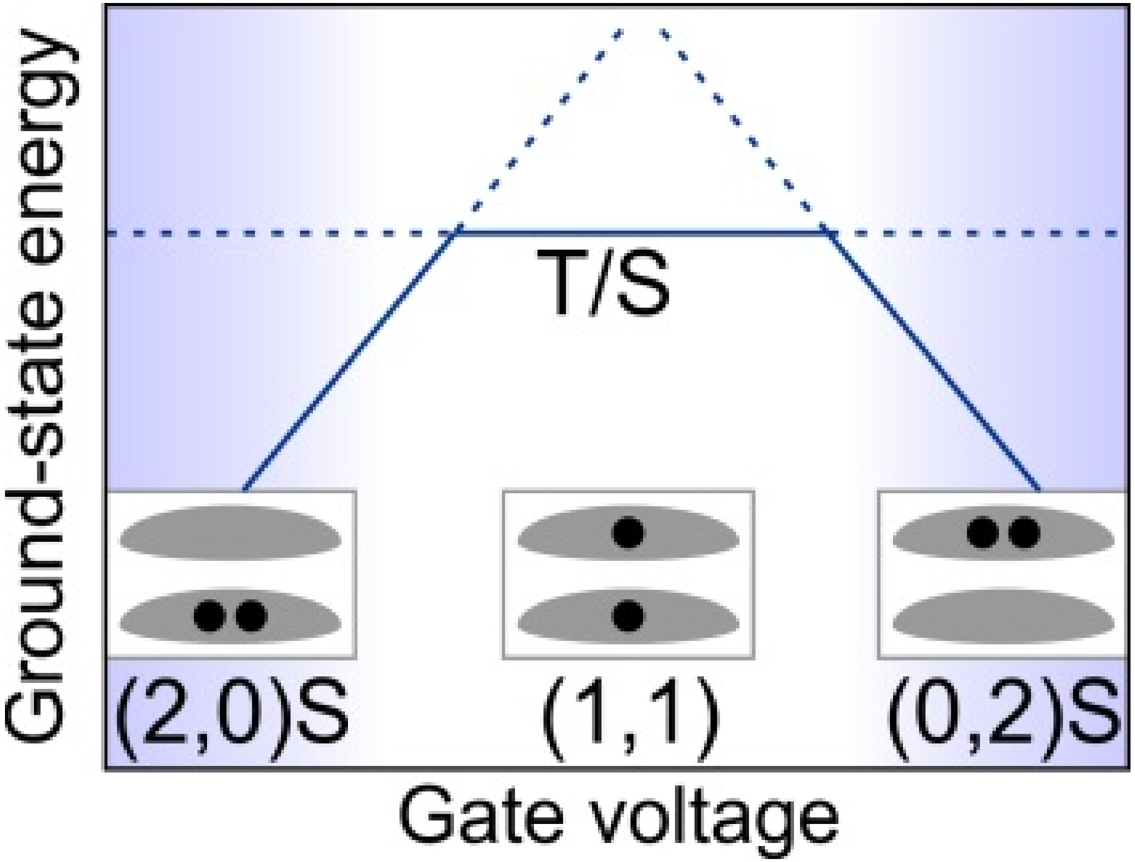
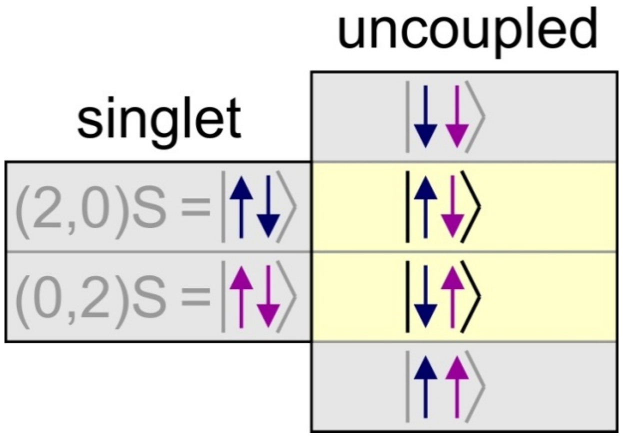
- Conclusions and outlook

Two-electron spin states





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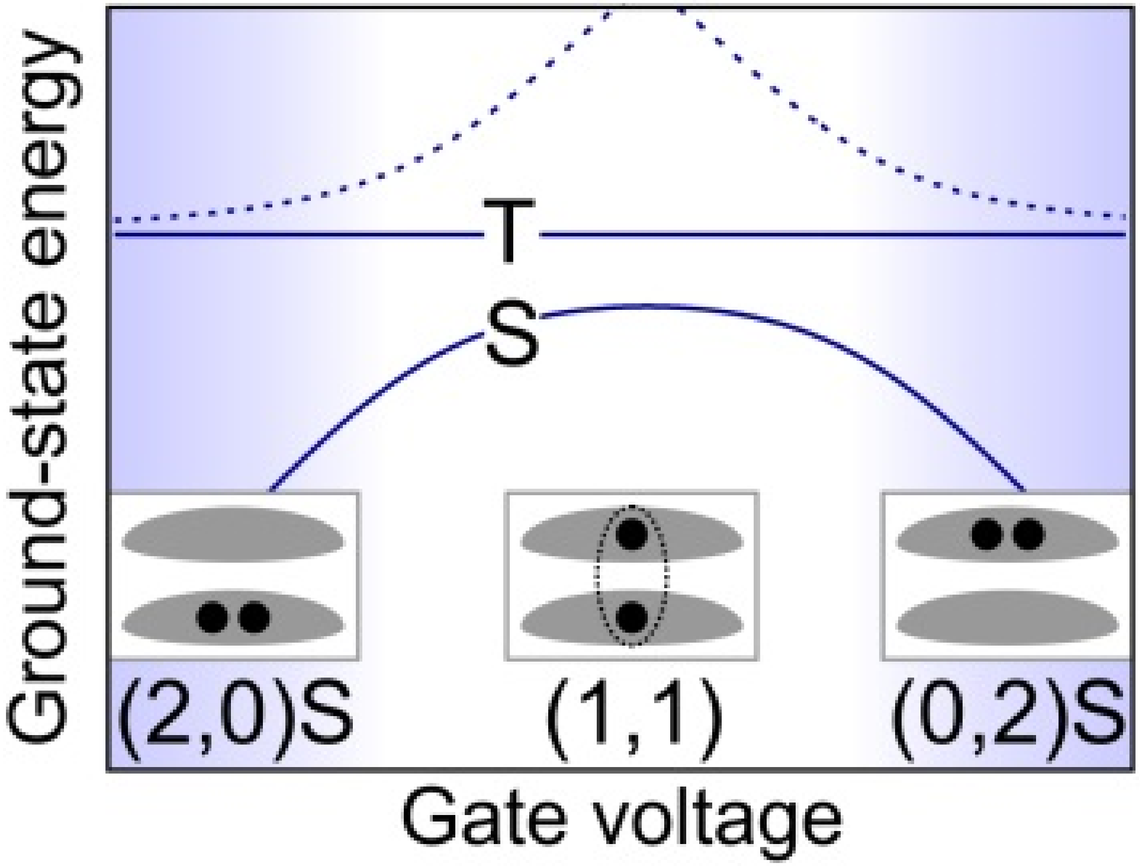
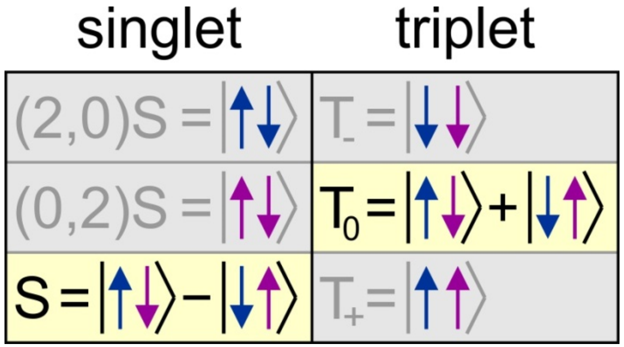
Two-electron spin states

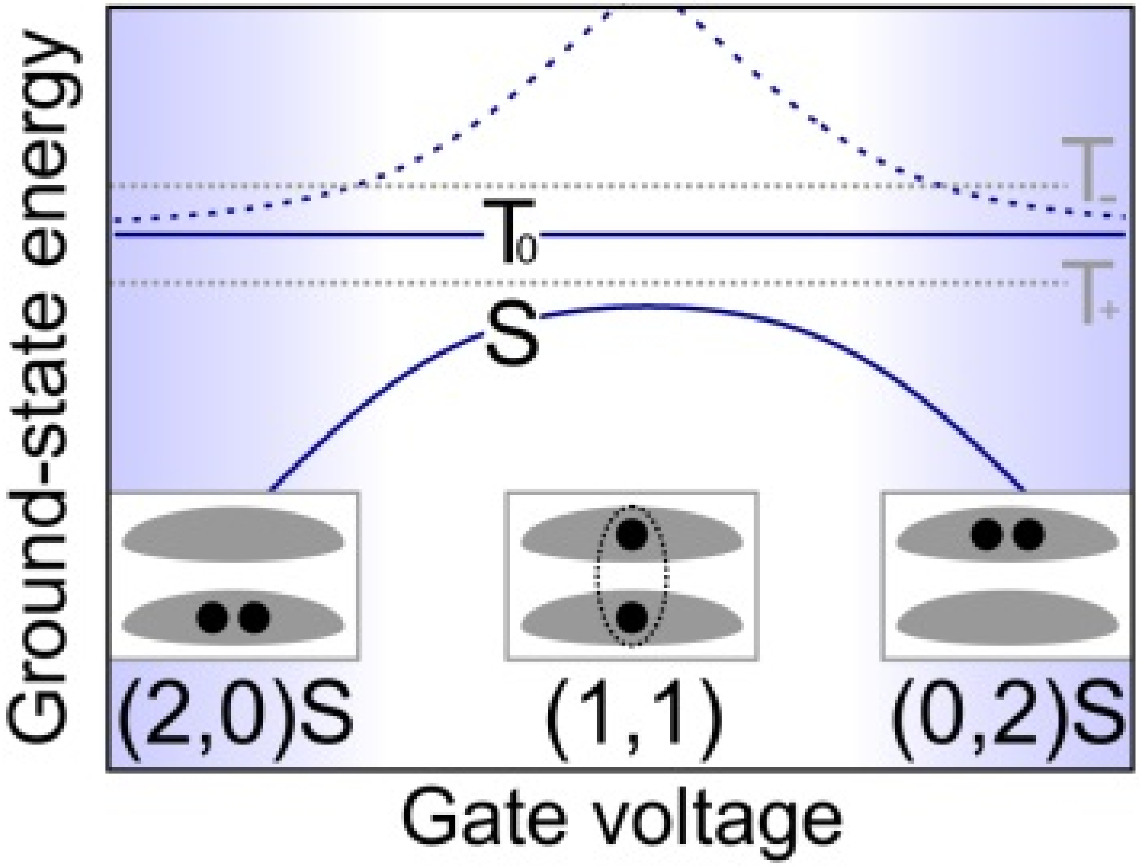


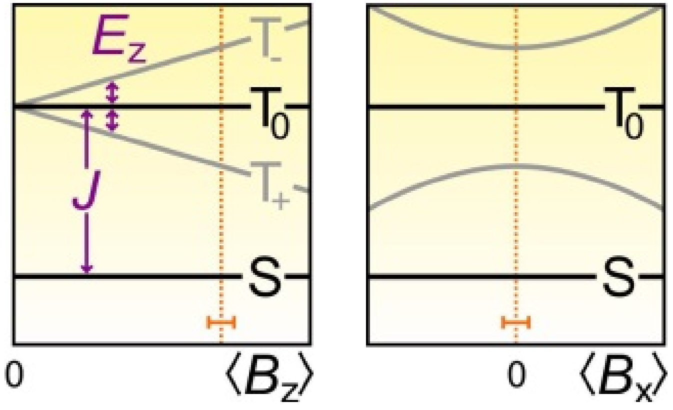


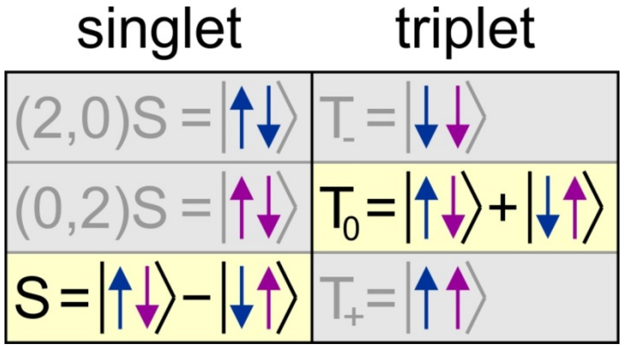
singlet	triplet
$(2,0)S = \lvert\uparrow\downarrow\rangle$	$T_- = \lvert\downarrow\downarrow\rangle$
$(0,2)S = \lvert\uparrow\downarrow\rangle$	$T_0 = \lvert\uparrow\downarrow\rangle + \lvert\downarrow\uparrow\rangle$
$S = \lvert\uparrow\downarrow\rangle - \lvert\downarrow\uparrow\rangle$	$T_+ = \lvert\uparrow\uparrow\rangle$

- No tunneling: (1,1) spin states degenerate, (0,2) and (2,0) spin states not
- With tunneling: (1,1)S splits from (1,1)T by *V*-dependent exchange energy

Two-electron spin states







singlet	triplet
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$(0,2)S = \lvert\uparrow\downarrow\rangle$	$T_0 = \lvert\uparrow\downarrow\rangle + \lvert\downarrow\uparrow\rangle$
$S = \lvert\uparrow\downarrow\rangle - \lvert\downarrow\uparrow\rangle$	$T_+ = \lvert\uparrow\uparrow\rangle$

- No tunneling: (1,1) spin states degenerate, (0,2) and (2,0) spin states not
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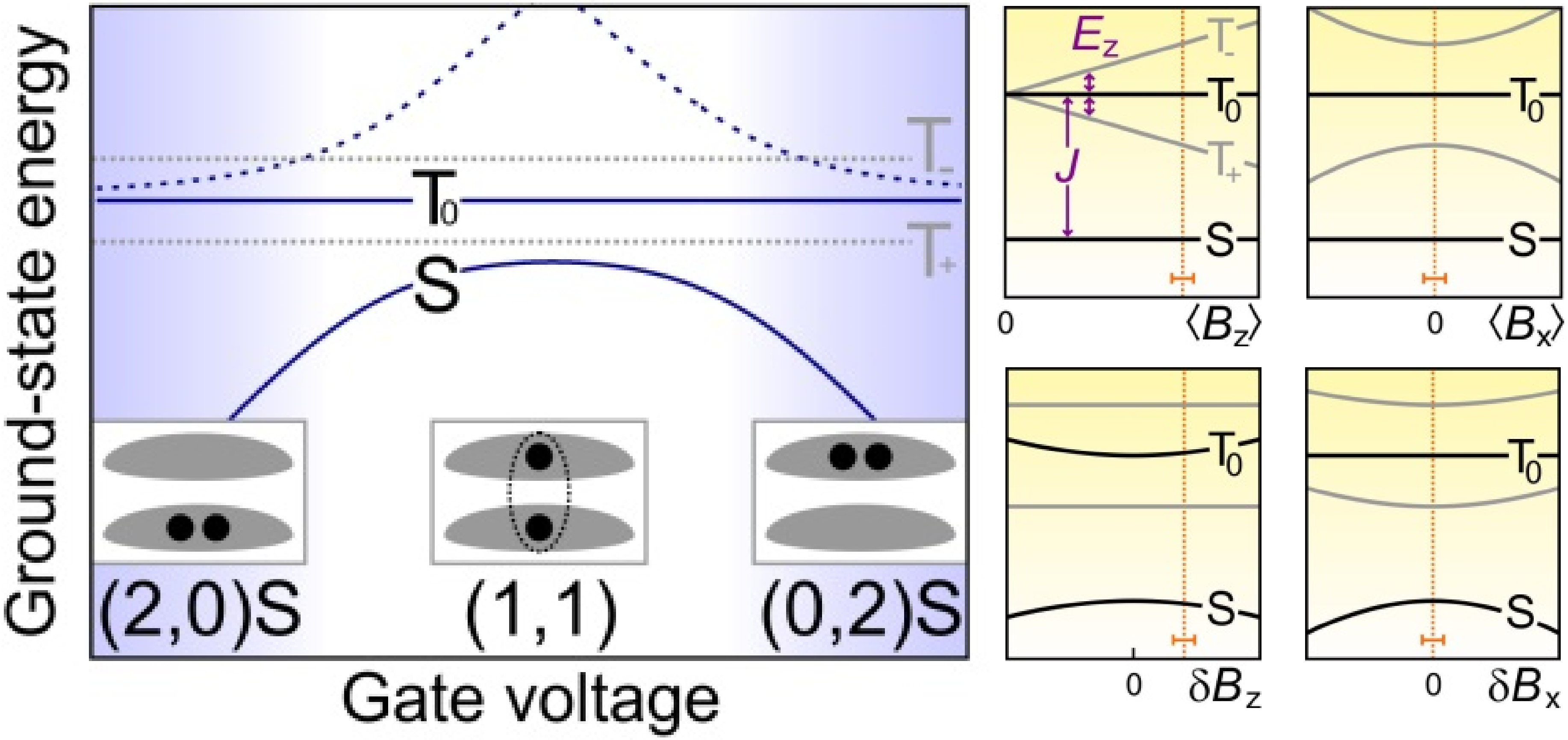
- Homogeneous *B*-field: (1,1)T split by Zeeman energy, S and T_0 unaffected

Two-electron spin states



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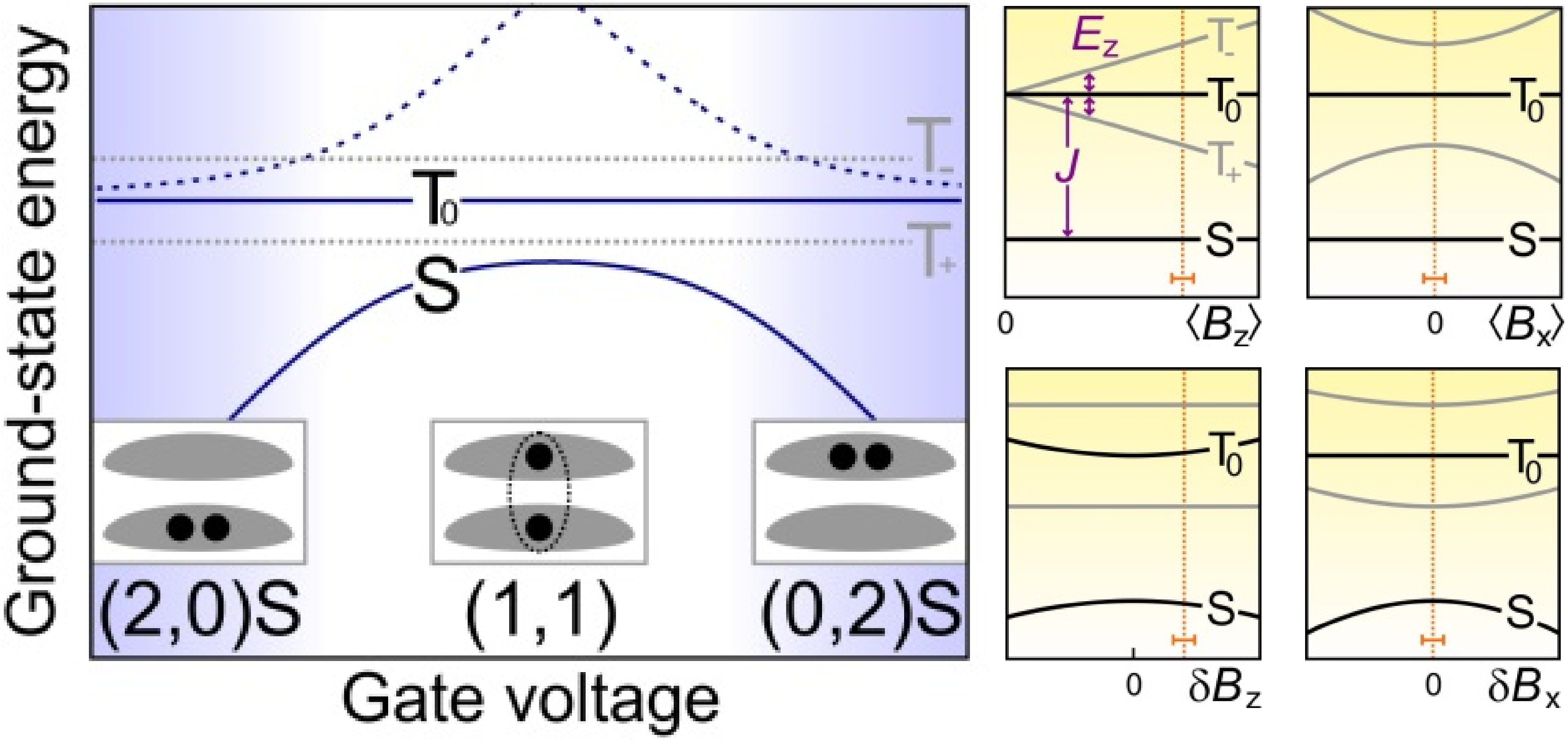
- ***Atomic clock states: insensitive to first order to nuclear spin fluctuations***

Two-electron spin states



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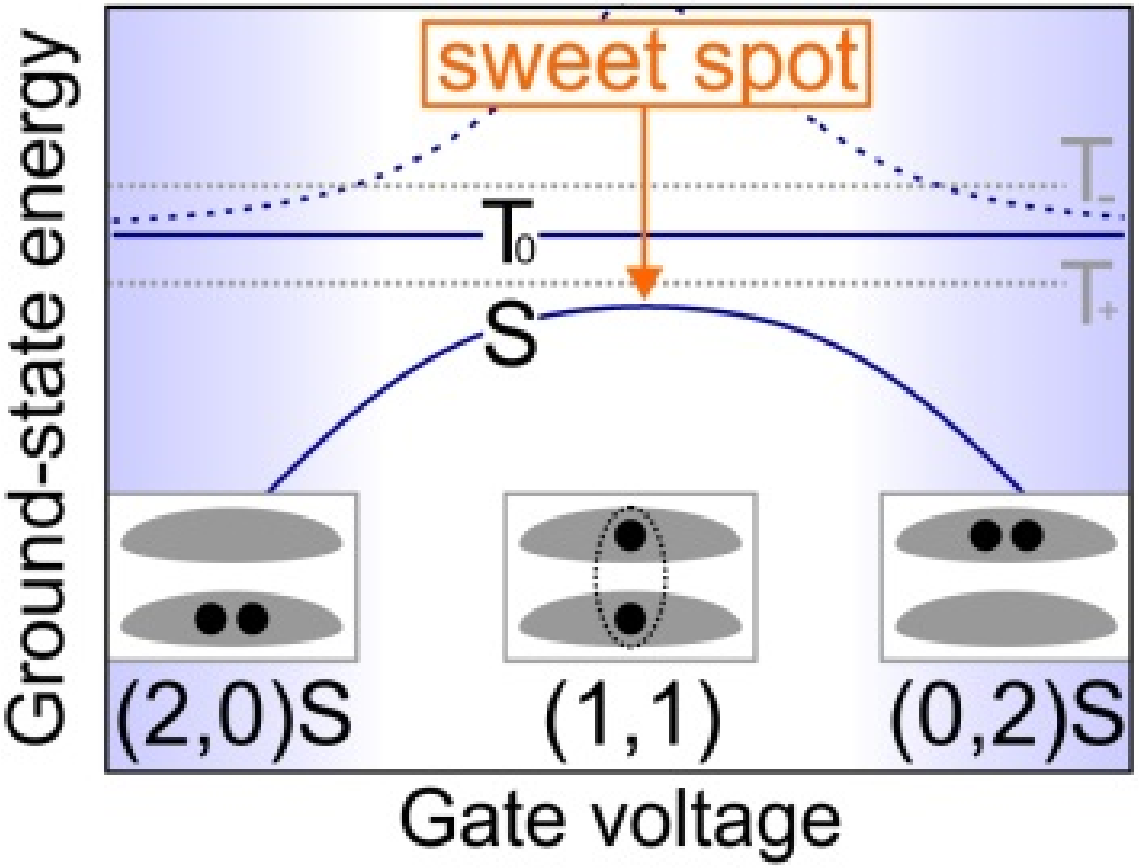
- ***Atomic clock states: insensitive to first order to nuclear spin fluctuations***
- ***BUT: exchange splitting depends on V*** $\Rightarrow$ ***sensitive to charge noise!***

Two-electron spin states



At "sweet spot":
S/T0 qubit states (to first order) insensitive to charge fluctuations!

D. Vion *et al.*,
Science **296**, 886 (2002)

J. Koch *et al.*,
PRA **76**, 042319 (2007)

D.A. Lidar *et al.*,
PRL **81**, 2594 (1998)

- No tunneling: (1,1) spin states degenerate, (0,2) and (2,0) spin states not
- With tunneling: (1,1)S splits from (1,1)T by *V*-dependent exchange energy
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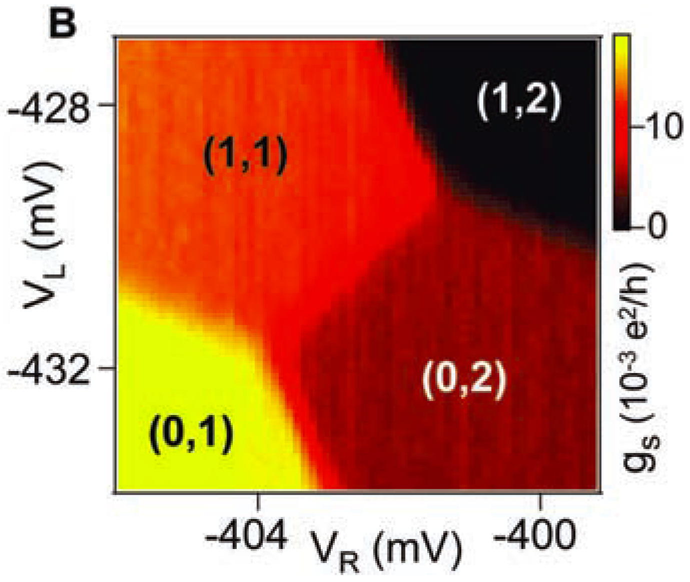
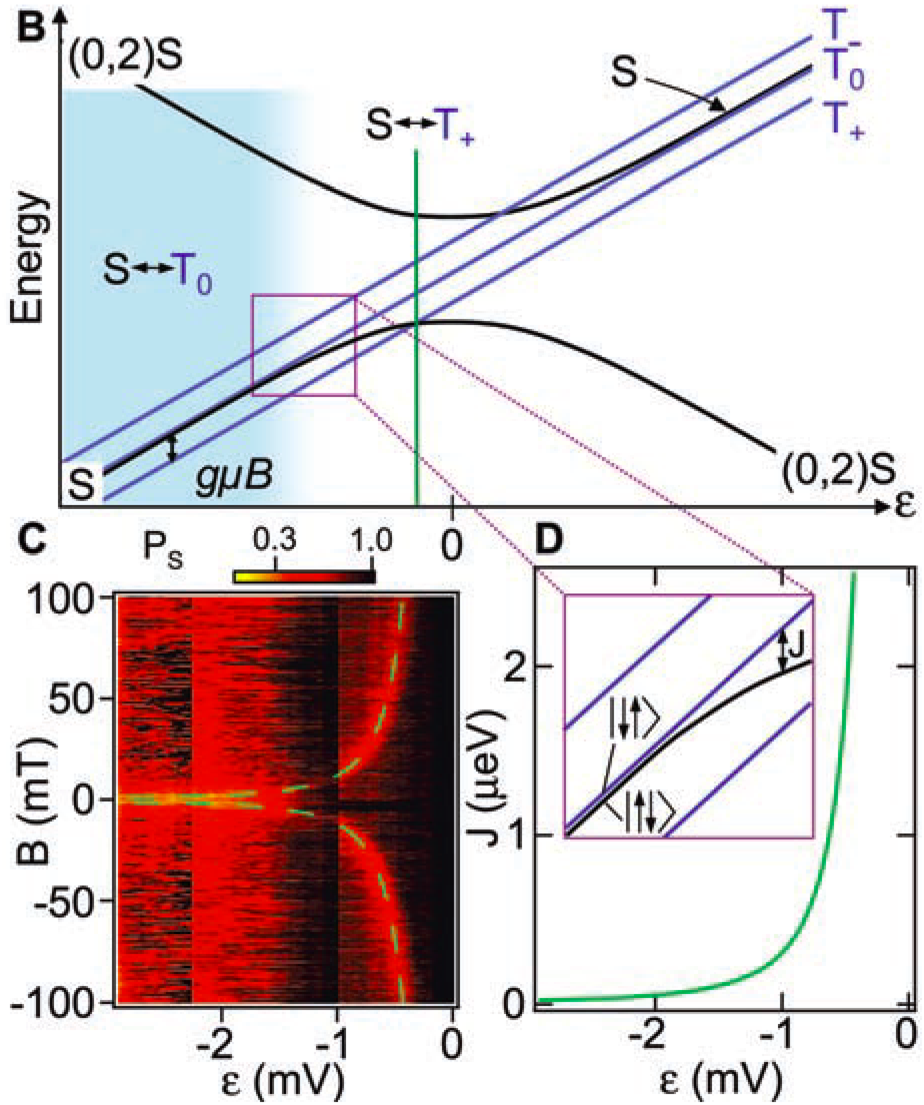
- ***Atomic clock states: insensitive to first order to nuclear spin fluctuations***
- ***BUT: exchange splitting depends on V*** $\Rightarrow$ ***sensitive to charge noise!***

ST qubits in electrically defined CQDs

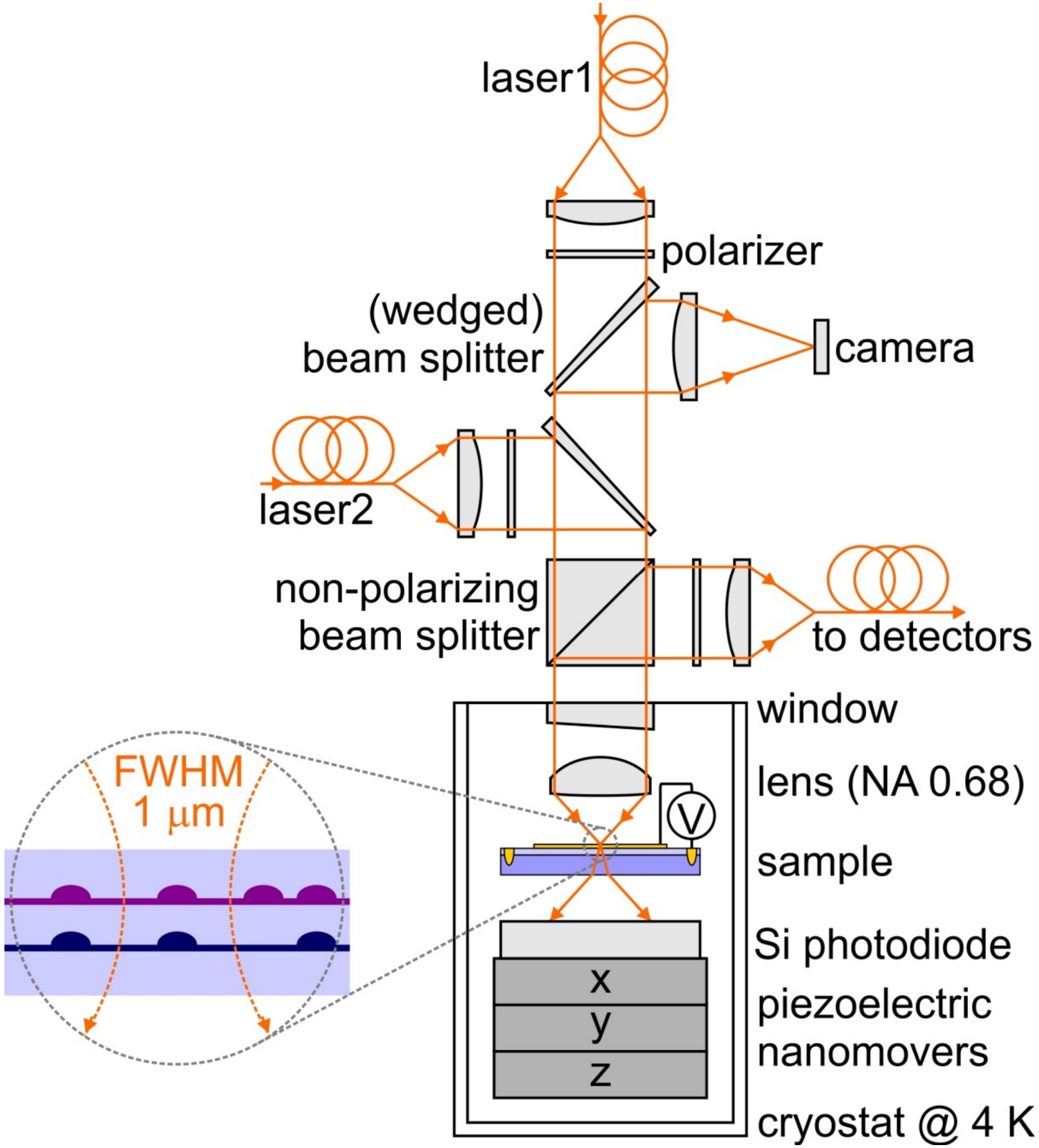
J.R. Petta *et al.*, Science **309**, 2180 (2005)
H. Bluhm *et al.*, Nature Physics **7**, 109 (2011)

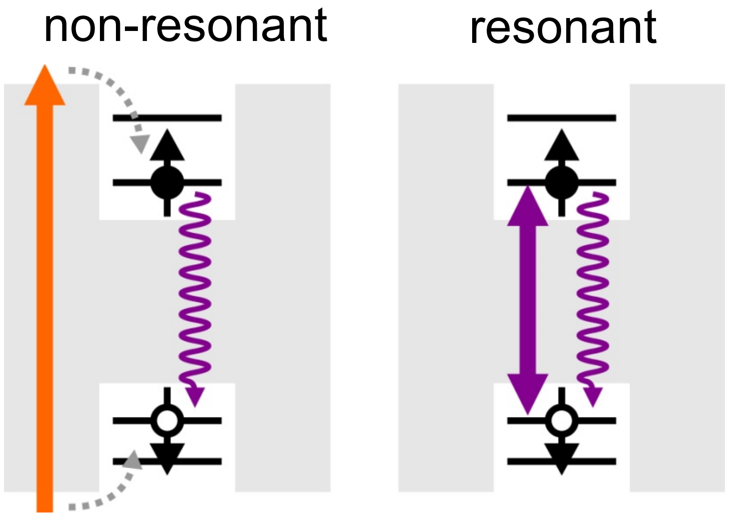




- Operate in spin blockade regime
  (1,1) ↔ (0,2)
  far away from sweet spot
- ST splitting smaller than hyperfine (gradient) fields
- Necessary for manipulation!

Experimental setup at the LCN





- Confocal dark-field microscope with device in liquid-helium bath cryostat (4K) with B_z = 0–10 T
- ***Nonresonant measurements***
  Excite above GaAs band gap and send emission from QD to grating spectrometer and CCD
- ***Resonant measurements:***
  Excite resonantly using tunable diode laser and detect interference between laser and QD emission
- OR: suppress laser using crossed polarisers to detect only QD light

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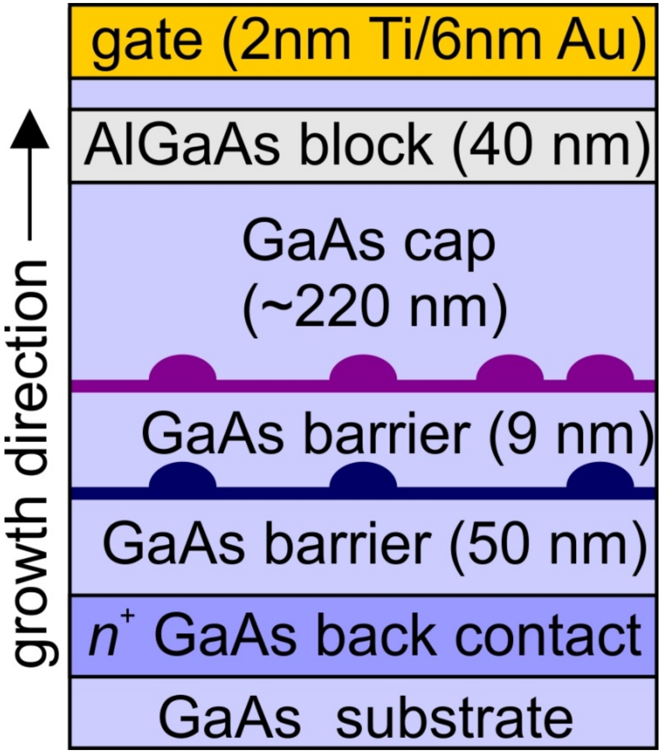
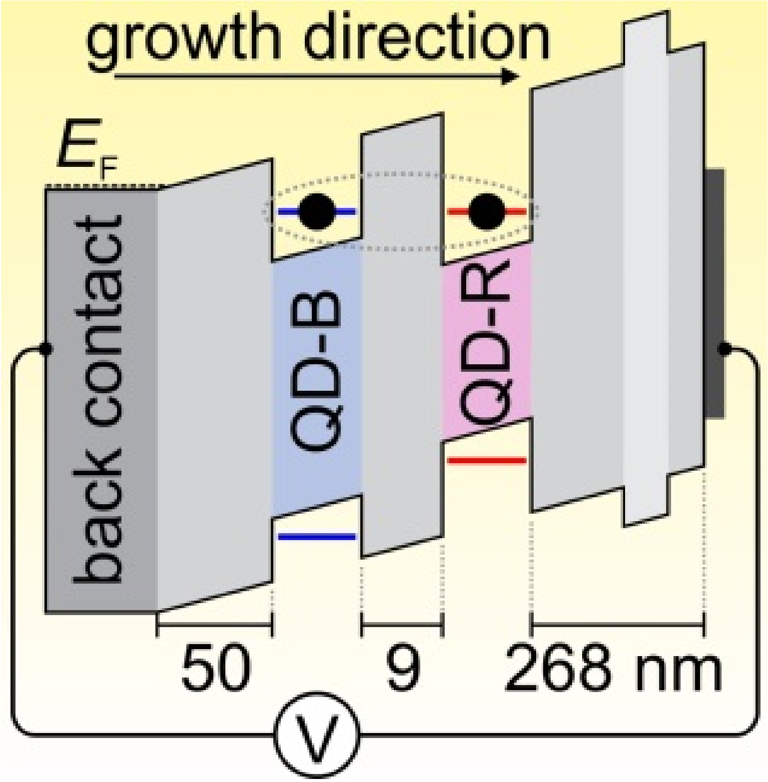
  K. Weiss, J. Miguel-Sanchez & JME,
  Scientific Reports **3**, 3121 (2013)

- Conclusions and outlook

Device layout and bandstructure

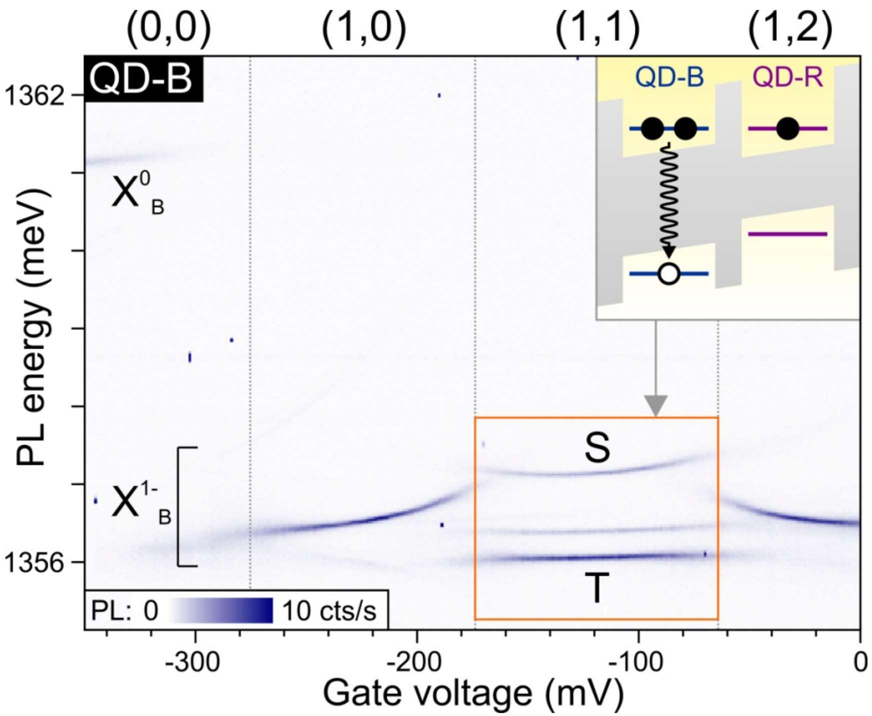


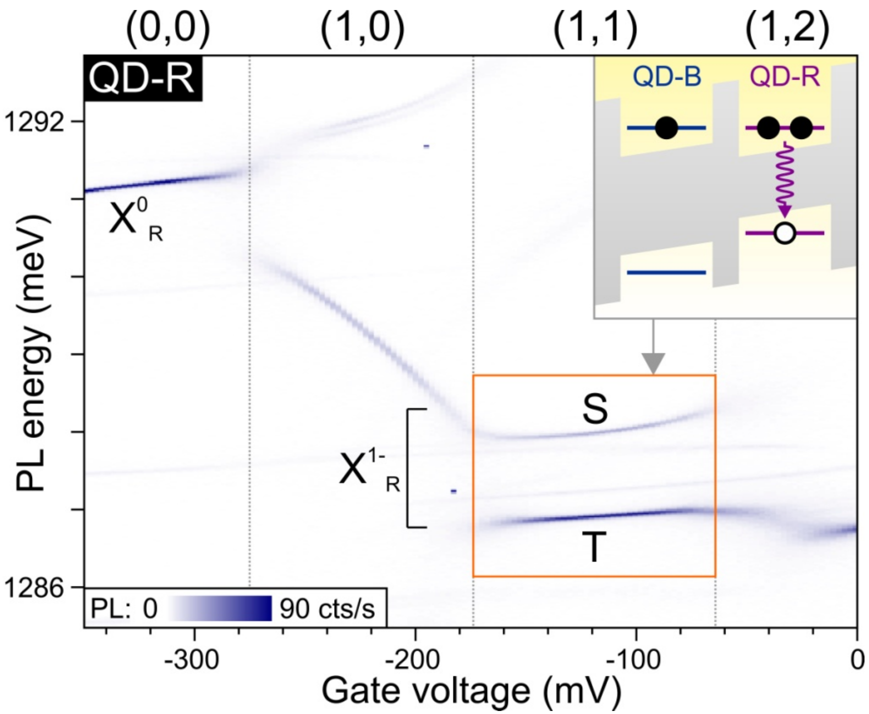


- 2 layers of self-assembled InAs QDs in GaAs Schottky diode
- QDs in top and bottom layers form vertical stacks due to strain
- Emission QD-B ~940 nm and QD-R ~970 nm (shifted by PCI technique)

- Tune *V* to charge each QD with single electron: (1,1) regime
- Requires accurate design of QD-B & QD-R wavelengths
- Strong tunnel coupling due to thin GaAs tunnel barrier

Identifying (1,1) charging regime using PL

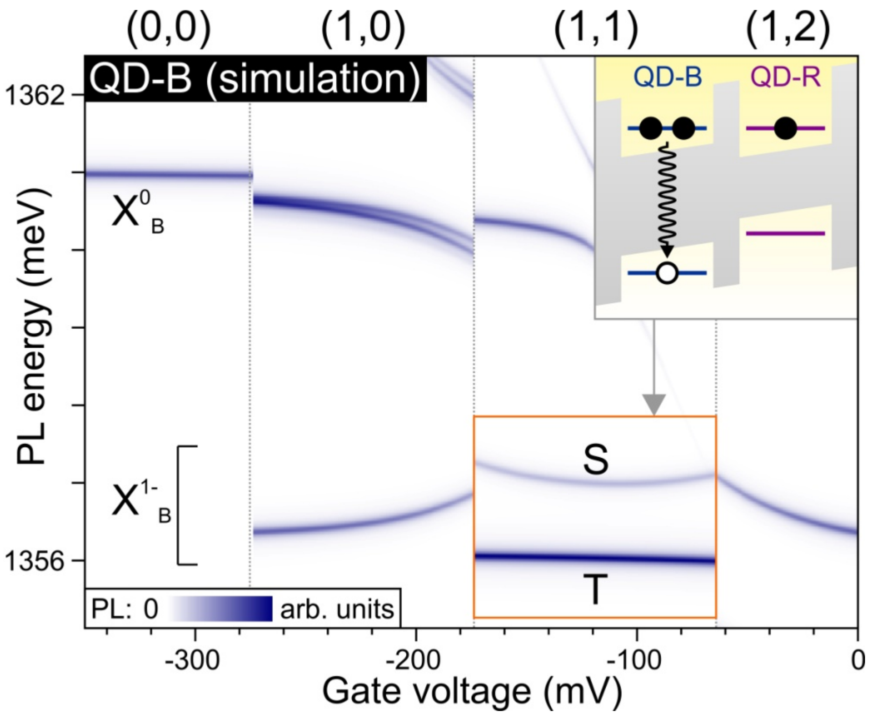


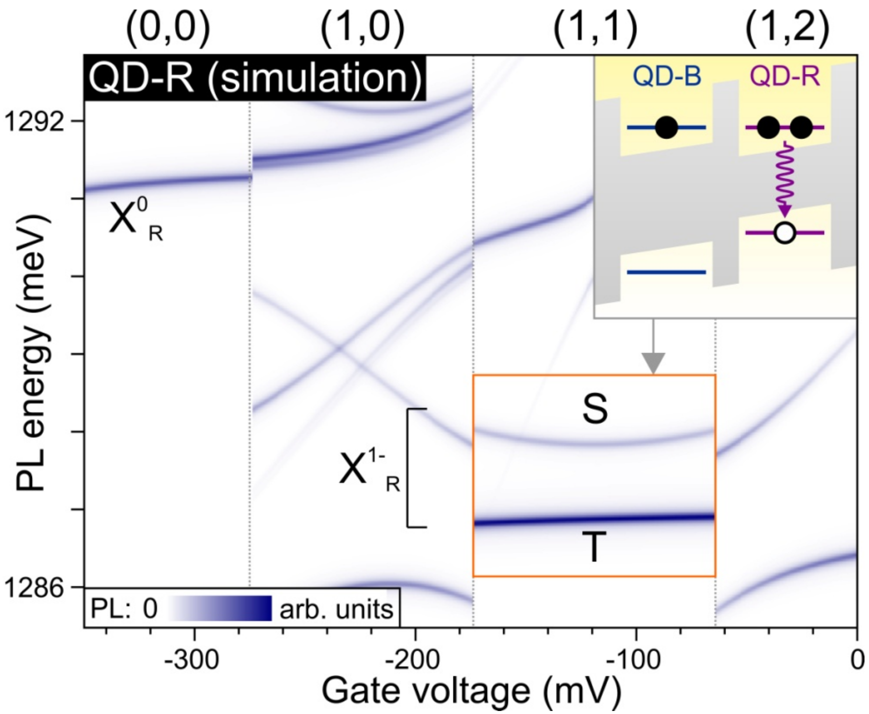


- PL versus gate voltage shows characteristic plateaus
- Shape of plateau influenced by electrons in partner QD
- Charging sequence: (0,0) > (1,0) > (1,1) > (1,2)

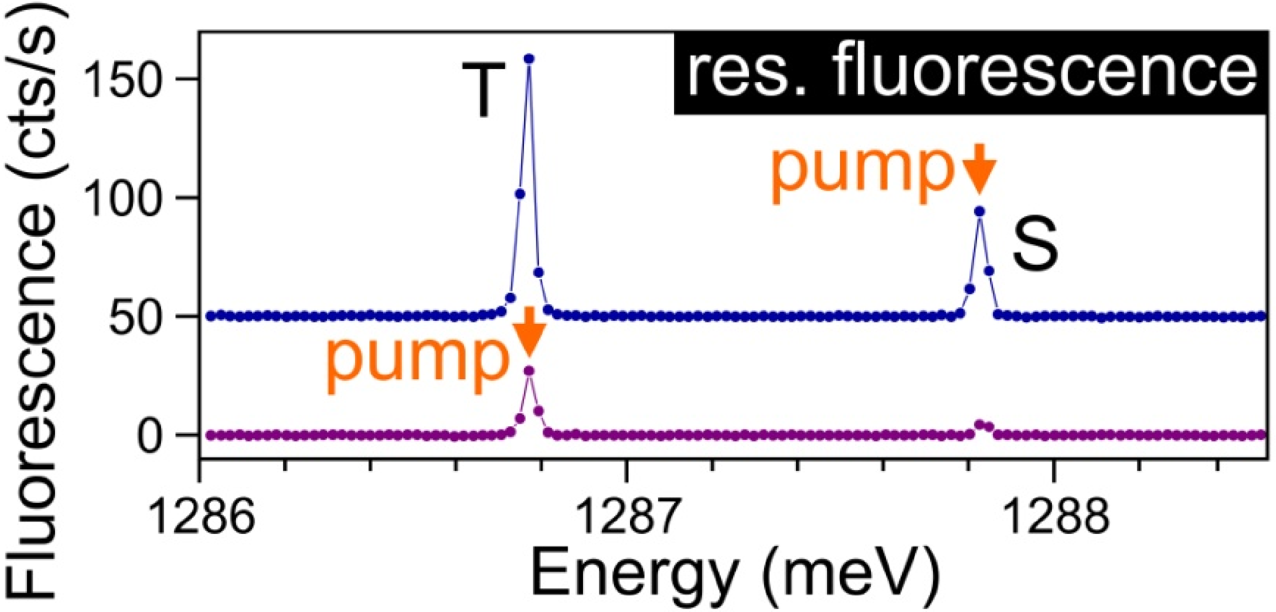
- (1,1)S shows typical curvature and 3 times lower PL intensity
- Very large 1.1 meV exchange splitting between S and T
- ***Sweet spot can be reached by tuning gate voltage!***

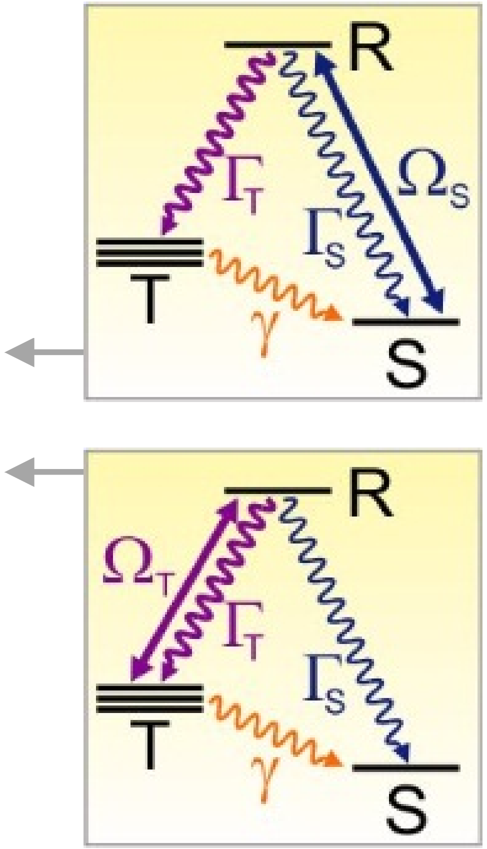
Numerical simulation of PL plateaus





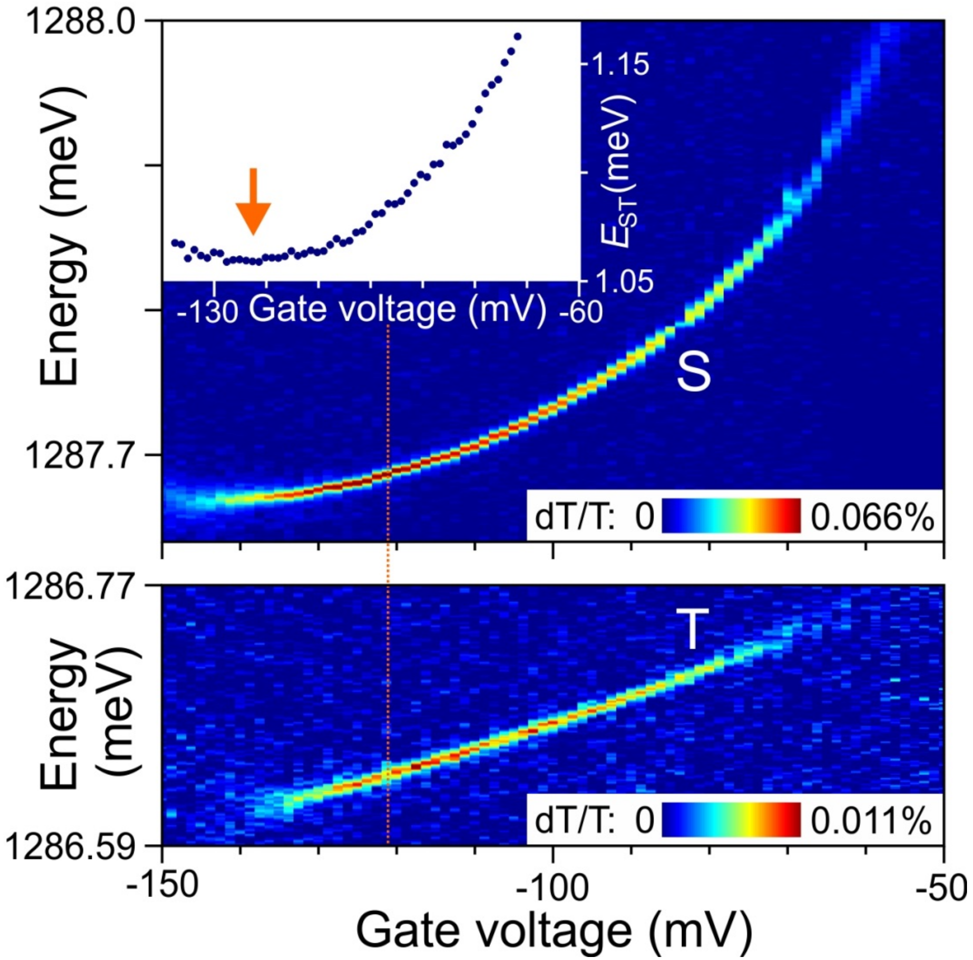
Resonance fluorescence reveals Λ scheme





- Resonantly drive S transition ⇒ fluorescence at S (Rayleigh) and T (Raman)
- T fluorescence ~3 times stronger ⇒ justifies simple lambda system picture
- Pumping T gives ~8 times less fluorescence than pumping S

Resonant spectroscopy reveals sweet spot



- Scan single laser through S or T resonance and measure differential transmission (dT) or differential reflection (dR)
- Sweet spot can be reached
- ***BUT:***
  no spin pumping even in middle of (1,1) regime
- Indicates strong spin-flip cotunnelling with back contact
- ***CONCLUSION:*** sample not suitable for studying spin coherence between S & T
- ***USE FOR:*** laser amplification

JME, K. Weiss, J. Miguel-Sanchez & A. Imamoglu, PRL **107**, 017401 (2011)

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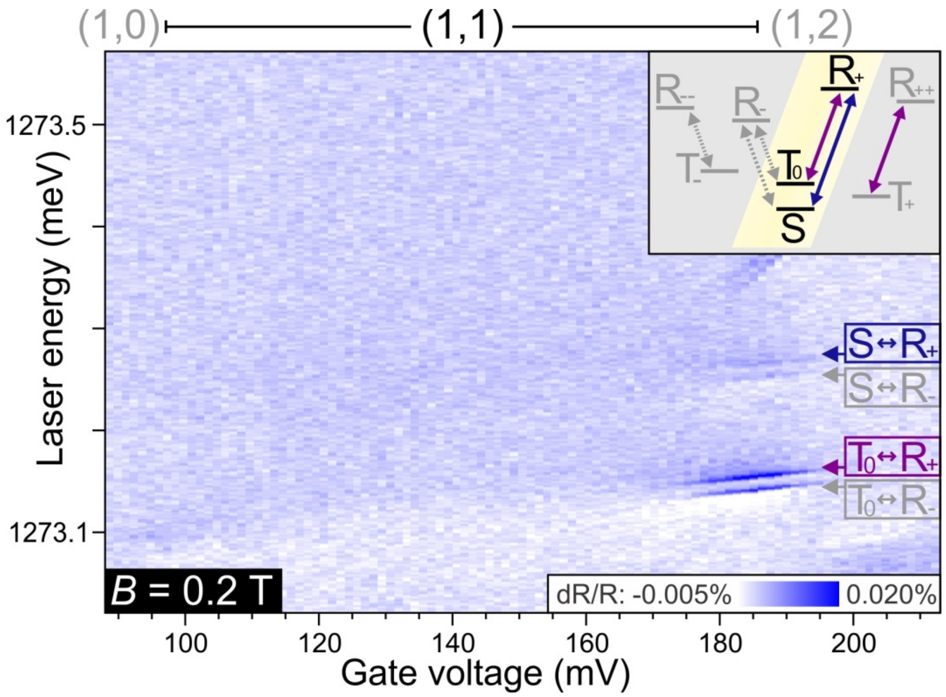
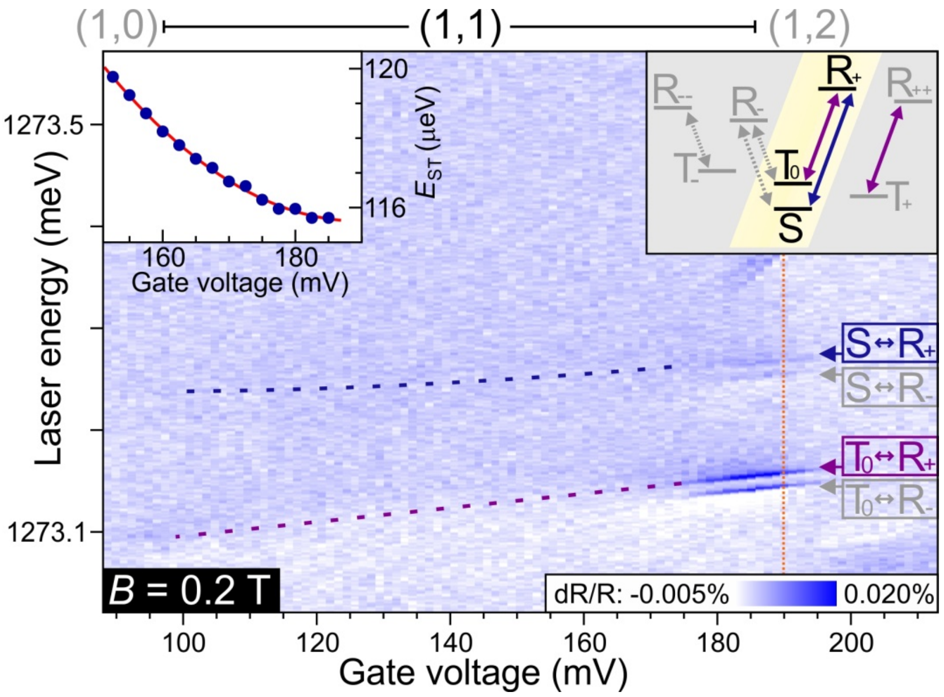
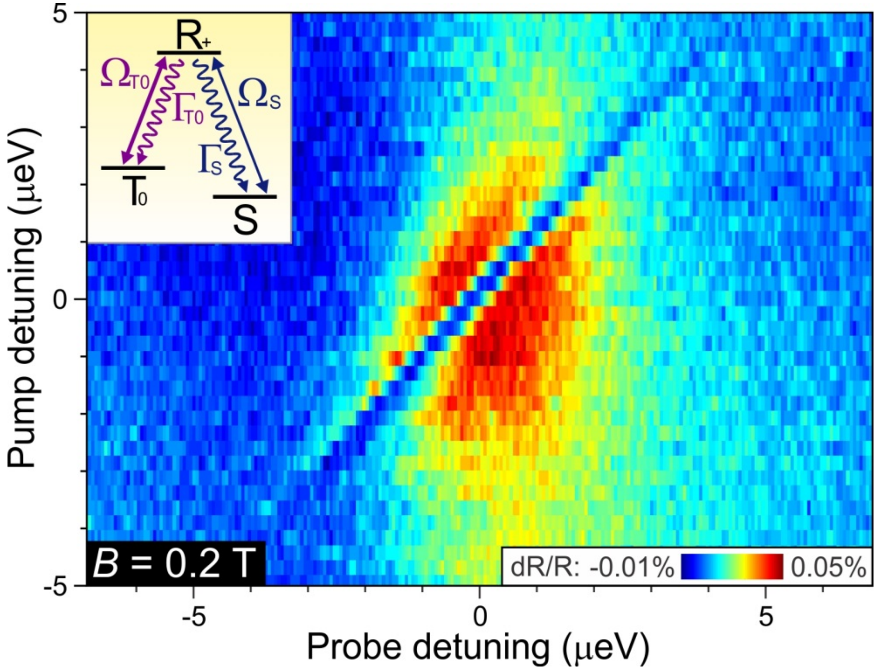
Improve device to show spin pumping



- B_z = 0.2 T ⇒ $T_\pm$ split off from T_0
- dR signal vanishes away from (1,1) plateau edge (spin pumping)
- Cotunneling rate reduced!

Improve device to show spin pumping

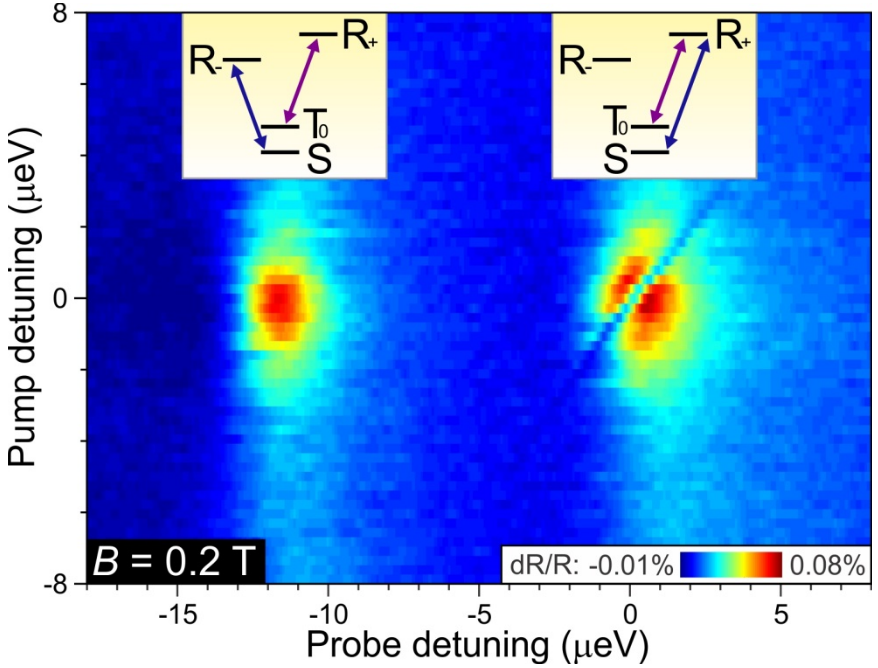


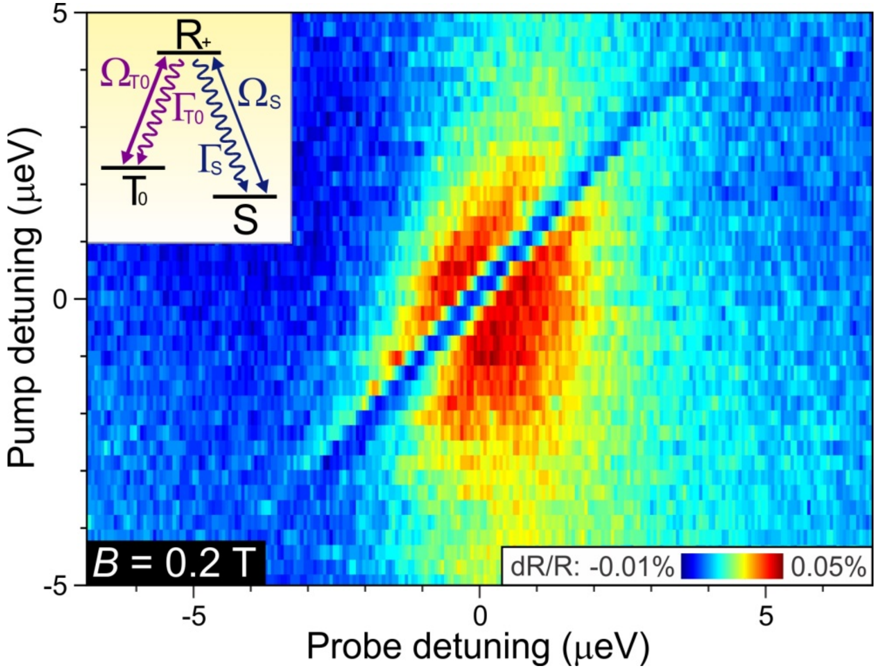


- B_z = 0.2 T ⇒ $T_\pm$ split off from T_0
- dR signal vanishes away from (1,1) plateau edge (spin pumping)
- Cotunneling rate reduced!

- dR signal restored by adding 2nd laser on other transition
- "Sweet spot":
  V_0 ~ 190 mV just outside (1,1)…

Coherent population trapping with two spins

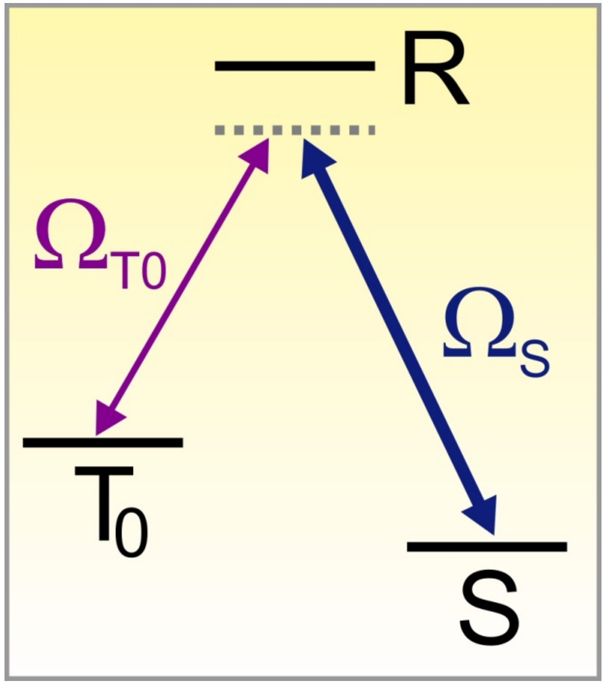




- Pump and probe orthogonal linear polarization ⇒ suppress reflected pump laser before detector
- Pump T_0 – R+ and probe S – R+
- CPT dip when probe hits S – R+ due to antisymmetric superposition of S and T_0

- Pump T_0 – R+ and probe S – R+ transition ⇒ clear CPT-dip at 2-photon resonance
- Large pump: dR signal vanishes completely, CQD fully transparent
- Weaker pump: depth of dip sensitive to dephasing between S and T_0

Coherent population trapping

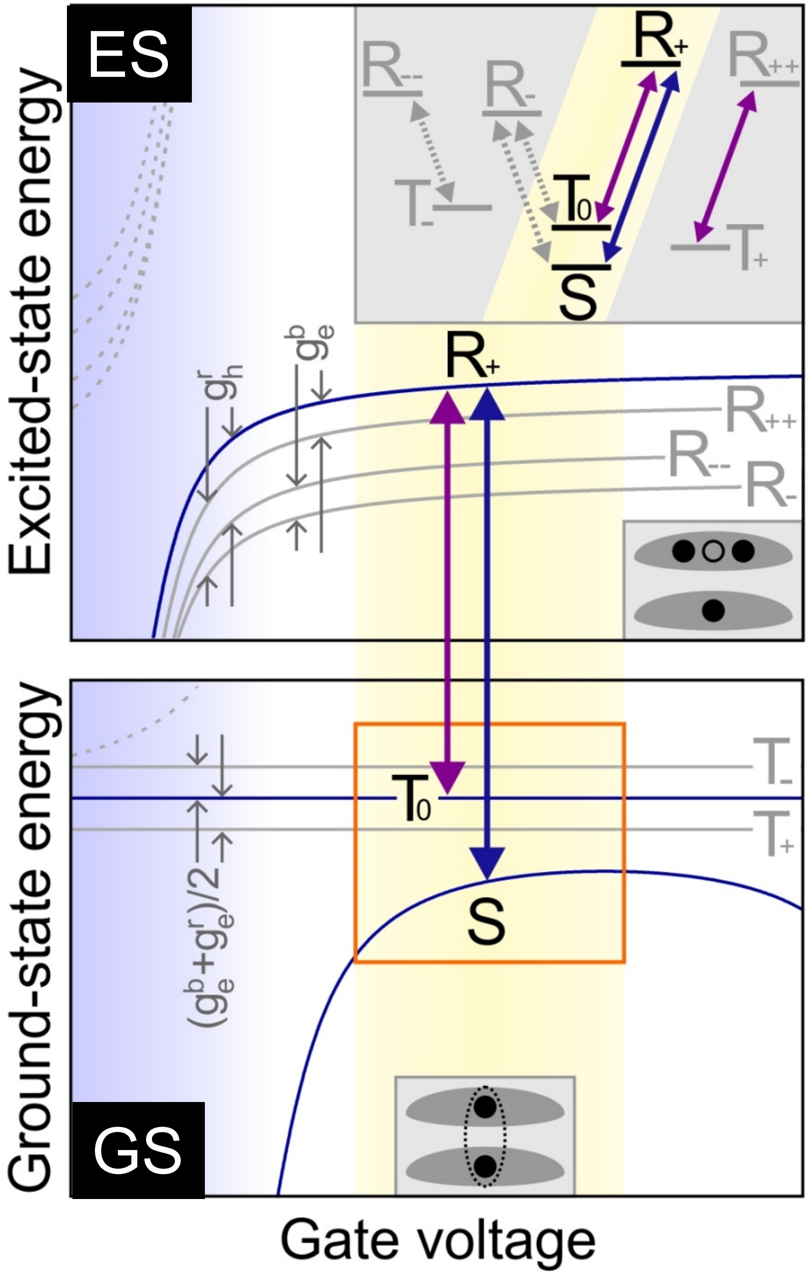


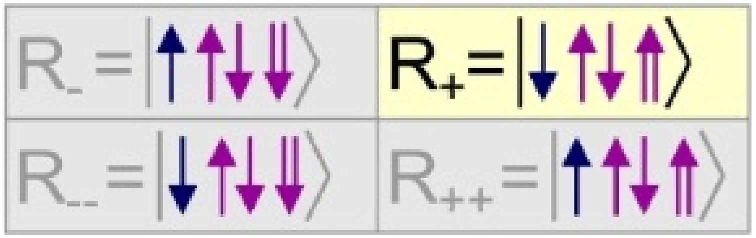
- Stimulated Raman scattering
- Three states in lambda system dressed by coherent laser fields
- One of the dressed states is linear superposition of two GSs without contribution from ES: ***dark state!***
- Dark state transparent to laser light: transmission dip
- CPT dip sensitive to coherence between ground states
- Similar to time-resolved coherent spin manipulation using short pulses: ***but in frequency domain!***

$$|\text{dark state}\rangle = \cos\theta\,|S\rangle - \sin\theta\,|T_0\rangle$$
$$\tan\theta = \Omega_S/\Omega_{T_0}$$

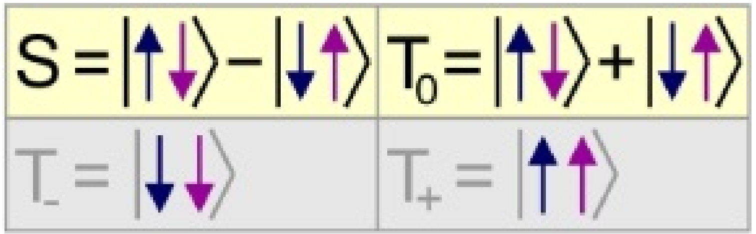
M. Fleischhauer, A. Imamoglu, J.P. Marangos, Rev. Mod. Phys. **77**, 633 (2005)

Numerical simulations of full 8-level system





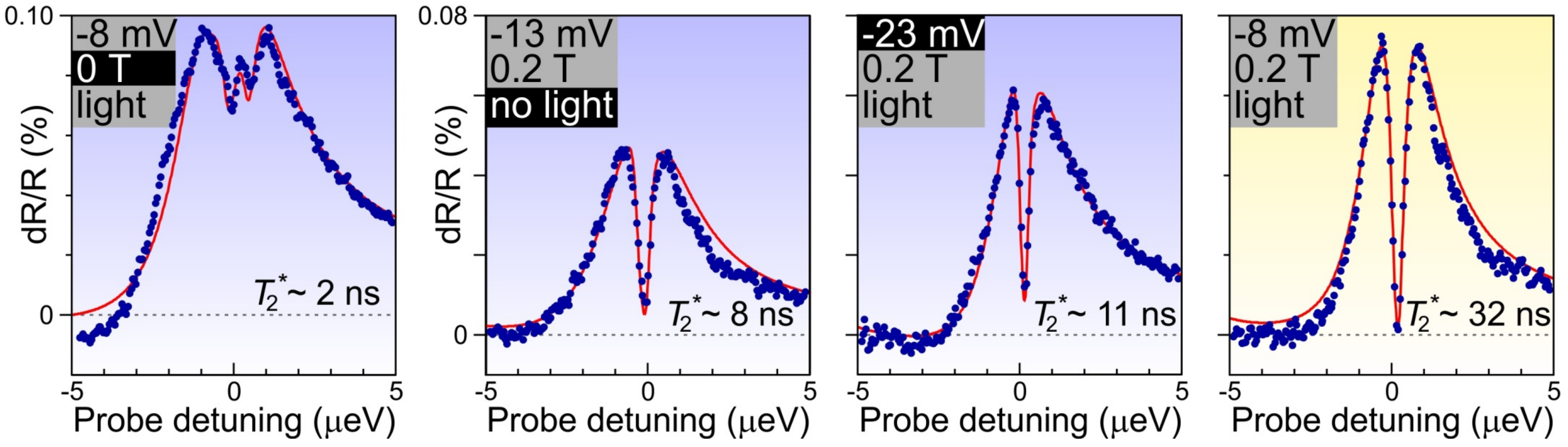
$R_- = \lvert \uparrow\uparrow\downarrow\Downarrow \rangle$	$R_+ = \lvert \downarrow\uparrow\downarrow\Uparrow \rangle$
$R_{--} = \lvert \downarrow\uparrow\downarrow\Downarrow \rangle$	$R_{++} = \lvert \uparrow\uparrow\downarrow\Uparrow \rangle$



$S = \lvert \uparrow\downarrow \rangle - \lvert \downarrow\uparrow \rangle$	$T_0 = \lvert \uparrow\downarrow \rangle + \lvert \downarrow\uparrow \rangle$
$T_- = \lvert \downarrow\downarrow \rangle$	$T_+ = \lvert \uparrow\uparrow \rangle$

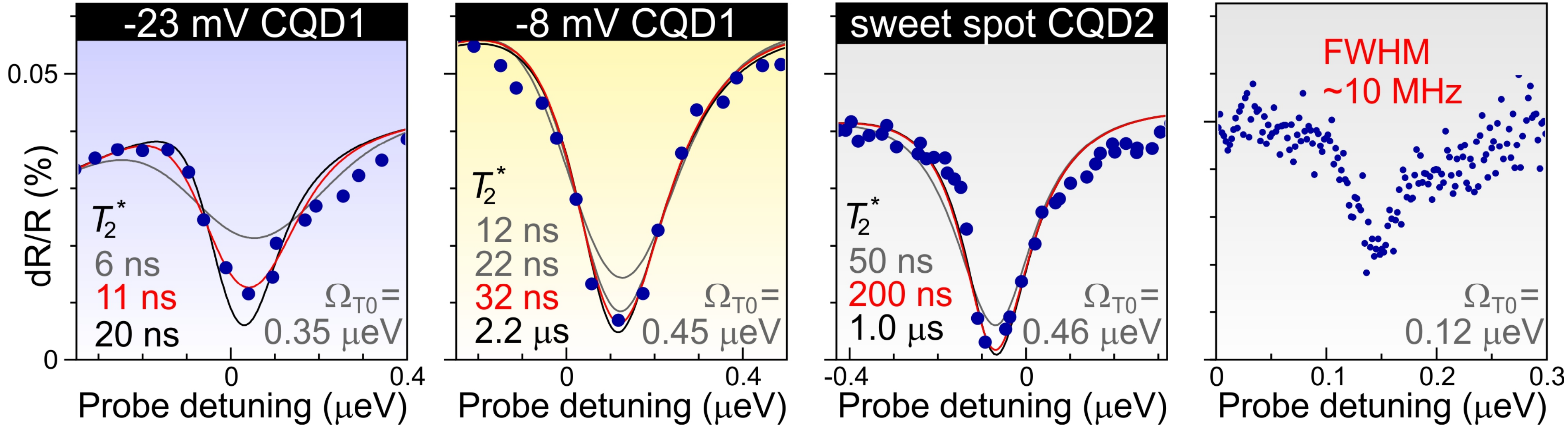
- Simulate full 8-level system (4 ground and 4 excited states) using master equation formalism
- Model fast processes (electron cotunneling with reservoir) in Lindblad form
- Model slow charge fluctuations by “Gaussian averaging” of multiple traces
- Extract T_2^* by simulated Ramsey

CPT dip as probe of S – T0 coherence



- At $B = 0$: in-plane component of nuclear field mixes all T states
  $\Rightarrow$ three CPT dips (one obscured by asymmetry)
- Without non-resonant (850 nm) laser: more charge fluctuations
- Tune closer to sweet spot: CPT dip becomes deeper
- Simulate full 8-level system (4 ground and 4 excited states) using master equation formalism
- Extract T_2* Model fast processes (electron cotunneling with reservoir) in Lindblad form
- Model slow charge fluctuations by "Gaussian averaging" of multiple traces
- Due to proximity of sweet spot to plateau edge: spin-flip tunneling limits spin coherence
- ***Find better CQD pair!***

Enhancement of T_2* close to sweet spot



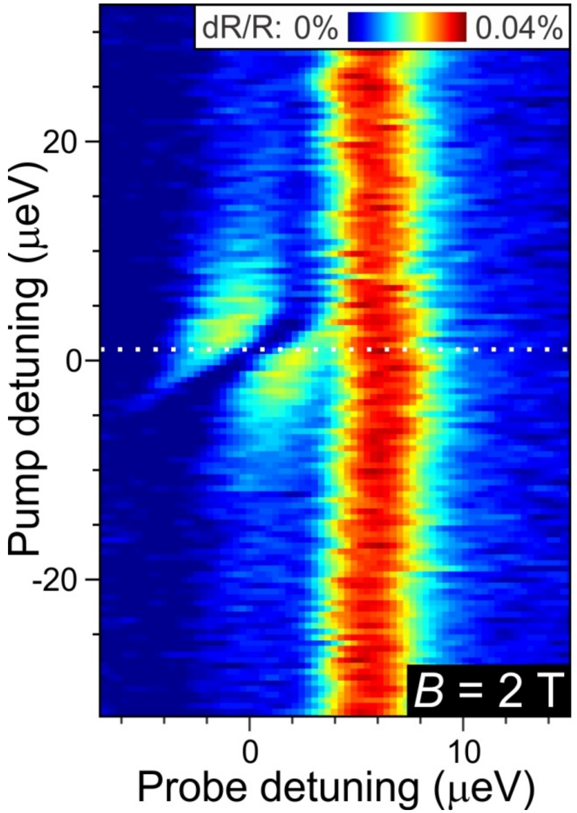
- Measure CPT dip for various pump powers
- Fit dip with full 8-level master equation in steady state, including two decoherence mechanisms:
  slow charge fluctuations (give Gaussian dip) plus
  fast spin-flip tunneling with back contact (Lorentzian dip)
- T_2* > 200 ns: ~100 times better than for single electron spin
- Imperfect sample: charge fluctuations $\delta V \sim 0.6\ \mathrm{mV}$

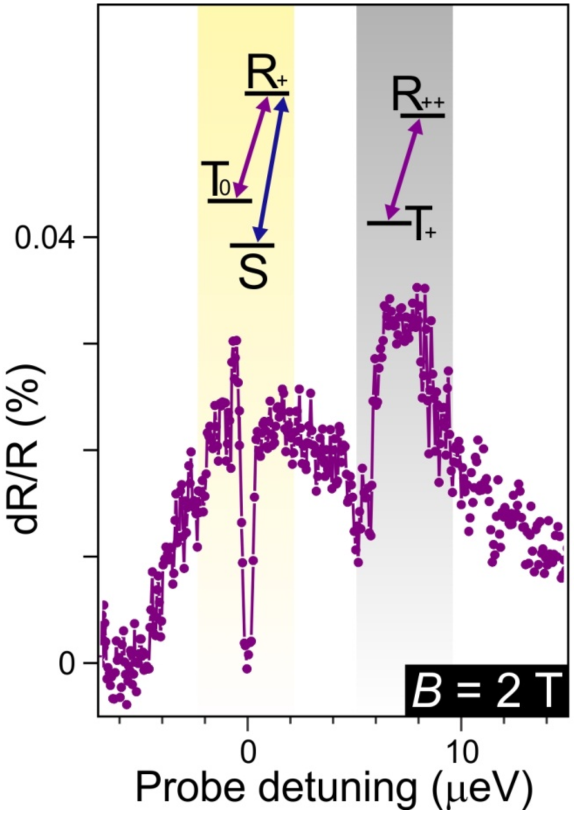
FWHM ~10MHz for lowest pump power used ⇒

high-resolution spectroscopy in solid state; probe for environment

K. Weiss, JME, Y.L. Delley, J. Miguel-Sanchez & A. Imamoglu, *PRL* **109**, 107401 (2012)

Opportunities beyond the lambda system





- Electronic g-factors for two dots ~10% different $\Rightarrow$ two σ+ transitions slightly detuned at *B* = 2 T
- One transition is part of lambda system $\Rightarrow$ very efficient spin pumping

- Other transition is quasi-recycling $\Rightarrow$ maintains dR contrast even away from pump resonance
- Could be useful for spin read-out or nuclear spin preparation

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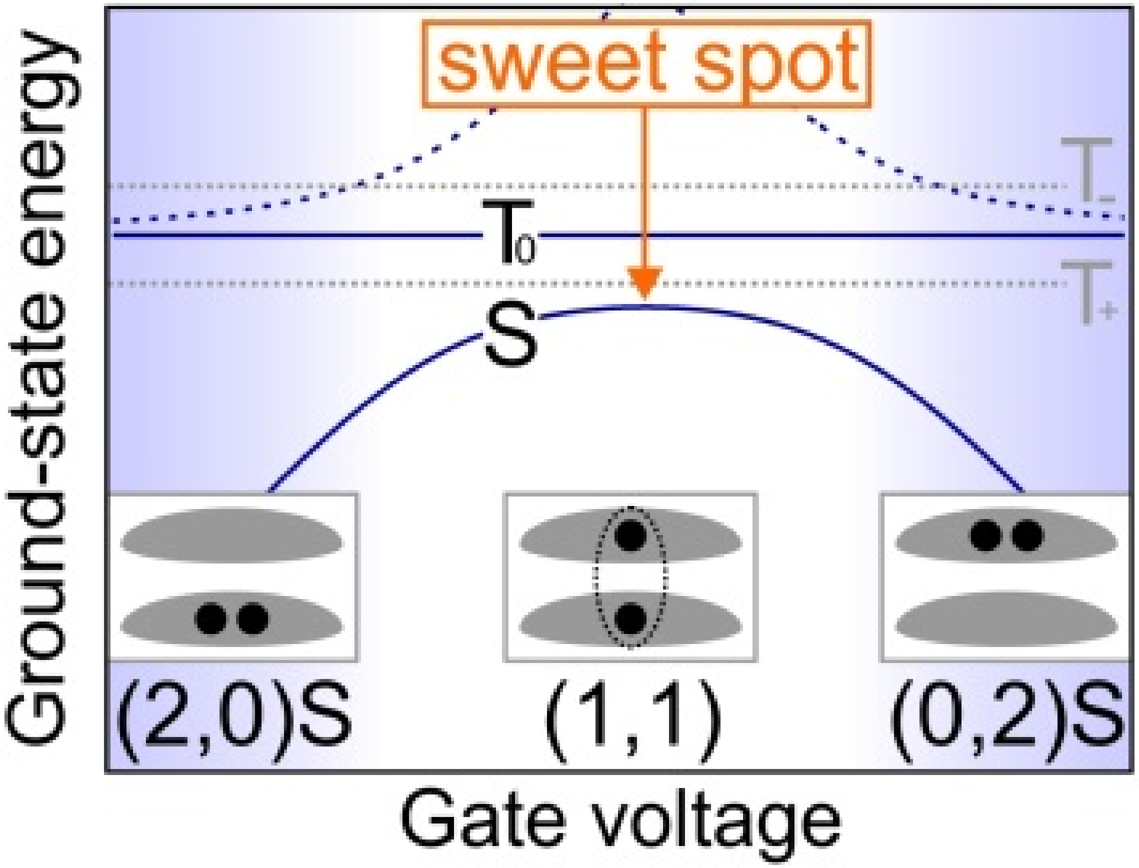
Two-electron spin states



At "sweet spot": S/T0 qubit states (to first order) insensitive to charge fluctuations!

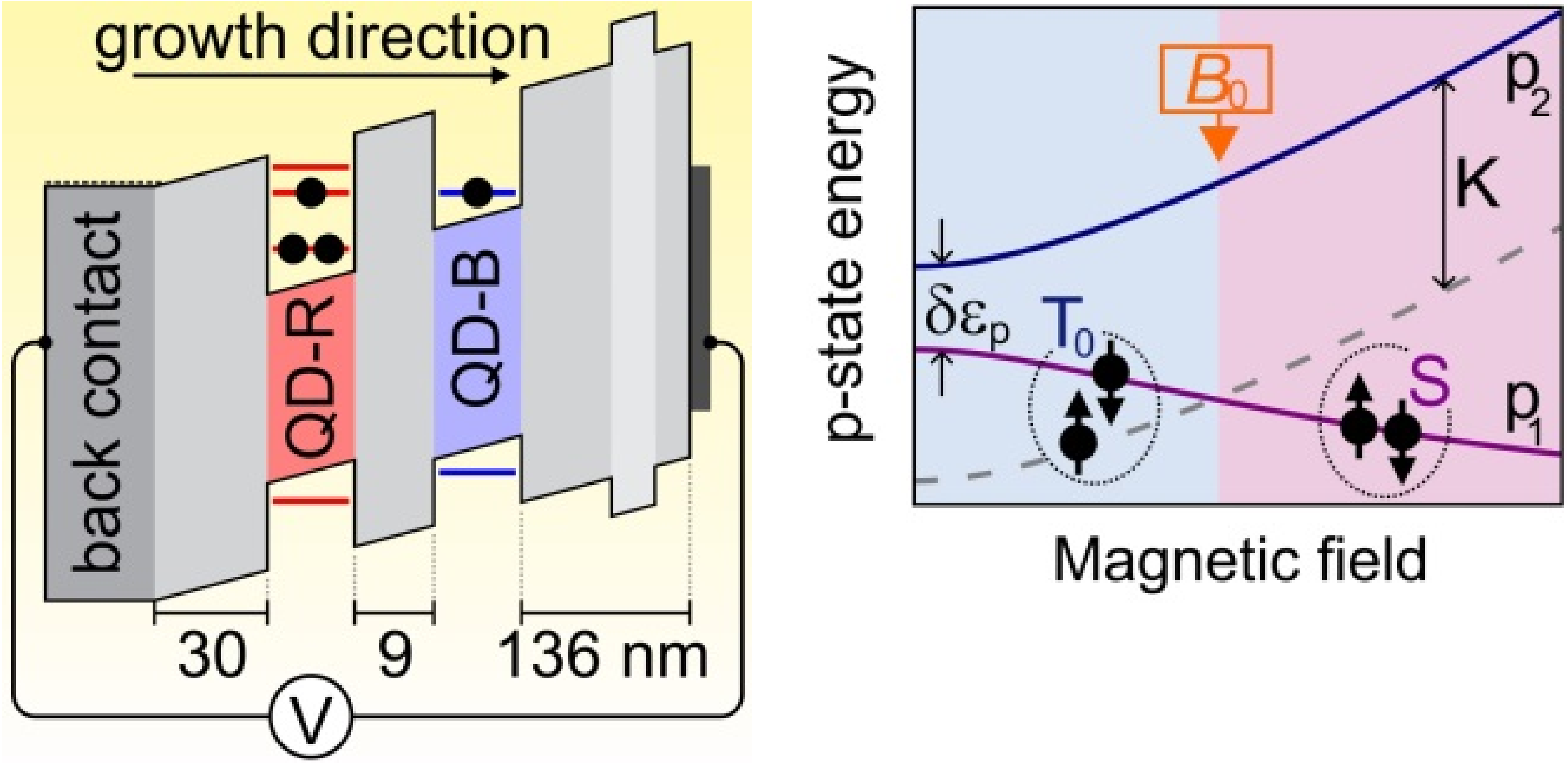
D. Vion *et al.*,
Science **296**, 886 (2002)

J. Koch *et al.*,
PRA **76**, 042319 (2007)

D.A. Lidar *et al.*,
PRL **81**, 2594 (1998)

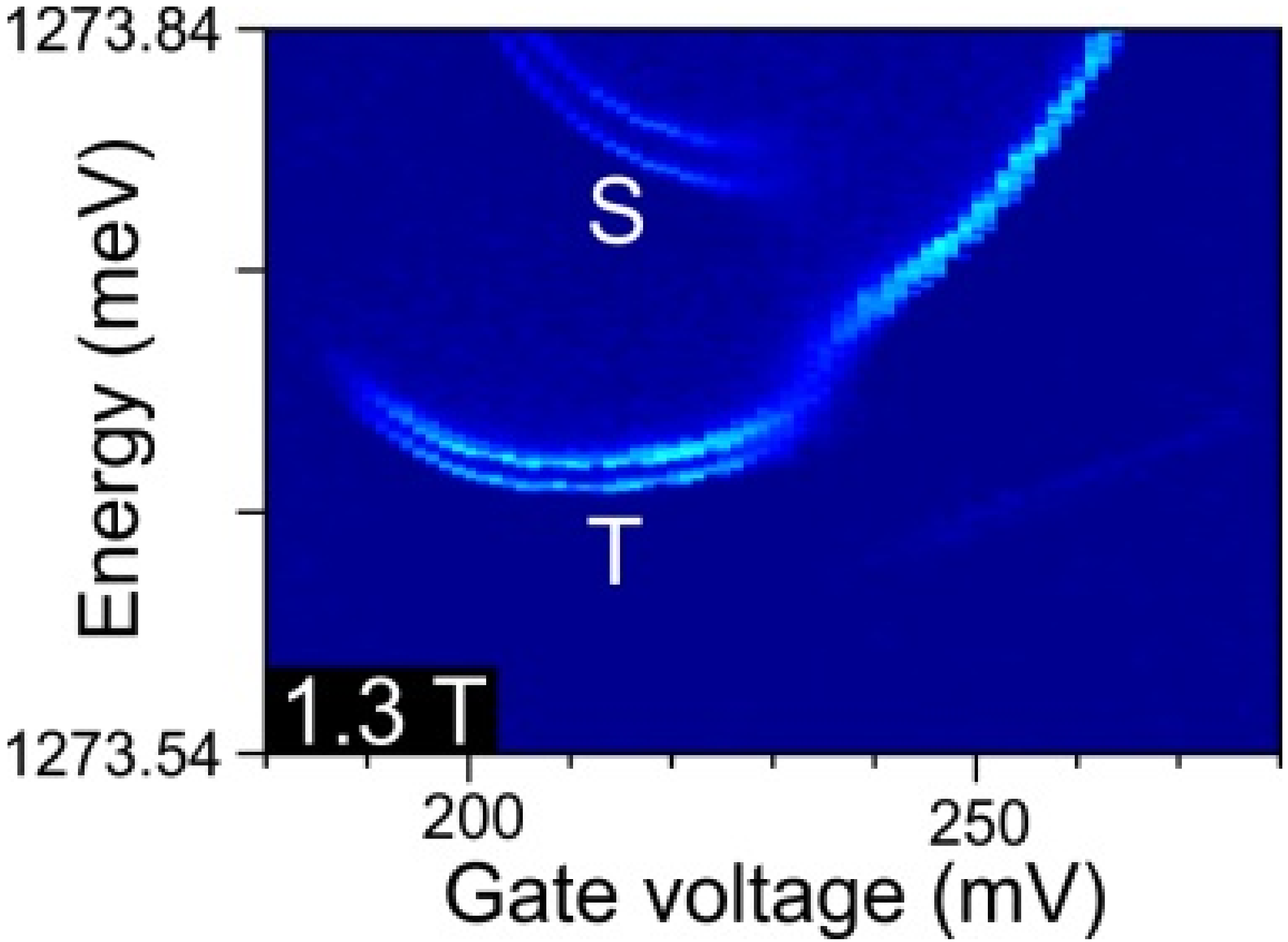
- ST splitting at sweet spot given by microscopic parameters of CQD
- ST splitting not tunable via B-field
- Not allowed to tune it via gate voltage
- Makes it hard to tune qubit into resonance with other qubits

Four-electron singlet-triplet qubit



K.M. Weiss, J. Miguel-Sanchez & JME, *Scientific Reports* **3**, 3121 (2013)

Four-electron singlet-triplet qubit



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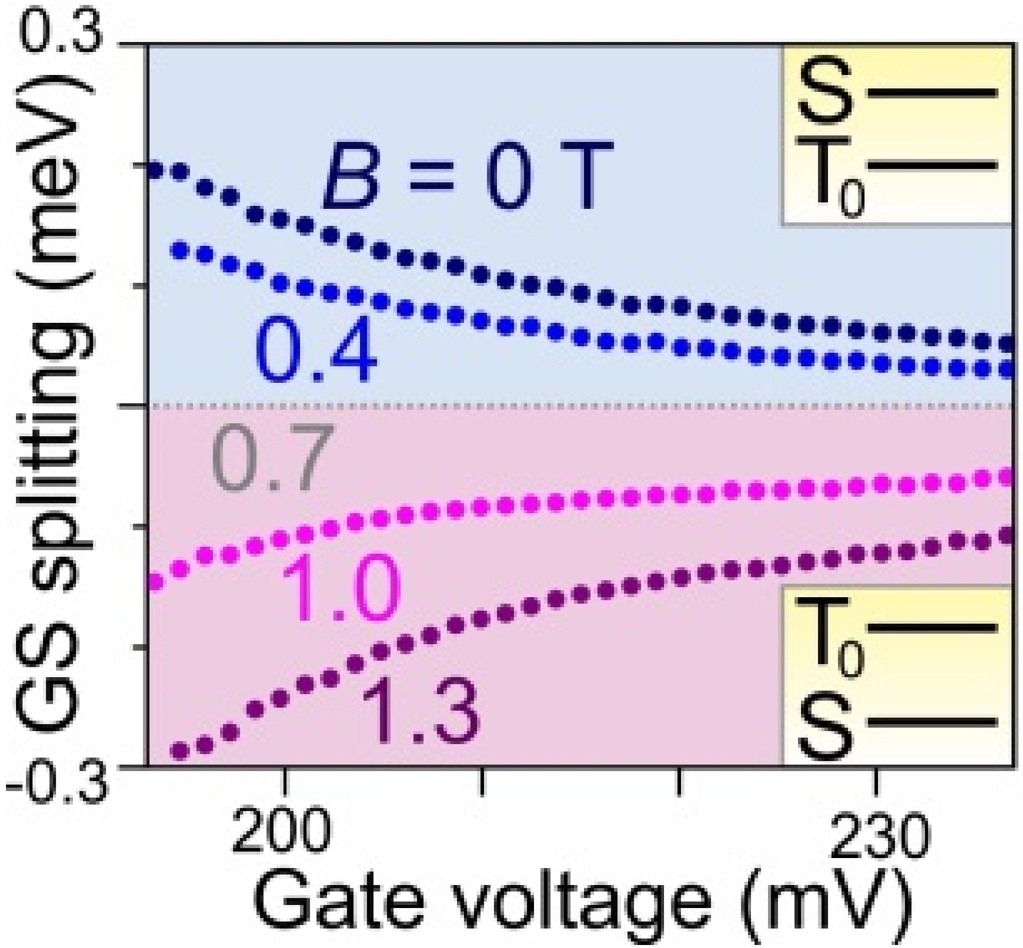

K.M. Weiss, J. Miguel-Sanchez & JME, *Scientific Reports* **3**, 3121 (2013)

Conclusions and outlook

Achievements:

- Coupled quantum dots can be used to engineer “atomic-clock states”
- Two-electron S and T_0 states can form qubit that is robust against both charge and nuclear spin fluctuations (to first order)
- At sweet spot and away from edge of charging plateau, coherence time is enhanced by (at least) two orders of magnitude
- Combining ST qubit at sweet spot with spin echo should lead to even longer T_2
- Four-electron ST qubit features magnetically tunable level spacing

Main challenge is scalability!

- Lateral positioning of QDs still challenging $\Rightarrow$ makes coupling several qubits hard
- Use novel single-layer materials (e.g. MoS_2, WSe_2) to engineer quantum dots
- Use single quantum emitter to generate highly entangled state of multiple *photonic* qubits (cluster state)