WALTER-SCHOTTKY-PREIS

Stopping spins spinning

Slowing of spin relaxation in semiconductor quantum dots

Jonathan J. Finley

The magnetic moment or "spin" of an individual electron is an unimaginably tiny quantity. It is so small that it was only during the past few years that it became possible to detect single spins in solids, manipulate them and directly study their behaviour. Special spin memory devices have been developed that enable the optical generation of single charges in semiconductor artificial atoms. Remarkably, the spin of these charges can be programmed by the polarisation of the photons used to create them. Localising spins in such nanostructures dramatically suppresses their coupling to the outside world, making them potentially useful for quantum information processing.

The discovery twenty years ago of Giant Magneto Resistance (GMR) [1] in ferromagnetic metal-insulator multilayers heralded the birth of a new paradigm for device functionality, spin electronics or *spin*tronics, where it is not the electronic charge but its spin that is used for information processing or magnetic sensing applications [2]. Since then a diverse array of commercially available spintronic devices have emerged, ranging from the sensitive magnetic read-out head in hard disk drives to magnetic random access memory (MRAM) and magnetic logic gates to name only a few examples [3]. In comparison to these mostly metallic spintronic devices, the spin of charge carriers in semiconductors (electrons and holes) generally has little or no direct relevance for the operation of conventional devices. Instead, much stronger *electric* interactions between charges are harnessed to store information, perform logical operations, and even to generate light in light emitting diodes (LEDs) or lasers. Much research effort is currently being focussed on understanding and exploiting spin related phenomena in semiconductors. This is motivated by the desire to combine their many remarkable properties, including tunable energy bandgaps, voltage switchable carrier densities, strong optical activity and mature nanofabrication technologies, with the possibility to controllably switch *on* and *off* magnetic effects in solids [4].

Whilst the idea to make use of magnetic phenomena in semiconductors is relatively new, the drive to realise ever smaller electronic and optoelectronic devices is already well established (**Fig. 1**). This downscaling trend is fuelled by the commercial pressures exerted by the microelectronics industry[1)] and the search for useful new effects in nanoscale materials. In particular, the past fifteen years have witnessed a huge upsurge of interest in semiconductor quantum dots (QDs); nanometre sized boxes that can localise charge carriers in all three spatial directions to lengthscales comparable to their de Broglie wavelength. Most semiconductor QDs confine carriers using one of two approaches: by locally modulating the chemical composition and

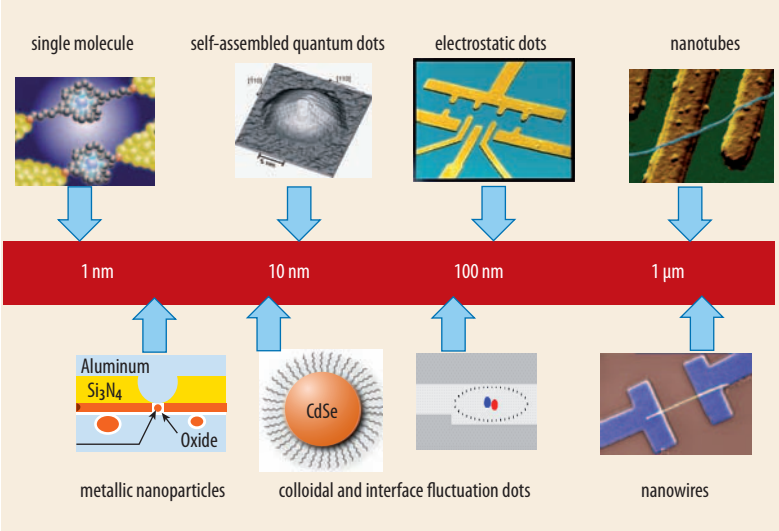

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Fig. 1 The typical motional confinement lengthscales for different quantum structures realisable in research laboratories can be as small as a few nanometres. Each of these systems is capable of confining the motion of charge carriers to lengthscales comparable to their de Broglie wavelength in two or more spatial directions. The physical dimensions of quantum dots range from the size of single molecules to that of nanotubes or wires.

1) See www.itrs.net

IN BRIEF

- The use of the spin degree of freedom in semiconductor devices has strong potential for spintronics and quantum information processing.
- To use a spin as a quantum bit it is necessary to initialise, manipulate and control the spin orientation and to suppress the strong coupling between the spin and its environment. To satisfy the last criterion quantum dot (QD) semiconductor nanostructures can be used.
- In charge memory devices optically generated holes are removed by an electric field, while electrons are stored and remain in the QDs for more than 1 ms.
- The spin orientation of optically generated charges can be programmed via the circular polarisation of the optical excitation. This allows for the investigation of mechanisms and timescales of electron spin flip relaxation.

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effective bandgap of a semiconductor crystal relative to its environment [5] or by electrostatically defining a potential minimum within a two-dimensional quantum film using metallic gates on the sample surface [6]. These two types of dots differ fundamentally in their ability to confine carriers: chemically defined dots can simultaneously trap both electrons and holes and are thus optically active, whilst electrostatically defined dots trap only one charge polarity (usually electrons) and are therefore optically inactive.

In a semiconductor quantum well, which consists of a layer of narrow bandgap semiconductor sandwiched between a wider bandgap material, the motion in one direction is quantised into a series of discrete states in full analogy to a particle trapped in a one dimensional potential well. In such systems the particle is free to move in the plane of the quantum well, leading to a two-dimensional system with a continuous spectrum of allowed energy levels. In contrast, the absence of any motional degrees of freedom in both types of QD discussed above gives rise to an energetically discrete, atomic like electronic structure. These ideas are illustrated in **Fig. 2a**, which shows a sketch of the typical confinement potential and discrete electronic structure for a chemically defined dot. The nomenclature s, p, d etc. is commonly adopted to describe the orbital character of these lowest lying quantum states, in analogy to the orbital states of atoms.

If one measures some property, such as the current flowing through the dot or the frequency of emitted photons, the result will depend on the number of charges already contained by the dot. Fundamentally, this arises from strong Coulomb interactions between the charges trapped inside the dots. In this sense the electrical or optical response of a QD is highly *non-linear*, even at the level of a single charge. This extraordinary sensitivity to charge occupation underpins the operation of many novel devices in the fields of nano-electronics and nano-photonics. Examples include the single electron transistor that can be switched on or off by adding or removing a single charge on the gate electrode [6] or deterministic single photon sources that produce streams of single photons for applications in the field of quantum cryptography [7]. Whilst single charges can now be successfully manipulated to build useful devices and single photons can be generated with an ever increasing efficiency, it is difficult to harness the *spin* of isolated charges in solids for applications. Indeed, it was only during the past few years that researchers gained experimental access to single spins [8–11] and developed the capabilities to initialise, manipulate and control their orientation using clever combinations of optical [9, 10, 12] and electrical [11] techniques.

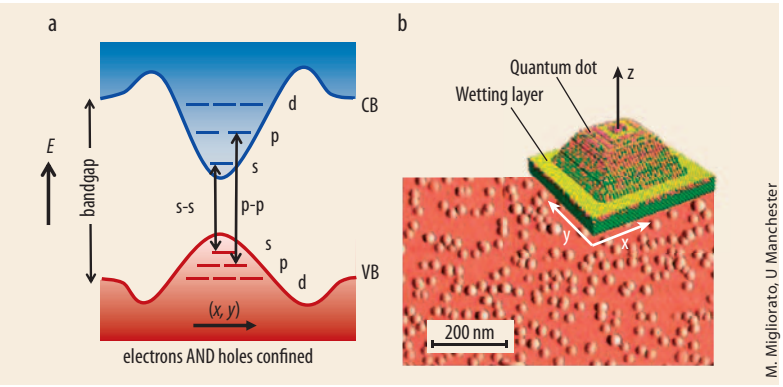

Fig. 2 (a) A series of energetically discrete electronic states is formed in the valence band (VB) and conduction band (CB) of self-assembled QDs due to a local reduction of the bandgap within the nanostructures. As a result, such QDs are optically active. In (b) the atomistic image of an InGaAs self-assembled QD is compared to a plan view atomic force microscope image. The formation mechanism of self-assembled QDs results in a dense array of QDs on the growth surface with inhomogeneities in size and, thus, the confinement energies.

Quantum dot charge and spin memory devices

From the fundamental physics perspective, a single spin in a magnetic field can be considered to be a prototypical two level quantum bit, or qubit – the basic logical unit of a quantum computer [13]. For a qubit to be useful a number of conditions must be satisfied; it must be possible to initialise it into one of its basis states (e. g. $|\uparrow\rangle = |0\rangle$ and $|\downarrow\rangle = |1\rangle$), generate arbitrary quantum superpositions ($\Psi = \alpha\,|\uparrow\rangle + \beta|\downarrow\rangle$) and allow these mixed states to evolve coherently under the action of some controlled interaction Hamiltonian. Moreover, for quantum logic operations it must be possible to manipulate and readout each qubit separately and switch on and off interactions between them using some convenient control parameter, such as the voltage applied to a gate electrode or a laser pulse tuned to a particular optical transition.

In reality, each of these requirements is very difficult to satisfy. The problem for spin qubits, indeed for all qubit implementations, lies with the unwanted and uncontrolled evolution of superposition quantum states due to coupling with their environment. If the timescale over which these couplings occur is shorter than, or even comparable to, the time required to controllably manipulate the system, then computation cannot proceed and the "quantum computer" will be useless! Thus, major efforts have been undertaken in recent years to understand the mechanisms by which spins in semiconductors and their nanostructures interact with their environment. Understanding these mechanisms and developing strategies to suppress them, would be a major step along the road towards a solid-state quantum processor.

We continue to discuss optical studies of spin relaxation in semiconductor QDs and show that the spins are largely prevented from interacting with their environment. After discussing the major physical, electronic and optical properties of optically active QDs, we describe how their non-linear response to laser excitation allows single charges to be optically generated. We show that the angular momentum of photons can be reversibly imprinted onto the spin of the generated charges, allowing investigation of the mechanisms and timescales for electron spin flip relaxation in QDs.

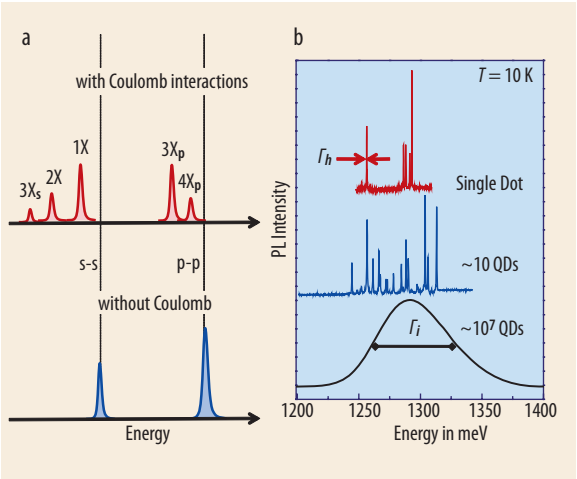

Fig. 3 Without Coulomb interactions the spectrum of a single QD consists of just a few inter-band transitions between orbital states (a, lower panel). Coulomb interactions lift the degeneracy of each few particle state (one e-h pair (1X), two e-h pairs (2X) etc.) resulting in a more complex spectrum. These effects can be seen in the optical emission from a single dot which reveals a large number of discrete transitions, each arising from a specific number of electrons and holes in the dot (b).

Of the many types of optically active QDs islands formed by strain driven self-assembly are amongst the most widely studied [5]. Self-assembled growth occurs when two semiconductors with significantly different lattice constants are grown epitaxially. For InAs growing on GaAs there is a lattice mismatch of $\Delta a/a = 7\,\%$. As a result, when InAs is deposited on GaAs during crystal growth it initially forms a strained two-dimensional layer referred to as the wetting layer. However, beyond a critical thickness of one or two atomic layers the growth transforms from two dimensional (layer by layer) to three dimensional (clustering) with the result that arrays of nanometre sized islands are formed on top of the wetting layer (**Fig. 2b**). This two to three-dimensional growth transformation is driven by a reduction in the elastic energy as the material in the islands is able to relax laterally, being unconstrained by surrounding material, although at a cost of an increased surface energy [14]. For optical applications these nanoscale islands are subsequently overgrown by a wider bandgap semiconductor to produce fully encapsulated nanostructures [5]. For GaInAs grown on GaAs, the typical heights of the QDs formed are of the order of 2 – 5 nm with base widths of approximately 20 nm. As shown by the atomic force micrograph in **Fig. 2b** areal densities are typically in the range of $10^9 - 10^{11}\,\mathrm{cm}^{-2}$ with the dots exhibiting high radiative efficiencies due to their excellent crystalline quality [5].

Without Coulomb interactions between the confined charge carriers, the absorption or emission spectrum of a single quantum dot would consist of a very small number of discrete lines arising from optical transitions between confined valence and conduction band states (**Fig. 3a**). However, upon switching on Coulomb interactions, each optically active configuration of electrons and holes (e. g. $1e + 1h$ = exciton, $2e + 2h$ = biexciton, $3e + 3h$ = triexciton, $2e + 1h$ = charged exciton etc.) has a distinct transition frequency as depicted schematically in **Fig. 3a**. At low temperature the homogeneous optical linewidth of each of these few-particle states is much smaller than the typical energy shifts between the states ($\Gamma_h \sim 5\,\mu\mathrm{V}$ c. f. $\Delta E \sim$ meV), and a laser tuned to the $1e + 1h$ single exciton transition of an empty dot will result in the generation of a maximum of one electron hole pair in each quantum dot illuminated.

The optical absorption spectrum of a large ensemble of dots is inhomogeneously broadened due to unavoidable fluctuations of the QD size, shape and morphology during growth. The inhomogeneous absorption linewidth is typically $\Gamma_i > 30$ meV, much larger than the homogeneous absorption linewidth of a single dot. This idea is illustrated by **Fig. 3b** which compares the emission spectrum measured for a single GaInAs-GaAs QD with a large ensemble of ~10^7 dots. This disparity between Γ_i and Γ_h means that a laser tuned into the ensemble absorption will interact only with a small fraction of all illuminated dots that have a single exciton transition in resonance with the laser source; optical excitation selects QDs from the ensemble via their inter-band absorption frequency.

Such optically pumped QD "charge memory" devices were realised by a number of groups over the past five to ten years. First studies focussed on the non-specific electrical sensing of the optically generated charges [16–18] followed closely by the development of methods for optical readout [19, 20]. The basic structure and operating principles of an electron storage device are summarised in **Fig. 4**. A single layer of self assembled GaInAs QDs is embedded into the undoped region of

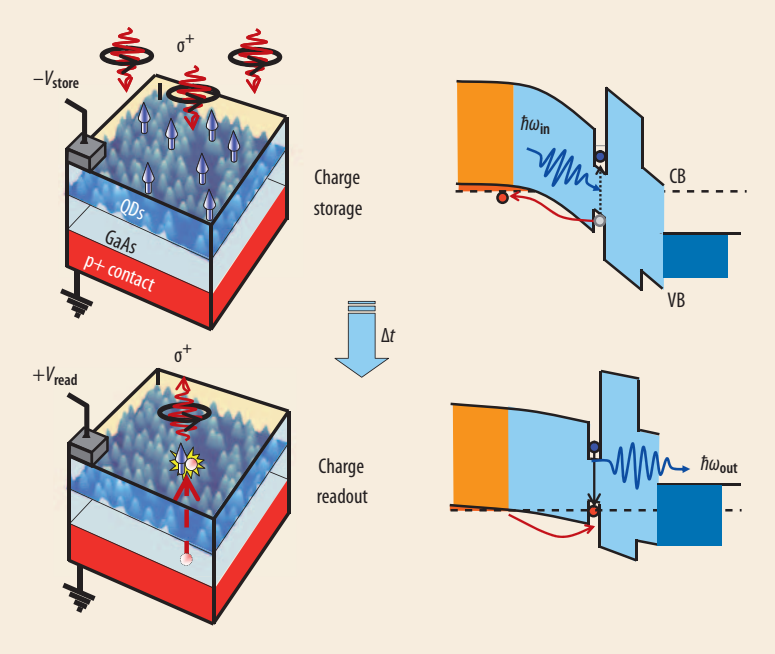

Fig. 4 In the charge *storage* configuration of the QD memory device a negative potential is applied between the semi-transparent surface gate electrode and the buried p-contact. This produces an electric field that removes optically generated holes from the QDs whilst electrons remain stored. To readout the stored charge, a positive potential is applied to the gate electrode resulting in holes being re-injected into the QD.

an (Al)GaAs semiconductor diode structure. In the charge storage condition, a negative potential ($V = -V_{store}$) is applied to the semi-transparent metal gate electrode on the sample surface with respect to the buried p-contact. This leads to the generation of a large static electric field along the growth axis of the dots. Single electron hole pairs are then optically pumped into the QDs at an energy $\hbar\omega_{in}$ and charge storage occurs when the hole tunnelling escape time is much shorter than the recombination lifetime of the exciton (~ 1 ns). This condition can be fulfilled by adjusting $|V_{store}|$ appropriately. Whilst the optically generated holes readily escape from the QDs by tunnelling, the electrons remain stored by virtue of a wider bandgap AlGaAs blocking barrier immediately above the QD layer (**Fig. 4**). After generation, the stored electrons can be tested after a well defined storage time Δt by positively biasing the Schottky junction $V = V_{read}$ for a short time. This results in a drift current of holes flowing into the negatively charged dots, thus neutralising the stored electrons and generating a time delayed electroluminescence (EL) signal. By switching on a sensitive single photon detector immediately prior to the reset voltage pulse, the intensity and spectrum of the emitted photons arising from the stored charge can be readily detected.

Typical charge storage EL spectra recorded at $T = 10$ K and for charge storage times of $\Delta t = 12$ µs are presented in **Fig. 5a** as the laser excitation energy is tuned through the inhomogeneously broadened QD absorption spectrum ($\hbar\omega_{in} = 1305 - 1370$ meV). A common feature in each spectrum is the pronounced peak which always appears close to $\hbar\omega_{in}$. This resonant peak arises from single electrons that are optically generated directly into the lowest orbital states of a small number of QDs and stored there until readout occurs as discussed above. Time dependent measurements show that electrons remain in the dots where they were created over very long (≫ 1 ms) timescales at low temperatures [21].

The spin orientation of the optically generated charges can be programmed via the circular polarisation of the optical excitation (see information panel), allowing for the study of spin flip relaxation dynamics in QDs. A few examples of the results of these measurements are presented in **Fig. 5b**, recorded at $T = 1$ K, $B = 8$ T and a storage time of $\Delta t = 2$ µs. Storage spectra following excitation with σ^+ and σ^- polarised light were recorded and analysed with σ^- (σ^+) discrimination in detection channel as shown by the red (black) curves on the figure. Following spin initialisation with circularly polarised light the storage EL is found to "remember" the polarisation of the light used to generate the electrons. The emitted light is predominantly co-polarised with a degree of polarisation $|P| = 65$ %, indicating that the spin orientation of the optically generated single electrons is preserved over timescales which are much longer than the $\Delta t = 2$ µs storage time. This observation demonstrates the reversible transfer from optical polarisation to electron spin orientation, followed by spin storage for a time of 2 µs and back-transfer from electron spin orientation into optical polarisation. The devices can, therefore, be considered to operate as a "spin memory"; storing the angular momentum of photons in the spin of stored electrons.

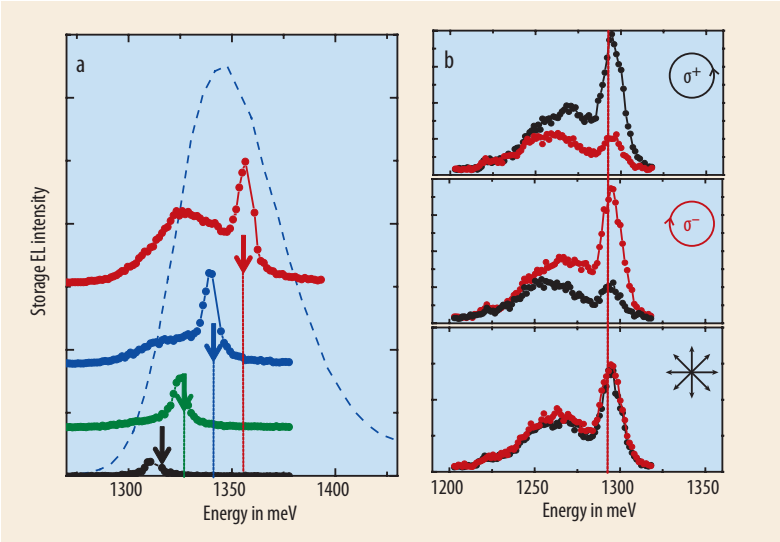

Fig. 5 (a) Examples of wavelength selective optical charge storage in a small ensemble of self-assembled GaInAs quantum dots for a storage time of $\Delta t = 12$ µs at $T = 10$ K and zero magnetic field. The spectra clearly reveal a pronounced peak close to the energy of the laser excitation (arrows) that arises from selectively generated charge within the ensemble. (b) The optical polarisation of the storage signal "remembers" the polarisation of the photons used to generate the charges in the quantum dots (upper pair of panels). These memory effects vanish following excitation with unpolarised light (lower panel).

Electron spin flip relaxation in quantum dots

Spin flip relaxation in bulk III-V semiconductor materials normally occurs over very short timescales (<10 ps), primarily due to scattering processes that can couple to the spin of the electron via the so-called spin orbit interaction (SOI). Coupling of the spin to the orbital motion is well known in atoms to have relativistic origins, arising from the interaction of the electron spin with an effective magnetic field experienced by the electron as it moves in the electric field $\vec{E}$ of the nucleus. Quite generally, an electron moving with momentum $\vec{p}$ in a vacuum experiences an effective magnetic field $\vec{B}_{eff} = (\vec{E} \times \vec{p})/2m_0c^2$, the origin of which can easily be appreciated by visualising the electron in its rest frame whereupon the positively charge nucleus executes orbital motion around it, generating a magnetic field at the position of the electron. In semiconductors, SOI will result in the electron spin precessing $\vec{B}_{eff}$ in as it propagates with a momentum $\vec{p} = m\vec{v}$ through the material. The direction of $\vec{p}$ and $\vec{B}_{eff}$ will be constant during uninterrupted ballistic motion but change upon scattering from phonons, impurities or other charge carriers. As a result, the randomisation of the electrons momentum by scattering will be accompanied by a scrambling of its spin orientation. Rather surprisingly the angle through which its spin is rotated whilst it executes ballistic motion turns out to be independent of the velocity $\vec{v} = \vec{p}/m$. This rather surprising result is a consequence of the fact that the precession frequency of the spin

depends on $|\vec{B}|_{\text{eff}}$ that itself is determined by $\vec{v} = \vec{p}/m$. Thus, if an electron moves with a larger velocity it also precesses faster and the net rotation over a given path length is the same. As a result, the strength of the SOI in any material can generally be characterised by the distance over which an electron must travel before its spin rotates by an angle π. This length is termed the spin-orbit length and is typically of the order of $l_{so} \sim$ 1–10 µm in Ga(In)As semiconductor heterostructures, defining the distance over which an electron should propagate before the spin can flip its orientation. As shown by **Fig. 2**, the typical dimensions of GaInAs QDs are far smaller than l_{so} and, thus, one would expect that SOI becomes ineffective. This expectation is supported by measurements of long spin lifetimes for excitons in quantum dots, which reveal no detectable decay of the exciton spin over their radiative lifetime (~1 ns), and also theoretical calculations that suggest that the spin flip rate in QDs should be suppressed by several orders of magnitude when compared with higher dimensional nanostructures (see e. g. [11]).

Spin memory devices such as those discussed here provide an ideal direct method to directly measure the electron spin flip relaxation time (T_1^e): electrons are optically initialised in the higher energy Zeeman level by exciting the system with σ^+ polarised light. The spin relaxation time is then directly measured by monitoring the intensity of the emission recorded with σ^+ helicity as a function of the storage time ($I_{+,+}(\Delta t)$). In the absence of spin relaxation over the time Δt one should observe a strong polarisation memory in the storage EL signal. In contrast, complete spin relaxation would result in steady state spin populations of the two spin states according to Boltzmann statistics, independent of the helicity of the optical excitation. For the low temperatures and high magnetic fields studied here, this would correspond to all electron spins occupying the lowest Zeeman level, or complete σ^- polarisation of the storage EL.

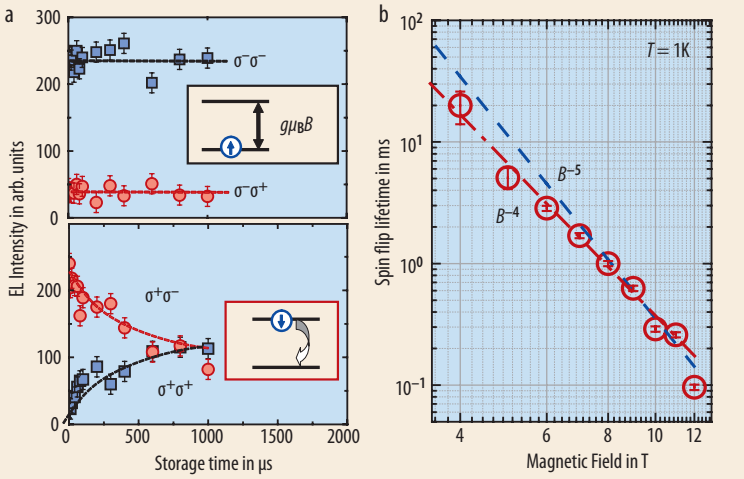

Fig. 6 (a) Intensity of storage luminescence recorded with circular polarisation discrimination in both the excitation and detection channels. The upper panel shows the result of optically pumping spins into the lowest Zeeman level, where no time evolution is observed. In contrast, pumping spins into the upper Zeeman level reveals a clear time evolution (lower panel), from which the spin flip lifetime is measured. (b) The electron spin lifetimes plotted as a function of magnetic field exhibits extremely slow spin relaxation in self-assembled QD nanostructure. Data reproduced from [12].

Examples of such time resolved spin storage measurements recorded at $T = 1$ K and $B = 8$ T are presented in **Fig. 6a**. The data show the temporal evolution of the storage luminescence intensity following $\sigma^{-/+}$ excitation over the time range $0.001\,\text{ms} < \Delta t < 1\,\text{ms}$. Following excitation with σ^- polarised light, to pump electrons into the lower energy Zeeman level, the storage EL is found to be predominantly σ^- polarised as expected. Furthermore, it exhibits no detectable evolution up to $\Delta t \sim$ 1 ms (**Fig. 6a**, upper panel) since the system is initialised close to thermal equilibrium. In contrast, following excitation using σ^+ polarised light, to generate electrons into the upper Zeeman level, a very marked time dynamics of the luminescence polarisation is observed.

OPTICAL ORIENTATION IN QUANTUM DOTS

The spin of the electron-hole pair in a QD can be optically aligned using circularly polarised light. As the photon is absorbed by the QDs its angular momentum ($\pm\hbar$ for photons of $\sigma^\pm$ helicity) is transferred to the total spin of the exciton. For an electron storage device, optical excitation with circularly polarised light having σ^- or σ^+ helicity results in the storage of spin up (e↑) or spin down (e↓) electrons, respectively, after the charge storage cycle is completed. These effects arise from the optical selection rules for III-V semiconductors: the conduction band quantum states are derived from s-like atomic orbitals and behave like spin ½ particles ($S_{z,e} = \pm\hbar/2$). In contrast, the valence band levels are derived from p-like atomic states and possess both spin and orbital angular momentum. For the heavy hole states, most important for self-assembled quantum dots, the valence band states then carry a total angular momentum $S_{z,h} = \pm 3\hbar/2$. These states combine to produce four energetically distinct exciton states with $J_{1e+1h} = S_{ze} + S_{zh} = \pm 1$ (e↓ h↑ and e↑ h↓) and ±2 (e↑ h↑ and e↓ h↓), respectively, as depicted in the figure. Since a circularly polarised photon conveys a single unit of angular momentum only the $J_{1e+1h} = \pm 1$ transitions are optically active, the $J_{1e+1h} = \pm 2$ states remaining dark. When the charge storage EL is read out after a storage time Δt the degree of circular polarisation of the emitted EL ($P = (I_{\sigma+} - I_{\sigma-})/(I_{\sigma+} + I_{\sigma-})$) provides a direct optical probe of the electron spin orientation.

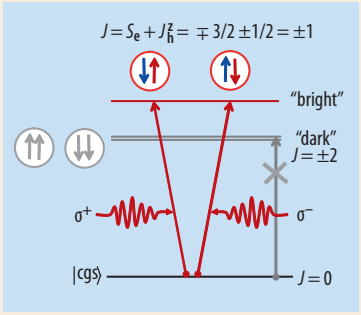

Fig. i The energy level diagram shows the optical selection rules for single exciton states in self-assembled quantum dots.

For $\Delta t = 0.001$ ms the measurement reveals $P(\Delta t) \sim +80$ %, decaying over time to a few percent at $\Delta t \sim 1$ ms as electrons flip their spin as depicted schematically on the figure. A very long spin lifetime of $T_1^e = 1.1 \pm 0.2$ ms is obtained, more than four orders of magnitude longer than spin flip times in quantum wells and seven orders of magnitude longer than the corresponding time in bulk III-V semiconductors!

The magnetic field dependence of T_1^e at $T = 1$ K is summarised in **Fig. 6b**. The decay time constants extracted using the above method are found to be very strongly dependent on the magnetic field, reducing dramatically from $T_1^e = 20 \pm 6$ ms at $B = 4$ Tesla, to only 0.1 ± 0.01 ms at 12 Tesla. Thus, spin relaxation in QDs is characterised by the rather unusual property that it becomes more efficient as the Zeeman levels move energetically further apart. The data presented in **Fig. 6b** suggest a clear power-law dependence ($T_1^e \propto B^m$), the least squares fit to the data yielding $m = -4.5 \pm 0.2$. As discussed in [11], this shows that spin relaxation in QDs is mediated by SOI combined with emission of a single acoustic phonon with an energy matching the Zeeman energy (ΔE_z). The absence of any observable saturation of T_1^e in **Fig. 6b** suggests that the relaxation time can even be much longer at lower magnetic fields. For example, extrapolating the observed ($T_1^e \propto B^{4.5}$) dependency would indicate $T_1^e = 80$ ms at ~ 3 Tesla, reaching ~ 1 s at ~ 1.8 Tesla. Very recent measurements on GaAs QD have shown T_1^e can even exceed one second in GaAs QDs with precisely the same characteristic magnetic field dependence described here.

We have seen how by trapping electrons in QDs their spin degree of freedom is largely protected from its solid state environment. The use of QDs to stop spins spinning opens the way for a wide range of spintronic applications based on single spins. For example, spin qubits are now checking off the various criteria required for viable quantum hardware: single electron spins can be initialised, manipulated and even readout using electro-optical methods. When combined with the future possibility to incorporate QDs into electrically and optically active devices with ever increasing complexity, the prospects for achieving even finer levels of control are very bright, indeed.

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