

USING LIGHT TO PREPARE AND PROBE AN ELECTRON SPIN IN A QUANTUM DOT

A.S. Bracker, D. Gammon, E.A. Stinaff, M.E. Ware, J.G. Tischler, D. Park, A. Shabaev, and A.L. Efros
Electronics Science and Technology Division

INTRODUCTION

Electrical charge is the fundamental physical quantity that makes modern electronic devices possible. However, recent advances in nanofabrication and materials physics have created the opportunity to use electron spin for next-generation devices. Spin is a quantum mechanical property with great potential to bring about entirely new technologies for computing, communications, and encryption. All of these applications require the ability to prepare electrons in specific spin states and to measure those states following a sequence of operations. We have recently demonstrated optical pumping and measurement of individual electron spins in quantum dots (QDs).¹ Circularly polarized laser light excites a specific spin state of the dot. We achieve spin-selective detection through the energy and magnetic field response of subsequent luminescence, which reveals clear signatures of our prior spin state preparation. These results lay the groundwork for using individual semiconductor electron spins as addressable logic elements for quantum information processing and quantum communication.

COHERENT SEMICONDUCTOR SPINTRONICS

Transmitting and processing information are essential to the success of military and intelligence operations, and the security of that information is naturally of prime importance. A fundamentally new class of optoelectronic devices that exploit quantum mechanical phase could enable a revolution in communications, encryption, and data processing. In a solid-state device, electron spin is a natural choice for the carrier of quantum phase, because we can leverage established materials and device technologies that already use electrons as charge carriers. “Spintronics” is a broad interdisciplinary research field that seeks to exploit this spin degree of freedom. While the invention of the transistor started a revolution in solid-state electronics by providing an elegant means to manipu-

late electrical charge, spintronics is expected to open still broader frontiers for electronics and optoelectronics.

Spin is an inherently quantum mechanical property, yet we often visualize it as if it were a classical angular momentum with the electron rotating about an axis. We usually depict the spin as a vector pointing along its rotation axis (Fig. 1). An arbitrary classical angular momentum is a vector that can point in any direction, while the analogous state of a quantum mechanical spin is a coherent sum $a|\uparrow\rangle + b|\downarrow\rangle$ of two basis states, spin up $|\uparrow\rangle$ and spin down $|\downarrow\rangle$, with some relative phase described by the coefficients a and b . Spin is therefore a two-level system, and some forms of spintronics approach spin as an additional binary degree of freedom. However, arbitrary superposition states have character of both “up” and “down.” In coherent spintronics, we take advantage of these quantum superposition states, which provide a great advantage as a form of information. While electrical charge readily provides a binary form of logic (i.e., a charged or uncharged bit), each spin quantum bit or “qubit” provides a continuum of states, and the degree to which we can use this information is determined by how precisely we can write or read the coefficients a and b that describe the superposition. By entangling a collection of many such spins, the information processing power increases greatly.

Quantum communication may be best achieved with photons of light or other electromagnetic waves, because they readily propagate over long distances, and their polarization states are robust to environmental perturbations. Indeed, basic quantum communication hardware is already available commercially, with information carried over hundreds of meters by the polarization state of light in an optical fiber. Someday, these approaches will be extended to longer distances through the use of optical repeaters and perhaps adapted to free-space transmission over much longer distances, including satellite communication.

While pure optics may be an excellent approach for communication, quantum information processing and storage are better achieved with electrons or holes

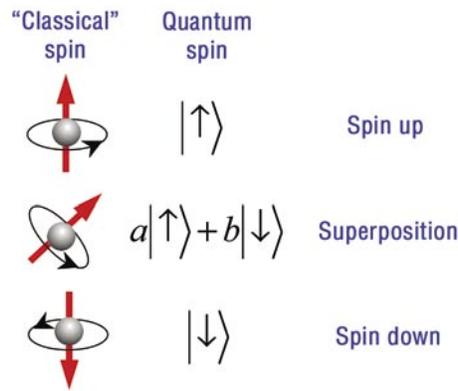


FIGURE 1
Schematic depiction of classical angular momentum with different projections and analogous quantum mechanical spin wavefunctions.

in a solid material, just as conventional electronics and logic are today. A complete communications and data processing system must therefore incorporate a hybrid technology that combines photonic and electronic spin qubits, and it must provide a way to convert between them. The research that we describe in this article addresses materials science and basic spin physics that can enable solid-state electronics based on spin coherence. Optical techniques are central to our work, as the ability to prepare and measure individual spins is critical for coupling spin-based electronics with communication.

SEMICONDUCTOR QUANTUM DOTS

Just as conventional logic circuits consist of ordered networks of transistors, one must likewise specify the spatial position, energies, and mutual interactions of electron spins in a multispin circuit. We use semiconductor QDs for this purpose. QDs are very small particles of crystalline material with discrete energy levels that correspond to confined orbital states of electrons in the QD. A typical QD in our research program consists of 10^4 to 10^5 atoms of a III-V compound semiconductor such as InAs, which is embedded within a host material of larger bandgap such as GaAs or AlGaAs. We can control their energy levels by modifying their composition, size, and shape. Advanced crystal growth and nanofabrication techniques offer ways to position QDs into ordered arrangements.

We fabricate QDs by molecular beam epitaxy (MBE), an ultrahigh vacuum technique for growing extremely pure semiconductor crystals from elemental precursors. Our QD fabrication was carried out in the NRL Epicenter, an MBE facility in Building 208 that is staffed by scientists and engineers from four NRL divisions. The facility contains four MBE machines for growth of III-V and II-VI compound semiconductors and magnetic materials, material processing

capabilities, scanning probe microscopy, and surface spectroscopy.

Our work on spin preparation and measurement involves two types of QDs. The first type consists of a thin planar GaAs quantum well sandwiched between AlGaAs barriers (Fig. 2(a)). Although the GaAs layer is nominally two-dimensional, with confinement in the growth direction, the interfaces between GaAs and AlGaAs have roughness on the scale of one or two atomic layers. This roughness is sufficient to confine electrons within the plane of the quantum well, thereby producing a QD. The second type of QD is formed through Stranski-Krastanov self-assembly (Fig. 2(b)). Here, a material such as InAs is grown on a flat substrate surface of GaAs, which has a smaller lattice constant. When the growth first starts, a very thin planar “wetting layer” forms, but only a few atomic layers of planar InAs can be grown before a critical threshold is reached. When the strain between the two materials becomes too great, most of the wetting layer coalesces into small solid droplets of InAs. These QDs are shaped like domes or pyramids, 10 to 50 nm wide and 3 to 6 nm high. For many of our samples, the newly formed dots are buried under a capping layer of GaAs or other materials. Figure 2(c) shows a typical plan-view atomic force microscope image of these dots (taken without the capping layer).

In order to controllably insert a single electron into each QD, we embed the QD layer in the insulating region of a Schottky diode heterostructure (Fig. 2(d)). This structure consists of an insulating GaAs layer grown by MBE on a heavily doped GaAs substrate and buffer layer. With a metal layer evaporated on the surface and wire contacts to the front and back of the sample, we apply an electric field across the insulating layer that contains the QDs. Figure 2(e) shows a heterostructure band-edge diagram. At low or forward bias (dark blue), the electric field is small, and the discrete energy levels of the QD lie below the Fermi sea of electrons, so that the dots each contain

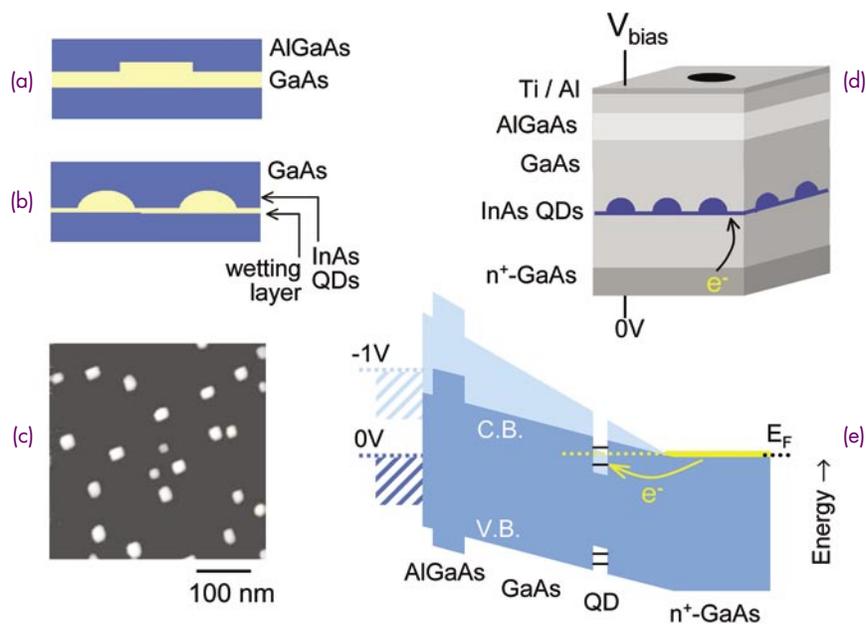


FIGURE 2 (a) Schematic side view of QDs formed by interface roughness in a GaAs/AlGaAs quantum well. (b) Schematic side view of Stranski-Krastanov self-assembled QDs and two-dimensional wetting layer, embedded in GaAs. (c) Plan-view atomic force microscope image of self-assembled QDs. (d) Schematic structure of InAs QD Schottky diode for tuning the charge state of QDs. (e) Conduction and valence band-edge profile of InAs QD Schottky diode for two typical biases.

one or more electrons. In reverse bias (light blue), the electric field increases, and electrons are driven out of the QDs. As we will show below, the small size of each QD allows electrons to be injected one at a time.

OPTICAL SPECTROSCOPY OF CHARGED QUANTUM DOTS

Photoluminescence (PL) spectroscopy is a powerful method for examining the energy levels and spin properties of electrons in QDs. In this technique, we excite a QD sample with a laser, which creates electron-hole pairs (e^-h^+) in the material (Fig. 3(a)). An electron-hole pair (exciton) confined in a QD will

eventually relax to the lowest unfilled energy level of the dot, where it can recombine and emit a photon. The energies of these luminescence photons, which we disperse in a grating spectrometer, provide information on the energy level structure of the QD. Figure 3(b) shows a typical PL spectrum obtained when the laser is focused to a $50\text{-}\mu\text{m}$ spot on the sample, illuminating thousands of QDs. Because these QDs all have slightly different energies, the spectrum is inhomogeneously broadened. Nevertheless, we can resolve a discrete energy level structure with a progression of S-, P-, D-, and F-like orbital states, confirming that we have three-dimensional atom-like quantum confinement in these QDs.

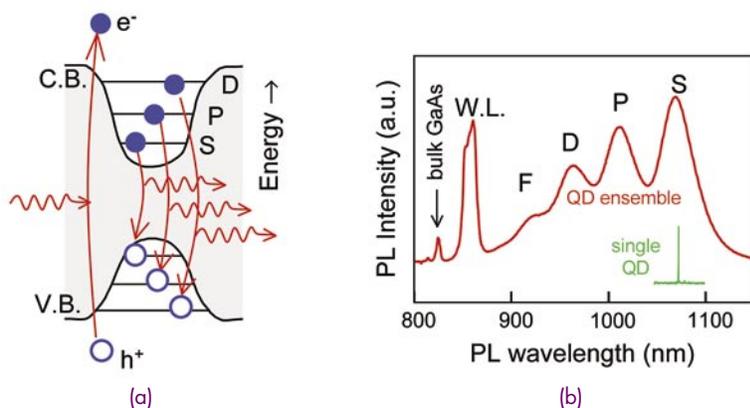


FIGURE 3 (a) Band-edge profile, level structure, and optical transitions (absorption and PL) in a neutral QD. Electrons are solid circles, and holes are open circles. (b) PL spectrum of a QD ensemble showing inhomogeneously broadened atomic-like level structure (red curve) and a single QD spectrum (green).

The inhomogeneous broadening in this spectrum obscures many details that would be clear if the peaks were much sharper. We get around this problem by using the powerful techniques of single-QD spectroscopy, which were developed at NRL and other institutions in the mid 1990s. With electron beam lithography and liftoff techniques, we fabricate a metal mask containing an array of tiny apertures, some as small as 200 nm, on the sample surface (Fig. 4(a)). Each aperture contains a small number of QDs and sometimes only one, so that if we point the laser beam at the aperture, we measure PL from only those QDs (Fig. 4(b)). The green curve in Fig. 3(b) illustrates the power of this technique, where a single, very sharp spectral peak from one QD inside an aperture is shown below the much broader ensemble spectrum.

By combining single-dot spectroscopy with bias-controlled charging in a diode heterostructure, we see a much richer picture of the energy level structure and charge states of the QD. Figure 5(a) is an intensity contour plot that shows how the PL spectrum of an

InAs QD evolves as we vary the bias across the diode. A vertical slice through this graph would correspond to single PL spectrum, and each colored band in the two-dimensional plot represents the evolution of a particular spectral peak with bias. Each PL peak corresponds to an exciton (electron-hole pair) recombining in the presence of one or more additional electrons or holes. Figure 5(b) depicts the possibilities. Going from left to right in Fig. 5(b), or from negative to zero bias in Fig. 5(a), we see excitons with increasing negative charge: first an extra hole (X^+), then a neutral exciton (X^0), then an extra electron (X^-), and finally two extra electrons (X^{2-}). When the diode is unbiased (0V), the QD and the wetting layer are filled with electrons, and the single QD PL spectrum at the right of the figure is smeared out. Each charge complex has a distinct PL energy, because the addition of new charge changes the net Coulomb energy (attraction or repulsion). These Coulomb shifts and the wide energy spacings of the orbital levels (Fig. 3(b)) provide energy discrimination that allows us to control the charge state in the QD.

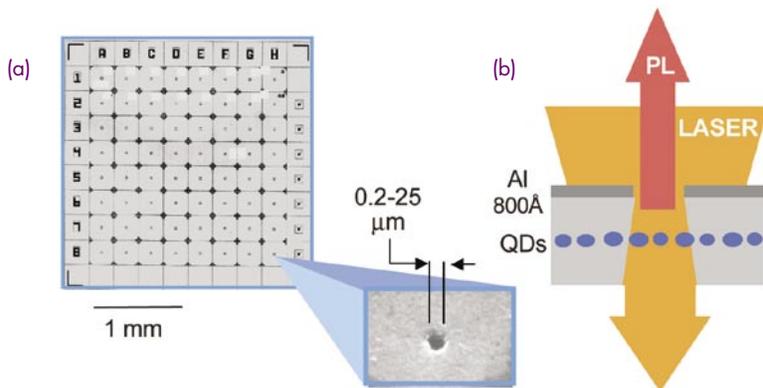
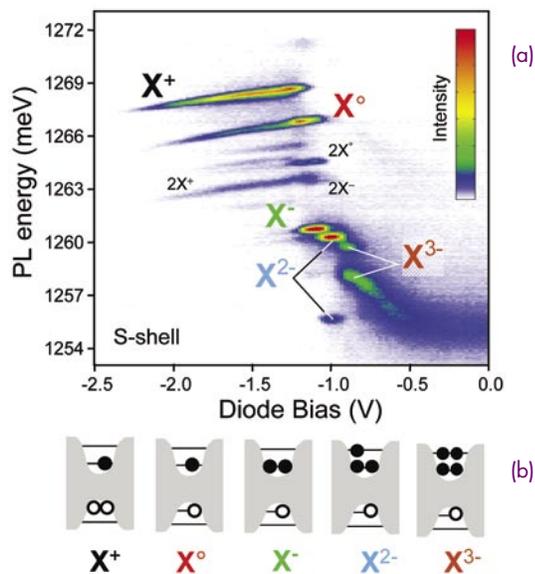


FIGURE 4
 (a) Image of an aluminum aperture mask on a GaAs QD sample used for single QD spectroscopy. (b) Cross-sectional schematic of laser excitation and PL of a small number of QDs through a submicron aperture.

FIGURE 5
 (a) PL intensity map as a function of PL energy and diode bias. PL features from various charge states of excitons (X symbols) and biexcitons ($2X$ symbols) are labeled accordingly. (b) Electron and hole configurations for the various excitons as labeled in (a).



Finally, we note that the pairs of peaks for X^{2-} and X^{3-} can be attributed to exchange interactions, while biexciton peaks (e.g., $2X^-$ and $2X^+$) appear due to the fairly high laser intensities used in this work.

SPIN SPECTROSCOPY AND OPTICAL PUMPING

We consider the spin properties of a QD containing a single electron in a confined conduction band level. This simple system serves as a model for a semiconductor spin qubit. The two degenerate spin states of this electron (up and down) are depicted with arrows at the bottom of the level diagram of Fig. 6. The optically excited state of this electron is the three-particle negative “trion” (X^-), which consists of the original electron, plus an additional exciton. The electrons in the trion are paired with opposite spins in a singlet state; therefore, they do not contribute to the overall spin of the trion. Instead, the spin of the trion is determined by whether the unpaired hole spin points up or down. The trion is an important focus of our work, because it can be used as an optically addressable intermediate state for initializing, rotating, and reading out the ground-state electron spin.

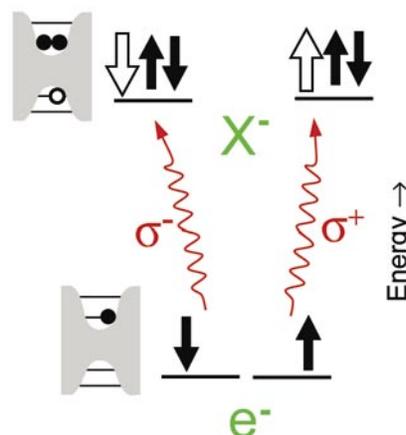
As shown in Fig. 7(a), our starting point for spin spectroscopy is a technique called optical orientation. In optical orientation, a photon of circularly polarized light is absorbed by a QD, where it is converted into a polarized electron-hole pair, with the hole spin “up” and the electron spin “down.” Through subsequent relaxation, this polarization may transfer to other spin degrees of freedom such as the electron that was initially in the QD. This process is known as “optical pumping,” in analogy with a similar process from the field of atomic physics. Optically pumped electron spin can persist long after recombination of excitons is over, providing the type of spin memory that is needed

for storage and processing of information. Optical pumping may provide a way to initialize spins prior to quantum logic operations. After exciting a QD sample with right-circularly polarized light to initiate optical orientation, we measure the degree of circular polarization “memory” in the PL. This polarization memory reveals the electron and hole spin dynamics that are caused by optical excitation and relaxation, including the possibility of optical pumping.

Figures 7(b) and 7(c) show the PL and PL polarization memory as a function of diode bias for a GaAs QD. We see distinctive behavior for all three types of excitons, but here we focus on the remarkable properties of X^- . In particular, its polarization memory has a negative value between 4 V and 4.3 V bias. A negative polarization memory implies that the PL emitted from the QD has the opposite helicity from the laser beam that excited the sample in the first place. This effect can be seen in an even more striking way if we measure the raw PL polarization directly as a function of the laser polarization (Fig. 7(d)). We can vary the laser polarization continuously from left circular to right circular and back, including intermediate linear and elliptical polarizations. When we do this, we see that the X^- raw PL polarization changes in the opposite sense from the laser polarization, while for X^0 and X^+ it changes in the same sense. We show next that this behavior is a consequence of optical pumping of the original QD electron spin.

Optical pumping of electron spins should occur more readily at higher laser intensities, where it can dominate competing processes. Under modest laser intensity, the polarization memory of X^- is negative only near the bias value of 4 V, but it becomes positive at higher bias, due to competing processes related to electron injection (Fig. 7(c) and Fig. 8(a) (blue)). However, if we increase the laser intensity by a factor of 15, the PL polarization becomes negative for all

FIGURE 6
Total energy spin configurations of a ground state electron (e^-) and its optically excited state (negative trion). Open and filled arrows represent hole and electron spins, respectively. Optical transitions are achieved with right (σ^+) and left (σ^-) circularly polarized light.



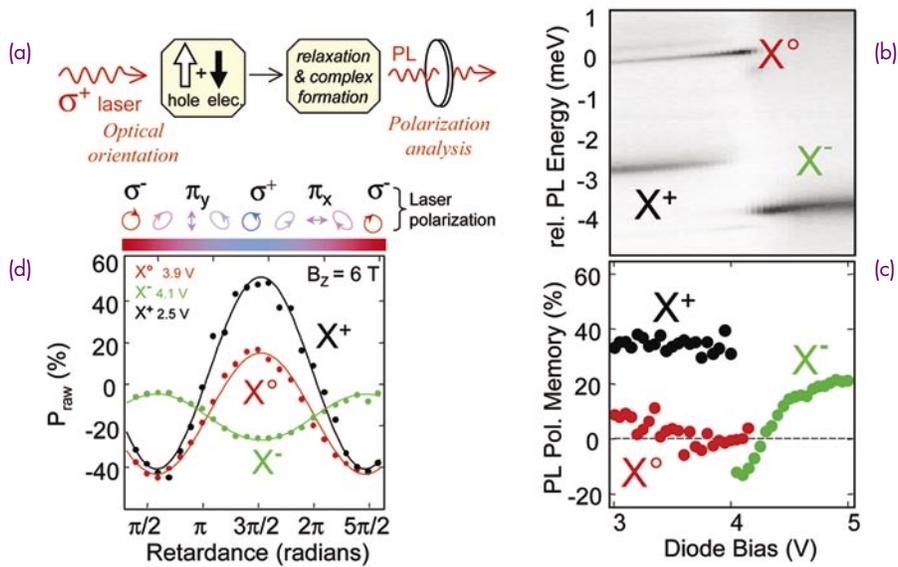


FIGURE 7
 (a) Schematic of an optical orientation experiment. (b) PL intensity map for a GaAs QD as a function of PL energy and diode bias. (c) Circular polarization memory for the same features as in (b). (d) Raw PL polarization as a function of laser polarization.

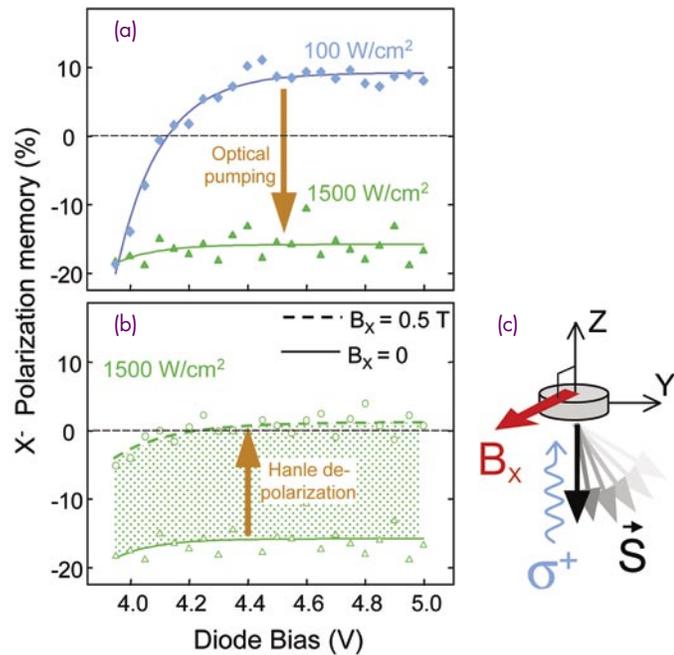


FIGURE 8
 (a) Polarization memory of negative trion (X^-) for two laser excitation intensities. (b) Elimination of negative polarization memory for X^- through Hanle depolarization of optically pumped electrons. (c) Geometry of Hanle effect: optically pumped electrons are initially oriented parallel to Z axis, but are depolarized by precession about transverse magnetic field B_x .

values of the bias (Fig. 8(a) (green)). This behavior is suggestive of optical pumping becoming more efficient than the high-bias competing process. The second piece of evidence comes from the Hanle effect. In the Hanle effect, the sample is placed in a small transverse magnetic field, where electron spins will precess around the field axis (Fig. 8(c)). If the magnetic field is high enough, this precession will erase the spin polarization that is caused by optical pumping. Indeed, application of a small magnetic field causes the negative polarization to disappear (Fig. 8(b), upper curve), proving that the negative polarization is caused by optical pumping.

SUMMARY

We have used polarized laser light to optically pump an electron spin in an individual semiconductor quantum dot. We detect this prepared spin state through an unusual negative spin memory that is

imprinted on the luminescence polarization of a charged exciton. These processes represent very basic forms of electron spin read and write operations. The critical aspect of this research is the ability to fabricate charge-tunable quantum dots that can be injected with a single electron and measured individually using microphotoluminescence spectroscopy. Together with new nanofabrication methods and coherent optical spin control, these advances should enable a new generation of spin-based electronics, providing capabilities in computation and secure communication that are not achievable with existing technologies.

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Reference

¹A.S. Bracker, E.A. Stinaff, D. Gammon, M.E. Ware, J.G. Tischler, A. Shabaev, A.L. Efros, D. Park, D. Gershoni, V.L. Korenev, and I.A. Merkulov, "Optical Pumping of the Electronic and Nuclear Spin in Single Charge-tunable Quantum Dots," *Phys. Rev. Lett.* **94**, 047402 (2005). ★