Quantum dots (QDs) are often referred to as artificial atoms due to their anharmonic discrete energy level structure. Many key experiments with QDs such as single photon generation, spin pumping and coherent manipulation demonstrated their potential in quantum information processing. However, as compared to atoms and ions, QDs show a strong spectral dispersion which is at present a serious limitation for their usefulness, for example as sources of identical photons. While strong efforts are undertaken to engineer QDs with identical spectral properties, applications in linear optical quantum computing or quantum repeaters appear out of reach with existing QD technology. One key element of a quantum repeater protocol is the creation of entanglement between two distant quantum emitters by overlapping spectrally identical single photons from two different quantum emitters on a beam splitter. For ions, such an entanglement scheme has been demonstrated recently in a ground-breaking experiment by the Monroe group. The realization of a similar entanglement scheme based on solid-state emitters remains an elusive goal up to date.

In order to probabilistically entangle two QDs that are energetically close an independent knob to ensure spectral overlap of the emitted photons is needed. In this letter we demonstrate the first direct observation of all-optically tunable spontaneous Raman fluorescence from a single self-assembled QD. Using a cross-polarizer setup and a scanning Fabry-Perot interferometer to suppress the excitation laser light, we are able to detect the Raman-scattered photons on a CCD chip. First, we demonstrate the frequency tunability of Raman photons with magnetic field. While magnetic fields could in principle be used to tune the two emitters in resonance with each other, all-optical manipulation is more versatile, faster and allows spatial addressing of several QDs within one sample. For this reason, in a second experiment the magnetic field is fixed and we vary the excitation-laser frequency to optically tune the frequency of the emitted photons over a range of roughly 2.5 GHz. From the raw data, we extract the number of scattered photons, their center frequency and linewidth. As expected, the number of photons follows a Lorentzian as a function of detuning. Moreover, the data for the photon linewidths indicate a decrease down to the resolution limit of the Fabry-Perot as we tune off-resonance.

Optical frequency tuning in a QD has been demonstrated using the AC Stark effect in coherent QD spectroscopy and more recently using resonantly scattered photons. However, the latter experiments usually show at least a lifetime limited spectral width, off-resonant Raman photons can in principle be arbitrarily narrow and are only limited by the laser linewidth and the low-energy spin coherence. For singly charged QDs as studied here, the main decoherence mechanism of the metastable ground state is the interaction of the electron spin with the surrounding nuclear spins. Hence, in principle the spectral distribution of the Raman scattered photons can give valuable information about this interaction and the work in this paper could form the basis of a new technique to study spin-bath interactions in single QDs. We also note that resonantly scattered Raman photons were previously used for spin read-out in a singly charged QD.

The experiment is carried out with single self-assembled InGaAs QDs that are charged with a single excess conduction-band electron and that are subject to a magnetic field in Voigt geometry. In Fig. 1 (a), the corresponding level scheme is depicted. At zero magnetic field, ground and excited (trion) states are degenerate. A finite magnetic field induces Zeeman splittings according to the in-plane electron and hole g-factors, and . For large enough Zeeman splittings, each of the trion states forms an independent λ-system together with the two ground states. Optical selection rules determine the polarization of both the vertical and diagonal transi-
tions to be linear but orthogonal to each other (denoted H and V in Fig. 1). Experimentally this allows for efficient polarization separation of the excitation laser from the photons of interest. The recombination from the excited state takes place on a nanosecond timescale with equal probability for decay into each ground state. Note, that this is in stark contrast to the widely used Faraday geometry, where this branching ratio is typically on the order of $10^{-5}$.

When the QD is resonantly driven on one of the optical transitions, the electron spin is very efficiently pumped into the other ground state and further photon absorption or emission is stopped [3, 4]. Hence, for efficient photon production by Raman scattering the spin state of the electron needs to be restored on a short timescale. One possibility is to work at the edges of the voltage range for which the QD is singly charged. In this co-tunneling regime, the electron interacts with the electrons of the Fermi sea in the back contact which leads to spontaneous spin-flip events and effectively suppresses spin pumping [20]. Another possibility is the use of a second laser to optically re-pump the electron spin [3]. Both methods are applied in this work.

In the experiment, two different QD samples with 25nm and 35nm tunneling barriers between the QD layer and the n-doped back contact are investigated. A transparent Schottky gate on the sample surface allows for deterministic charging of the QD. The QD density is low (typically less than 0.1 per $\mu m^2$) such that individual QDs can be studied using a confocal microscopy setup with an almost diffraction-limited spot size of 1 $\mu m^2$. The sample is immersed in a liquid-helium bath cryostat at a temperature of 4.2 K, with magnetic fields up to 10 T. A stack of xyz nano-positioners allows for precise positioning of the sample. QDs are spatially and spectrally located by photoluminescence (PL) spectroscopy. The emission wavelength of the QDs studied is typically centered around 960 nm. A differential transmission (DT) technique [21, 22] is used to precisely determine the transition energies of the individual QDs. The transmitted signal is split into its H and V polarization components in order to separate vertical and diagonal transitions from each other. The Raman-scattered photons are collected through the confocal microscope and are sent through a fiber onto a CCD camera for counting. The excitation laser light is suppressed by a polarizer before the fiber. In addition, the collected light passes through a Fabry-Perot interferometer before hitting the CCD chip. This further suppresses the undesired background and serves as a spectral filter for the collected photons. The interferometer with parallel mirrors has a free spectral range of 15 GHz and depending on the exact alignment a finesse of typically 40 to 60 is achieved. The Fabry-Perot is stabilized using the transmission of an independent titanium sapphire laser at 905 nm. Both lasers are frequency-locked to a wavemeter (High Finesse, WSU-30) which has an accuracy of better than 30 MHz.

The ground and excited state splittings are given by $g_e \mu_B B$ and $|g_h| \mu_B B$ respectively. We find experimentally, that the electron g-factor $g_e$ is roughly the same for all the QDs investigated here ($g_e \approx 0.45$), while the in-plane hole g-factor $g_h$ shows a large distribution, in accordance with previously reported results in literature [19, 23]. In particular, 2 out of 15 QDs had a hole g-factor with approximately the same magnitude as the electron g-factor but opposite sign, i.e. $g_h \approx -g_e$, which ensures large enough splittings. Moreover, for this particular case the two diagonal transitions are degenerate and consequently they exhibit no spin pumping, since the excitation laser acts both as pump and re-pump at the same time. Figure 1(c) demonstrates this effect: The three polarization resolved DT traces were recorded at the two edges and in the middle of the charging plateau for a magnetic field B=0.6 T. Whereas the outer transitions are efficiently spin-pumped away from the plateau edges, the degenerate inner transitions (central dip) exhibit no spin-pumping. We confirmed that spin pumping is suppressed up to B=4 T.

The efficient optical restoration of the electron spin makes this QD a good candidate for observing resonantly scattered photons. Figure 2 displays the number of detected photons after the Fabry-Perot filter as a function of detuning from the driving laser frequency which was set on the diagonal transition. The experiment was performed again at a B=0.6 T. Even though the laser light is suppressed by a polarizer and the Fabry-Perot filter,
we obtained a splitting of 7.9 GHz which is consistent with the DT data from Figure 1(c) for which the splitting corresponding to photons emitted on the two vertical transitions. Within our experimental resolution, the splitting at -3.7 GHz and 3.9 GHz correspond to the two outer transitions. The solid line is a guide to the eye.

Besides this central line, two distinct peaks are visible at -3.7 ± 0.2 GHz and +3.9 ± 0.2 GHz detuning, corresponding to photons emitted on the two vertical transitions. Within our experimental resolution, the splitting is consistent with the DT data from Figure 1(c) for which we obtained a splitting of 7.9 ± 0.1 GHz. An obvious feature of the data in Figure 2 is the different height of the two fluorescence peaks. Comparing with the DT data of Figure 1(c), we note that the amplitude ratio for the two transitions is approximately reversed. The origin of this asymmetry is not clear. The data presented in Figure 2 were taken for an excitation laser power at around saturation for which the signal to noise ratio is optimal.

Next, we turn our attention to off-resonant Raman scattering. At typical powers used in the experiment, i.e. around saturation, resonantly scattered photons should have a linewidth determined by dephasing of both excited and ground state and potential inhomogeneous broadening due to e.g. charge fluctuations in the environment of the QD [24]. In contrast, for very low powers, we would expect a narrowing of the fluorescence down to a linewidth determined by the dephasing rates of the two ground states $\gamma_{11}$, which consists of the co-tunneling rate to the back-contact, $\kappa_{11}$, plus additional fluctuations due to magnetic impurities in the environment. For large detunings, i.e. $\Delta \gg \Omega, \Gamma, \gamma_{11}$, the linewidth is solely determined by $\gamma_{11}$. Hence, a photon source based on (far) off-resonant Raman scattering could in principle produce photons that are limited by spin-dephasing.

To demonstrate off-resonance Raman scattering we study a QD in the 25 nm sample that has splittings of outer and inner transitions of 10 GHz and 4.8 GHz, respectively, at $B \approx 1.2$ T. The excitation laser frequency is tuned close to the energetically higher outer transition. Measurements are performed in the co-tunneling regime to ensure efficient restoration of the spin-state by co-tunneling. Figure 4 (a) displays the number of scattered Raman photons for different detunings $\Delta$ of the excitation laser from resonance. When the laser is detuned from resonance (in steps of 0.5 GHz), the center frequency of the scattered photons shifts accordingly and at the same time the number of scattered photons decreases. Without changing the power of the excitation laser a tuning range of about 2.5 GHz around resonance is covered. In order to analyze the data quantitatively, we fit a Lorentzian to each curve and extract center frequency, amplitude and linewidth [25]. Results are displayed in Fig. 4(b) and (c). We expect the number of Raman photons to follow a Lorentzian in $\Delta$, i.e. to go as $1/\Delta^2$ for large detuning. The Lorentzian fit in Fig. 4(b) has a width of 1.5 GHz, coinciding with the linewidth measured in differential transmission.

Figure 4(c) demonstrates a decrease of the Raman frequency, amplitude and linewidth [25]. Results are displayed in Fig. 4(b) and (c). We expect the number of Raman photons to follow a Lorentzian in $\Delta$, i.e. to go as $1/\Delta^2$ for large detuning. The Lorentzian fit in Fig. 4(b) has a width of 1.5 GHz, coinciding with the linewidth measured in differential transmission.

Figure 3: Frequency tuning with magnetic field. The center frequency of the Raman photons shifts with magnetic field. Again, the solid lines are guides to the eye.
A testing follow-up experiment could be the demonstration that would need to compensate for this energy difference by tuning g-factors. In order to resonantly tune two QDs one needs to compensate for the electron ground state splitting will in interaction with the surrounding nuclei [17].

The Over-ness limit of the Fabry-Perot puts an upper bound to the dephasing rate resulting in a new tool for studying the interaction of a single electron spin with the surrounding solid state.

In conclusion, we have demonstrated all-optical tuning of Raman photons in a single QD. The present experiment opens up the possibility to optically gate voltage by repeating the experiment in different regions of the plateau. Moreover, moving away from the plateau edges to the middle of the plateau, we expect the cotunneling rate to become vanishingly small compared to other decoherence mechanism, such as the interaction with the nuclear spins.

In conclusion, we have demonstrated all-optical tuning of Raman scattered photons in a single QD. The present experiment opens up the possibility to optically tune the frequency of photons emitted from two different QDs into resonance with each other. This could be done for two QDs that are subject to the same external magnetic field. In addition to the Zeeman shift, the electron experiences a random Overhauser shift due to hyperfine interaction with the surrounding nuclei [17]. The Overhauser shift varies strongly from one QD to another and consequently the electron ground state splitting will in general differ between two QDs even for identical electron g-factors. In order to resonantly tune two QDs one would need to compensate for this energy difference by for example using two different driving lasers. An interesting follow-up experiment could be the demonstration of two-photon quantum interference as was done in ions and atoms [26,27]. This would constitute a first step towards a probabilistic entanglement scheme. Beyond its potential for applications in quantum information processing, we envision the detection of Raman photons as a new tool for studying the interaction of a single electron spin with the surrounding solid state.

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FIG. 4: (a) Optical tuning of Raman photons with the excitation laser detuning Δ. (b) Overall number and (c) linewidth (FWHM) of Raman photons (both as a function of Δ). The dashed line represents the resolution limit of the Fabry-Perot. The solid line is a guide to the eye based on a Lorentzian fit with the resolution limit of the Fabry-Perot as the offset.

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