

Modulation spectroscopy on a single self assembled quantum dot

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We present high resolution modulation spectroscopy on single quantum dots and discuss briefly the differences to other spectroscopy techniques. We use this technique to study the excitonic fine structure while charging the quantum dot and applying mechanical strain to it. We also show that the fine structure can be used as a polarization analyzer.

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1 Introduction

Self assembled semiconductor quantum dots (QDs) are traps for charge carriers in semiconductors. QDs form under certain growth conditions when a semiconductor is grown on a host semiconductor which has a smaller lattice constant. Because of this lattice mismatch three dimensional monocrystalline growth is not possible. If the deposited layer is thin enough strained islands without lattice defects will be formed. As the QD material has a smaller band gap these islands are three dimensional carrier traps if they are capped with further layers of the host material [1]. Because of its nanometer dimensions the trapped carriers can only occupy discreet energy levels like electrons trapped in the electrostatic potential of an atomic nucleus. Many effects known from atomic optics like the Zeemann effect, diamagnetic [2] and Stark shifts [3], many particle exchange interactions [4, 5], emission of single anti bunched photons [6] and the emission of entangled photons [7, 8] have been observed on QDs. But in contrast to atoms and ions QDs have the advantages that they are already embedded in a host material and do not need ultra high vacuum traps and they have a much stronger interaction with light fields as their oscillator strength is of the order of ten because of their larger size.

QDs are all unique in their physical properties as the trap potential is slightly different from dot to dot. This leads to an inhomogeneous broadening of the transition energies of an ensemble of QDs. Atoms of one type are all identical which leads to sharp transitions in energy for measurements on an ensemble like a gas. The absorption lines of gas are just broadened by external effects like Doppler broadening or scattering but not by the atoms them selves. Weak transitions on atoms can be studied by measuring on a sufficient large ensemble as the oscillator strengths add up. Because of the inhomogeneous QD distribu-

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tion it is important to study only one single QD as many effects can not be resolved in the inhomogeneous broadened ensemble spectrum.

2 Spectroscopy on quantum dots

The standard optical spectroscopy method for QDs is the Photoluminescence (PL) where a laser with energy larger than the host materials band gap is shined on the sample and lifts electrons from the valence into the conduction band (Fig. 1) [9, 10]. The so created free electrons and holes relax into states with lower energy and get partly trapped in QDs. There they form excitons which decay radiatively. These photons are collected and analyzed with a spectrometer. The subsequent relaxation process is incoherent. Therefore information about spin and phase is lost. A further draw back is that the non resonant excitation can lead to excitations in other QDs and impurities and by that to fluctuations of the potential landscape in the surroundings of the QD. On the other hand the PL is a versatile method to study ground and excited states in continuous wave and time resolved experiments.

In the Photoluminescence Excitation (PLE) experiment a tunable laser is tuned into resonance of a transition above the excitonic ground state in the dot and an excited exciton is created [11]. This will relax into its ground state where it will decay and can be analyzed like in the PL experiment. In addition to the exciton ground state energy excited states can be studied by PLE when the laser is swept while the exciton ground state intensity is monitored.

A way to study the resonant creation of ground state excitons is the Photocurrent (PC) experiment [12, 13]. Here a ground state exciton is created resonantly with a laser while a strong electric field is applied to the QD. This field leads to tunneling of the electron and hole out of the dot and by this to an ionization of the exciton. The resulting current is measured. The disadvantage of the PC is that due to the necessary tunneling an additional decay channel is opened which leads to a linewidth broadening. Further more the exciton must be destroyed to do a measurement and because of the applied electric field no charged excitons can be studied. Nevertheless Rabi oscillations of neutral excitons have been observed [13].

In the Transmission experiment none of these disadvantages exist. Laser light is brought into resonance with an excitonic transition and the change in transmission or reflection is measured. That has been demonstrated on neutral and charged ground state excitons as well as on excited ones [14, 15].

In the PL and PLE experiments the resonance is measured by a spectrometer and the spectral resolution is therefore limited by that. For InGaAs QDs the lifetime limited linewidth is of the order of 1 μeV while typical grating spectrometers have a resolution of a few tens of μeV . The PC and transmission experiments are laser spectroscopy experiments. Here the resonance is mapped by a narrow laser line

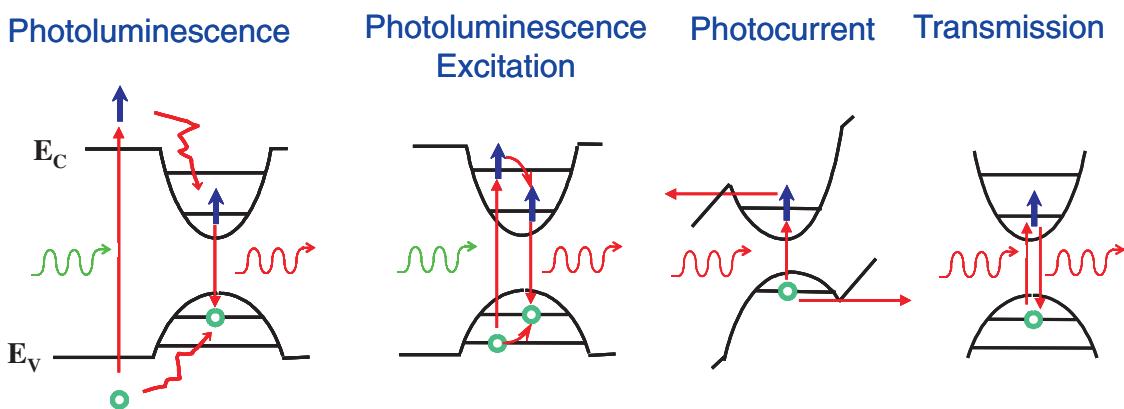


Fig. 1 (online colour at: www.pss-a.com) Schemes of four spectroscopy methods to probe a ground state exciton. The band edges of the host material (straight lines) and the QD (parabolas) are sketched as well as the energy levels for the electrons (in the upper parabolas) and the holes (in the lower ones).

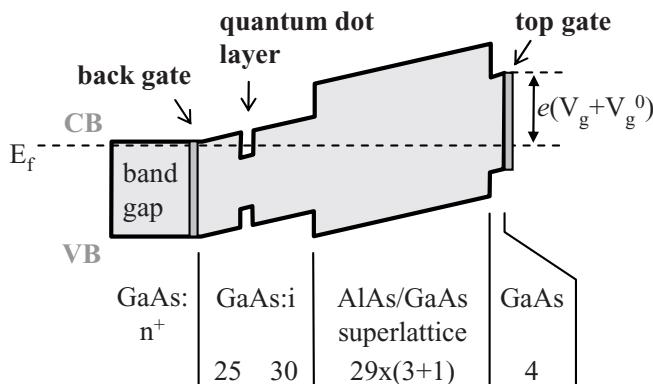


Fig. 2 Scheme of the sample band gap structure along the growth direction (left to right). The gate voltage V_g adds up with the Schottky voltage V_g^0 to a constant electric field between the n-doped back gate and the top gate. This is used to tune the QD's potential relative to the Fermi energy E_f which is pinned by the highly doped back gate. Through the 25 nm tunnel barrier between back gate and QD electrons tunnel in and out of the QD so that only the energy levels below E_f are occupied. The exact number of electrons in the QD can be adjusted by V_g .

with a linewidth much below the resonance width (for external cavity laser diodes about 0.01 μeV). Therefore laser spectroscopy provides information about the true lineshape.

3 Sample structure and setup

The studied QDs are InGaAs dots which are imbedded in a FET structure as shown in Fig. 2. The device was grown by MBE on a GaAs substrate. First a highly n-doped back contact was grown followed by a 25 nm GaAs tunnelling barrier and the QDs which were centered around 950 nm in PL energy [16]. Above the QDs a 30 nm GaAs spacer was deposited and a 116 nm AlAs/GaAs blocking barrier to prevent current flow from the top gate. The sample is capped by an 4 nm GaAs layer and a 5 nm semitransparent NiCr top gate was evaporated on top of it.

The n-doped back contact pins the Fermi energy to the GaAs conduction band edge and a Schottky voltage V_g^0 of -0.6 V is formed between the sample and the metallic top gate. This adds up with the externally applied gate voltage V_g to the total voltage drop between top and back gate. As the semiconductor material between the gates is not conducting the voltage drops linearly which leads to the tilt of the band edge seen in Fig. 2. By applying a more negative voltage to the gate the structure gets more tilted and electron states in the QD are moved above the Fermi energy. As only the states below the Fermi energy are occupied, the number of electrons, the charge of the dot can be adjusted with one electron accuracy [4].

All measurements were done in a bath cryostat at 4.2 K. The sample is mounted above a p-i-n diode on a three axis positioner system under an objective of a confocal microscope with a NA of 0.55. The sample and microscope are put in a tube of stainless steel. The tube is evacuated and filled with 5 mbar He as exchange gas. This system is top loaded into the cryostat. The optical connection between the low temperature microscope and the room temperature setup is made by a glass fiber. This fiber can be connected to a spectrometer and an 830 nm laser for PL experiments or to a tunable laser for the resonant experiments.

For resonance experiments a diode laser with a tunable external cavity is used. Its beam passes through a Faraday isolator to prevent back reflections into the laser and a beam splitter where 50% of the light is sent to a wavemeter to monitor the laser wavelength. The other 50% are coupled through neutral density filters to adjust the intensity and a polarization unit into the microscope's glass fiber. To improve the signal to noise ratio a lock-in amplifier is used. The gate voltage is modulated by a square wave volt-

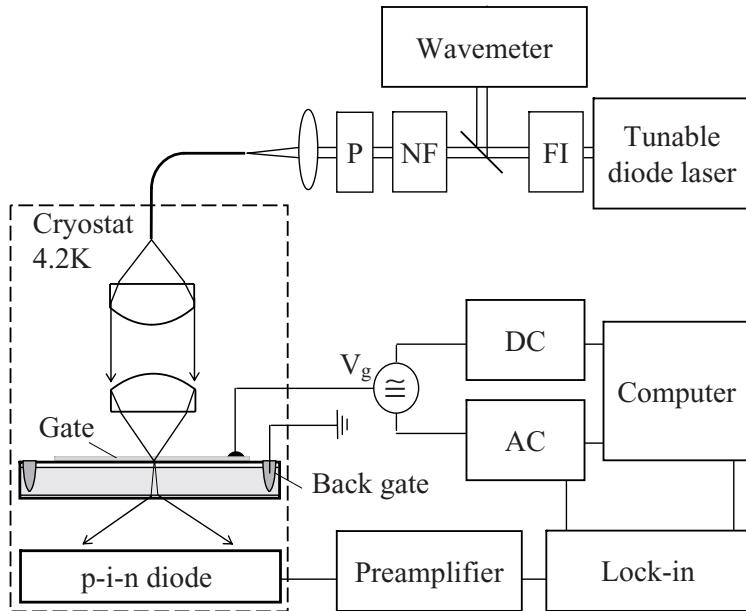


Fig. 3 Sketch of the experiment setup. A DC voltage is applied to the gate to adjust the charge of the QD and to shift its transition energy by the Stark effect. An AC voltage is added to modulate the signal which is recorded by a computer after amplification by a preamplifier and a lock-in. The QD is illuminated by a narrow laser line which is send through a Faraday isolator (FI), neutral density filters (NF) and a polarization unit (P). The wavelength is recorded by a wavemeter.

age with a peak to peak amplitude of about 100 mV. Because of the Stark effect, which corresponds to a shift of the resonance energy by about 1 μeV per mV this leads to an energy shift of 100 μeV . That is nearly two orders of magnitude larger than the linewidth. Hence the modulation is a complete switching on and off of the resonance and the true lineshape is revealed. This AC voltage is mixed with an DC one to bias the gate. The signal of the photo detector is amplified by a low noise preamplifier and fed into a lock in amplifier.

To prevent mode hops and intensity changes of the laser it is kept constant during the measurements. Before a scan is done the laser is tuned close to the excitonic transition frequency and then the exciton energy is tuned through the laserline by the Stark effect caused by the DC voltage applied to the gate.

4 Experimental results

4.1 Charge controlled fine structure

Figure 4 shows transmission spectra for a neutral exciton (X^0) where an electron hole pair is created in an empty QD and a negatively charged exciton (X^{1-}) where the electron hole pair is created in addition to a single electron occupying the QD. The X^{1-} shows one polarization independent resonance while the X^0 has a fine structure splitting.

The two lines of the X^0 are linearly polarized and the fine structure splitting for InGaAs QDs varies strongly even on neighboring QDs. On our sample the splitting is typically in the range of 10 to 40 μeV . It is caused by the lack of rotational symmetry of the QDs potential and the interaction of the total electron and hole spin [17, 18]. The X^{1-} consists of one hole and two electrons and the electron spins are anti parallel because of Pauli's exclusion principle. Therefore the total electron spin is zero and no splitting exists on the X^{1-} . Hence by charging the QD with a single electron the fine structure can be turned off. It

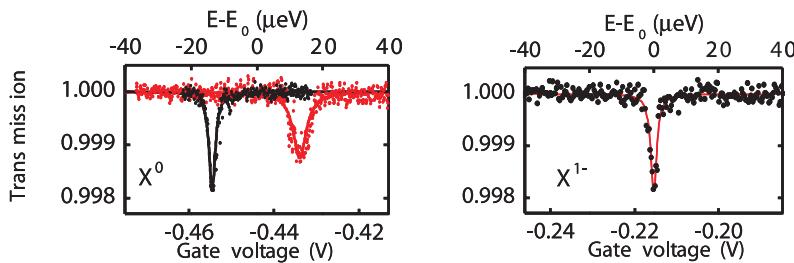


Fig. 4 (online colour at: www.pss-a.com) Transmission spectra of the neutral (X^0) and negatively charged (X^{1-}) exciton measured as a function of gate voltage and converted by the Stark effect into energy detuning.

has to be noted that the resonance energy of the transition changes by about $6 \mu\text{eV}$ when the dot gets charged. This shift is caused by the Coulomb interaction between electrons and hole.

4.2 Strain controlled fine structure

A reason to study the tuning of the fine structure without changing the charging state is the emission of entangled photons. It was recently shown that polarization entangled photons can be produced by neutral QDs where the fine structure is reduced below the linewidth of the resonances [8]. Hence an *in situ* control of the potential's rotational symmetry is of interest. In plane electric [19] and magnetic fields [20] have been used to manipulate the potential shape. We investigated the effect of strain [21].

To apply mechanical strain to the sample it is glued onto a piezostack as shown in Fig. 5. The stretching direction is aligned parallel to the [110] direction of the sample and to screen electric fields from the piezostack the back gate of the sample and the ground of the piezo voltage source are connected. A sample identical in dimensions is glued on the opposite side of the stack to prevent bending. The strain is calibrated by monitoring the shift of the GaAs photoluminescence as a function of piezo voltage and comparison to literature values [22]. A strain $\Delta L/L$ of 7.9×10^{-7} per volt is applied to the sample. As the sample is directly glued onto the piezostack and the stack is not transparent no transmission experiments can be performed. Therefore the QD is studied in reflectivity. A beam splitter is placed between the laser unit and the glass fiber to the cryogenic microscope so that the reflected light can be measured by a detector at room temperature (Fig. 5b).

Figure 6 shows two reflectivity spectra of a single QD. One at zero piezo voltage and one at 150 V. The fine structure splitting is clearly observed in both cases. Both resonances are shifted in energy by about $80 \mu\text{eV}$. To study the effect of the strain in detail the following procedure is chosen. The polariza-

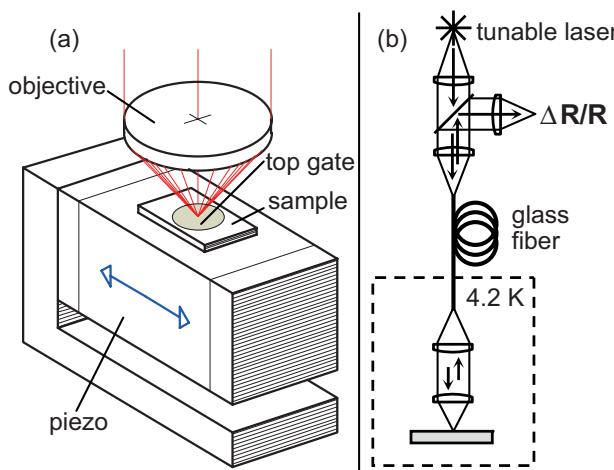


Fig. 5 (online colour at: www.pss-a.com) (a) Sample is glued onto a piezo with the [110] direction parallel to the piezo movement (arrow). The piezo is glued onto an L-shaped holder and mounted in the cryogenic confocal microscope. (b) A beam splitter is placed in the beam path to measure the reflection signal of a single QD.

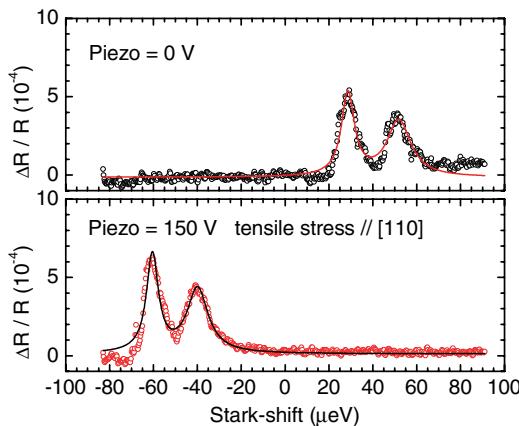


Fig. 6 (online colour at: www.pss-a.com) Reflectivity spectra of a single QD at piezo voltages of zero and 150 V. The two resonances seen in each spectrum are the linearly polarized exciton transitions, separated by the anisotropy fine structure splitting.

tion is aligned such that one of the X^0 resonances has its maximal amplitude and the other vanishes. Because of increased signal to noise the resonance position in energy can be determined with higher accuracy. After that the polarization is changed to the orthogonal one and the position of the other peak is determined for the same piezo voltage. After changing the piezo voltage the laser is detuned until the shift of the excitonic transitions in gate voltage caused by the strain (the shift to the left in Fig. 6) is compensated. This back tuning by the laser is done to ensure a constant Stark shift. To improve the data accuracy further, 15 individual spectra are taken at every piezo voltage and laser energy, fitted by a Lorenz function and the average value and the standard deviation for each data point is calculated.

The results of these measurements are shown in Fig. 7a. The data points could be well fitted by two linear lines with slopes of 11.3 ± 1.8 eV/MPa for the resonance with the higher energy and 11.6 ± 8 μeV/MPa for the lower one. As the lattice constant is reduced with pressure the band gap increases. This is in perfect agreement with the measurements. Figure 7b, shows a plot of the fine structure

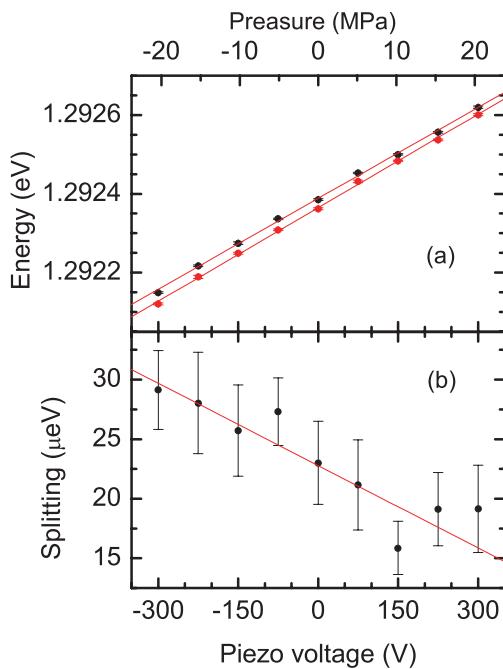


Fig. 7 (online colour at: www.pss-a.com) Piezo voltage dependence of the two fine structure resonances (a) and their splitting (b). Each data point is the average of 15 measurements, the error bars are the standard deviations and the linear lines are fitted.

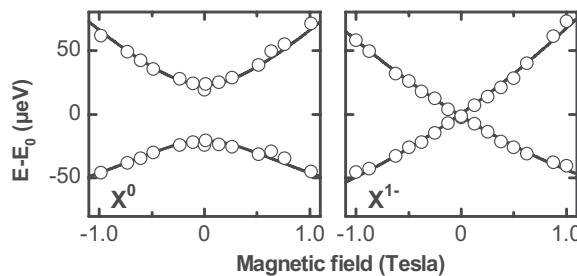


Fig. 8 Dispersion of the X^0 and the X^{1-} in magnetic field. The X^{1-} data is fitted by the function $\pm 1/2g^*\mu_B B + \beta B^2$. The resulting parameters are $g^* = 1.8$ and $\beta = 8.7 \mu\text{eV/T}^2$. These are used in combination with the fine structure splitting $\Delta_{\text{FS}} = 42 \mu\text{eV}$ to describe the dispersion of the X^0 with $\Delta E = \pm \frac{1}{2} \sqrt{\Delta_{\text{FS}}^2 + (g^* \mu_B B)^2} + \beta B^2$.

splitting. The fitted line has a slope of $-34 \pm 08 \mu\text{eV/MPa}$. That is 3% of the shift in energy. We can decrease and increase the splitting in situ, both by a value of $6.9 \mu\text{eV}$.

It has to be emphasized that the applied strain in this experiment was limited by the piezo and not by the material properties of the sample. Therefore it can be speculated that a control of the fine structure splitting over a much bigger energy range is possible and even zero splitting might be achievable.

4.3 Quantum dot as polarization analyzer

Unlike the X^0 the X^{1-} does not have a fine structure or polarisation dependence. However when a magnetic field is applied along the growth direction of the sample which is also the optical axis in the setup (Faraday configuration) a Zeeman splitting occurs. Figure 8 shows the dispersion of the neutral and charged exciton of the same QD in magnetic field. The data points of the X^{1-} are fitted by two parabolas. One for the resident electron spin in the upward direction and the other one for the downward direction. The quadratic term in the fit function is the diamagnetic shift which is $8.7 \mu\text{eV/T}^2$ for that QD. The Zeeman term has a excitonic g -factor of 1.8. These values are in good agreement with photoluminescence measurements done on the same sample [2]. The X^0 (Fig. 8 left) has at zero magnetic field a fine structure splitting of $42 \mu\text{eV}$. This splitting has to be included into the dispersion function as a hyperbolic that becomes asymptotically the fit function of the X^{1-} for high magnetic fields and is plotted as solid line to the X^0 data points. This curve has no free parameters.

The two Zeeman resonances of the X^{1-} are circularly polarised as can be seen in Fig. 9b. A magnetic field of 0.45 T is applied to the sample and spectra for left-handed circular, linear and right-handed circular laser light are shown. In Fig. 9a, the linear polarisation dependence of the X^0 at zero magnetic field is shown. The two resonances of the X^0 become asymptotically circular polarised at high magnetic fields.

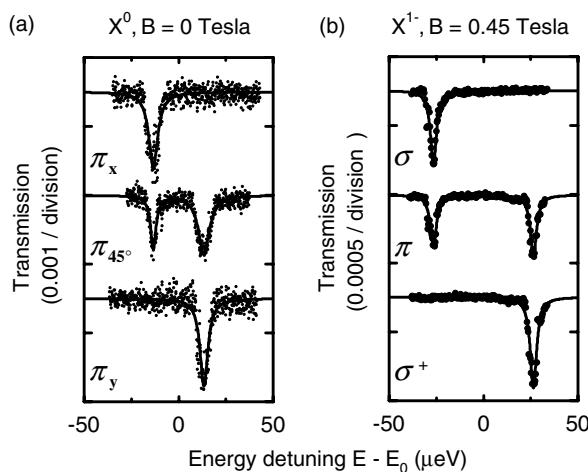


Fig. 9 Two X^0 resonances are linearly polarized (a) while the Zeeman split lines of the X^{1-} are circularly polarized (b).

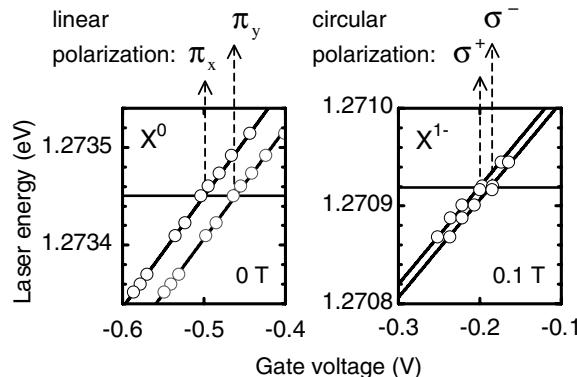


Fig. 10 Resonance gate voltages for different laser energies are plotted for X^0 and X^{1-} (circles). The voltage dependence of the resonances energies is caused by the Stark effect. The horizontal lines represent a monochromatic light source whose polarization can be analyzed in the basis of the diagonal lines (linearly for the X^0 and circularly for the X^{1-}).

The coefficient of the Stark effect has to be measured to convert the gate voltage detunings (e.g. resonance linewidth or fine structure splitting) into an energy scale. Therefore the experiment is repeated several times for different laser wavelength. The laser energy is plotted against the resonance voltages (Fig. 10). The data points are fitted by linear curves. The slope of these curves is the Stark coefficient. Fig. 10 shows those data for X^0 and X^{1-} . While no magnetic field was applied for the X^0 measurements 0.1 T were applied for X^{1-} to resolve a Zeeman splitting. It can be seen that the excitonic transition can be brought into resonance with the laser not only for one laser energy but for an energy range of order 100 μ eV because the QD is imbedded into the FET-structure. As this energy range is larger than the fine structure splitting of the X^0 both resonances can be detected for one fixed laser energy just by changing between the two resonance voltages. These resonances have orthogonal linear polarizations. Therefore the QD can be used as a linear polarization analyzer for monochromatic light in the basis set of the dot potential. To analyze the polarization in a circular basis set the X^{1-} can be used in the same way when a small magnetic field is applied (horizontal lines in Fig. 10).

5 Conclusion

The presented results were measured by Stark effect modulation spectroscopy. This method offers a high spectral resolution so that the asymmetry fine structure splitting of the exciton could be resolved. This splitting could be switched off by charging the QD with an electron. But that process destroys the possibility to emit entangled photons. We showed that the splitting can be reduced by changing the QD potential by applying uniaxial strain. Further we presented measurements that the neutral exciton can be used as an analyzer for linear polarizations and the charged exciton for circular polarizations if a magnetic fields is applied.

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