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Effect of uniaxial stress on excitons in a self-assembled quantum dot

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The fine structure of the neutral exciton in a single self-assembled InGaAs quantum dot is investigated under the effect of an applied uniaxial stress. The spectrum of the excitonic Rayleigh scattering was measured in reflectivity using high-resolution laser spectroscopy while the sample was submitted to a tunable uniaxial stress along its [110] crystal axis. We show that using this stretching technique, the quantum dot potential is elastically deformable such that the exciton fine structure splitting can be substantially reduced. © *2006 American Institute of Physics*.
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Entangled photons have attracted considerable theoretical and experimental interests in recent years.[1] Semiconductor quantum dots (QDs) have been proposed as sources of polarization-entangled photons.[2] The commonly proposed scheme is to exploit the biexciton which has two pathways for radiative decay. The practical limitation is presently the existence of an anisotropic exchange splitting (AES) in the intermediate state, the single exciton ground state.[3] This splitting originates from the lack of cylindrical symmetry in the quantum dot confining potential and leads to two distinct linearly and orthogonally polarized emission lines.[4,5] The AES leads to several interesting effects such as quantum beats on optical excitation with circularly polarized light[6,7] and the possibility of an all-optical quantum gate.[8] Additionally, the AES has important consequences for the generation of polarization-entangled photon pairs. When the AES is smaller than the radiative broadening of the transitions, the two single exciton decay paths are no longer distinguishable and interference between the two pathways is expected. In the other limit, when the AES exceeds the radiative broadening, biexction decay results in polarization-correlated but not polarization-entangled photon pairs. In practice the radiative broadening of the exciton is typically $\sim 1\ \mu$eV in an InGaAs self-assembled QD, yet the AES is a few tens of μeV. While the lack of inversion symmetry in the III-V lattice is itself a source of AES,[4] the AES in a self-assembled QDs is dominated by symmetry breaking through a structural anisotropy, as through the piezoelectric field.[9] For instance, an InGaAs QD is typically elongated along the [110] crystal axis as a result of the self-assembly growth process.[10] In this case, the generation of polarization-entangled photon pairs without manipulating the AES is impossible.

A clear challenge is to develop techniques to tune the AES with the ultimate goal of reducing it below the radiative broadening limit. The use of postgrowth annealing[11,12] shows a clear reduction of AES but is unlikely to lead to its cancellation and tuning cannot be carried out *in situ*. An in-plane electric field has been shown to reduce the AES in a tunable way but unfortunately a large field is presently required to approach zero AES and at these large fields, there is a dramatic reduction in the excitonic emission intensity as a result of field-induced reduction of the electron-hole overlap.[13] So far, the most successful approach has been the use of an in-plane magnetic field[14] in combination with a small and negative AES achieved with optimized growth, allowing the AES to be tuned through zero. Strong evidence for polarization-entangled photons has very recently been reported.[15] In this letter we present an alternative approach. We show that applying a tunable uniaxial stress on the QD along its major axis partly restores the cylindrical symmetry of the excitonic wave function without reducing the exciton oscillator strength.

Our technique for deforming the sample ($4 \times 3 \times 0.5$ mm^3) in a controlled way is to glue it tightly onto a piezoelectric lead zirconic titanate (PZT) ceramic stack[16] as shown in Fig. 1.[17,18] The stretching direction of the piezostack is aligned parallel to the [110] axis of the crystal. One end of the piezostack is attached to an L-shaped holder mounted in a cryogenic (4.2 K) confocal microscope operating with a spot diameter of 1.3 μm. The microscope is equipped with a three-axis Attocube positioner allowing us to focus a laser beam onto a single quantum dot. The microscope is introduced into a vacuum tight thin-walled stainless-steel tube which is in turn top loaded into a liquid He bath cryostat. The whole microscope along with the sample is cooled using He exchange gas at low pressure. The microscope is routinely used in a standard photoluminescence (PL) configuration in order to select a single QD spectrally well separated from the neighboring peaks. This setup is thermally stable, allowing an alignment-free investigation of a single dot over several weeks. The QDs in the sample are embedded in a field-effect heterostructure containing a highly *n*-doped region in the sample which acts as a back gate. The sample is processed with Ohmic contacts to the

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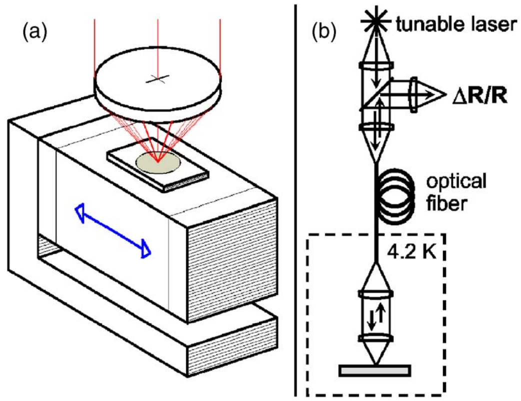

FIG. 1. (Color online) Schematic of the experimental setup. The sample is tightly glued onto a piezostack so that its [110] axis lies along the direction of the piezostress (shown by the arrow). The piezostack is attached by one of its ends to an L-shaped holder which is in turn mounted in a low-temperature confocal microscope. Laser light is coupled through a fiber beam splitter into the microscope. The microscope objective has NA of 0.55. The reflected light is collected with the same optics.

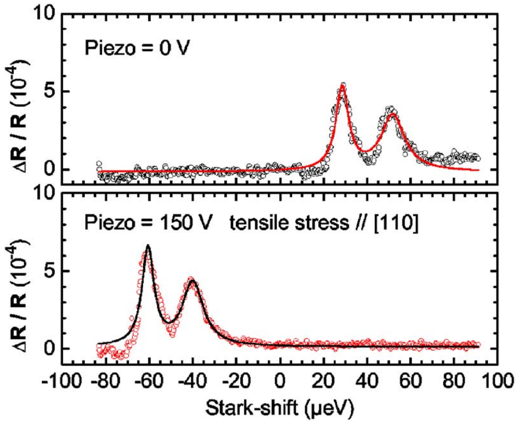

FIG. 2. (Color online) Low-temperature reflectivity spectra showing resonances from the neutral exciton in a single QD at piezostack voltages of 0 V (top) and 150 V (stress=10.2 MPa) (bottom). The laser wavelength is 964.644 nm. The two resonances seen in each spectrum are the two linearly polarized transitions of the exciton, separated by the anisotropic exchange splitting (AES).

back gate and a NiCr semitransparent Schottky contact on the sample surface.[19] In the experiment, the back gate is connected to the ground of the piezostack in order to screen fringing fields generated in the 100 μm periodically poled stacks. A voltage is applied to the top gate and is used to shift the QD exciton energy through the Stark effect. The QD energy is tuned through the energy of a narrow-band laser, detecting the resonance through the Rayleigh scattering. In this way, we can perform spectroscopy with high resolution, less than 0.1 μeV, considerably less than the limit determined by radiative broadening, ~ 1 μeV. Previously, we detected the resonant Rayleigh scattering in transmission.[20] In the experiment reported here, the piezostack prevents access to the transmission signal and we have therefore turned instead to measuring the reflectivity.[21] In the reflectivity experiment, the laser beam is focused down to the diffraction limit onto the QD. The light reflected off the sample surface and the light backscattered through the Rayleigh process off the QD add coherently and retrace their path through the microscope objective towards a beam splitter that directs 99% of the reflected light to a standard silicon detector located at room temperature. The current induced in the detector is transformed into a voltage using a very low noise current amplifier.[22] A weak square modulation voltage is applied on the gate (80 mV, 77.3 Hz) in order to tune the exciton in and out of resonance with the laser energy allowing for a lock-in detection of the Rayleigh scattered light (2 s integration time per data point). A dc gate voltage is superimposed and slowly ramped (1 mV/s) in order to record a spectrum. The gate voltage is converted into a detuning by repeating the experiment at several closely spaced laser wavelengths where in each case the laser wavelength is determined with a wave meter.

In order to symmetrize the load on the stretching device, a piece of sample, identical in size and thickness to the QD sample, is mounted on the opposite side of piezostack. A resistive strain gauge[23] is glued onto the surface of this second sample. The strain is monitored with an ac resistance bridge in combination with a lock-in detection. Evidently the strain gauge is not glued directly onto the QD sample as in this case it would block the light path to the QDs. In addition to the strain gauge, the uniaxial stress was measured through the energy shift it induces to the GaAs band gap of the QD host material. The GaAs PL emission was measured along with the QD PL from the same micron-sized surface region allowing us to measure the local stress experienced by a particular QD. In practice, we find that the GaAs PL consists of four peaks with slightly different dependencies on the piezostack voltage. The peaks correspond to excitons bound in different ways. We assume that the highest energy GaAs PL peak behaves in the most similar way to the GaAs band gap as it has the smallest binding energy. This peak shifts with piezostack voltage with rate of 0.82 eV/V. With the known dependence of the GaAs band gap on the [110] uniaxial stress, namely, 12 eV/MPa,[24,25] and the Young's modulus of GaAs (86 GPa),[26] we find that the dot experiences a strain of $\Delta L/L$ of 7.9×10^{-7}/V applied to the piezostack. This value agrees well with similar experiments performed with the same geometry but with a different GaAs heterostructure at 4.2 K.[17] In the following, we choose the convention that positive voltages correspond to a compressive stress of the piezostack.

Figure 2 shows two typical reflectivity spectra exhibiting resonant Rayleigh scattering. One was measured with a piezostack voltage of 0 V and the other with 150 V which corresponds to an added compressive uniaxial stress of 10.2 MPa. The fine structure splitting of the exciton is clearly visible in both cases. The major effect is that at constant laser energy, the uniaxial stress shifts the resonance lines to lower gate voltage. This corresponds to an increase of the fundamental QD gap with stress. When the scans are repeated we find that the peak positions fluctuate slightly. This effect is also observed in transmission spectroscopy and might be caused by charge reconfiguration near the QD.[17] A typical standard deviation in the peak value of ± 2 μeV was determined over 15 individual spectra. In order to determine the full stress dependence, we adjust the laser energy at each piezostack voltage such that the resonances always appear at the same gate voltage, 385 ± 2 mV. In this way, we can be sure that the changes in the optical response of the QD arise from the applied stress and not from a changing vertical electric field. In addition, to make sure that the presence of both resonances in the spectrum does affect the individual peak positions, we measured each exciton resonance separately by

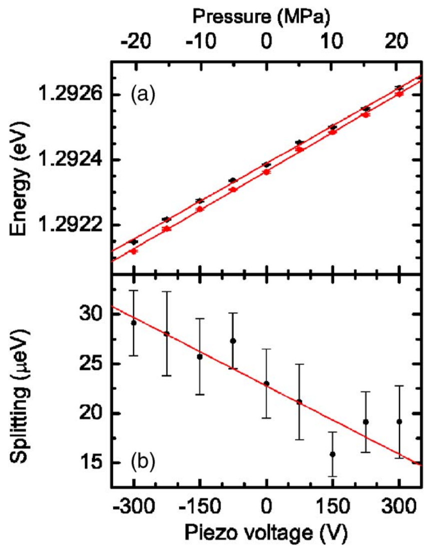

FIG. 3. (Color online) Piezovoltage dependence of (a) the two neutral exciton resonances and (b) the splitting between the two resonances. In both (a) and (b), the solid lines are linear fits to the measured data points. Each data point is the average of 15 measurements and the error bar represents the standard deviation.

selecting the appropriate linear polarizations. In each case, the peak maximum is determined by fitting a Lorentzian line shape to the reflectivity spectrum. The peak positions are plotted in Fig. 3(a) as a function of piezostack voltage and the corresponding optically determined stress. We find that the slope for the peak with the higher (lower) transition energy is $11.3\pm1.8(11.6\pm1.8)$ μeV/MPa. To within the measurement error, these values are the same as the response of the GaAs band gap to the [110] uniaxial stress.

The crucial result concerns the AES: Fig. 3(b) shows the dependence of the AES on the piezostack voltage. We find that the AES changes linearly with stress in this range with a slope of -0.34 ± 0.08 μeV/MPa, corresponding to 3% of the average shift in exciton energy. With +300 V (−300 V) on the piezostack we were able to decrease (increase) the AES by 6.9 μeV. The important result is therefore that we have achieved an *in situ* tuning of the fine structure splitting through application of a uniaxial stress.

This experiment demonstrates the principle of AES tuning with uniaxial stress. As yet, we have not managed to tune the splitting towards zero. The maximum change in AES is presently limited by our upper limit on the stress. This is limited by the piezostack and possibly also by the quality of the gluing. The sample itself should withstand uniaxial stresses up to the breaking limit of 1.0 GPa,[24] a stress much higher than the ones applied here. Hence, in order to reduce the AES further, a larger stress needs to be applied and we believe that a convenient technology allowing an *in situ* variation could be developed to achieve this. An alternative strategy would be to start with a preannealed QD sample with a typical AES of about 10 μeV and then to manipulate the AES with stress. In this case, the stress generated in the present experiment may be sufficient to tune the AES to zero. It remains an open question whether it is possible to cancel all the QD asymmetry and also the residual asymmetry in the atomic lattice.

As a final point, we note that the fact that the QD exciton energy can be adjusted through the piezostack voltage allows for an alternative scheme for modulation spectroscopy. The exciton in the QD can be tuned into resonance with a narrow-band laser by changing the voltage applied to the piezostack. Such a feature can be used to advantage when modulation through the Stark shift is not possible. We propose modulating the resonance by modulating the stress with an ac piezostack voltage while slowly ramping the resonance position with a dc piezovoltage.

In conclusion we have shown that the symmetry properties of the confining potential of a single QD can be fine tuned *in situ* at low temperature by applying a uniaxial stress to the sample. We have demonstrated in this way that the fine structure splitting of the exciton can be substantially lowered.

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