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Polarization spectroscopy of positive and negative trions in an InAs quantum dot

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Abstract

Using polarization-sensitive photoluminescence and photoluminescence excitation spectroscopy, we study single InAs/GaAs self-assembled quantum dots. The dots were embedded in an n-type, Schottky diode structure allowing for control of the charge state. We present here the exciton, singly charged exciton (positive and negative trions), and the twice negatively charged exciton. For non-resonant excitation below the wetting layer, we observed a large degree of polarization memory from the radiative recombination of both the positive and negative trions. In excitation spectra, through the p-shell, we have found several sharp resonances in the emission from the s-shell recombination of the dot in all charged states. Some of these excitation resonances exhibit strong coulomb shifts upon addition of charges into the quantum dot. One particular resonance of the negatively charged trion was found to exhibit a fine structure doublet under circular polarization. This observation is explained in terms of resonant absorption into the triplet states of the negative trion.

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1. Introduction

Semiconductor quantum dots (QDs) have been studied intensely in recent years as attractive components for implementing quantum computation schemes [1]. In particular, the unpaired spin of an electronically charged semiconductor QD is especially exciting, because of its relatively long spin-dephasing time, and because its spin can be controlled and measured optically [2]. Knowledge of the discrete energy spectrum of these QDs and

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the ability to control the individual charges in the QDs is vital for future progress in this field.

A great deal of progress has been made recently in understanding the behavior of confined charges by studying single QD photoluminescence (PL) with charge controlled by a field effect device containing the QDs [3–5]. These studies have readily identified exciton states which exhibit characteristic coulomb shifts in energy when a QD is charged by countable numbers of positive and negative charges. This, then, lays the groundwork for further study of specific charged exciton states. The present work has concentrated on the positively charged exciton or positive trion (X^+), the neutral exciton (X), the negative trion (X^-), and the doubly negatively charged exciton (X^{2-}).

In addition to being able to controllably charge the QD, a requirement for quantum information processing is the ability to control and measure the spin of the charge carriers in the QD [1]. This is accomplished with circularly polarized light which couples to certain spin states of the carriers. In particular, if one defines the quantization axis as that of the incident photon, a spin-up heavy hole and spin-down electron pair are photogenerated by a right circularly polarized photon (σ^+), and the opposite spins pair is photogenerated by a left circularly polarized photon, (σ^-) [6]. The reverse process, recombination, involving these same spin polarized pairs will in turn result in the creation of a σ^+ and σ^- photon. In isolated QDs, recombination of single electron–hole pairs can be readily identified spectrally [7]. This allows one to probe the spin of the recombining charge carriers by detecting the polarization of the emitted photon. When a single electron (hole) is present in the dot, optical excitation of an electron–hole pair (exciton) results in a pair of electrons (holes) in the ground state and an unpaired hole (electron) in its ground state. By exciting with circularly polarized light and analyzing the degree of circular polarization of the emitted light, one can measure the polarization memory of the QD, which is related to the rate by which the unpaired photogenerated charge carriers flip their spin. By combining these optical orientation techniques and the charge selectivity with single-dot excitation spectroscopy [8], we hope to gain an understanding of the excited state resonances of the dot and how they influence the spins of the charge carriers confined to the dot.

2. Experiment

InAs QDs are grown by molecular beam epitaxy and are embedded in a n^+-intrinsic-Schottky (NIS) diode in order to control the QD charging. The structure growth and fabrication are described in detail in Ref. [9].

Using a tunable cw Ti-Sapphire laser, PL and PL excitation (PLE) was excited and detected through metal apertures. The light was collected and dispersed with a 0.5 m single-grating spectrometer and a charge-coupled device array. The exciting light was circularly polarized and the degree of circular polarization of the emitted light was analyzed with variable liquid crystal retarders. The excitation power for the polarization memory measurements was chosen such that multiexciton spectral lines were minimized.

3. Results

Fig. 1 shows representative PL of a single QD as a function of diode bias, where the coulomb shifts due to charging of the dot are clearly visible [3]. The X^+ is excited by sub-wetting layer excitation through the photogeneration of a confined hole which is discussed at length for these types of samples in Ref. [9]. Increasing the bias turns off the hole generation resulting in PL emission from neutral excitons. Further increase in the bias results in addition of negative charges to the QD. With 1, 2, and 3 electrons, emission lines from X^- (negative trion), X^{2-}, and X^{3-}, respectively, are observed in the PL spectra.

By examining the PL and measuring the polarization memory as a function of bias for several different excitation energies below the wetting layer energy, we can gain a general understanding of the relaxation rates of the spin states of the photogenerated carriers. Fig. 2 summarizes the results for these experiments. The polarization for each excitation energy is recorded

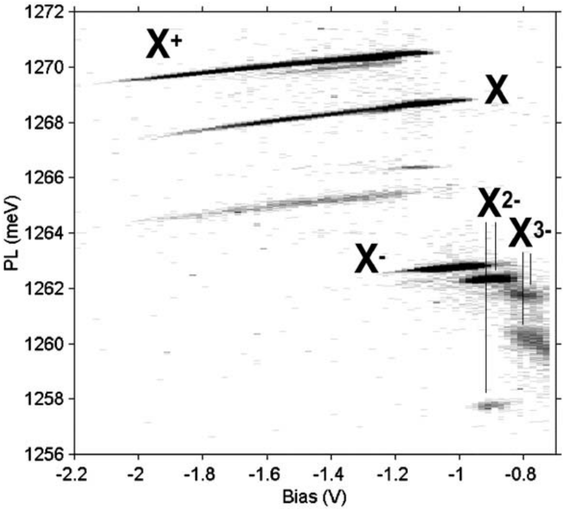

Fig. 1. Intensity plot of PL versus diode bias at ~10 K with excitation just below the wetting layer at 1425 meV for dot 1. The exciton is labeled with an X, and the various charge states are labeled by a superscript.

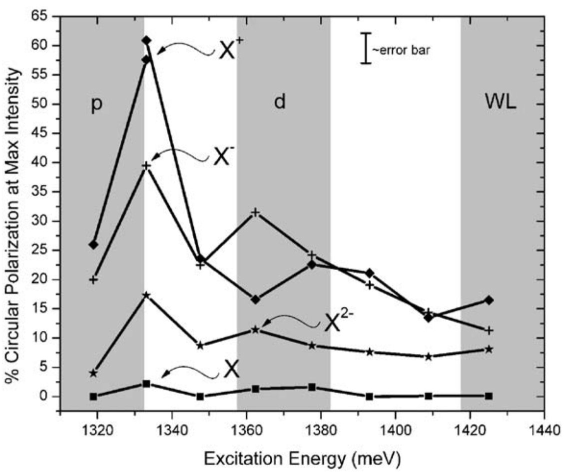

Fig. 2. Circular polarization memory as a function of excitation energy for the PL lines from Fig. 1. The polarization values are taken at the bias which maximizes the PL intensity of each respective line. These bias values are not constant with excitation energy and therefore must be optimized at each laser step. The grayed regions denote the range of energies through which this dot emits light in the p- and d-shells, and the approximate location of the wetting layer (WL) emission.

at the bias where the emission intensity is maximum for that line. These biases, for which the intensity is maximum, change with excitation energy most likely due to background absorption altering the local electrostatic field at the QD. The data show that the polarization memory of the charged lines generally increases as the laser energy decreases. The circular polarization of the neutral exciton is found for all conditions to be vanishingly small. Indeed, we expect the exciton to be linearly polarized due to the anisotropic electron–hole exchange [10]. However, the overall increase in polarization of the trions is convoluted with large fluctuations as the laser is tuned into the excited states of the QD as shown by the highlighted regions labeled p and d in Fig. 2.

In order to understand better the polarization memory at resonant excitation into the QD excited states, we first present in Fig. 3 PLE spectra at fixed biases through the range of energies of the p-state for the observed PL lines. Quite noticeably, there are several sharp resonances for each of the different charge states. Identification of each of the resonances is beyond the scope of this report. We note, however, that recent calculations indicate that several of the spectral features may be explained by forbidden optical transitions between

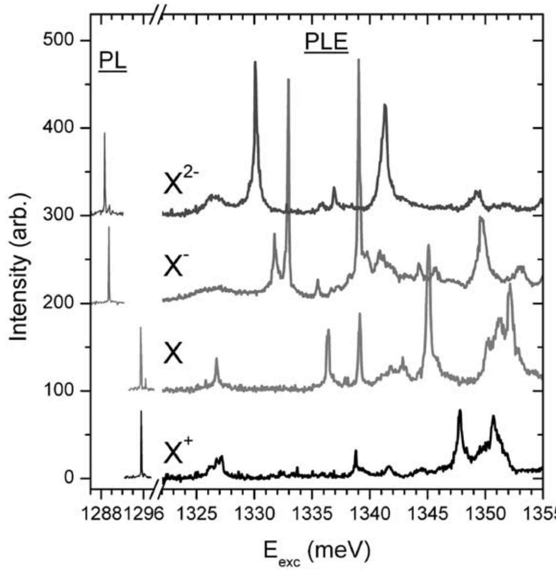

Fig. 3. p-Shell PLE spectra at ~10 K for each of the available charge states of the exciton for dot 2. The monitored PL lines are shown to the left. The bias was fixed at: −0.67 V, −0.81 V, −0.98 V, and −2.17 V for the X^{2-}, X^{-}, X, and X^{+}, respectively.

QD-confined electron and hole states and unbound states in the wetting layer [11]. Nevertheless, the polarization of the resonances follows the non-resonant values in general. The negative trion in particular exhibits large polarization fluctuations at its strongest PLE resonance. We briefly comment below on this observation.

The strong resonance of the negative trion around 1339 meV is composed of two spectral lines, which are clearly resolved in circular polarization (Fig. 4a). For σ^+ or right circularly polarized excitation, the low-energy resonance emits with a very high degree of σ^+ polarized light, ~70%. Conversely, the high-energy resonance emits with a high degree of σ^- polarized light, i.e., opposite that of the exciting light, resulting in a negative polarization of ~−30%. A plot of the polarization is presented in Fig. 4b. It shows a characteristic line shape which has been identified in every QD that we have studied so far. The energy difference between the two lines, on the order of 100–200 μeV, is similar to the electron–hole exchange seen in, for example, the fine structure of the X^{2-} and the $2X^-$, both of which are caused by the triplet state of an electron in the s-shell and an electron in the p-shell interacting through exchange with a hole [12]. The fine-structure doublet in the polarized PLE of the negative trion has been proposed as being the result of absorption into two states of the triplet separated by the electron–hole exchange [13].

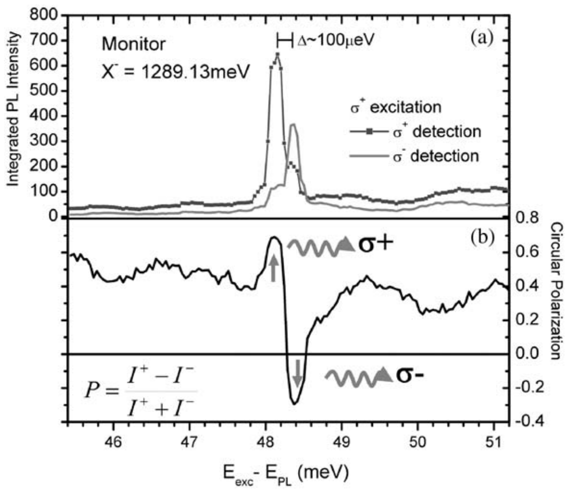

Fig. 4. (a) Polarized PLE spectra of the X^- resonance at ~1339 meV in Fig. 3. (b) Calculated polarization memory for the doublet in (a). The spectra were taken at ~10 K.

In summary, non-resonant, sub-wetting layer excitation has been used to demonstrate a significant positive polarization memory of up to 60% for the X^+ and up to 40% for the X^-. These values vary strongly, depending on the excitation energy and slightly on the particular QD. PLE has been performed on the various charged states of the dot and several sharp resonances are evident. This PLE has revealed a fine structure doublet in the excitation spectra of the X^-. The low-energy component of the doublet is positively polarized, while the high-energy component is negatively polarized. This observation is compatible with a simple model of resonant absorption into the triplet state of the negative trion.

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