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Emission line instabilities of single quantum dots of InAs in GaAs

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Abstract. We have measured the energy levels of quantum dots of InAs in GaAs as a function of time. On a very slow timescale there is a shift in energy for some dots. This shift can be enhanced by annealing the sample at different temperatures up to room temperature. The data are consistent with a model based on defects interacting with the dots.

Introduction

Semiconductor quantum dots grown by the Stranski–Krastanow (SK) technique [1] have recently become the focus of intense interest. The discrete density-of-states in these structures allows detailed spectroscopy to be performed, in particular if individual quantum dots are studied. Recent studies of individual quantum dots have demonstrated the importance of few-particle effects [2, 3], highly non-linear exciton–phonon coupling [4], and random telegraph noise in emission [5]. The most commonly studied system is InAs quantum dots in GaAs. We will here show that the emission lines of InAs quantum dots shift in energy with time. The shift is temperature dependent. Similar spectral shifts have previously been observed for colloidal quantum dots [6] but have not been reported for SK-dots and not for dots formed by interface fluctuations in quantum wells [7].

1. Experimental

The quantum dots were grown by chemical beam epitaxy [8] and a sample containing low-density dots was selected for measurements. The emission from single dots was selected using a microscope, dispersed using a monochromator and detected by a cooled CCD-camera. Typical integration times were 5 minutes at a temperature of 4 K. In order to exclude possible strain effects, the sample was very lightly glued (in one corner) on the horizontal sample holder. In order to check the stability of the detection system (monochromator and CCD-camera) we repeatedly measured the emission from a spectral lamp, often simultaneously with measurements on dots. A second source of errors comes from the temperature stability at the measurement temperature and from possible strain effects. This was checked by measuring shifts of several quantum dots at the same measurement occasion. These dots were situated within less than about $1\ \mu\text{m}$ from each other. We found that the shifts were uncorrelated between different dots, which shows that temperature differences do not cause these shifts. Since the sample is $300\ \mu\text{m}$ thick we also exclude strain effects, which should be homogeneous over $1\ \mu\text{m}$ as a source of systematic error.

2. Results and discussion

In Figure 1 we show spectra from one quantum dot, both from the ground state, at an energy of 1.329 eV, and from the excited state at an energy of 1.378 eV. After heating the sample to room temperature for one hour the experiment was repeated and the emission lines had

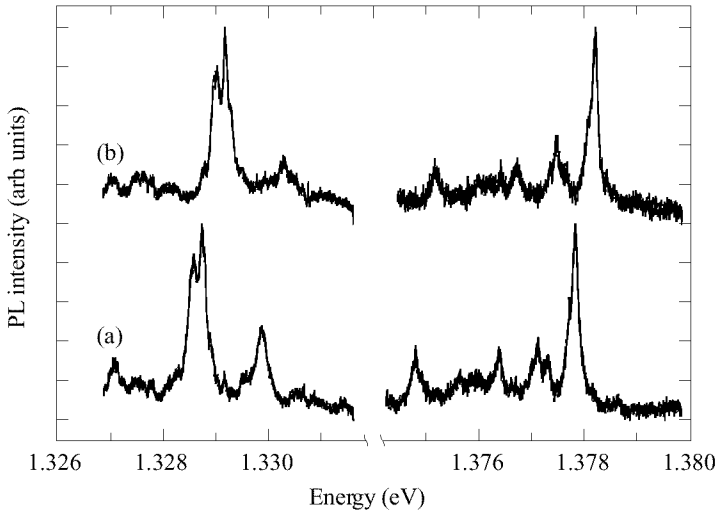


Fig. 1. Spectra of one quantum dot at two different occasions. Inbetween the measurements the sample was heated to room temperature for about one hour. Both the ground state emission at an energy of about 1.33 eV and the excited state emission at an energy of about 1.38 eV shift.

now shifted about 0.4 meV to higher energy. The shift was the same for both the ground state emission and the excited state emission. Such spectral shifts, after heat-treatment to room temperature, have been observed for every dot investigated (ten).

In Figure 2 we show the shift of the emission energy as a function of measurement occasion. Inbetween each measurement different heat-treatments were performed, indicated in the figure. Most dots have no measurable spectral shift at 4 K, but there are exceptions. In the insert of Fig. 2 we show the energy-shifts of a dot which was kept at a constant temperature of 4 K. The spectral shifts of this dot appear not as sudden jumps but rather as a slow shift with time.

In Figure 3 we show histograms of the distribution of shifts with energy for different temperatures and for different times of the heat treatment. It appears from the data that there is only a weak correlation of the shifts with annealing time. The correlation with annealing temperature is also weak, except that annealing at 4 K usually gives no shift. Concerning the cause of the spectral shifts, we can only speculate. One possibility is that the surface of the sample is charged differently at each measurement occasion. We have tested this possibility by measuring several quantum dots which were situated within $1\ \mu\text{m}$ (i. e. within our spatial resolution limit) of each other.

The shifts of each dot are uncorrelated with the shifts of other dots, as shown in Fig. 4. This indicates that surface charging is not responsible, since in that case we would expect shifts in the same direction. We rather attribute the shifts to changes in the crystalline environment of the dots which is a much more local effect than surface charging. In Figure 4 we also show the spectral shifts of two different emission lines originating from one quantum dot. The shifts are in this case highly correlated. An attractive origin of the shifts is defects in the GaAs. Such defects could be mobile and in addition they could be metastable in a variety of ways. In any case they should be activated by high temperatures, in agreement with experiment. For the dot which experienced spectral shifts at 4 K (see Fig. 2) we observed a slow shift in the spectral position, consistent with the expected behaviour of a defect passing by. Due to the absence of a clear activation energy and the

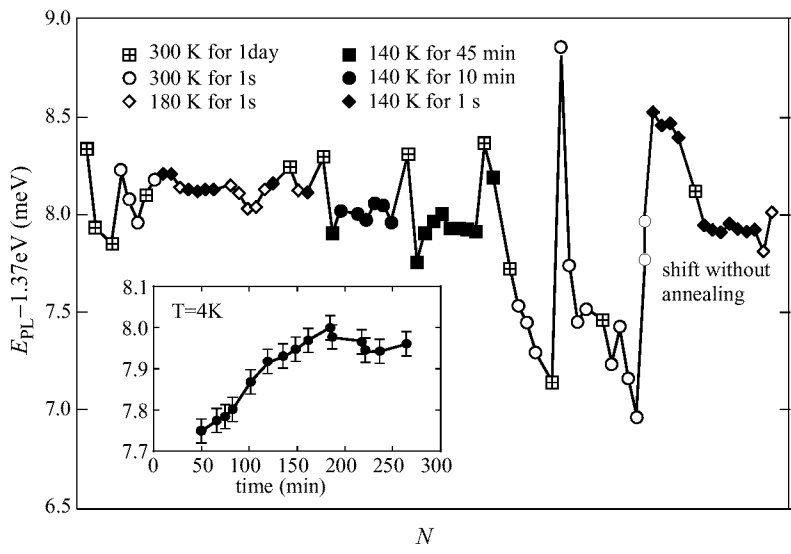


Fig. 2. Energy position of a spectral line as a function of measurement occasion (N). Different annealing temperatures were performed between each measurement, indicated in the figure. In the inset we show the energy shift for a dot at a constant temperature of 4 K.

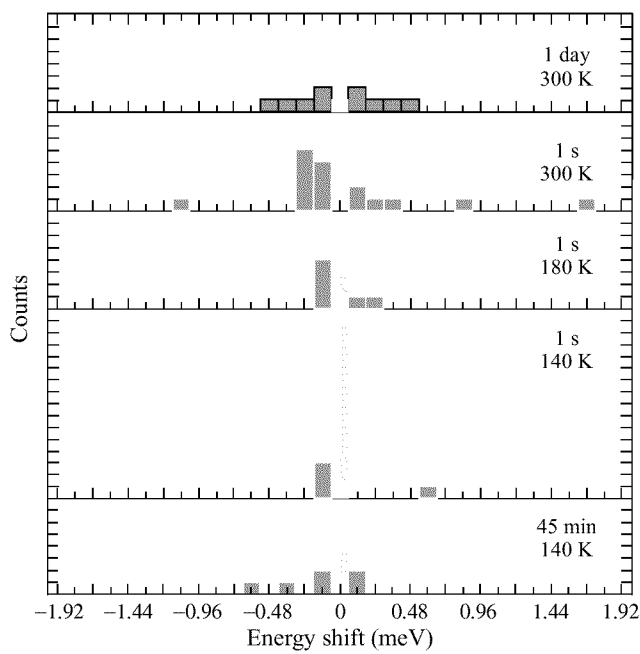


Fig. 3. A histogram of the energy shifts for different annealing times and temperatures.

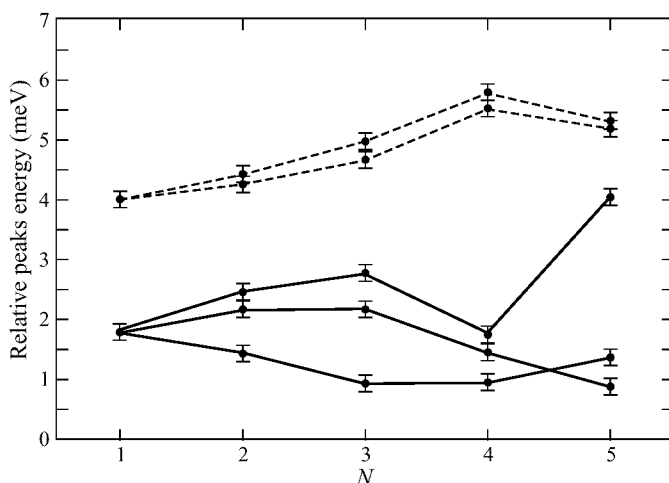


Fig. 4. A plot of the energy positions for four different dots. Three dots situated within $1\ \mu\text{m}$ are indicated with solid lines, showing that the shifts are uncorrelated. The dashed lines are indicating two different emission lines originating from one dot showing a correlated shift.

non-existent dependence on the annealing time, we believe that we actually observe the effect of several defects passing by. This instability of the emission lines from individual quantum dots may be a serious problem for the use of such structures in advanced devices. Present (conceptual) designs of quantum logic using quantum dots typically involve two coupled dots and the alignment and stability of the energy levels of each dot are critical for the operation of the gate. Other types of devices such as quantum dots in a high-Q microcavity also rely on the stability of the emission lines over time.

Acknowledgement

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