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Magnetoluminescence of Annealed Self-Organized InGaAs/GaAs Quantum Dots

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We have investigated the effects of annealing a self-assembled InGaAs/GaAs quantum dot sample between 580 and 700 °C by magnetoluminescence measurements at 2 K and fields up to 15 T. By using a high power density of about 5 kW/cm^2 for the excitation of the luminescence we were able to observe up to three features in addition to the ground-state emission arising from radiative recombination processes between excited states of the quantum dots. With increasing annealing temperature all emission lines shift to higher energies while varying their splittings. The diamagnetic shift of the ground-state emission as well as the Zeeman splittings of excited-state transitions exhibit a strong dependence on annealing and clearly speak for an increase in volume and Ga content of the dots.

In the past few years self-organized InGaAs/GaAs quantum dots (QDs) have been increasingly used as the active medium in semiconductor lasers because of their peculiar optical and electronic properties [1]. In particular, the zero-dimensional (0D) density of states with its sharp peaks at the discrete energy levels, and the enhanced excitonic binding due to carrier confinement, largely contribute to improving the lasing performance of quantum dot structures [2]. Although the energy spectrum of a quantum dot resembles that of an atom [3], a detailed description of quantum confined states for the InAs/GaAs system, for instance, ought to take into account the effects of dot shape, built-in strain distribution, and piezoelectric fields [4]. One way of modifying these parameters for a single structure, which, in turn, might lead to an optimization of QD growth procedures, consists in thermally annealing a sample at different temperatures while doing the optical characterization [5,6]. In this respect, magnetoluminescence measurements at high excitation powers can yield valuable information about the electronic structure of 0D systems by comparing the observed shifts and splittings of optical transitions with model calculations of QDs [7 to 10].

In this paper, we present photoluminescence (PL) measurements for a series of five thermally annealed InGaAs/GaAs QD samples in magnetic fields up to 15 T, and by using a high power density for excitation. From the changes in the diamagnetic shift of the ground state emission, the energy separation between PL peaks, and the magnetic field splittings, we infer an increase in Ga content and an increase in size of the dots with increasing annealing temperature.

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The as-grown sample essentially consists of a single sheet of InGaAs dots embedded in GaAs grown by low-pressure MOCVD at a temperature of 485 °C in the Stranski-Krastanow mode, as described elsewhere [5]. The nominal In concentration of 42% in the InGaAs layer is expected to be substantially higher in the QDs due to In segregation and migration effects. For the as-grown sample, the dots have an average base size of 14 to 19 nm and a height of 6 to 8 nm, as deduced from transmission electron micrographs [5]. Individual pieces of the same sample were reinserted into the MOCVD reactor and annealed for 30 min at temperatures between 580 and 700 °C under an AsH_3 atmosphere. Magnetoluminescence measurements up to 15 T were done at 2 K, and in the Faraday configuration, with circularly polarized light by inserting the sample in the cold bore of a superconducting split-coil magnet. PL spectra were excited with $\approx$5 kW/cm^2 incident power density from an Ar^+ ion laser (514 nm line), and analyzed by a 0.75 m double-grating spectrometer equipped with a LN_2-cooled Ge detector. The luminescence spectra were corrected for the spectral dependences of the various optical components.

Fig. 1 shows low-temperature PL spectra at zero magnetic field for the complete series of annealed samples in the energy region of the dot emission. The peak labeled E_0 corresponds to the ground-state exciton recombination in the QDs. At high excitation power densities, up to three additional features are apparent from the spectra at energies higher than E_0, which arise from optical transitions between excited confined states of the dots. With increasing annealing temperature, we observe a significant blueshift of the ground-state emission by about 150 meV, whereas the energy separation between the excited states is reduced by a factor of two. The wetting-layer lumines-

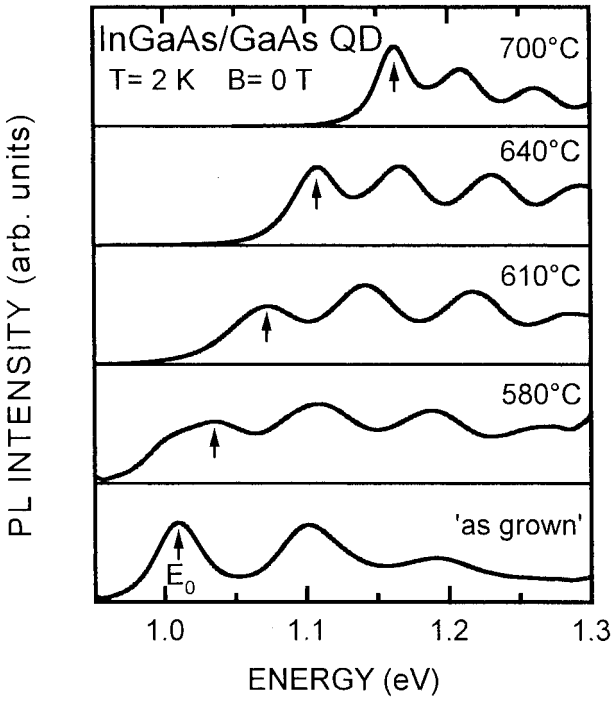

Fig. 1. Photoluminescence spectra of an InGaAs/GaAs quantum dot structure annealed at different temperatures measured using high excitation powers of 5 kW/cm^2 at 2 K and without magnetic field. The arrows indicate the position of the ground state emission maximum for each sample

cence (not shown in Fig. 1) is almost independent of annealing [5]. The PL line at 1.005 eV observed in the spectrum of the sample annealed at 580 °C (this peak is also seen in several spectra of the 610 °C sample) has not been identified yet. Nevertheless, its energy position is not altered by thermal annealing in contrast to the dot-related emission, thus it will not be further discussed.

The blueshift of the emission energies with annealing clearly speaks for an additional incorporation of Ga into the dots. By comparing the average shift of the dot emission with the band gaps of $In_xGa_{1-x}As$ alloys, we obtain an increase by ≈20% of the Ga content [11]. This value is obviously strongly underestimated, since there is a simultaneous reduction of the built-in strain due to Ga incorporation. In a first approximation, one can estimate the contribution of strain relaxation to the band gap shift from the hydrostatic part of a tetragonal stress produced by the lattice mismatch for a given Ga concentration. This results in a 45% increase in Ga concentration for the 700 °C sample, as compared with the as-grown one. In addition, thermal annealing causes an enhancement of the dot size, as indicated by the smaller energy separation between the PL peaks at higher annealing temperatures. We show below that this volume change cannot occur isotropically, but consists most probably of a stronger increase of dot height with respect to base size.

With increasing magnetic field, the emission at E_0 displays for all the samples of the series the characteristic diamagnetic shift of an excitonic ground state, and a very small Zeeman splitting of the order of 1 meV. In contrast, the excited state PL lines exhibit large splittings of up to 30 meV at 14 T depending on the circular polarization of the emitted light. In Fig. 2 we show the magnetic field dependences of the two lowest PL

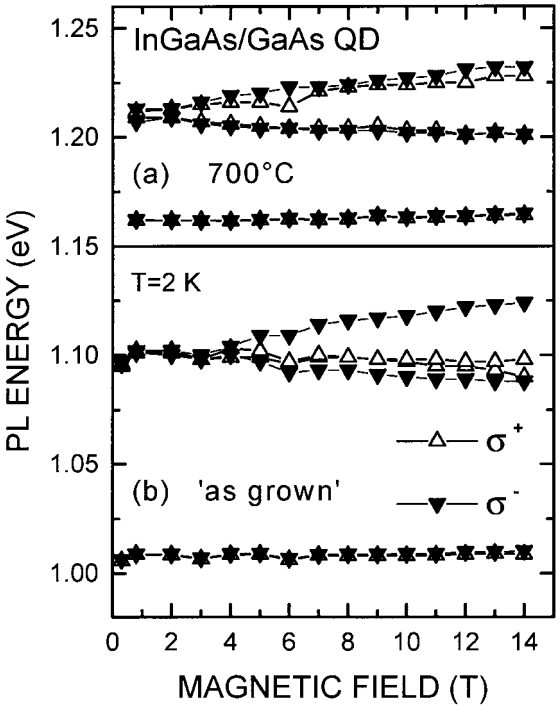

Fig. 2. Magnetic field dependence of the peak energy of the two lowest PL lines for a) the sample annealed at 700 °C and b) the as-grown sample. Solid (open) symbols correspond to σ^- (σ^+) circular polarization

peaks for the two limiting cases, i.e. for the *as-grown* and the 700 °C annealed samples. In the framework of the effective mass approximation, the energy shift of the excitonic ground state in an external magnetic field B can be written as

$$\Delta E_0 \approx \frac{e^2}{12\mu} \langle r^2 \rangle B^2 \pm g^* \mu_B B . \quad (1)$$

The quadratic term in the field corresponds to the diamagnetic shift which is proportional to the square of the average exciton radius $\langle r^2 \rangle$ and the inverse of the reduced effective mass $1/\mu = 1/m_e + 1/m_h$, where m_e and m_h are the electron and hole effective masses, respectively, calculated within $\mathbf{k} \cdot \mathbf{p}$ theory [12]. The Zeeman term is linear in B and represents the splitting between the σ^+ and σ^- circularly polarized emission lines. Its magnitude is determined by an effective Landé factor g^* for the exciton times the Bohr magneton μ_B. The quantity g^* contains both the contributions from the electron and the hole Zeeman splittings.

From the slope of a plot of E_0 versus B^2, and using Eq. (1), we obtained values for the spatial extension of the exciton wave function given by the mean diameter $2\sqrt{\langle r^2 \rangle}$. The results are shown in Fig. 3a for the five samples studied here. In agreement with the size increase of the dots derived from the PL measurements at zero field, we observe an increase by roughly a factor of two in the effective exciton diameter with annealing. For the *as-grown* sample the exciton diameter appears to be determined by the dot height, whereas for the samples annealed at the highest temperatures the exciton size seems to have reached a constant value which is in the range of the dot base size for the as-grown sample.

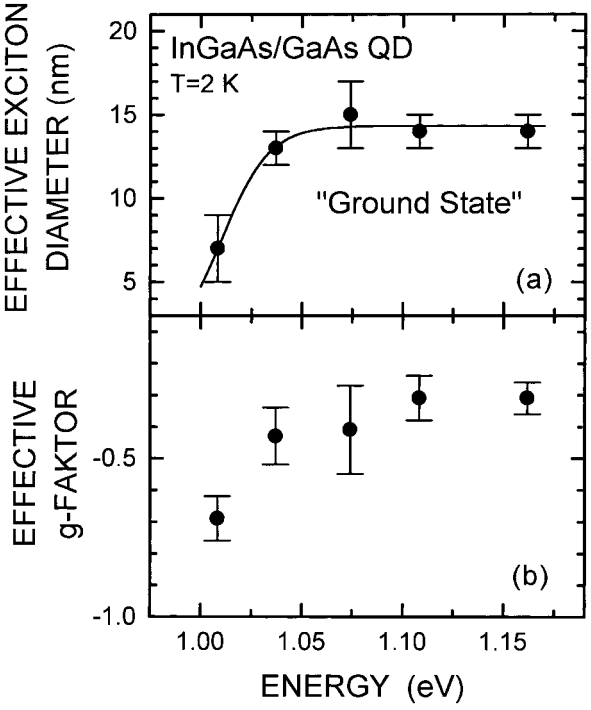

Fig. 3. a) Average exciton diameter and b) effective g^*-factor of the dots for the exciton ground state as a function of its emission energy E_0. Line is a guide to the eye

Fig. 3b shows the effective g^*-factors of the annealing series as obtained from the splitting between both circular polarization components of the ground state emission in the magnetic field (1.5 to 0.7 meV at 14 T). Similar splittings have been recently reported for the same material system from single-dot spectroscopy [13]. The g-factor of the as-grown sample is smaller than that of excitons in bulk GaAs (≈ -2), and it decreases in magnitude with annealing, i.e. with increasing Ga concentration in the dots. This tendency is in accordance with the expected behavior of the electron g-factor, which changes from -15 to -0.44 in bulk InAs and GaAs, respectively, mainly due to the increase of the electron effective mass m_e and the reduction of the ratio Δ_0/E_g between spin–orbit splitting and band gap [14]. In the case of quantum dots, the dependence of the hole g-factor on dot size and morphology might play a much more important role than in the bulk for explaining the observed changes of g^* with annealing.

Finally, we turn to the discussion of the magnetic-field induced splitting of the first excited state emission (see Fig. 2). These splittings are too large to be of atomic origin. They rather arise from the Zeeman coupling of the angular momentum associated with the excitonic envelope function in the dots and the external magnetic field [9]. This allows us to obtain important information about the electronic structure of the self-organized dots. For instance, we notice that, in contrast to the behavior of the 700 °C sample, for the as-grown one the σ^+ component of the first excited state emission does not split at all, whereas the σ^- luminescence splits into two lines separated by $\approx$30 meV at 14 T. This clearly speaks for a change in shape or symmetry of the dots. Since the overall splitting is essentially the same for all five samples, we infer that the annealing of the QDs leads mainly to an increase in dot height by Ga incorporation.

In summary, we have varied the Ga concentration and the shape of self-assembled InGaAs/GaAs QDs by annealing a given sample at different temperatures between 580 and 700 °C. From our magnetoluminescence measurements at high excitation powers, we conclude that the effect of annealing consists in the gradual incorporation of Ga into the dots, while the dot shape changes mainly by growing in height.

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