Temperature dependence of photoreflectance in InAs/GaAs quantum dots

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Temperature dependent photoreflectance (PR) and photoluminescence experiments of the InAs/GaAs quantum dot (QD) structures were performed. At 20 K, effective band-gap transitions due to the InAs QDs, wetting layers, and GaAs buffer and cap layers were identified. Transition energies of the ground state and four excited states with nearly equal interlevel spacings (75–80 meV) were observed. The linewidth of the ground-state transition decreased as the temperature increased from 20 K to 100 K while the linewidth became broader at temperatures above 100 K. Energy features of the PR spectra originating from QDs and relating to the in-plane parabolic potentials were discussed. © *2003 American Institute of Physics.* [DOI: 10.1063/1.1581003]

InAs–GaAs-based quantum dot (QD) lasers have attracted considerable interest during the last two decades.[1] QDs provide some unique physical properties, such as the delta functionlike density of state due to the three-dimensional quantum confinement. In the future, QD lasers promise to improve various features of laser performances, such as low threshold current and high characteristic temperature.[2] Although the potential advantages of QD lasers have been predicted theoretically,[2] high performance QD lasers have not been developed because of the difficulties in the fabrication process.[3] Recently, Marzin *et al.*[4] overcame some of these difficulties and fabricated QD lasers by using the Stranski–Krastanov growth method with molecular-beam epitaxy. The QDs formed in the Stranski–Krastanov phase transition are called self-organized or self-assembled dots (SADs) and have been studied extensively.[5] We realize that a long wavelength at 1.3 μm is suitable for fiber optic communications because of low loss. It has been demonstrated that InAs or InGaAs QD lasers near 1.3 μm have a continuous-wave with a low threshold current density of 19 A/cm^2 at room temperature and a single lateral-mode maximum power of 210 mW.[6] In these long-wavelength InAs–GaAs QD lasers, the deep confinement potentials lead to multiple energy levels and lasing from well-resolved higher-energy transitions. Several important effects obtained from the electroluminescence are related to the energy levels of the spatially lateral simple harmonic oscillators in the QDs.[7] Also, the temperature dependent optical gain is important because nonlinear gain can degrade the efficiency of a laser and cause level switching, thus hindering the performance of a QD laser.[8] Although the optical properties of InAs QD laser structures were widely studied by photoluminescence (PL),[3–5,9] PL excitation spectroscopy,[9–11] time-resolved PL,[12,13] and calorimetric absorption spectroscopy,[14,15] little work has been done using modulation spectroscopy.[16–18]

In this letter, we report the results of our investigation on the photoreflectance (PR) of the optical transitions in QD structures at various temperatures (20, 100, and 200 K). Signals were observed from all relevant portions of the sample including QDs, wetting layers, and GaAs layers. The PR spectra from the QDs and the wetting layers were fitted to the first derivative of a Gaussian profile to accurately determine the energies and linewidths of the observed features.[16] The signals from the GaAs layers were fitted to the third-derivative function based on the band-to-band transition.[17,18] Our PR results provide evidence of optical transitions in the ground state and the higher excited states in the QDs. We observed that the linewidth of the ground state decreased to a minimum as the temperature increased to 100 K.

The sample was grown by the gas-source molecular-beam epitaxy in the Stranski–Krastanov growth mode on an n^+-doped GaAs (100) substrate. After desorbing the oxide with hydrogen plasma at 610 °C, GaAs buffers with a thickness of 500 nm were grown. The growth temperature of GaAs was 590 °C while the InAs QDs and the InGaAs overgrowth layers were both grown at 500 °C. Two monolayers (MLs) of InAs were deposited on GaAs and then covered with an overgrowth layer which consisted of 9 MLs of $In_{0.33}Ga_{0.67}As$. The growth rate of InAs QDs was 0.085 ML/s and the V/III ratio was equal to 2. The InGaAs layers were grown using an alternating submonolayer deposition method with 0.5 ML GaAs, As_2, and 0.25 ML InAs, respectively. The schematic structure of the sample is shown in Fig. 1. The morphology of the specimen was examined with the field-emission scanning electron microscopy. The density of QDs was about $7.64\times10^{10}/cm^2$.

For the PR and the PL temperature dependent experiments, the sample was mounted in a continuous-flow He cryostat. For the PR experiments, a tungsten lamp dispersed by a 0.30 m grating monochromator served as a probe beam and a low excitation density (<0.7 mW/cm^2) light source of a 543.2 nm line He–Ne laser served as a pumping beam. The PR experiment has been described previously in literature.[18] The high power excitation of the PL measurement at 20 K

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9 ML	InGaAs
2 ML	InAs QDs
500 nm	undoped GaAs
6 nm	LT undoped GaAs
9 ML	InGaAs
2 ML	InAs QDs
500 nm	undoped GaAs
	n^+ GaAs substrate

FIG. 1. Schematic structure of InAs QDs and growth parameters.

TABLE I. Fitted results of PR spectra of InAs QD samples. The unit of energy E in eV and broadening parameter Γ in meV.

		Energy features							
		A	B	C	D	E	F	G	H
20 K	E	1.026	1.114	1.193	1.286	1.354	1.458	1.506	1.516
	Γ	31	15	18	14	12	25	9	4
100 K	E	1.018	1.084	1.170	1.252	1.337	···	1.507	1.515
	Γ	20	17	18	15	14	···	4	3
200 K	E	0.973	1.047	1.129	1.199	1.275	1.415	1.462	1.484
	Γ	24	20	23	19	13	24	3	3

was performed using an Ar^+ ion gas laser with a wavelength of 488 nm and a power density ~ 17.7 W/cm^2. The emitted photons were detected with a 0.5 m double-grating monochromator using a cooled InGaAs detector.

The upper (lower) part of Fig. 2 shows the PR (PL) spectrum of the InAs QDs at 20 K. The fitted energies of the optical transitions are marked with arrows and labeled alphabetically (A to H) from low to high energies. Five energy features (A, B, C, D, and E) were observed and the fitted energies and the broadening parameters or linewidths (Γ) were listed in Table I. Transition energy feature A located at 1.026 eV with a linewidth of 31 meV originates from the ground-state transition of the InAs QDs. Because the measured linewidth is so narrow, we believe that the quality of the samples is reasonably good and the dot size distribution must be uniform. The features marked B, C, D, and E come from the optical transitions of the excited states of the InAs QDs. The lower part of Fig. 2 displays the PL spectrum of the optical transitions of the ground state (E_0) and the first two excited states (E_1 and E_2). Similar PL results of the excited state levels were observed and reported in literature.[9–11,19]

The fitted energy feature F is 1.458 eV and is the optical transition energy from the InAs wetting layer covered with the $In_xGa_{1-x}As$ overgrowth layer (the nominal value of x is 0.33). Because the interdiffusion across the InAs/InGaAs interface occurs within a few MLs (~ 8 Å) and causes a band-gap shrinkage in InAs, the energy feature F (1.458 eV) is higher than the optical transition energy of the InAs wetting layer (~ 1.425 eV).[14] The energy features G and H are located at 1.506 eV and 1.516 eV, respectively, and originate from the fundamental band-gap transitions of the GaAs of the n^+ substrate[18] and the buffer and cap layers, respectively.

Detailed information of the PR spectra of the InAs QDs measured at temperatures of 20, 100, and 200 K are shown in Fig. 3. In order to focus our study on the fine structures of the optical transitions in the spectra, the optical energies range from 0.9 eV to 1.4 eV. The features are marked with arrows and labeled A, B, C, D, and E, as in Fig. 2. We realize that the energy band gap shrinks as the sample temperature increases.[20,21] Therefore, we see that all features (A, B, C, D, and E) show redshifts as the temperature increases as depicted in Fig. 3. Detailed discussions of the temperature de-

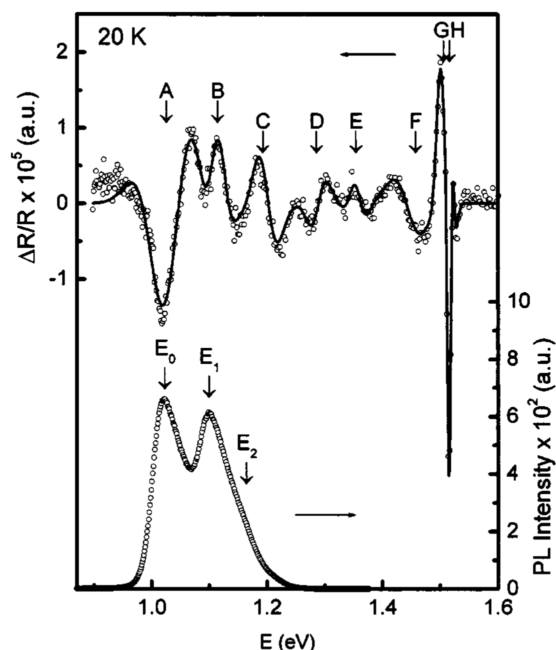

FIG. 2. PR and PL spectra of InAs QDs at 20 K.

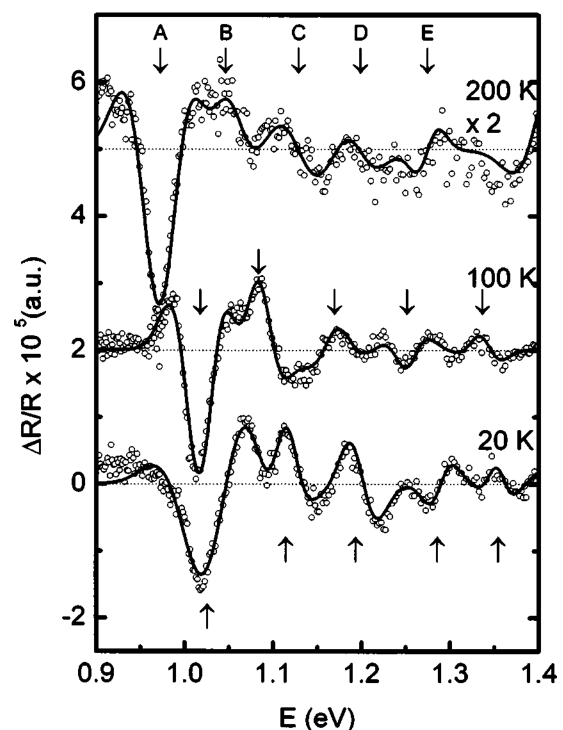

FIG. 3. PR spectra of InAs QDs at 20 K, 100 K, and 200 K.

pendent transition energies and the broadening parameters will be reported elsewhere. The line shapes of the PR spectra are fitted to the first derivative of the Gaussian function and plotted with the solid curves. The open circles in Fig. 3 represent the experimental data. The interlevel spacings between the energy features (A to E) in Fig. 3 are nearly equal (75–80 meV) and are higher than those reported in literature.[20,22,23]

The reason why nearly equal spacing in energy occurs is because spatially lateral (in-plane) simple harmonic potentials exist in the QDs as mentioned earlier[7] and reported in literature.[24,25] Using Eq. (4) from Ref. 7, we calculated the energy levels in the QDs with a lateral harmonic potential. The shape of each QD is characterized by a height and a diameter. In our calculations, we used an effective diameter of 33.0 nm, which is slightly larger than the base diameter of 30 nm measured by field-emission scanning electron microscopy. For InAs, the effective masses of the electron and the heavy hole used in our calculations were 0.03 and 0.51, respectively.[26] The interlevel spacing in energy was estimated to be 79.4 meV and is in good agreement with our experimental results. From our temperature dependent PR experiments on the InAs SADs, we observed not only the transition energies of the ground state and the excited states of a harmonic oscillator but also the unusual linewidth narrowing. The narrowest linewidth was 20 meV at a temperature near 100 K as reported in Table I. This particular phenomenon could be interpreted as the thermal activated carrier and the interaction between the intradots.[8,27–29]

In summary, we have characterized the optical properties of the InAs–GaAs QD structures by the PR and the PL spectroscopies at various temperatures. Detailed energy features from well-fitted PR spectra were analyzed. Signals from a ground state, four excited states, wetting layers, and the fundamental band gap of GaAs were identified and discussed. The linewidth decreased as temperature increased to 100 K. Energy features of the PR spectra originating from QDs and relating to the in-plane parabolic potentials of QDs were discussed.

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