Research Paper

Temperature-Dependent Optical Properties and Bandgap Characteristics of InAs/GaAs Sub-monolayer Quantum Dot Investigated by Photoreflectance Spectroscopy

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Abstract

InAs/GaAs submonolayer quantum dots (SML-QD) were investigated by temperature dependent photoreflectance (PR) spectroscopy. To investigate the optical properties of SML-QD, GaAs and InAs SML-QD related PR spectra were monitored at different temperatures. Two notable signals were observed in the SML-QD and GaAs regions. The PR spectra of SML-QD region were interpreted by the third-derivative functional form method. We observe the oscillatory signal above the GaAs band gap energy (E_g) due to the Franz-Keldysh effect caused by an interface electric field (F). At room temperature, the PR transition of SML-QD was obtained at near ~ 1.3 eV with a broadening of 29.5 meV. The F was obtained from the Aspnes' numerical PR analysis. The F was changed from 14 to 12 kV/cm by decreasing the temperature from 300 to 140 K causing a thermal induced carrier distribution near the interfaces.

Keywords: Submonolayer quantum dot, Photoreflectance, InAs/GaAs

I. Introduction

Photoreflectance (PR) spectroscopy is a powerful non-destructive optical tool for the investigation of electrical and optical properties at surfaces and interfaces of multilayer structures [1]. It is widely used for the determination of surface or interface electric fields and optical transitions energies in various semiconductor materials.

Conventionally, the Stranski-Krastanov (S-K) mode is used as a method of quantum dots formation (QDs) [2]. However, in InAs/GaAs QDs grown by S-K method, strain-related defects caused by interface strain between InAs and GaAs are unavoidable. These defects act as carrier trapping and recombination centers, resulting in the degradation of the optical properties and current transport performance [3,4]. Therefore, it is important to suppress the generation of defects and maintain the crystal quality of QDs to improve the performance of the semiconductor device. To fabricate InAs QDs with low defect density caused by strain, a submonolayer (SML) deposition method obtained through intersections of InAs and GaAs below critical

thickness (over 1.7 ML) has been recently suggested as an alternative method for the S-K growth mode [5-7].

In this study, we investigated the optical properties of SML-QD grown by molecular beam epitaxy (MBE) method through PR spectroscopy at various temperatures [6,8,9]. In section II, we briefly introduce PR theory applied to semiconductors. In Section III, we describe the InAs/GaAs SML-QD structure and the PR experimental method. In section IV, the experimental results are analyzed. Finally, the conclusions obtained from experiments and theories are summarized.

II. Theoretical models

The theoretical model begins with the definition of the modulated PR [10]

$$\frac{\Delta R}{R} = a\Delta\epsilon_1 + b\Delta\epsilon_2, \quad (1)$$

where a and b are the Seraphin coefficients, given by the indices of diffraction and absorption $n(E)$ and $\kappa(E)$, respectively, of the semiconductor. Using the Aspnes form of $\Delta\epsilon$, the change of the dielectric function ($\Delta\epsilon = \Delta\epsilon_1 + j\Delta\epsilon_2$) is expressed as [11,12]

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$$\Delta\varepsilon_1 = B \cdot \sqrt{\hbar\theta} \operatorname{Im} \left[\frac{H(\eta)}{(E - j\Gamma)^2} \right], \quad (2a)$$

$$\Delta\varepsilon_2 = B \cdot \sqrt{\hbar\theta} \operatorname{Re} \left[\frac{H(\eta)}{(E - j\Gamma)^2} \right], \quad (2b)$$

$$H(\eta) = \pi[Ai'^2 - \eta Ai'^2] + j\pi[Ai'Bi' - \eta AiBi] + j\sqrt{\eta}, \quad (2c)$$

where B is a constant related to the polarization and transition strength, $H(\eta)$ is the broaden electro-optic function, $E = \hbar\omega$ is the photon energy, Γ is a broadening parameter at the band-gap energy, and $\eta = \frac{(E_{cp} - E + j\Gamma)}{\hbar\theta}$, where E_{cp} is the transition energy. Ai and Bi are Airy functions of the first and second kinds, respectively. Ai' and Bi' are differential forms of Ai and Bi , respectively. The electro-optic energy $\hbar\theta$ with a constant electric field (F) is defined as

$$\hbar\theta = \left(\frac{e\hbar F}{\sqrt{2}\mu} \right)^{2/3}, \quad (3)$$

where e is electron charge and μ is the interband reduced effective mass. Except for impurity contributions and Coulombic effects (i.e., excitons), the PR signal can be simulated using Eq. (1), including Eqs. (2) and (3) for a single bulk layer with a uniform electric field.

Furthermore, transitions involving degenerate valence bands should be considered separately by replacing μ by μ_{lh} and μ_{hh} because of transitions including the light and heavy hole. This creates nesting in the PR spectra as

$$\frac{\Delta R}{R} = A_{hh} \left(\frac{\Delta R}{R} \right)_{hh} + A_{lh} \left(\frac{\Delta R}{R} \right)_{lh}, \quad (4)$$

where A_{hh} and A_{lh} are relative amplitudes of the heavy

Figure 1. Theoretical PR spectra of n-GaAs at room temperature using Eq. (4). $F = 10$ kV/cm, $\Gamma = 3$ meV, and the relative contribution of heavy hole and light hole is used as $A_{hh}/A_{lh} = 1.336$.

and light hole transitions.

Figure 1 shows the theoretical intermediate-field PR spectrum of n-GaAs. It should be emphasized here, that the light and the heavy holes contributions in the Eq. (4) are able to produce a beat in the damping region of the Franz-Keldysh oscillation (FKO).

In a small electric field region, Aspnes defined the PR signal as a third derivative functional form (TDFF) as follows [13]

$$\frac{\Delta R}{R} = C \operatorname{Re} [\exp(i\varphi) (E - E_{cp} + i\Gamma_{TD})^{-n}], \quad (5)$$

where C is an amplitude parameter, E_{cp} is the critical point energy, meaning the band gap energy, φ is the phase parameter, and $n = 2, 2.5$, and 3 represent the exciton, the three-dimensional band-to-band transition, and the two-dimensional (2D) band-to-band transition, respectively. In this study, therefore, 2D and 3D TDFF (i.e., $n = 2.5$ and 3) are not used to represent the PR spectra of the InAs QD, but $n = 2$ TDFF was used, which can analyze the energy states of the medium.

III. Experimental details

Figure 2 shows a schematic structure of the InAs/GaAs SML-QD samples used in this study. Samples were grown on a semi-insulating (SI) GaAs (100) substrate by a solid MBE source system. An undoped-GaAs buffer layer of thickness 400 nm was grown on the SI-GaAs substrate. To form a SML-QD, a 0.5 ML thick InAs layer was grown on the GaAs buffer layer and a 2.5 ML thick GaAs layer was employed to cap the InAs layer; this sequence was repeated five times to form a SML-QD. After formation of the SML-QD, a 10 nm-thick GaAs layer was used as a spacer layer. Eight periods of SML-QD layers were formed, each of which consists of five cycles of InAs (0.5 ML)/GaAs (2.5 ML) and is covered with a 10 nm-thick GaAs layer.

For the PR experiments, a laser diode (637 nm) was used as an excitation light source. The excitation intensity (50 and 400 mW/cm², at 800 Hz) was controlled using a

Figure 2. Schematic structure of InAs/GaAs SML-QDs.

Figure 3. Temperature -dependent PR spectra of InAs/GaAs SML-QD. Blue and red holes represent transition energy of SML-QD and E_g of GaAs epilayer.

neutral density filter. The probe light was a monochromatic beam, obtained from a tungsten-halogen lamp dispersed through a monochromator. The probe beam was incident on the sample surface, and the reflected beam was measured using a Si detector. A closed-cycle He refrigerator was used to control the sample's temperature.

IV. Results and analysis

Figure 3 shows the InAs/GaAs SML-QD PR signal at different temperatures. At low temperatures, the signal is larger than GaAs, but at 300 K it is lower than GaAs. This phenomenon can be attributed to the fact that, as the temperature increases, the carriers confined in the InAs SML-QD have enough thermal energy to move to the GaAs conduction band.

Figure 4. Experimental and theoretical PR spectra of InAs/GaAs SML-QD at (a) 300 and (b) 140 K. Low energy interference oscillation (LEIO) is a signal that appears as the interference of the PR signal inside and on the surface of the medium.

In the regions below the transition energy of the SML-QD and the E_g of the GaAs, the PR signals are observed from 100 and 60 to 300 K, respectively. These signals are caused by interference between the beam reflected from the sample surface and the internal PR signal. In general, interference is affected by thickness, refractive index, and transition energy. The transition energies and refractive indices of InAs SML-QD and GaAs were changed by temperature. Figure 3, therefore, shows that the coherence length varies with temperature.

Figure 4 (a) shows the PR spectrum of InAs/GaAs SML-QD measured at 300 K. The observed PR spectrum can be fitted with Eqs. (4) and (5). For signals related to SML-QD at 300 K, the fitting parameters of the TDFF gave $n = 2$, $E_{CP} = 1.333$ eV, and $\Gamma_{TD} = 10.17$ meV. These are lower than in the previously reported S-K method of InAs/GaAs

Table 1. Theoretical parameters (Eqs. (4) and (5)) used to fit the experimental PR signal (Fig. 4). The m_{hh} and m_{lh} are the effective mass of heavy and light holes.

	T [K]	C	ϕ [rad]	E_{cp} [eV]	Γ_{TD} [meV]	n
InAs SML-QDs	300	4.11×10^{-10}	5.044	1.333		2
	140	4.88×10^{-10}	4.468	1.393		2
	T [K]	A_{hh}/A_{lh}	F [kV/cm]	E_g [eV]	Γ [meV]	m_{hh}/m_{lh}
GaAs	300	1.55	14	1.409	3	0.45/0.087
	140	1.375	12	1.465	3	0.45/0.087

QDs growth. The signals related to GaAs were fitted to $E_g = 1.409$ eV and $\Gamma = 3$ meV.

Figure 4(b) shows the InAs SML-QDs signal measured at 140 K and the result of fitting with TDFF of $n = 2$. The fitting result shows two transition signals (QD1 and QD2) of the InAs SML-QDs. The QD1 (1.393 eV), which was observed at 300 K, is the ground state of the InAs SML-QDs and the QD2 (1.423 eV), which was observed at low temperature, is the excited state of the InAs SML-QDs. This energy implies that two energy bands are formed in the conduction band of InAs SML-QD. At 300 K, QD2 is difficult to observe in the PR signal because the electrons in the excited state have enough thermal energy to easily migrate to the conduction band of the GaAs. Table 1 shows the material parameters used to fit SML-QD and GaAs PR signals.

V. Conclusions

Temperature-dependent PR measurements of the InAs/GaAs SML-QD were analyzed using theoretical PR equations in TDFF. The LEIO signals of the InAs SML-QD and GaAs were observed below the transition energies. The TDFF fitting results at 300 K show one transition signal in the InAs SML-QD QDs, while two transition signals were obtained at 140 K. These two transitions are attributed to the ground (1.393 eV) and excited (1.423 eV) states of the InAs SML-QD QDs. Furthermore, the signal of GaAs is well explained with the theoretical value of temperature. At high temperature, it was difficult to observe the signal of the QD2 because the carriers in the

excited state have enough thermal energy to easily migrate to the conduction band of the GaAs.

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References

- [1] H. Shen and M. Dutta, *J. Appl. Phys.* 78, 2151 (1995).
- [2] P. B. Joyce, T. J. Krzyzewski, G. R. Bell, B. A. Joyce, and T. S. Jones, *Phys. Rev. B* 58, R15981(R) (1998).
- [3] A. Martí, E. Antolín, E. Cánovas, N. López, and A. Luque, *Proceedings of the 21st European Photovoltaic Solar Energy Conference*, (Dresden, Germany, Sept. 4-8, 2006), pp. 99-102.
- [4] A. Nasr, *Opt. Laser Technol.* 48, 135 (2013).
- [5] S. Krishna, D. Zhu, J. Xu, K. K. Linder, O. Qasimeh, P. Bhattacharya, and D. L. Huffaker, *J. Appl. Phys.* 86, 6135 (1999).
- [6] Z. Xu, D. Birkedal, J. M. Hvam, Z. Zhao, Y. Liu, K. Yang, A. Kanjilal, and J. Sadowski, *Appl. Phys. Lett.* 82, 3859 (2003).
- [7] T. D. Germann, A. Strittmatter, J. Pohl, U. W. Pohl, D. Bimberg, J. Rautiainen, M. Guina, and O. G. Okhotnikov, *Appl. Phys. Lett.* 92, 101123 (2008).
- [8] I. L. Kresnikov, N. N. Ledentsov, A. Hoffmann, and D. Bimberg, *Phys. Stat. Sol. (a)* 183, 207 (2001).
- [9] N. N. Ledentsov and D. Bimberg, *J. Crystal Growth* 255, 68 (2003).
- [10] B. O. Seraphin and N. Bottka, *Phys. Rev.* 145, 628 (1966).
- [11] D. E. Aspnes, *Phys. Rev.* 153, 972 (1967).
- [12] C. Huber, C. Krammer, D. Sperber, A. Magin, H. Kalt, and M. Hetterich, *Phys. Rev. B* 92, 75201 (2015).
- [13] D. E. Aspnes, *Surf. Sci.* 37, 418 (2003).