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1997 J. Phys.: Condens. Matter 9 L13

(http://iopscience.iop.org/0953-8984/9/1/003)

J. Phys.: Condens. Matter **9** (1997) L13–L17. Printed in the UK PII: S0953-8984(97)77871-8

LETTER TO THE EDITOR

Photoluminescence study of spin–orbit-split bound electron states in self-assembled InAs and $In_{0.5}Ga_{0.5}As$ quantum dots

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Received 17 September 1996

Abstract. The photoluminescence of electron excitations was measured in InAs and $In_xGa_{1-x}As$ self-assembled quantum dot systems. For the first time spin–orbit-split excitons were observed in resonant photoluminescence when the excitation energy was close to the exciton energy; off-resonant photoluminescence spectra reveal heavy-hole excitons.

Self-assembled semiconductor quantum dot (QD) structures are formed during heteroepitaxy of strongly mismatched compounds such as InGaAs/GaAs, AlInAs/GaAs and InSb/GaSb [1–3]. One of the greatest advantages of these systems is the possibility of growth of high-quality (defect-free) QDs. Self-assembled QDs have been widely studied by atomic force microscopy, transmission electron microscopy and scanning tunneling microscopy [1, 4, 5]. Photoluminescence (PL) was frequently used to characterize electron excitations in these QDs. Up to now only electron transitions between the confined states of the conduction band and the heavy-hole valence band have been detected. However, the perfect structure of self-assembled QDs giving rise to the lifetimes of electron and hole states created by light makes it possible to use PL to study the high-energy electron excitations, such as those involving the spin–orbit-split electron states, which until now have had to be studied in different semiconductors by reflectance or electroreflectance [6].

In this paper we apply the PL spectroscopy to investigate the spin–orbit-split electron states confined in $In_xGa_{1-x}As$ QDs embedded in GaAs. The QDs were obtained by Stranski–Krastanow growth with molecular-beam epitaxy of the strained $In_xGa_{1-x}As$/GaAs heterostructures with $x = 1.0$ and $x = 0.5$ on (111)-oriented GaAs substrates; six and 15 monolayers of InAs and $In_{0.5}Ga_{0.5}As$ respectively were deposited in order to form QDs. The total structure contained 20 periods of the GaAs/AlAs (2 nm/2 nm) superlattice, 0.5 μm of the GaAs buffer layer and 3 nm of the $In_{0.2}Ga_{0.8}As$ reference quantum well (QW). The QDs were separated from the QW by 100 nm of the GaAs barrier. A 50 nm thick GaAs cap layer was grown at the top of the structure. The substrate temperature was 600 °C during the superlattice growth and 500 °C (for the InAs QDs) and 520 °C (for the $In_{0.5}Ga_{0.5}As$ QDs) when the other layers were grown. The interface smoothness was introduced by growth interruptions during InAs deposition at both interfaces. The QDs nucleation was directly seen by reflection high-energy electron diffraction as used in [1].

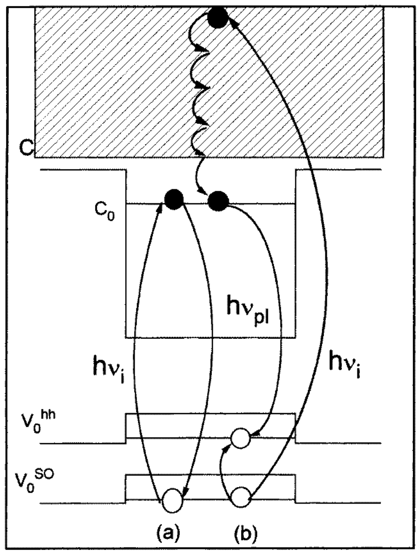

Figure 1. The scheme of the electron transitions involved in a quantum dot at resonant excitation (a) and at off-resonant excitation (b).

The PL measurements were performed at $T = 10$ K and at $T = 22$ K. The 6520 Å line of a DCM DYE laser pumped with an Ar^+ ion laser was used for excitation close to the resonance with electron excitations between the spin–orbit-split bound electron states and those bound in the conduction band of a QD, while the 5145 Å line of an Ar^+ ion laser was used for excitation out of resonance.

In order to detect the PL involving the spin–orbit-split electron states we used the excitation energy close to the energy gap between the spin–orbit-split valence band and the conduction one. The scheme of electron transitions in a QD participating in PL is shown in figure 1. The process (a) corresponds to the resonance conditions which allowed us to shorten the thermalization time of excited electrons in the conduction band (C); thus, during the relaxation of electrons to the conduction band bound state (C_0), holes remaining in the spin–orbit-split valence band (V_0^{so}) would not have time enough to scatter in the top heavy-hole valence band state (V_0^{hh}) and as a result they would recombine with electrons giving rise to PL. In the case of the off-resonant excitation (process (b)) both electrons and holes will lose their energies due to the thermalization and recombination between the final C_0 and V_0^{hh} states will occur.

The PL spectra of the InAs and $In_{0.5}Ga_{0.5}As$ QDs measured with the excitation energy of 1.801 eV (6898 Å) are depicted in the right panel of figure 2; they reveal PL lines at 1.72 eV and at 1.65 eV (denoted as E_{so} in the spectra), which we assign to the recombination of electrons in conduction band confined states with holes in the spin–orbit-split valence band bound states. The left panel of figure 2 shows the off-resonant PL spectra of the same samples measured with the excitation energy of 2.41 eV (5145 Å) which present the PL lines originating from the recombination of an electron in the conduction band confined state with a hole in the heavy-hole valence band confined state (shown as E_{hh}).

In addition, both close-to-resonance and off-resonance PL spectra of the GaAs/InGaAs superlattice, which consisted of 40 periods of isolated ($W_b = 10$ nm) quantum wells with the same thicknesses (3 nm) and compositions ($X_{In} = 0.2$) as a reference quantum well,

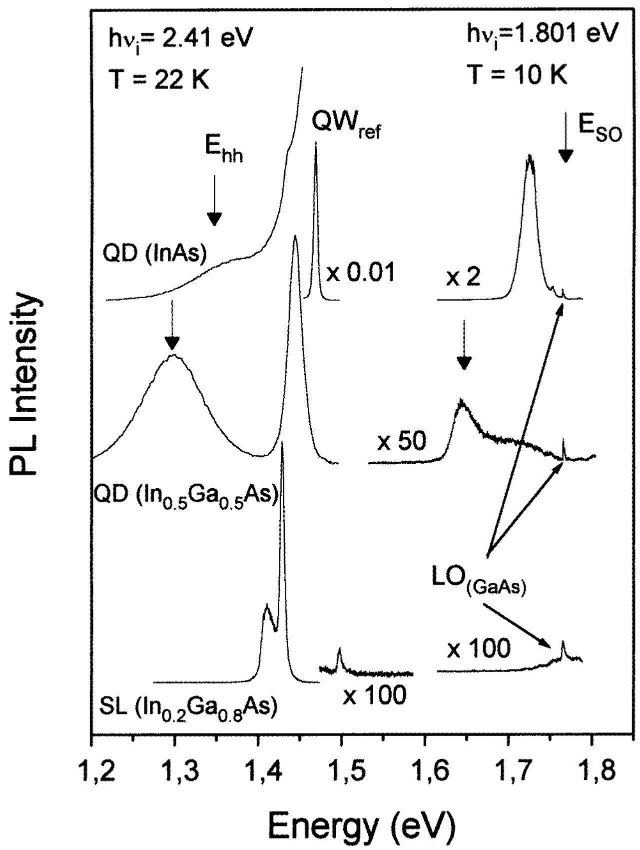

Figure 2. The PL spectra of the InAs and $In_{0.5}Ga_{0.5}As$ quantum dots together with the spectrum of the GaAs/InGaAs superlattice measured with the excitation energy of 2.41 eV (off-resonance) and 1.801 eV (close to resonance).

are shown in figure 2. These PL spectra reveal the heavy-hole exciton line around 1.43 eV (the split low-energy line is probably due to defects) and the Raman line caused by the LO phonon of GaAs at 1.76 eV (the LO Raman lines were found even in the spectra of samples with quantum dots). No line was observed in the spectral range where the spin–orbit-split excitons contributed in QDs. This proves that the line which we assign to the spin–orbit-split exciton indeed originates from the quantum dots.

The E_{so} lines were found even in the off-resonant PL spectra, although with much weaker intensities (about 400 times less) than in the resonant spectra. Red shifts of both heavy-hole and spin–orbit-split PL peak energies were observed in the $In_{0.5}Ga_{0.5}As$ QDs relative to the InAs QDs which are caused by the increase of the QDs size in the alloy.

The narrow PL lines observed around 1.45 eV are due to the reference quantum wells.

In order to determine the electron recombination energies in the QDs here studied we calculated the eigenvalues of the bound ground states in a three-dimensional square well potential of finite depth (V_0) with the radius R_0. The effect of a strain was not included in the calculations because of its complexity; as shown in [7], the strain in the InAs pyramidal quantum dots is strongly inhomogeneous giving components of different sign. Thus, we could expect even a compensation of different components of a strain. In such a case, attempts to simplify a problem by taking into account only constant strain will be far from reality. Following [8] we found the solutions of the radial Schrödinger equation for the ground state:

$$\frac{\mathrm{d}^2\Psi_0(r)}{\mathrm{d}r^2} + k^2\Psi_0(r) = 0 \tag{1}$$

inside the QD ($r < R_0$) and

$$\frac{\mathrm{d}^2\Psi_0(r)}{\mathrm{d}r^2} - \chi^2\Psi_0(r) = 0 \tag{2}$$

outside the QD ($r > R_0$) in the form of a spherical Bessel function

$$\Psi_0(r) = Aj_0(kr) \qquad r < R_0 \tag{3}$$

and in the form of a spherical Hankel function of imaginary argument

$$\Psi_0(r) = Bh_0(\mathrm{i}\chi r) \qquad r > R_0 \tag{4}$$

where $k^2 = k_0^2 - (2m/\hbar^2)|E|(1+|E|/E_g)$, with $k_0^2 = (2m/\hbar^2)V_0$, and $\chi^2 = (2m/\hbar^2)|E|(1+|E|/E_g)$. Here the effect of nonparabolicity was included in the dispersion relation following the Kane theory [9].

The matching of the logarithmic derivatives $(1/\Psi_0)\mathrm{d}\Psi_0/\mathrm{d}r$ at the interfaces ($r = R_0$) allows us to eliminate the A and B constants and as a result to obtain the eigenvalue relation

$$\tan(kR_0) = -\frac{x}{\sqrt{1-x^2}} \tag{5}$$

where $x = k/k_0$.

The observation of the PL due both to the heavy-hole and to the spin–orbit-split bound states enabled us to determine very accurately the effective radius of the QD because the fitting of the calculated energies to the experimental ones was restricted by use of only the single parameter R_0 for both cases.

The results of numerical calculations together with the experimental data are presented in table 1. The parameters of InAs and GaAs used in calculations were taken from [6] and they are listed in table 2. Linear interpolation was used to obtain the effective masses and the spin–orbit splitting for the alloy. The value of the gap in the alloy was taken from [6]. The offset ratio (Q_e) of 0.85 was used for both InAs and $In_{0.5}Ga_{0.5}As$ QDs.

Table 1. The calculated and measured recombination energies (in eV) from the conduction band bound ground state to the heavy-hole valence band bound ground state (E_{hh}) and to the spin–orbit-split valence band bound ground state (E_{so}) respectively in the InAs and in the InGaAs quantum dots of radius R_0.

QD	$E_{hh}^{\mathrm{exper.}}$	E_{hh}	$E_{so}^{\mathrm{exper.}}$	E_{so}	R_0 (Å)
InAs	1.37	1.347	1.72	1.768	27
$In_{0.5}Ga_{0.5}As$	1.3	1.297	1.65	1.647	34

A rather good correspondence was obtained between the measured and the calculated recombination energies with the parameters relating to the unstrained $In_{0.5}Ga_{0.5}As$ QDs, while a slightly higher-energy position of the spin–orbit-split PL peak was obtained for the InAs QDs with the parameters of unstrained bulk InAs. In the case of the InAs QDs the fitting could be improved by introducing the strain. Thus, no strong influence of strain in the electron recombination energies was detected in the self-assembled $In_{0.5}Ga_{0.5}As$ QDs

Table 2. The parameters of GaAs and InAs used in calculations.

	m_e^*/m_0	m_{hh}^*/m_0	m_{so}^*/m_0	E_g (eV)	Δ_0 (eV)
GaAs	0.068	0.475	0.17	1.52	0.33
InAs	0.023	0.33	0.14	0.42	0.38

embedded in GaAs, while the InAs QDs reveal a shift of the PL peaks which can be attributed to the effect of strain.

Financial support from CNPq, FAPESP and CAPES is gratefully acknowledged.

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