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Γ -X mixing in type-II GaAs/AlAs short period superlattices

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Abstract: We have measured the absolute absorption coefficient of optical transitions in type-II short period GaAs/AlAs superlattices, on a broad spectral range, at low temperature. The transmission experiments have been performed in a waveguiding configuration. Photoluminescence excitation experiments show as well the characteristics of the pseudodirect HH1-X_z excitonic transition. Theoretical calculations of the effective dielectric tensor and absorption coefficient in the vicinity of the exciton resonance frequency are presented, taking explicitly into account the Γ -X mixing of electronic states at heteroboundaries. From comparison between the experimental and theoretical values of the absorption coefficients, we have deduced a value of the Γ -X coupling coefficient for the studied superlattices.

1. INTRODUCTION

In short period GaAs/AlAs superlattices (SL), a transition from type-I to type-II occurs, when the GaAs layer width is smaller than 35 Å and the mean Al concentration is greater than 36 % [1]. The carriers are then spatially separated, electrons occupying AlAs X-states, while holes remain confined in GaAs Γ -wells. For AlAs layer thickness less than 55 Å and GaAs layer thickness larger than 15 Å, the lowest confined electron-state is an X_z state [1, 2].

The SL translational symmetry allows mixing between the X_z-valley states and the states lying at or near the Γ conduction band edge. The Γ -X_z mixing is the origin of the weakly allowed HH (or LH)-X_z optical transitions in type-II SLs. However, their oscillator strength is expected to be reduced as compared to type-I transitions because of the carriers spatial separation. The amount of Γ -X mixing has been evaluated with a disparity of values by several groups [3, 4, 5, 6]. Some of the authors [3, 5] proposed to consider as a mechanism responsible for mixing not only the superperiodicity of the ideal SL but also the random potential due to the thickness fluctuations at the interfaces.

In this paper, we present absolute absorption experiments carried out on optical planar waveguides formed by type-II GaAs/AlAs short period SLs, at low temperature. The absolute absorption coefficient was determined by a study of the luminescence reabsorption [7, 8]. A complete theoretical calculation is performed within the generalized envelope function approximation proposed by Ando and Akera [9] and corrected by Aleiner and Ivchenko [10].

2. OPTICAL WAVEGUIDING EXPERIMENTS

The advantages of the waveguiding geometry have been discussed in details in [8]. It is worth noticing that we observe the transmission, on a broad spectral range, in both polarization directions, parallel and normal to the plane of layers and that this geometry allows us to measure absorption coefficients varying from a few cm⁻¹ to several tenths of thousands of cm⁻¹ in the case of a single quantum well [7, 8].

2.1 Samples

The samples used in the absorption experiments are optical planar waveguides made of GaAs/AlAs short period SLs grown by MBE at 630° C on a (001) oriented GaAs substrate and characterized by X-ray diffraction. Sample 1 is formed by 2.148 μm of a (6/4) monolayers GaAs/AlAs short period SL (labelled SL1), grown on a GaAs substrate. A 200 Å GaAs cap layer is grown on top of the sample. This sample is a leaky guide since the refractive index of the GaAs substrate is the highest. The losses in the substrate are of the order of 50 cm^{-1} for the fundamental mode [8]. The guide is designed to be quasi-monomode.

Sample 2, grown on a GaAs substrate, is formed by 1 μm of a (2/15) monolayers GaAs/AlAs SL, 0.4 μm of a (4/9) monolayers GaAs/AlAs SL (labelled SL2), 0.5 μm of a (2/15) monolayers GaAs/AlAs SL. A 200 Å GaAs capping layer ends the growth sequence. Sample 2 is designed in such a way that the optical mode propagating along the structure is localized in the SL of interest (SL2). Therefore, this sample is a guide with very weak losses and the absorption experiment gives directly the absorption of the SL itself. It is also a monomode structure. SL1 and SL2 are both type-II SLs. SL1 is close to the type-I to type-II transition [1]. The lowest confined electron-state is an X_z electron-state in AlAs, lying 30 to 40 meV below the Γ electron-state in GaAs. SL2 is also in the X_z region. The energy difference between the X_z and Γ electron states is ~ 350 meV. One expects that the absorption coefficient in SL2 is much weaker than in SL1.

2.2 Experimental setup

The experimental setup has been described in details in [8]. The transmission experiments are done with a quartz halogen lamp at low temperature. The coupling between light and sample is obtained by focusing the incident beam on the sample with a microscope objective of great aperture. The transmitted light is collected by a mirror-objective which has also a great aperture. The transmitted intensity is detected through a spectrometer by a photomultiplier, followed by a lock-in amplifier.

The PL experiments are performed at 4 K with the 514.5 nm line of a cw Ar^+ laser and the PLE spectra are recorded using a tunable Ar^+ -pumped-dye-laser (Rhodamine-6G).

2.3 Experimental results

Fig.1 shows at low temperature the absolute absorption of SL1 in the polarization perpendicular to the growth axis z , the PL and the PLE spectra. One sees clearly the onset of absorption at 1.954 eV followed by a plateau attributed to the excitonic transition $\text{HH1-}X_z$, involving electrons in X_z states in AlAs layers and heavy-holes at Γ -point in GaAs layers. Let us notice that no excitonic peak is clearly resolved. The steep rise of absorption at higher energy is attributed to the direct $\text{HH1-}\Gamma$ transition. The absorption coefficient of the $\text{HH1-}X_z$ excitonic transition on the plateau is measured to be equal to 360 cm^{-1} . This value is by two orders of magnitude weaker than the absorption coefficient in a type-I SL with the same period. A tail of absorption remains on the low-energy side of the absorption curve and is likely due to localized exciton states. The PLE spectrum shows the same features as the absorption experiment.

The PL consists of a main zero-phonon line attributed to the recombination of the X_z -HH1 localized excitons, shifted by 11 meV from the absorption line. Two phonon replicas appear at lower energy, at 34 meV and 48 meV with respect to the zero-phonon line.

In Fig.2 are presented the absorption spectra of SL2 in the parallel and perpendicular polarizations and the PL spectrum. The PL curve shows like in SL1, a main zero-phonon line at 1.989 eV assigned to type-II excitons. On the low energy side of the main line, appear two phonon replicas of the transition, at 1.964 and 1.942 eV. The PL zero-phonon line is Stokes shifted from the absorption peak by about 20 meV. This shift is explained, as in the case of SL1, by the localization of excitons.

The absorption spectrum in the perpendicular polarization shows a "hump" at 2.01 eV, attributed to the $\text{HH1-}X_z$ excitonic transition of SL2 (Fig.2). The absorption coefficient is very weak, a few cm^{-1} . On the high energy side of the $\text{HH1-}X_z$ transition we observe a quasi-linear variation of the absorption versus energy which extrapolates roughly at 30 meV higher than the exciton peak. This absorption is attributed to indirect absorption processes with phonon emission. This indirect absorption is expected indeed to be a linear function of energy since the 2D-density of states is a constant. The phonons involved in the process are difficult to identify and several can participate to the absorption. Let us note that in the parallel polarization, we observe a linear variation of the absorption versus energy, shifted by 30 meV from the linear curve in the parallel polarization. Since the X_{xy} states lie at 20-30 meV above the X_z states, we attribute this absorption to the phonon assisted $\text{HH1-}X_{xy}$ transitions. This unexpected polarization selection rule can be explained if we assume that the electron-phonon interaction connects the

valence band extrema at $\mathbf{k}=0$ and at the zone boundary. In this case, the optical transition should occur at the zone boundary with modified selection rules.

Fig.1. (a) Absorption in the $E_{\perp z}$ polarization, (b) PLE and (c) PL spectra of SL1 at low temperature. The detection energy was set at 1.894 eV and the scanned energy range was between 1.989 and 2.17 eV.

Fig.2. (a) Absorption in the $E_{\perp z}$ polarization, (b) in the $E_{\parallel z}$ polarization and (c) PL spectra of SL2 at low temperature.

3.THEORY AND DISCUSSION

In the approximation of an optically-homogeneous effective medium [11], the absorption coefficient $\alpha_{\perp}$ in the $E_{\perp z}$ polarization, can be represented near the HH1- X_z exciton resonance frequency, ω_o , as

$$\alpha_{\perp}(\omega) = \frac{\omega}{c} \sqrt{\epsilon_b} \frac{\omega_{LT} \Gamma}{(\omega_o - \omega)^2 + \Gamma^2} \quad (1)$$

Here, Γ is the exciton damping considered as a phenomenological parameter, ϵ_b is the background dielectric constant, ω_{LT} is the exciton longitudinal-transverse splitting (or oscillator strength) given by

$$\omega_{LT} = \frac{8e^2 p_{cv}^2}{\hbar \omega_o^2 m_o^2 \epsilon_b d a_{\perp}^2} \left(\int C_e(z) \phi_h(z) dz \right)^2, \quad (2)$$

where p_{cv} is the interband matrix element $\langle S | p_x | X \rangle$, d is the SL period, a trial envelope function describing the relative electron-hole motion is chosen in the exponential form $(2/(\pi a_{\perp}^2))^{1/2} \exp(-\rho/a_{\perp})$, ρ being the in-plane electron-hole distance and $a_{\perp}$ being the effective Bohr radius. We also use in Eq. (2) the following notations: $\phi_h(z)$ is the envelope of the heavy-hole wave function, $C_e(z)$ is the envelope of the Γ state contribution to the wave function of an electron in the lowest subband $e1X_z$. Here we assume the indirect optical transition HH1- X_z to be allowed due to the Γ - X_z mixing at the heteroboundaries. In the generalized effective mass approximation [9, 10] (see also [12]), $C_e(z)$ is determined from the continuity requirement at the interface and the second boundary condition that relates the derivative $\partial C_e / \partial z$ to an interface value of the X_z envelope function. The dimensionless constant $\tau_{\Gamma X}$ of the Γ - X mixing is considered as a free parameter which should be estimated from comparison between the theory and the measured absorption spectra. The calculations of ω_{LT} are made (i) in the 2D limit where $a_{\perp}$ is taken as the 2D Bohr radius and (ii) in the simple quasi-2D exciton model where $a_{\perp}$ is found using the variational procedure with a one parameter trial function. Taking $\tau_{\Gamma X} = 0.1$ and $\sqrt{\epsilon_b} = 3.6$ for both SLs, $\Gamma = 5$ and 10 meV for SL1 and SL2 respectively, we find in the 2D limit the following results: $\omega_{LT} = 8 \text{ ns}^{-1}$, $\alpha_m \equiv \alpha_{\perp}(\omega_o) = 365 \text{ cm}^{-1}$ and $\omega_{LT} = 12 \text{ ns}^{-1}$, $\alpha_m = 565 \text{ cm}^{-1}$ for SL1 with (6/4) and (7/4) monolayers respectively. This is in agreement with the experimental value of $\alpha_m = 360 \text{ cm}^{-1}$ obtained for SL1. For the same value of $\tau_{\Gamma X}$, we also obtain a good agreement for SL2. The theoretical values are $\omega_{LT} =$

0.2 ns^{-1} , $\alpha_m = 4.25 \text{ cm}^{-1}$ and $\omega_{LT} = 0.07 \text{ ns}^{-1}$, $\alpha_m = 1.75 \text{ cm}^{-1}$ for (4/9) and (4/10) monolayers respectively, while the experimental absorption coefficient is of the order of a few cm^{-1} . In the quasi-2D model, one can fit the data with a larger value, $t_{rx} \sim 0.2$. Thus, the real value of t_{rx} lies between 0.1 and 0.2.

In well defined type-II $(\text{GaAs})_N(\text{AlAs})_M$ SLs, the resonant absorption coefficients are independent on the parity of monolayer numbers N, M . However, the theory predicts a noticeable M -parity dependence for SLs near the type-I to type-II transition.

We also measured the photoluminescence decay in the two SLs. If we assume that some delay after the excitation pulse, the decay is mainly due to the radiative recombination, then the determination of both the absorption coefficient and the radiative lifetime, allows us to estimate the excitonic localization radius as a few tens Å for the two studied SLs.

4. CONCLUSION

We have measured for the first time the absolute value of the absorption coefficient in type-II GaAs/AlAs SLs in an optical waveguiding experiment. We have shown that the mechanism of Γ -X mixing of electronic states by perfect interfaces can be successfully used for the description of the measured resonance absorption spectra and we have determined the value of the Γ -X coupling coefficient for the studied SLs.

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