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Detection of lateral composition modulation by magnetoexciton spectroscopy

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Abstract

An experimental signature for detecting spontaneous lateral composition modulation in a $(InAs)_n/(GaAs)_m$ short period superlattice on a InP substrate based on magnetoexciton spectroscopy is described. We find by aligning the magnetic field in three crystallographic directions, one parallel to and the other two perpendicular to the composition modulation direction, that the magnetoexciton shifts are anisotropic and are a good indicator for the presence of composition modulation.

Keywords: Composition modulation; Photoluminescence; Magnetoexciton; Quantum wires

Self-assembled structures that are arranged in quantum-size configurations are used to obtain high densities of quantum wires and dots. Spontaneous lateral composition modulation (CM) resulting from the deposition of short period superlattices (SPS) have produced quantum-wire like structures [1–6]. Recently, a review for this phenomenon was presented by Mirecki Millunchick et al. [7]. This type of lateral CM in short period superlattices occurs for a range of material systems such as $(InP)_m/(GaP)_n$ on GaAs [1, 2], $(InAs)_m/(GaAs)_n$ on InP [3, 4], and $(AlAs)_m/(InAs)_n$ [5, 6]. Here m and n are the number of monolayers (ML) of each binary compound deposited. For these materials and growth conditions, the strong lateral CM wave is observed long the [1 1 0] direction.

The principal experimental verifications for lateral CM have been transmission electron microscopy (TEM) [1–6], optical absorption [8], and polarized photoluminescence (PL) techniques [9]. Recently, X-ray reciprocal space analysis has also proven to be a valuable diagnostic tool for the detection and measurement of CM [6]. In this paper we report a new diagnostic tool using an old technique, magnetoexciton spectroscopy, for the detection of spontaneous lateral CM. Here, magnetoexciton shifts are studied for three orientations of the magnetic field with respect to the direction of the CM, giving a relatively fast and unambiguous determination for the presence of composition modulation. The structure used to demonstrate the utility of magnetoexciton

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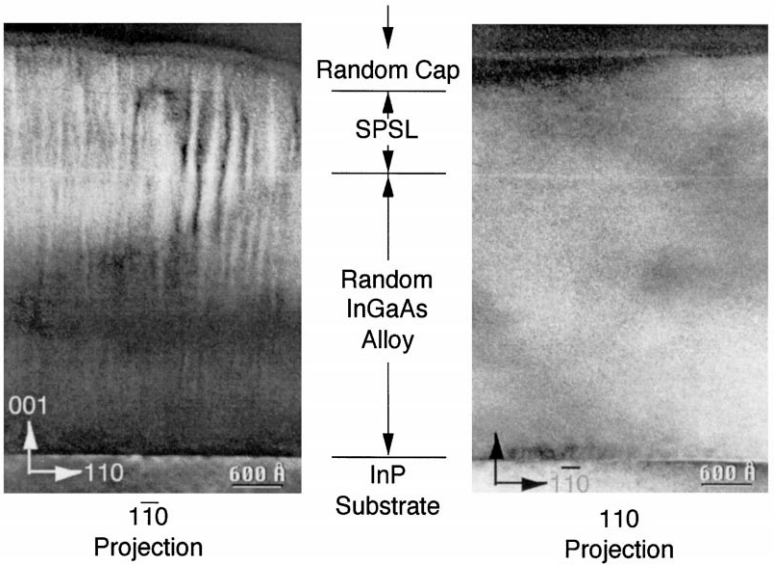

Fig. 1. Dark field images of the $(InAs)_n/(AlAs)_m$ SPS structure for the $[1\,\bar{1}\,0]$ and $[1\,1\,0]$ projections. The left side of the figure shows the lateral contrast variation due to showing lateral composition modulation but not in the orthogonal projection shown on the right side.

spectroscopy is an $(InAs)_n/(GaAs)_m$ SPS on a InP substrate that has spontaneously formed a lateral superlattice, with modulation, perpendicular to the growth direction. The microstructure was confirmed by cross-sectional TEM, high-resolution electron microscopy, and low-temperature polarized photoluminescence spectroscopy.

The SPS structure (#G1517) was grown on an n-type InP (0 0 1) substrate by molecular beam epitaxy. A 3.0 μm-thick $In_{0.5}Ga_{0.5}As$ buffer layer was deposited on the InP substrate. A 150-period undoped $(InAs)_n/(GaAs)_m$ SPS with $n+m\approx 2$ was deposited on top of the InGaAs buffer. Finally, a 0.5 μm cap layer of undoped $In_{0.5}Ga_{0.5}As$ was added to the top of the SPS. The growth temperatures and rates were, respectively, 500°C and 0.8 ML/s. Double crystal X-ray diffraction measurements show that the resulting buffer and SPS layers have a nominal indium concentration of 48%, hence, the resulting SPS structure is under tension.

Two TEM images of the CM structure are shown in Fig. 1. The left side of the Fig. 1 shows the formation of the lateral CM while the orthogonal direction shows a uniform distribution characteristic of a random alloy. The modulation wavelength λ is asymmetric and when averaged over the structure $\lambda\approx 13$ nm. As can be seen in the figure, there may be evidence for CM in the buffer layer. The root causes for spontaneous lateral CM are currently being pursued, and the presence of the composition modulation in the buffer layer is not understood.

The magnetoexciton spectroscopy measurements were made at 1.4 K and the magnetic field was varied between 0 and 14 T. The sample was attached to the end of a 100 μm-core-diameter optical fiber and located in a variable-temperature dewar ($1.4\,K\leqslant T\leqslant 300\,K$) insert placed in the middle of the superconducting magnet. The sample–fiber combination could be oriented in the three principal magnetic field directions, i.e., magnetic field along the growth direction, magnetic field perpendicular to the CM direction, and magnetic field along the CM direction. The PL measurements were made with an Argon-ion laser operating at 514.5 nm. The laser was injected into the optical fiber by means of a optical beam-splitter and the returning photoluminescence signal was directed to a 0.27 m, $f/4$ optical monochromator and a IEEE488-based data acquisition system. Typical laser power densities on the sample were of the order of $1\,W/cm^2$ and a NORTH COAST EO-817

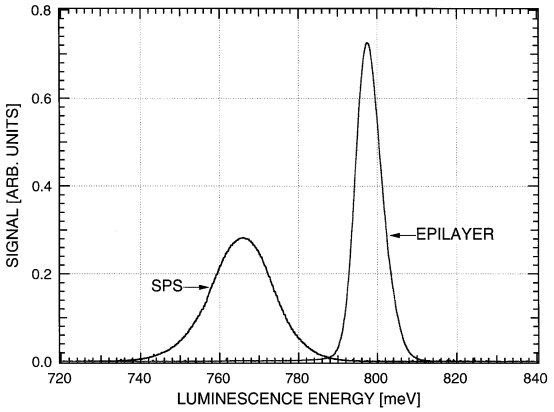

Fig. 2. Two 1.4 K zero-field PL spectra for the CM SPS structure (left) and the InGaAs-epilayer (right).

germanium detector was used to detect the ~800 meV (1.6 μm) infrared energy photons.

Two 1.4 K zero-field PL spectra are shown in Fig. 2. The spectrum for the CM SPS structure is shown on the left while the right-hand spectrum is that from an unintentionally doped, nominally lattice-matched random alloy epilayer of $In_xGa_{1-x}As$ on InP (#G1872.) The full-width at half-maximum (FWHM) of the CM SPS structure is 18.5 meV and for the random alloy epilayer, FWHM ≈ 8 meV. The indicated relative signal strengths of the two spectra are approximately correct. Neglecting strain effects, the 1.4 K band-gap energy of $E_g \approx 762$ meV for the SPS sample is considerably lower than the expected band-gap energy $E_g \approx 870$ meV for an $In_{0.48}Ga_{0.52}As$ alloy. A band-gap reduction of nearly 100 meV may be a measure of the presence of strong composition modulation; however, until the actual shape, composition modulation amplitude, valence-band offsets, etc., of the composition modulated region is known, it is difficult to make quantitative interpretations. The epilayer's PL spectrum peak energy of 798 meV is also somewhat lower than the accepted band-gap energy of 812 meV for lattice matched InGaAs/InP epilayer. This difference may be attributed to small composition differences. A general rule of thumb for the InGaAs ternary alloys system nominally lattice matched to InP is a change to the band-gap energy ΔE_g of about 11 meV per 1% change in composition. Nominally lattice-matched InGaAs epilayers on InP substrates grown at various times and conditions in our laboratories show exciton-peak energies ranging from 790 to 820 meV. Double crystal X-ray diffraction measurements of the lattice constant, and hence composition, for these other epilayer specimens, also show a similar variation. The SPS band-gap energy of 762 meV is clearly outside this range.

In undoped semiconductors, the coulomb interaction between conduction-band electron and a valence-band hole can form a hydrogenic-type bound state which is referred to as an exciton. The observed magnetoexciton diamagnetic shifts can be fitted using a simple hydrogenic model and from first-order perturbation calculations, the diamagnetic shift of the 1s ground state is given by [10] in cgs units as

$$\Delta E = \tfrac{1}{2} R^* \gamma^2, \tag{1}$$

where $\gamma = (\hbar\omega_c/2R^*), \hbar\omega_c = (2\mu_\beta B/\mu c)$ is the cyclotron resonance energy, $R^* = (\mu e^4/2\hbar^2\varepsilon^2)$ is the effective rydberg, $\hbar$ is Planck's constant over 2π, μ is the reduced mass ($1/\mu = 1/m_c + 1/m_v$), m_c and m_v are the conduction and valence-band masses, ε is the static dielectric constant, μ_β is the Bohr magneton, c is the velocity of light, and B is the applied magnetic field. Combining all of the above, the magnetoexciton diamagnetic shift ΔE given in Eq. (1) depends upon the reduced mass μ and magnetic field B as $\Delta E \propto \mu^{-3}B^2$, i.e., quadratic in magnetic field and inversely proportional to the cube-power of the reduced mass.

The experiment is thus performed by measuring the exciton diamagnetic shift, Eq. (1), in the three principal directions, i.e., along the growth axis, and two orientations perpendicular to the growth axis. A schematic for these orientations is shown in Fig. 3, where a three-dimensional view of the band-gap energy associated with the CM wave is shown as a spatially varying amplitude. The orientation B_1 is along the growth axis, B_2 is along the CM direction, and B_3 is perpendicular to both the growth and CM directions. The plane of exciton orbits for the three directions are the "donut" shapes shown for each magnetic field orientation. Because the zero-field exciton radius for InGaAs/InP is nearly 200 Å, the exciton orbits for two of the orientations B_1 and B_3, along the two perpendicular directions to the $\lambda = 130$ Å composition modulation wave, will include areas of varying composition and hence areas of varying band-gap energies. As is obvious from Fig. 3, the only orientation

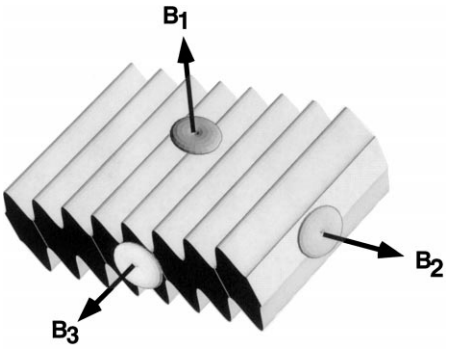

Fig. 3. Schematic drawing showing the lateral spatial variation of the band-gap energy from the CM wave. The three principal orientations of the magnetic field are B_1, B_2, and B_3 where B_1 is the growth direction. The "donut" shapes represent the plane of the aligned exciton orbits.

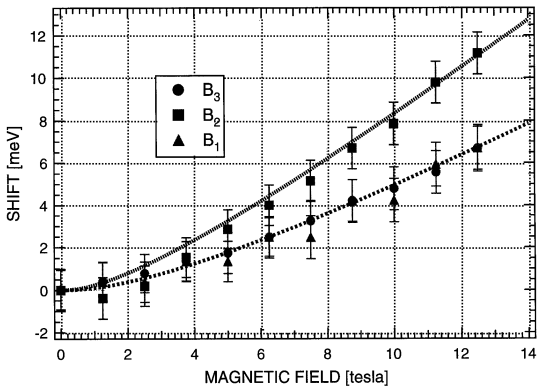

Fig. 4. Diamagnetic shifts for the CM SPS structure. The magnetic field orientations for the three shifts correspond to the alignments shown in Fig. 3. The best-fit curves of Eq. (1) to the data yield reduced masses $\mu(B_2) = 0.042$ and $\mu(B_1) = \mu(B_3) = 0.056$.

where the plane of the exciton orbit remains in a non-varying composition region (non-varying band-gap energy) is the B_2 direction.

Fig. 4shows the magnetoexciton diamagnetic shifts taken for the three principal directions B_1, B_2, and B_3 at a temperature of 1.4 K. As one can see from the figure, the diamagnetic shift for two directions, B_1 and B_3, are nearly identical while the diamagnetic shift for the B_2 direction is about 50% larger than the other two orientations. The lines through the data are best fits of Eq. (1), with $\varepsilon \approx 12.8$, with the result $\mu(B_2) = 0.042$ and $\mu(B_1) = \mu(B_3) = 0.056$. All masses are in units of the free electron mass. The conduction-band mass for lattice-matched InGaAs/InP is generally accepted to be 0.041, thus our measured exciton reduced mass of 0.042 indicates that the valence-band mass is large. A large valence-band mass can arise from bulk-band behavior where the two "heavy-hole" and "light-hole" valence-bands are degenerate, or from valence-bands experiencing tensile strain. The former explanation for a heavy-hole mass contribution to the reduced exciton mass μ is possibly the more correct assumption, especially in view of the optical absorption data obtained by Roura et al. [8].

If we attribute all of the variation to the reduced mass from 0.042 to 0.056 to the conduction band electrons, one can perform the following analysis about the change in composition. The conduction band-masses for InAs and GaAs are, respectively, 0.023 and 0.067. Assuming a linear variation for the conduction-band mass between InAs and GaAs predicts a change in mass $\delta\mu$ of 4.4×10^{-4} per 1% change in composition. Using this value, the lattice matched $In_{0.54}Ga_{0.46}As$/InP mass would be 0.047 which is reasonable in view of the mass uncertainties. The 0.014 mass difference between the three orientations then suggest a 30% change in group-III composition which may appear as a large value, however, recent STEM measurements [11] on a CM structure $(InAs)_n/(AlAs)_m$ SPS on InP gave a similar variation for the indium content. Until the exact nature of the energy band structure for these kinds of materials can be quantified, we will not be able to make quantitative predictions about the amplitude of the CM wave. However, the qualitative nature of the anisotropic diamagnetic magnetoexciton shifts does provide a good signature for the presence of composition modulation.

In conclusion, we have shown that magnetoexciton spectroscopy in three different directions can provide a rapid diagnostic tool for the documenting the presence of composition modulation. When quantitative detail concerning the size, shape, etc., of the composition modulation region becomes available, then these kinds of studies will provide insight about the local band structure properties.

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