

Energy levels and exciton oscillator strength in submonolayer InAs-GaAs heterostructures

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We have studied monolayer (ML) and submonolayer InAs insertions (1–0.08 ML) in a GaAs matrix by photoluminescence, photoluminescence excitation, and optical-reflectance spectroscopy. Linewidths of heavy-hole and light-hole exciton peaks as narrow as 0.15 meV are observed. A surprisingly high exciton oscillator strength, its weak dependence on the average thickness of the InAs layer, and the pronounced anisotropy and splitting of heavy- and light-hole exciton peaks are all revealed in the optical studies and are attributed to the formation of ordered arrays of InAs wirelike islands. Furthermore, from photoluminescence and reflectance-anisotropy measurements, we confirm that the wire arrays are elongated along the $[01\bar{1}]$ direction.

Growth of highly strained InAs-GaAs quantum-well heterostructures has attracted much attention due to their intriguing properties.¹ More recently, there arises an interest toward structures with further reduced dimensionality:^{2–17} quantum wires and dots. InAs quantum wires and dots have been grown selectively on SiO₂-patterned GaAs substrates² and on terraced GaAs surfaces with InAs monolayer (ML) coverage.³ It has also been reported that the growth of highly strained semiconductor layers (InAs, InGaAs) onto a substrate (GaAs, InP) could lead to the spontaneous formation of semiconductor nanometer-scale clusters.^{4–10} Very sharp emission lines from InAs-GaAs heterostructures prepared from this method have been observed recently,^{11,12} directly representing the δ -functionlike density of states in the zero-dimensional (0D) system. However, the integrated photoluminescence (PL) spectrum representing the dot average size distribution is still broad. The critical InAs thickness for cluster formation in the InAs-GaAs system has been found to be ~ 1.5 – 1.7 ML.^{11,12} We have also reported previously that long growth interruption in monolayer-thick InAs induced layer transformation to InAs wirelike clusters.¹³ More recently from optical anisotropy studies, we found that uniform array of InAs wires elongated along the $[01\bar{1}]$ direction is formed in the case of submonolayer InAs depositions on GaAs surfaces.¹⁷ Scanning-tunneling-microscopy studies have

shown that the growth of an InAs submonolayer (< 1 ML) on (100) and on vicinal GaAs surfaces results in the spontaneous formation of coherent InAs monolayer-high wirelike islands having a width of approximately 40 Å and an elongation along the $[01\bar{1}]$ direction.¹⁴ In this paper, we report a systematic study of energy levels, optical anisotropy, and exciton oscillator strength of nm-sized wire arrays formed by submonolayer InAs growth on GaAs (100) surface. We have found extremely narrow PL linewidths (down to 0.15 meV) indicating very good average uniformity of the island array. We have also found a remarkably high exciton oscillator strength characteristic of these structures even in the case of extremely diluted (0.08 ML) InAs coverage.

The structures are grown by conventional molecular-beam epitaxy (MBE) at 480 °C on GaAs (100) substrates. Growth rates are calibrated by reflection high-energy electron diffraction intensity oscillations and are 0.1 ML/s for InAs and 1 ML/s for GaAs growth. (2×4) surface reconstruction is maintained prior to the InAs deposition. Growth interruptions of 10 s are introduced before and after the InAs deposition. The structure consists of a 0.2- μm -thick GaAs buffer layer, six periods of 20-Å GaAs/20-Å AlAs forming a short-period superlattice (SPSL). A single InAs submonolayer (SML) is inserted in the center of 0.2- μm -thick GaAs layer. Another similar SPSL is grown on the top of the GaAs layer. Fi-

nally, a 100-Å GaAs cap layer is grown to protect the surface.

For a precise calibration of the InAs growth rate in the submonolayer deposition mode, a superlattice structure was grown composed of InAs submonolayers ($\frac{1}{16}$ ML) separated by a 230-Å-thick GaAs layer. The superlattice was characterized using double-crystal x-ray diffraction. Rocking curves are measured for the (004) reflection using Cu $K\alpha$ radiation as a source. The x-ray diffraction spectrum of the InAs SML-GaAs superlattice (SL) is shown in Fig. 1. X-ray diffraction measurements give a precise value of the average indium composition in the InAs-GaAs superlattice obtained from the separation of the 0th SL peak (SL_0) and the GaAs substrate peak. A precise value of the superlattice period was derived from the separation of superlattice (SL_{-1} , SL_{+1}) high-order diffraction peaks and the SL_0 peak.¹⁵ The period of the superlattice derived from the separation between satellite peaks (230 Å) agrees well with the calibrated GaAs growth rate. The average InAs layer thickness derived from the SL period and the shift of the SL_0 peak is 0.53 Å (± 0.02 Å) assuming fully pseudomorphic growth. This is in excellent agreement with the value estimated from the InAs growth rate calibration (0.5 Å). One should note, however, that conventional x-ray diffraction measurements do not reveal any definite information on the character of InAs distributions over the surface.

Photoluminescence-excitation (PLE) measurements were carried out at 5 K in a He-flow cryostat. A tunable Ti-sapphire laser was used as the excitation source. Samples were chemically polished from the substrate side and mounted in paper bags to avoid any source of external strain. Optical-reflectance and reflectance-anisotropy studies were carried out at 83 K using the same technique reported elsewhere.¹⁶

In Fig. 2, we show 5-K PLE spectra under circular and linear polarization of the structures with a 0.08 ML of InAs. These InAs submonolayers in a GaAs matrix have been shown¹⁷ to result in narrow PL peaks with energies between the energy of 1-ML InAs peak and the GaAs

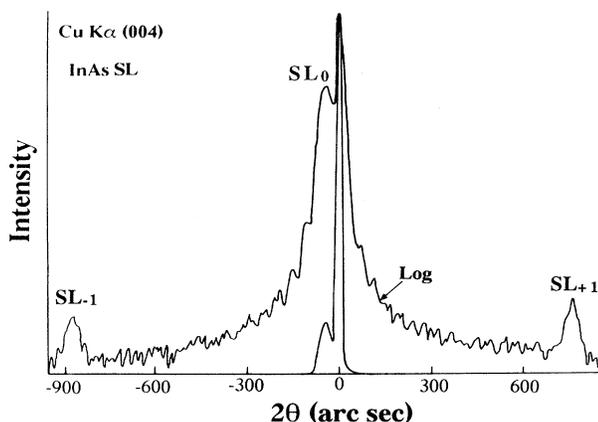


FIG. 1. Double x-ray rocking curve of InAs-GaAs superlattice.

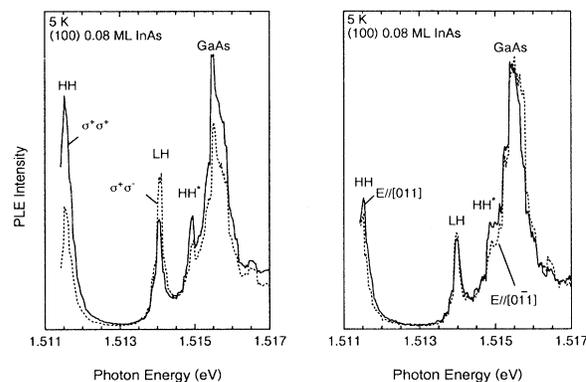


FIG. 2. (a) Circularly polarized PLE spectra of an 0.08 ML of InAs in (100) GaAs. (b) The linearly polarized PLE spectra of the same sample. The optical anisotropy is clearly observed in submonolayer InAs embedded in a GaAs matrix, indicating that InAs chains are elongated along $[01\bar{1}]$ directions. HH^* indicates excited-state transitions of the heavy-hole exciton.

free exciton peak. Such a shift to low energies may be explained either by an extremely uniform distribution of InAs molecules over the surface or by the formation of 1-ML-high InAs islands having diameters smaller than the exciton Bohr radius. The latter case seems to be much more probable taking into account the low-temperature MBE growth mode, which encourages island formation. At the same time, the extremely narrow linewidth of the PL and PLE peaks [with full width at half maximum of 0.15 meV, see Fig. 2] indicates the surprisingly high uniformity of the array of InAs islands. This may be explained by the formation of an ordered array InAs monolayer-high wirelike islands having a uniform width of about 40 Å and an elongation along the $[01\bar{1}]$ direction directly revealed in a recent scanning-tunnel-microscope (STM) study.¹⁴ These observations indicate the possibility of partial exciton lateral quantization in submonolayer InAs. Heavy- and light-hole exciton peaks in the PL and PLE spectra are weakly polarized parallel to $[011]$ and $[01\bar{1}]$ directions, respectively. This indicates that the optical anisotropy is controlled by the anisotropic strain directed along the wires, in agreement with the theoretical work.¹⁸ The anisotropy is stronger for the excited exciton states indicating that lateral confinement effects are more important for the ground state leading to the reduction of the strain-induced optical anisotropy.¹⁹

Figure 3 shows the optical-reflectance (OR) spectra for structures at 83 K with submonolayer InAs coverage. The experimental details have been reported previously.¹⁶ Dashed lines show a theoretical fit to experimental data assuming a realistic structure geometry, which will be discussed later in the paper. The intensity minimum at 1.507 eV can be attributed to a GaAs free exciton. The characteristic width of this feature is only 0.6–0.8 meV and its shape is similar in all the structures with InAs SML. Some difference in the shape of the GaAs free ex-

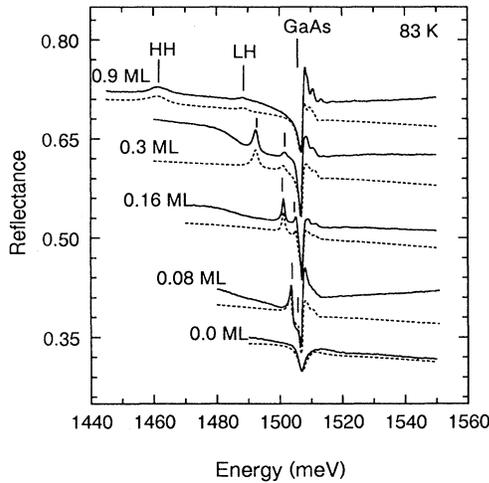


FIG. 3. Optical-reflectance spectra of submonolayer InAs at 83 K. Spectra are vertically shifted by 0.1 for clarity.

citon feature in the upper curves may be explained by slightly larger GaAs layer thickness in these samples. All the samples with InAs SML exhibit heavy-hole (HH) and light-hole (LH) exciton peaks in the OR spectra with energies corresponding to those revealed in PL and PLE spectra. Figure 3 demonstrates a remarkably strong modulation of OR caused by excitons bound to InAs islands even in the case of very dilute InAs coverage. The width of InAs-related exciton peaks decreases from 7 meV in a 0.9-ML sample to 0.6 meV in a 0.08-ML InAs sample. At the same time, the amplitude of the modulation increases. The effect is so strong that one may conclude from Fig. 3 that the oscillator strength of InAs-related excitons remains fairly high even in the case of very dilute InAs coverage. One should also note the appearance of the peaks on the high-energy side of the GaAs free exciton peak. These structures become progressively stronger with increasing average thicknesses of the InAs SML. They can be explained by the above-barrier states expected both in an isoelectronic impurity model²⁰ and in a quantum-well case.²¹

Strain-induced optical anisotropy was also readily observed in OR measurements. In Fig. 4, we show the reflectance-anisotropy (RA) spectrum of the 0.3-ML InAs sample. The RA signal is defined as $2(R[011] - R[01\bar{1}]) / (R[011] + R[01\bar{1}])$. The RA spectrum of pure GaAs (0-ML InAs coverage) was checked for the accuracy of experimental setup and exhibited no RA signal. From Fig. 4, one can see a definite anisotropy signal in the region of HH and LH exciton resonances as well as in the vicinity of the GaAs free exciton peak. Moreover one sees that the polarization contour of HH and LH lines has a complex shape: the low- and high-energy sides of each of the lines were polarized in different directions. However, the opposite polarization in one peak was hardly resolved in PL and PLE studies as only the low-energy part of the PL spectrum is monitored in polarized PLE

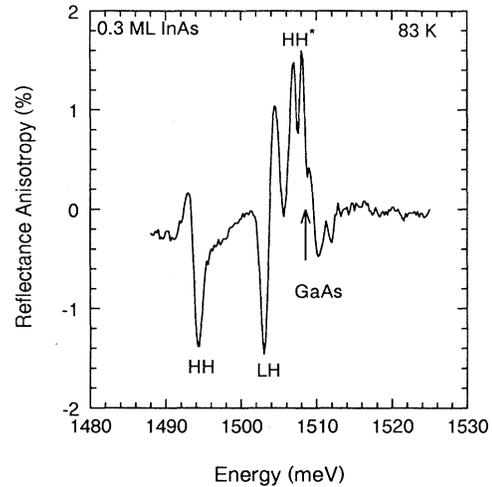


FIG. 4. Reflectance anisotropy (RA) in 0.3-ML InAs. Note the doublet structure in HH, LH, and excited HH exciton transitions. RA is defined as $2(R[001] - R[01\bar{1}]) / (R[011] + R[01\bar{1}])$. HH* indicates excited-state transitions of the heavy-hole exciton which is close to the GaAs band-gap transition.

measurements (see Fig. 2). A combination of the in-plane exciton localization and anisotropic strain within the same plane, which results in a splitting of the doubly degenerate light- and heavy-hole states, is believed to be responsible for this observation. To fit the experimental curve, we have to assume a splitting of 0.1 meV and 5% difference in the exciton oscillator strength for two oppositely polarized transitions. These results are in good agreement with the highly anisotropic shape of InAs wirelike islands on the GaAs surface.

In Fig. 5(a), we show the InAs HH and LH exciton peak positions observed in the PLE and OR experiments at 83 K. The assignment of the transitions was based upon circular polarization measurement.¹⁷ Solid lines show the calculated exciton transition energies assuming that the submonolayer growth results in a uniform distribution of InAs molecules, i.e., assuming that a 1-ML-thick $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well has an average In composition derived from the fraction of the surface covered with InAs molecules. Calculated conduction- and valence-band offsets were obtained using the effective-mass approximation assuming a 1-ML-thick $\text{In}_x\text{Ga}_{1-x}\text{As}$ quantum well of finite potential. This approximation was shown previously to agree well with experimental results in the case of monolayer-thick InAs quantum wells.³ These are shown in Fig. 5(b). The lateral strain relaxation was not included in the strain modification of the band gap. The exciton binding energy (E_{ex}) was assumed to be equal to the GaAs bulk value. This assumption is not valid in the case of InAs layers close to or thicker than 1 ML, where E_{ex} may exceed the bulk value by a factor of 2–3.²² However, in the case of 1-ML-thick InGaAs quantum wells with a diluted In composition,

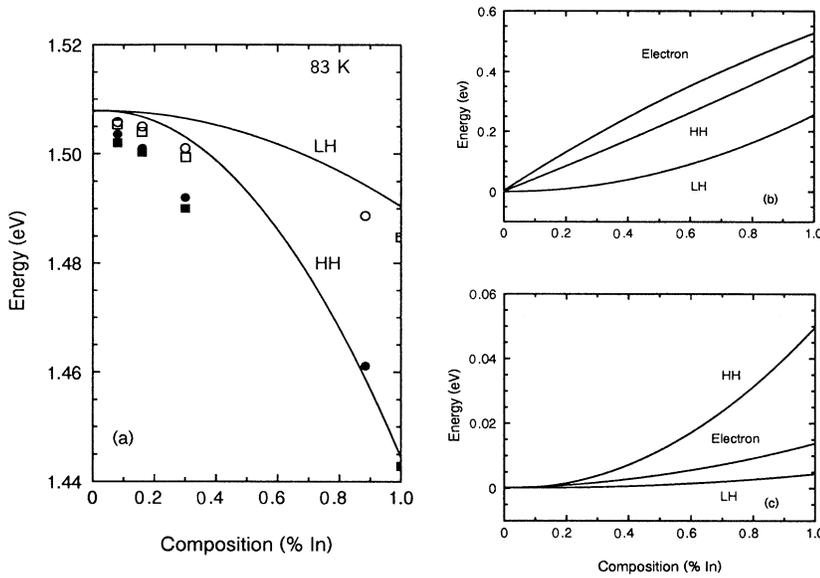


FIG. 5. (a) Theoretical calculations of exciton transitions (solid line) based on effective-mass approximations. The filled and open squares are HH and LH exciton transitions measured from PLE spectra at 77 K, respectively. The linewidth at 77 K did not change very much as compared to that of 5-K data. Filled and open circles are HH and LH exciton transitions determined by OR measurements at 83 K, respectively. (b) Theoretical calculations of conduction, HH, and LH band offset as a function of In composition. (c) Calculation of the electron, HH, and LH confinement energies with respect to the GaAs matrix.

where the conduction-band offset should be smaller, this assumption seems to be reasonable. In Fig. 5(c), we show the dependence of the electron and heavy- and light-hole energy levels with respect to the GaAs barrier energy calculated using the uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer model.

From the comparison of theoretical²³ and experimental data presented in Figs. 5(a)–5(c), one can conclude that there exists a marked disagreement between the calculated energies and the experimental values for submonolayer coverage, indicating that the model of a uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ layer is not suitable. Both LH and HH exciton peaks are shifted to lower photon energies. One can see from Fig. 5(c) that the strongest contribution to the confinement energy of the excitonic transition is expected from the heavy-hole localization energy in an $\text{In}_x\text{Ga}_{1-x}\text{As}$ alloy quantum well (QW). One would expect that the heavy-hole localization is even stronger in an InAs quantum-wire case. For lateral quantization, the heavy-hole wave function is confined mostly in InAs regions and partly propagates in GaAs regions, thus the PL is shifted to lower energy with respect to the position expected for a uniform $\text{In}_x\text{Ga}_{1-x}\text{As}$ QW (solid curves in Fig. 5) with an average In composition. Therefore, experimental results in Fig. 5(a) indicate the increase in the HH localization energy in agreement with the formation of InAs 1-ML-high wirelike islands. Light-hole excitons, which are weakly localized, are shifted to lower energies due to smaller contributions of uniaxial strain caused by partial strain relaxation in lateral directions resulting in an increase in the potential well for the light hole, which is otherwise negligibly shallow.

The InAs SML related exciton oscillator strength is deduced from a model of the reflectance spectrum (Fig. 3). To fit the experimental data we calculated the optical-reflectance spectrum assuming a multilayer structure (GaAs cap layer, superlattices, GaAs layers, InAs layer)

using parameters deduced from the growth rate calibration. The exciton oscillator strength and the broadening were introduced as variable parameters. Excited exciton states and the states above the GaAs continuum are not included in the fitting. In general, the dielectric response is given by

$$\epsilon(E) = \epsilon_{\text{bkg}}(E) + \sum_j \frac{S_j E_j}{E_j - E - i\Gamma_j}, \quad (1)$$

where S_j , E_j , and Γ_j are oscillator strength, energy, and damping of the j th exciton transition. $\epsilon_{\text{bkg}}(E)$ is the background dielectric function resulting from the GaAs band-gap transitions.¹⁶

The contribution of the InAs SML to the reflectance spectrum was calculated using two approaches, giving essentially the same results.

In the first approach²⁴ we introduced the dielectric susceptibility of the InAs SML and calculated the amplitude coefficient of the optical-reflection r_{QW} from an InAs SML quantum well with an effective 1 ML thickness of $d_{\text{QW}} = 3.03 \text{ \AA}$

$$r_{\text{QW}} = r \frac{\exp(-i\Psi) - \exp(i\Psi)}{\exp(-i\Psi) + r^2 \exp(i\Psi)}, \quad (2)$$

where r is the amplitude coefficient of the OR corresponding to GaAs-InAs SML interfaces, and Ψ is the phase shift of the lightwave in the layer; r and Ψ are given as

$$r = \frac{n_b - n_{\text{QW}}}{n_b + n_{\text{QW}}}, \quad \Psi = d_{\text{QW}} n_{\text{QW}} \frac{2\pi}{\lambda}, \quad (3)$$

where $n_{b,\text{QW}} = \sqrt{\epsilon_{b,\text{QW}}(E)}$ is the refractive index, and λ is the light wavelength in the crystal.

Using the second approach, the OR was calculated as

TABLE I. Results and fitting parameters from optical-reflectance calculations.

	0.9 ML	0.3 ML	0.16 ML	0.08 ML
		Heavy hole (hh)		
E_j (eV)	1.461	1.492	1.501	1.504
$S_j \times 10^{3a}$	130	90	50	60
τ_j (ps) ^b	38	51	82	59
		Light hole (lh)		
E_j (eV)	1.489	1.501	1.505	1.506
$S_j \times 10^3$	25	30	30	30

^aDetermined from the first approach discussed in the text.

^bDetermined from the second approach discussed in the text and in agreement with calculations from Eq. (5).

$$r_{\text{QW}} = \sum_j \frac{-i(2\tau_j)^{-1}}{E_j - E - i\Gamma_j}, \quad (4)$$

where τ_j is the exciton radiative lifetime²⁵ in the InAs SML.

In the second approach, τ_j is a fitting parameter and it is found to agree very well with τ_j calculated from the exciton oscillator strength S_j found using the first approach:

$$(\tau_j)^{-1} = 2\pi S_j E_j [\epsilon_{\text{bkg}, \text{QW}}(E_j)]^{-1/2} d_{\text{QW}} / \lambda. \quad (5)$$

The results of these calculations and the fitting parameters are summarized in Table I.

Surprisingly, both Fig. 3 and Table I show remarkably high exciton oscillator strengths even in the case of ultrasmall InAs coverage. This high exciton oscillator strength is directly revealed in the OR spectra as modulations of the signal caused by the InAs resonances. A high exciton oscillator strength was reported previously for quantum wells.²⁶ It was shown theoretically that the GaAs-Al_xGa_{1-x}As quantum-well exciton oscillator strength increases with the decrease of well width from 28.7 ps for a 150-Å-thick QW to 16.3 ps for a 50-Å-thick QW. These results are in excellent agreement with the PL decay times experimentally observed for a high-quality 45-Å-thick GaAs-AlAs quantum well under pulsed resonant excitation.²⁷ Similar lifetimes and exciton oscillator strengths are also found from OR spectra line-shape fitting²⁵ with the advantage of less complicated experimental setup and weaker requirements for the structure homogeneity. For very thin (or very shallow) quantum wells, the situation is reversed. In this case, energy levels of confined carriers become weakly localized. Their wave functions expand into the barrier and the exciton oscillator strength decreases due to the decreased wave-function overlap.²⁸ The effect of a dramatic (by an order of magnitude) decrease of the exciton oscillator strength with the decrease of the SL layer thickness (from 80 to 25 Å) was observed for GaAs-Al_xGa_{1-x}As superlattices²⁹ (estimated from longitudinal-transverse splitting). In the InAs SML case, heavy-hole exciton radiative lifetimes are relatively close to the case of a quantum well of moderate thickness (150–250 Å) and show practically no dependence on the average InAs layer width (composition).

To explain our results, we may consider several possible interpretations. First of all, a model of a diluted array

of InAs monolayer-high islands acting as localization centers for GaAs bulk excitons should be mentioned. This corresponds to the bound exciton complexes model.³⁰ In a GaAs case, where electron effective mass is much smaller than the heavy-hole effective mass, both theory and experiment³⁰ give radiative lifetimes of 1–2 ns for exciton localization energy of the order of 1 meV. Such lifetimes should increase with the increase of localization energy. In a narrow QW model, the reduction of the surface fraction covered by InAs monolayer islands with lateral sizes larger than the exciton Bohr radius should result in an effective decrease of the exciton oscillator strength normalized to the total surface area. Thus, our results are in marked disagreement with both models.

We believe that our results can be explained by assuming a lateral quantization model.¹⁶ The formation of quantum wire and dot arrays may provide an exciton oscillator strength comparable to that of quantum wells even in the case of their very small fractional coverage of the surface area.³¹ This effect is more pronounced for the case of quantum dots than for quantum wires. In fact, the rather short exciton radiative lifetime suggests a strong lateral exciton confinement in the structures under investigation and agrees with the formation of elongated uniform InAs islands revealed in STM studies.¹⁴ In our case, it corresponds to a gradual transition from quantum-well to quantum-wire array and, finally to a quantum-dot array. One should also note that our observation of high exciton oscillator strength agrees with a high exciton binding energy observed for an InAs submonolayer (0.3 ML).²² Although the InAs island is rather small, giant differences in band-gap energies between GaAs and InAs make lateral confinement of excitons very effective.

In conclusion, we have studied the optical properties of InAs submonolayers inserted in a GaAs matrix. Extremely narrow linewidth, high exciton oscillator strength, and optical anisotropy are found in PL, PLE, and RA spectra and attributed to the formation of a uniform array of wirelike InAs chain structures. This indicates the importance of this type of structure both for fundamental research and possible device applications.

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