

Photoluminescence of undoped InAs autoepitaxial layers

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Abstract. Photoluminescence (PL) properties of undoped InAs autoepitaxial layers are studied at various temperatures (9-120 K) and excitation power density (0.2-12.5 W/cm²). The studied structures have been grown on highly doped n⁺⁺-InAs substrates by chloride-hydride vapour phase epitaxy method. PL spectra measurements were carried out with an FTIR spectrometer. The samples exhibit several luminescence peaks in the 2.9-3.3 μm range, which are attributed to free exciton transitions, deep donor bound excitons, and donor-acceptor pairs. A correlation is revealed between the PL intensity, and the background doping level of InAs epilayers – as supported by the free carrier concentration and mobility measurements, as well as SIMS data. A decrease of donor-bound exciton PL peak is observed for InAs sulfidized by Na₂S.

1. Introduction

Indium arsenide (InAs) autoepitaxial structures grown on highly doped substrates (n⁺⁺-InAs) are widely used [1] as materials for backside-illuminated infrared image detectors operating at wavelengths about $\lambda \sim 3 \mu\text{m}$. Such structures can selectively cover a broad spectral range due to variable transmission of differently doped n⁺⁺-InAs [2], while the epitaxial n-InAs acts as an active region. To obtain high sensitivity of such photodetectors, powerful diagnostic techniques are required to control optical properties and growth quality of fabricated [3] InAs epitaxial layers. One of the available methods for contactless determination of epitaxial layers' quality is photoluminescence spectroscopy.

2. Studied samples and experimental setup

The studied structures containing undoped autoepitaxial InAs layers were grown by chloride-hydride vapour phase epitaxy method on highly doped n⁺⁺-InAs substrates. Epitaxial layer thickness, determined from the interference pattern in infrared (IR) reflectance spectra [4, 5], is varied from 3 μm to 10 μm . Free carrier concentration in the layers, obtained with magnetoresistance measurements, was found to be in the $6 \cdot 10^{13}$ - $6 \cdot 10^{15} \text{ cm}^{-3}$ range. The parameters of the structures are listed in Table 1.

The studied samples include three series (I, II, III) differentiated by epitaxial growth parameters. Series I samples with carrier concentration less than $1 \cdot 10^{15} \text{ cm}^{-3}$ were grown under the condition of In mole fraction X_{In} being greater than As mole fraction X_{As} . The shortage of arsenic leads to formation of structural defects based on As vacancies in the epitaxial InAs [6]. In combination with various dopants, these defects can result in donor or acceptor levels within the InAs band gap. A reduction of carrier mobility, observed alongside the decrease of concentration values, implies the predominant formation of acceptor-type defects in our structures.



Table 1. Sample parameters

Sample series	Sample number	Free carrier concentration at 77 K (magnetoresistance), cm^{-3}	Carrier mobility at 77 K (magnetoresistance), $\text{cm}^2 \text{V}^{-1} \text{s}^{-1}$	Comments
I	I-0	$6.2 \cdot 10^{13}$	$1.5 \cdot 10^4$	$X_{\text{In}}/X_{\text{As}} > 1$
	I-1	$2.0 \cdot 10^{14}$	$3.3 \cdot 10^4$	$X_{\text{In}}/X_{\text{As}} > 1$
	I-2	$1.3 \cdot 10^{15}$	$7.9 \cdot 10^4$	Optimal
II	II-1	$5.4 \cdot 10^{15}$	$4.3 \cdot 10^4$	After In source refilling
	II-2	$4.0 \cdot 10^{15}$	$4.7 \cdot 10^4$	In source annealed
III	III-1	$2.3 \cdot 10^{15}$	$7.3 \cdot 10^4$	Sulfidized by Na_2S
	III-2	$2.3 \cdot 10^{15}$	$7.3 \cdot 10^4$	Reference sample

The second sample series was grown to examine the influence of residual donor doping originating from the metallic In source. A concentration increase above $5 \cdot 10^{15} \text{ cm}^{-3}$ was generally observed after refilling the source with a new supply of indium. A prolonged (above 30 hours) annealing of the refilled source in hydrogen atmosphere at $T = 850^\circ \text{C}$ results in electron mobility increase, and stabilization of carrier concentration in the $(1-4) \cdot 10^{15} \text{ cm}^{-3}$ range.

Sulfidization of the sample surface by Na_2S water solution was used to remove the natural oxide layer and the surface-related impurities (sample III-1). A reference sample of non-sulfidized epitaxial InAs was taken from the same wafer.

The photoluminescence (PL) experimental setup was based upon a VERTEX 80 Fourier-transform infrared spectrometer (FTIR) operating in continuous scan mode. The spectrometer was equipped with a liquid nitrogen-cooled HgCdTe photovoltaic detector sensitive in the $1.3-16 \mu\text{m}$ range. PL measurements were carried out at various temperatures (9-120 K) using a closed-cycle He cryostat. Excitation was performed with an 809 nm laser diode of variable power (1 mW - 300 mW).

3. Results and discussion

An example of low temperature PL spectra of an InAs autoepitaxial layer (sample I-0) is shown on Fig. 1. The main PL peak at $3.0 \mu\text{m}$ (0.413 eV) is rather close to the direct band gap of InAs ($E_g = 0.415 \text{ eV}$ at 12 K [7]). Due to the relatively small exciton binding energy of InAs ($E_{\text{ex}} \approx 1 \text{ meV}$ [8]), the peaks observed at $3.1 \mu\text{m}$ (0.400 eV) and $3.25 \mu\text{m}$ (0.382 eV) can not be attributed to weakly bound exciton recombination (as they are apart from the main peak by 13 meV and 31 meV, respectively). To determine their origin, we analyzed the dependence of peak intensity on laser excitation power density, as shown on Fig. 2.

The intensity of the main peak at 0.413 eV is demonstrating a linear increase proportional to laser power density, which is characteristic of free exciton recombination [9]. The long wavelength peaks exhibit a much slower change in intensity, and therefore should be connected to impurity-based states [10]. According to [11], the peak at 0.382 eV originates from donor-acceptor pairs (DAP). This is confirmed by the characteristic blue shift of the peak with increasing excitation density (Fig.1). The 0.4 eV peak, which has shown a quick decrease at higher temperatures, can be attributed to excitons bound to deep donors in InAs (presumably carbon) [12].

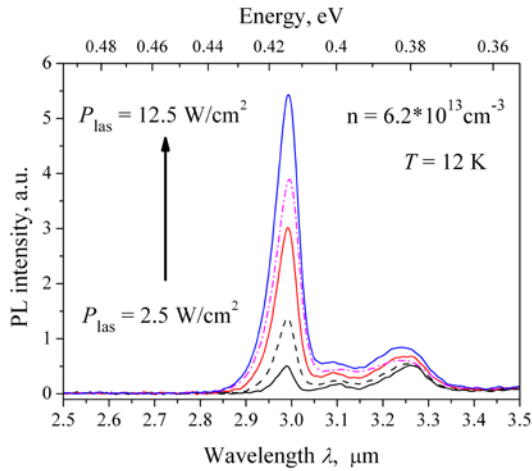


Figure 1. PL spectra of a typical InAs/n⁺⁺-InAs structure (Series I) at various laser excitation power densities P_{las} .

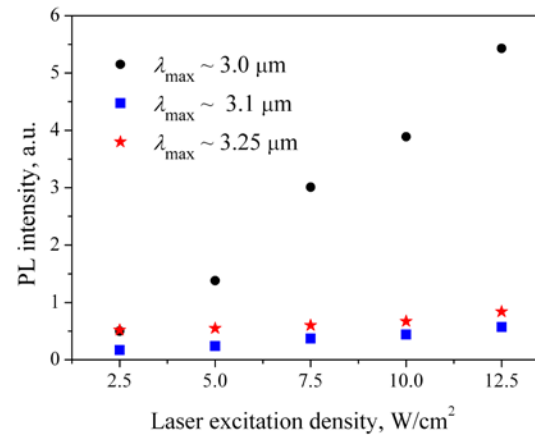


Figure 2. Dependence of PL intensity on excitation power density for 3.0 μm (dots), 3.1 μm (squares) and 3.25 μm (stars) peaks, shown on Fig. 1.

A comparison of the PL spectra for the series I samples has revealed that the donor-acceptor pair luminescence peak is more prominent for structures with lower carrier concentration in the epitaxial layers. Therefore, it can be concluded that such structures have a greater degree of impurity compensation.

The low-power PL spectra of the second series of n-InAs epitaxial layer samples are depicted on Fig. 3. Both samples demonstrate the absence of the 3.25 μm DAP peak, which implies a considerably reduced acceptor concentration. This means that the free carrier concentration in the layers should be proportional to the amount of background donor doping.

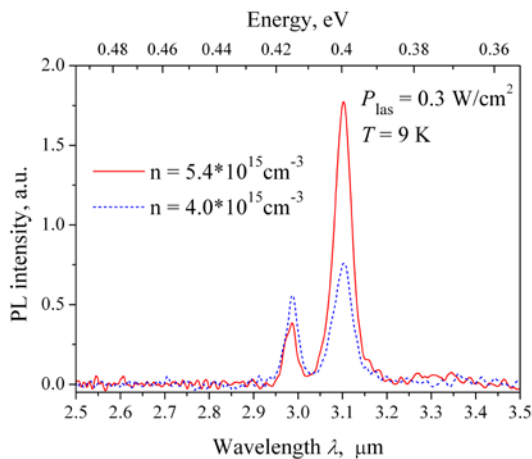


Figure 3. Comparison of PL spectra for n-InAs layers of the series II samples with different free carrier concentration.

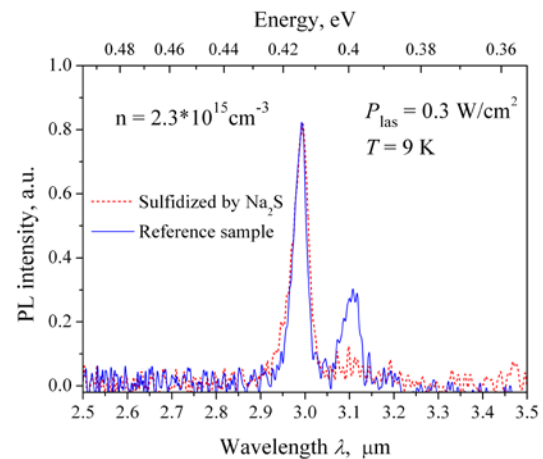


Figure 4. The influence of surface sulfidization on the PL spectra of an n-InAs/n⁺⁺-InAs structure (series III).

Although the intensity of the free exciton recombination peaks at 3.0 μm is roughly the same, the peak at 3.1 μm is noticeably more intensive for the sample with a higher value of free carrier concentration.

The last part of this study is dedicated to evaluating the impact of surface-based impurities on the photoluminescence of InAs epitaxial layers. Fig. 4 shows the PL spectra of an n-InAs structure treated with Na₂S water solution, and of a reference sample taken from the same wafer.

It has previously been reported [13] that the Na₂S treatment of the InAs surface should increase the main PL peak (80 K) due to removal of surface defect states promoting nonradiative recombination. However, as can be seen from Fig. 4 (9 K), the Na₂S passivation has resulted in a considerably weaker donor-bound exciton PL peak, which should suggest a decreased concentration of donors.

This can be explained if we consider the impurity profiles for n-InAs/n⁺⁺-InAs structures obtained by secondary ion mass spectrometry (SIMS). An order-of-magnitude increase in carbon and oxygen concentration was observed in a thin (<30 nm) layer at the sample surface. According to [14], the sulfide passivation of InAs, in addition to oxide removal, can also result in limited etching of InAs. This might lead to removal of the unintentionally doped surface layer, thus severely decreasing the donor-bound exciton luminescence – as was observed in our spectra.

In conclusion, we have observed a correlation between the PL properties of InAs autoepitaxial layers, and the residual donor and acceptor concentrations. These results can potentially be used to develop a contactless method of determining impurity concentration in InAs on the basis of photoluminescence measurements.

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