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Observation of photoluminescence from InAs surface quantum wells grown on InP(100) by molecular beam epitaxy

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Photoluminescence (PL) measurements are presented for thin epitaxial layers of InAs, 2.5 $\text{\AA} < d < 36 \text{\AA}$, grown on InP(100) by molecular beam epitaxy. The combination of efficient carrier capture and PL redshift with increasing InAs thickness clearly indicate the formation of InAs quantum wells on the InP surface. Data are also presented for InAs/InP structures capped with strained layers of either GaAs or In_{0.5} Al_{0.5} As. Since radiative recombination within the InAs layers can be distinguished from PL arising from both bulk and surface defects, this system allows us to monitor the quality of both the InAs/InP and InAs/air interfaces via their influence on the InAs quantum well luminescence.

Clean and well-ordered InP(100) surfaces can be prepared, for use in growth by molecular beam epitaxy (MBE), by annealing InP under an arsenic over-pressure.¹ Originally, such thermal cleaning was thought to leave a layer of As on the substrate surface. However, recent studies^{2,3} have shown that oxide removal under As stabilization of the InP surface can result in the exchange of P for As atoms, leading to the formation of a thin pseudomorphic InAs layer on the InP surface. Furthermore, photoluminescence (PL) measurements on such layers have revealed⁴ PL transitions consistent with the existence of InAs quantum wells on the surface of InP. In this letter we report the first observation of PL from InAs surface quantum wells grown epitaxially on InP(100) by MBE.

Growth of InAs was carried out in two separate MBE reactors, a VG Semicon V80H and a Riber 2300, using slightly different conditions. Good overall agreement was found between similar structures grown in either machine. Since the results to be presented in this letter pertain, on the whole, to material grown in the VG Semicon machine, we will restrict our discussion of growth details to this system.⁵ Fe-doped, semi-insulating InP(100) substrates were obtained from Nippon Mining and prepared by etching in a mixture of H₂SO₂:H₂O₂:H₂O in the ratio 7:1:1. Prior to growth, the substrate temperature was slowly increased from ambient, using a stabilizing As₄ over-pressure of 8×10^{-5} mbar, while the InP surface was observed in situ by reflection high-energy electron diffraction (RHEED) measurements. At low temperature the RHEED pattern exhibited only bulk streaks; however, at 500 °C, an abrupt transition was seen to a (2×1) surface reconstruction indicative of oxide removal from the InP. The substrate temperature was then increased to 510 °C and epitaxial growth of InAs initiated using a growth rate of 0.52 μ m/h (previously calibrated using a combination of double-crystal x-ray diffraction measurements and scanning electron microscopy) and a V:III pressure ratio of 120:1. As noted earlier, oxide removal under an As overpressure results in the creation of a thin InAs layer, ~ 5 Å thick, on the InP surface. The thickness of this InAs layer, formed by the exchange of P for As, obviously needs to be added to the deposited thickness of epitaxial InAs to give the total InAs thickness. Throughout the text, we will only refer to the thickness of InAs deposited epitaxially by MBE when distinguishing between samples. The total InAs thickness will only be stated explicitly when we seek to identify a trend for the whole range of samples grown, e.g., the variation of PL peak energy versus InAs layer thickness. PL measurements were performed in the temperature range 4.2-200 K inside a cryostat, using an excitation density of $\sim 50 \text{ mW/cm}^2$ at 632.8 nm. A Ge detector was used for PL energies greater than 0.7 eV, while a PbS cell was used for PL energies below 0.7 eV. The spectral resolution of the system always remained better than 2 meV.

Figure 1 compares 4.2 K PL spectra for 10-, 5-, and 2.5-Å-thick layers of InAs deposited on InP(100), with the PL spectrum obtained from an InP substrate chemically etched with choline. These deposited InAs thicknesses were checked by ex situ x-ray photoelectron spectroscopy (XPS). The form of the XPS attenuation plots for the P core levels indicated average layer-by-layer growth of the InAs on InP, for these thicknesses. All the spectra for InAs/InP in Fig. 1 exhibit more PL from the InAs layer than from the InP substrate, even though the absorption length at this excitation energy is around 170 nm for InP.⁶ PL spectra for 20 Å and 30 Å layers of InAs/InP show a continuous redshift of the PL emission to lower photon energies, together with an accompanying decrease in PL linewidth, with increasing layer thickness. The combination of efficient carrier capture and PL redshift with increasing InAs thickness clearly indicate the formation of InAs quantum wells on the InP surface. While the PL



FIG. 1. Comparison of 4.2 K PL Spectra for (a) 10 Å InAs/InP, (b) 5 Å InAs/InP, (c) 2.5 Å InAs/InP, and (d) InP etched in choline.

lineshape for the 10 Å InAs/InP surface quantum well can be adequately described by a single Gaussian, the PL lineshapes observed for the thinner InAs layers are similar to those previously⁷ attributed to monolayer fluctuations in ultrathin quantum well structures. Since the bulk lattice constants of InP, InAs are $a_0 = 5.8687$ Å and a = 6.0583Å, respectively, the 2.5, 5, and 10 Å epitaxial InAs layers differ in thickness by ~ 1 monolayer (ML). We have found that all the InAs associated luminescence in Fig. 1 can be fitted with weighted contributions from three simple Gaussians located at energies of ~ 0.83 , ~ 0.97 , and ~ 1.08 eV. The full width half maximum (FWHM) value of the PL lineshape obtained for 10 Å InAs/InP is 93 ± 1 meV, which can be compared with a linewidth of 10-14 meV obtained⁸⁻¹⁰ for 1-3 ML. InAs/InP quantum wells grown by metalorganic vapor phase epitaxy (MOVPE). For InAs quantum wells of this thickness, the PL FWHM will be influenced¹¹ by the material quality of the well and the barriers, as well as by interfacial roughness. Neither the InAs/InP nor InAs/air interfaces provide abrupt potential barriers. The smoothness of the InAs/InP interface is controlled by the As/P exchange reaction, which itself depends on the substrate preparation and exact conditions of oxide removal prior to growth. The nature of the InAs/air interface is determined by the oxidized InAs surface. We have attempted to examine the effect which this oxide layer has on the quantum well emission by depositing strained "capping" layers of either GaAs or In_{0.5}Al_{0.5}As directly after stopping InAs growth, thus moving the oxide layer further away from the outermost InAs interface.

Figure 2 compares the 4.2 K PL spectrum for 10 Å InAs/InP with PL spectra obtained from "capped" surface quantum wells, 5 Å GaAs/10 Å InAs/InP and 200 Å $In_{0.5}Al_{0.5}$ As/6 Å InAs/InP. In all three cases, the struc-



FIG. 2. 4.2 K PL spectra for (a) 10 Å InAs/InP, (b) 5 Å GaAs/10 Å InAs/InP, and (c) 200 Å $In_{0.5}Al_{0.5}As/6$ Å InAs/InP, showing effect of capping layer.

tures were still observed to be strained after growth. The main thing to note for the GaAs-capped layer is that the PL FWHM has decreased from 93 ± 1 meV for 10 Å InAs/ InP to 63 ± 1 meV for 5 Å GaAs/10 Å InAs/InP, while there is little change in peak position. The PL emission for 200 Å In_{0.5}Al_{0.5} As/6 Å InAs/InP is centered at ~0.97 eV, with a FWHM of 98 ± 1 meV. In this case, the broad lineshape reflects the sensitivity of the InAs quantum well emission to alloy brcadening in the capping layer. Furthermore, the lack of any appreciable shift in PL energy between an InAs surface quantum well and a similar structure capped with either GaAs or InAlAs suggests that any coupling¹² between the confined states in the InAs quantum well and the InAs surface states is not a significant factor.

Figure 3 plots the observed PL energy (left-hand axis) together with the integrated InAs-to-InP PL intensity ratio (right-hand axis) versus total InAs thickness (epitaxial InAs + 5 Å InAs due to As/P exchange). The solid curve has been calculated assuming the PL transition to be e_1 hh₁ within strained InAs quantum wells with InP barriers. The parameters used in the calculation are listed in Table I. The value of conduction-band offset chosen, $\Delta E_c = 0.4$, is that recently used to model¹⁰ InAs quantum wells with InP barriers grown by MOVPE. The theoretical calculation provides quite a good representation of the experimental data, particularly as we have: (i) assumed that the potential wells are exactly rectangular, (ii) not included any strain relaxation for the thicker wells, and (iii) not corrected for the exciton binding energy. The integrated PL intensity in Fig. 3 is observed to reach a maximum value for a total InAs thickness of 15 Å and thereafter decreases with increasing InAs thickness. This decrease in PL inten-



FIG. 3. Variation of PL peak energy (+) and InAs-to-InP PL intensity ratio (\blacksquare) with total InAs layer thickness. Solid line is calculation for strained InAs/InP quantum wells, taking $\Delta E_c = 0.4$.

sity is almost certainly a sign of strain relaxation via the generation of dislocations. *In situ* RHEED measurements of the InAs surface lattice constant have confirmed the onset of relaxation for InAs thicknesses around 17 Å. Our results are in good agreement with PL measurements for InAs/InP single quantum wells grown by MOVPE,⁹ where evidence for strain relaxation has been found for well

TABLE I. Parameters used to calculate e_1 - hh₁ transition energy for strained InAs/InP quantum wells, taking $\Delta E_c = 0.4$.

Parameter	InAs	InP
a_0 (Å)	6.0583	5.8687
C_{11} (10 ¹⁰ Pa)	8.33	10.22
C_{12} (10 ¹⁰ Pa)	4.53	5.76
a (eV)	- 6	- 6.4
b (eV)	- 1.8	- 2
E_g (eV)	0.472 (strained)	1.423
$m_{e}(m_{0})$	0.0325 (strained)	0.085
$m_{\rm hh}$ (m_0)	0.35 (strained)	0.45

thicknesses greater than 5 ML InAs.

In conclusion, we have reported the first observation of PL from InAs surface quantum wells grown on InP by MBE. Since radiative recombination within the InAs layer can be distinguished from PL arising from both bulk and surface defects, this system allows us to monitor the stability of both the InAs/InP and InAs/air interfaces via their influence on the InAs quantum well luminescence. These InAs structures still exhibit efficient carrier capture after ten months exposure to air. A fuller discussion of both the thermal and temporal evolution of the PL lineshape will appear shortly.

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