## Influence of growth temperature on crystalline quality and Raman property of InAs<sub>0.6</sub>P<sub>0.4</sub>/InP<sup>\*</sup>

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InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers grown by low-pressure metal organic chemical vapor deposition (LP-MOCVD) on InP (100) substrate are investigated. The influence of growth temperature on crystalline quality of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayer is characterized by scanning electron microscopy (SEM), Hall measurements, photoluminescence (PL) spectra, and the Raman properties are analyzed by Raman scattering spectrum. The characterization results show that the crystalline quality and Raman property of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers have close relation to the growth temperature. It indicates that 530 °C is the optimum growth temperature to get good quality and properties of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers.

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Due to the wide range of attainable band-gap energy from 0.36 eV to 1.35 eV,  $InAs_xP_{1-x}$  materials are very important for photodiodes<sup>[1,2]</sup>, high electron drift velocity devices, infrared emitting and detecting devices<sup>[3-6]</sup>, 1.3 µm laser diodes<sup>[7]</sup>, etc. The quality of materials is believed to be important for heterostructure device performance. However, in growth process of  $InAs_xP_{1-x}/InP$ materials, using dissimilar materials with different lattice constants and band gaps may lead to poor structures and degrade the electrical and optical properties. The influence can be reduced by the growth of buffer layers, in which step-graded buffer layers are often used to inhibit the dislocations towards the active layer of  $InAs_xP_{1-x}$  materials<sup>[8]</sup>. Optimizing InAs<sub>x</sub>P<sub>1-x</sub>/InP with different growth conditions is very important. However, the influence of growth temperature on properties of  $InAs_xP_{1-x}/InP$  single layer is rarely reported.

In this paper, we investigate the influence of growth temperature on properties of  $InAs_xP_{1-x}/InP$  heterostructures grown by low-pressure metal organic chemical vapor deposition (LP-MOCVD) with the mole fraction of As fixed as 0.6. The scanning electron microscopy (SEM), Hall measurement, photoluminescence (PL) and Raman scattering spectrum are employed to study the properties of the materials.

All the samples were grown on semi-insulating (100) InP substrates in a horizontal reactor by MOCVD at a pressure of  $1.01 \times 10^4$  Pa. Palladium-diffused hydrogen was used for carrier gas at a total flow of 2.0 L/min. The

growth was performed using trimethylindium (TMIn), arsine (AsH<sub>3</sub>) and phosphine (PH<sub>3</sub>) diluted to 10% in H<sub>2</sub> as precursors. The substrates were heated by inductively coupling radio frequency (RF) power, and temperatures were detected by a thermocouple. The growth temperatures of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers were selected between 500 °C and 580 °C. The thicknesses of all the samples were fixed at 1  $\mu$ m. The epilayers grown at 500 °C, 530 °C, 550 °C and 580 °C are named as samples A, B, C and D, respectively.

The general morphologies of the epilayers were investigated by SEM. The electrical property was measured by Hall measurement using van der Pauw technique at room temperature, and the optical property was studied by PL spectra at 77 K. Raman property of the epilayers was discussed by Raman scattering spectrum. The Raman scattering measurements were made in the backscattering geometry with the 488 nm line of an  $Ar^+$  laser at room temperature.

SEM is used to study the surface morphologies of four samples which are shown in Fig.1. A cross-hatched pattern of sample A which is typical characteristic of strained-layer heterostructures is presented in Fig.1(a). For Fig.1(b), the cross-hatched patternsed of sample B become obvious, and the surface becomes rough than that of sample A. From the surface morphologies of samples C and D shown in Fig.1(c) and (d), a lot of big pits appear in the surface of epilayer, and the surface morphology is degraded seriously. It can be seen that the

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surface morphology is degraded with the increase of sample B has





Fig.1 Surface morphologies of  $InAs_{0.6}P_{0.4}$  epilayers grown on InP substrates for (a) 500 °C, (b) 530 °C, (c) 550 °C and (d) 580 °C

Lattice mismatch and growth temperature are two important factors for the surface morphology. The lattice mismatch between the InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers and InP substrate is about 1.94%. When the InAs<sub>0.6</sub>P<sub>0.4</sub> layer is grown on InP substrate, strain energy is created in the InAs<sub>0.6</sub>P<sub>0.4</sub> layer<sup>[9]</sup>. The appearance of cross-hatched patterns is ascribed to the arrays of misfit dislocation networks produced by the large mismatch which have propagated through the structure to the surface<sup>[10]</sup>. The growth temperature mainly provides the migration power for the surface atoms. Hereby, decreasing the growth temperature can reduce the surface atom diffusion and delay the 3-dimensional growth mode<sup>[11]</sup>. Since the epilayers of samples A and B are grown at relatively low temperature, the surface atom migration and nucleation or motion of dislocations in these films are significantly restricted during the growth. For samples C and D grown at relatively high temperature, the compressive strain in the films is relaxed by the introduction of misfit dislocation as the appearance of the big pits. It indicates that the growth of InAs<sub>0.6</sub>P<sub>0.4</sub> at an appropriate temperature is essential for improving the surface morphology of its epilayer.

Hall measurements are used to further characterize the crystalline quality of  $InAs_{0.6}P_{0.4}$  epilayers. The results of Hall measurements are shown in Fig.2. On one hand, the carrier concentration of samples decreases from  $6.756 \times 10^{15}$  cm<sup>-3</sup> to  $3.46 \times 10^{15}$  cm<sup>-3</sup> with growth temperature rising from 500 °C to 530 °C, and then it increases to  $1.27 \times 10^{15}$  cm<sup>-3</sup> with further increasing growth temperature to 580 °C. On the other hand, the mobility of samples increases from 3 973 cm<sup>2</sup>/Vs to 4 661 cm<sup>2</sup>/Vs with increasing growth temperature from 500 °C to 530 °C, and then it decreases to 1 828 cm<sup>2</sup>/Vs with further increasing growth temperature to 580 °C. It is clear that

sample B has the lowest carrier concentration and the highest electron mobility. The changes of carrier concentration and mobility of the samples may be attributed to the growth temperatures. As mentioned above, the growth temperature may restrict the migration and nucleation or motion of dislocations in the epilayers which can serve as scatting centers for electrons and limit the electron mobility. Therefore, the density of misfit dislocations in the epilayer of sample B is the least.



Fig.2 Carrier concentration and mobility for InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers with different growth temperatures

The optical property of the samples is investigated by PL spectrum<sup>[12,13]</sup>. Fig.3 shows the PL spectra of the four samples at 77 K. The band gap energy of the samples is about 0.794 eV which is consistent with the value of 0.795 eV calculated in Ref.[14]. The full width at half maximum (FWHM) values of PL for the four samples are shown in Fig.4, which are 14.6 meV, 11.8 meV, 31.7 meV and 53.6 meV corresponding to the samples A-D, respectively. It is clear that the PL FWHM of sample B is the minimum. Generally, the luminescent efficiency for the band edge emission increases when the purity of the crystal increases<sup>[15]</sup>. Some defects, such as misfit dislocations, are known to quench luminescence due to nonradiative recombination as well as to broaden PL linewidths. So the change of the FWHM of PL is related to the growth temperature of the epilayers. The PL results also indicate that sample B has better crystalline quality.



Fig.3 PL spectra of four samples with different growth temperatures

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Fig.4 FWHM of PL for four samples with different growth temperatures

Fig.5 shows the Raman spectra for the four samples. It can be seen that  $InAs_{0.6}P_{0.4}$  alloys show a typical two-mode behavior, with InAs- and InP-like phonons of the four samples<sup>[16]</sup>. Two main phonon bands of the InAs-like region 200-250 cm<sup>-1</sup> and the InP-like region 300-350 cm<sup>-1</sup> can be observed in the Raman spectra. In the InAs-like optical region, a main peak appears at the energy 235 cm<sup>-1[17]</sup>. This peak is considered to be the InAs-like longitudinal optical (LO) phonon. Meanwhile, an asymmetric broad band appears in the InP-like optical region. This band shows an asymmetric shape, which is due to the shouldering of the InP-like transverse optical (TO) mode<sup>[18]</sup>. Another broad band with Raman shift 150 cm<sup>-1</sup> of all samples is observed. A similar band has been reported by R. Carles<sup>[16]</sup>. This band is identified due to disorder-activated longitudinal acoustic (DALA) phonons.



Fig.5 Raman scattering spectra of the four  $InAs_{0.6}P_{0.4}$  epilayers with different growth temperatures (The inset shows  $\Gamma_a$  and  $\Gamma_b$  which are used in the asymmetry ratio of Raman scattering spectra.)

Raman scattering is an indirect way to characterize the crystalline quality of material. Raman scattering is determined by the overlap integral of electrons, phonons and photons, and the finite phonon mode will lead to the broadening and asymmetry of the Raman scattering line shape<sup>[19]</sup>. Therefore, the FWHM and the asymmetry ratio  $(\Gamma_a/\Gamma_b)$  of Raman scattering spectra can determine the

alloy disorder, and then characterize the crystalline quality of samples.  $\Gamma_a$  and  $\Gamma_b$  are two half-widths at halfmaximum of the InP-like Raman scattering peak, which are shown in the inset of Fig.5. The FWHM and the  $\Gamma_a/\Gamma_b$ of Raman scattering spectra for samples A–D are 14.6 cm<sup>-1</sup>, 13.3 cm<sup>-1</sup>, 18.0 cm<sup>-1</sup>, 21.1 cm<sup>-1</sup> and 12.1 cm<sup>-1</sup>, 12 cm<sup>-1</sup>, 13.1 cm<sup>-1</sup>, 13.6 cm<sup>-1</sup>, respectively. The asymmetry ratios of InP-like Raman spectra of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers are shown in Fig.6. It is clear that the sample B has the minimum FWHM and  $\Gamma_a/\Gamma_b$ . Considering the results with SEM, Hall measurement and PL spectrum, the growth temperature of 530 °C is an effective temperature for improving the crystalline quality of the epilayers on InP substrate.



Fig.6 The asymmetry ratios ( $\Gamma_a/\Gamma_b$ ) of Raman scattering spectra of InAs<sub>0.6</sub>P<sub>0.4</sub> epilayers at different growth temperatures

In summary, in order to investigate the optimum growth temperature, the  $InAs_{0.6}P_{0.4}$  epilayers are grown by LP-MOCVD with growth temperatures of 500 °C, 530 °C, 550 °C and 580 °C, respectively. The influence of growth temperature on crystalline quality and Raman property of  $InAs_{0.6}P_{0.4}$  epilayers is investigated carefully. The results from SEM, Hall measurement and PL spectrum show that the crystalline quality can be improved by using a proper growth temperature. The result from the Raman spectrum can further confirm that the optimum growth temperature of the epilayer is 530 °C. It is expected that the results and analyses may help us to further improve our understanding of the growth mechanism for governing  $InAs_xP_{1-x}$  alloys.

## References

- C. A. Tran, R. A. Masut, J. L. Brenbner, R. Leonelli, J. T. Graham and P. Cova, Journal of Crystal Growth 124, 596 (1992).
- [2] M. D. Birowosuto, A. Yokoo, G. Q. Zhang, K. Tateno, E. Kuramochi, H. Taniyama, M. Takiguchi and M. Notomi, Nature Materials 13, 279 (2014).
- [3] T. J. Lu, S. H. Kim, P. A. Postigo, P. Regreny, C. Seassal and A. Scherer, Electronics Letters 49, 1633 (2013).
- [4] M. A. Meeker, B. A. Magill, T. R. Merritt, M. Bhow-

mick, K. McCutcheon, G. A. Khodaparast, J. G. Tischler, S. McGill, S. G. Choi and C. J. Palmstrøm, Applied Physics Letters **102**, 222102 (2013).

- [5] J. S. Byun, T. J. Kim, S.Y. Hwang. Y. R. Kang, J. C. Park and Y. D. Kim, Thin Solid Films **558**, 438 (2014).
- [6] L. Ji, S. L. Lu, Y. M. Zhao, M. Tan, J. R. Dong and H. Yang, Journal of Crystal Growth 363, 44 (2013).
- [7] S. H. Kim, J. K Lee, H. S. Jeon, S. Callard, C. Seassal, K. D. Song and H. G. Park, Physical Review A 88, 023804 (2013).
- [8] M. K. Hudait, Y. Lin, C. L. Andre, P. M. Sinha, C. A. Tivarus, J. P. Pelz, D. M. Wilt and S. A. Ringel, Applied Physics Letters 82, 3212 (2003).
- [9] R. People and J. C. Bean, Applied Physics Letters 47, 322 (1985).
- [10] M. K. Hudait, Y. Lin and S. A. Ringel, Journal of Applied Physics 105, 061643 (2009).
- [11] Y. Tatsuoka, M. Uemura, T. Kitada, S. Shimomura and S. Hiyamizu, Journal of Crystal Growth 227-228, 266 (2001).

- [12] Z. X. Ma, Q. S. Li, L.C. Zhang, F. Z. Zhao and Y. Ren, Journal of Optoelectronics Laser 23, 819 (2012). (in Chinese)
- [13] B. J. Zheng and W. Hu, Journal of Optoelectronics Laser 24, 1942 (2013). (in Chinese)
- [14] M. Wada, S. Araki, T. Kudou, T. Umezawa, S. Nakajima and T. Ueda, Applied Physics Letters 76, 2722 (2000).
- [15] W. Gao, P. R. Berger, M. H. Ervin, J. Pamulapati, R. T. Lareau and S. Schauer, Journal of Applied Physics 80, 7094 (1996).
- [16] R. Carles, N. Saint-Cricq, J. B. Renucci and R. J. Nicholas, J. Phys. C: Solid State Phys. 13, 899 (1980).
- [17] K. P. Jain, R. K. Soni, S. C. Abbi and M. Balkanski, Physical Review B 32, 100 (1985).
- [18] A. Sayari, N. Yahyaoui, A. Meftah, A. Sfaxi and M. Oueslati, Journal of Luminescence **129**, 105 (2009).
- [19] S. D. Wu, L.W. Guo, Z. H. Li, X. Z. Shang, W. X. Wang, Q. Huang and J. M. Zhou, Journal of Crystal Growth 277, 21 (2005).