Invited

Atomic-Scale Chemical Characterization, Control and Synthesis Semiconductor Surfaces and Interfaces

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The problem of lattice-mismatched or strained layer epitaxy is particularly interesting in view of its promise for the design and fabrication of new electronic and optoelectronic materials. Molecular Beam Epitaxy (MBE) is a powerful crystal growth technology which particularily well suited to the growth of novel artificially structured electronic materials since it provides the method whereby thin films can be fabricated with atomic monolayer-by-monolayer precision. In this technology, the control of the real-time surface chemical reactions literally defines the physical and electronic structure of the epitaxial film. Because the growth front in MBE is open and exposed to the Ultra High Vacuum ambient, surface analytical spectroscopies can be used for dynamic chemical control and characterization of the process. In addition to the interest in the novel properties of these systems, our understanding of the dynamics of the MBE growth promises to strongly evolve through an appreciation of the contribution of strain to the kinetic process. Generally, lattice-mismatched materials grow in registration to the substrate, giving rise to a tetragonal distortion of the local coordination in the specific case of the InAs/GaAs system. This modification of the bonding and symmetry of the growing film has serious ramifications for the physisorbed and chemisorbed species which establish the growth front. The surface transport of In or Ga, the chemical incorporation of As2 or As4, and the details of the terrace and step distributions all show a sensitivity to bond strain and lattice mismatch. The interfacial roughness of heterojunction boundaries in these superlattice systems is likewise affected by local strain and potential variations.

The surface phase diagram which describes the MBE growth of InAs on GaAs, and GaAs on InAs on the (100) surface has a complex dependence on temperature, component flux ratios, surface reconstruction and substrate topography. Recent insights in the Si/Ge system have emphasized the importance of surface free energies in establishing the

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wetting and non-wetting transitions which change the observed growth modes in MBE from layer-by-layer to islanded or cluster growth. Recent calculations have suggested that the surface free energy in the III-V compounds is strongly determined by the stoichiometry, surface reconstruction, temperature and local strain. The surface free energy of in is considerably less than that of Ga, but elemental As can have many values. as can the various reconstructed surfaces of nominal and strained GaAs and InAs. The proper choice of temperature and growth front stoichiometry for the nucleation of InAs on GaAs, the growth of InAs on strained InAs, and the nucleation of GaAs on strained InAs are all different because of these energetics. In addition to these thermodynamical considerations, the kinetics of epitaxy are also impacted by these issues as well as by the presence of segregating or incorporating impurities. Low temperature growth in conjunction with dynamic control of Group III and V fluxes, make possible the synthesis of metastable films and interfaces. Because of the possibility of variation of the surface energetics of this process from terrace to terrace on the surface, we anticipate areal variations in optical, electrical and chemical properties of GaAs/InAs/GaAs Quantum Wells and InAs/GaAs interfaces. In our work, time-resolved RHEED has been used to control In:As stoichiometry at 1:1 and provide uniform GaAs layers with an excess flux of 5 to 15% of the Group V element. The interface for InAs on GaAs is therefore nucleated under metal-stabilized conditions and that for GaAs on strained InAs/GaAs is initiated under Arsenic-stabilized conditions. We also use Reflection Mass Spectrometry(RMS) to follow and compensate the substantial changes in sticking coefficient of Group III and V species during strained layer and Quantum Well growth. In this paper, we report several new results on the systematic study of controlled variations in MBE growth front stoichiometry. and their effect on the photoluminescence, polarized optical absorption, and cathodoluminescence of thin pseudomorphic Quantum Wells with InAs layer thicknesses of 1 to 8 monolayers grown on GaAs(100) substrates. The data is correlated with high resolution TEM micrographs taken with a 200 keV ABT 002B microscope.

Strained InAs is grown on GaAs (100) within the temperature window of 325- 435 C. The surface lattice constant of the InAs films is within 0.03% of that of the GaAs buffer layer. TEM results show low dislocation density material for 1 through 4 monolayer InAs films. QW thickness values calibrated by RHEED and RMS are compared with TEM micrographs. Optical characterization of the QW structures shows substantial variation with growth conditions. PL peak positions shift systematically to longer wavelengths and

peak widths broaden with increasing QW thickness. The optical absorption spectra obtained using a polarized multiple reflectance geometry resolve, for the first time, 1E-1H and UE-1H transitions for 1 through 4 monolayer InAs QW structures. The splitting between the two transitions increases with thickness as anticipated. The curves are modeled with envelope calculations using a partition ratio of 72/28 for the conduction/ valence band discontinuities. The peak of the PL lines shifts with excitation power density. The maximum of the PL occurs at higher energy than the optical absorption spectra, and the FWHM at 4.2 K for the 3 ML QW varies from 8 to 45 mev with growth conditions. The data is interpreted with a model with variable QW thicknesses on the scale of the exciton diameter (~ 150 Å). The maximum thickness variation for the 4 ML QW is less than 1 ML.

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