GROWTH AND CHARACTERIZATION OF Si-Ge MULTILAYER STRUCTURES ON Si(100)

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1. INTRODUCTION

Heteroepitaxy of Si$_{1-x}$Ge$_x$ on Si by molecular beam epitaxy (MBE) has been a subject of increasing interest in recent years (1). In spite of the large lattice mismatch between Si and Ge, high quality films of Si$_{1-x}$Ge$_x$ covering the entire range of alloy compositions have been grown utilizing strained-layer epitaxy methods. In this heteroepitaxial system, the misfit strain has dramatic effects on the band structure of the materials and can be used for tailoring their optoelectronic properties. This has been exploited for production of novel Si-based devices such as infrared photodetectors (2) and modulation-doped field-effect transistors (3). For these applications alternating layers of Si and Si$_{1-x}$Ge$_x$ were grown to form a superlattice structure. This has the advantage of increasing the total thickness of strained material attainable before plastic relaxation occurs. As the Si-Ge MBE technology matures, more complex heterostructures are being synthesized. For example, growth of Si$_{1-x}$Ge$_x$/Si Fibonacci superlattices has been reported (4). Such a superlattice, built according to the Fibonacci sequence is quasi-periodic and exhibits unusual structural properties (5). Resonant tunneling of holes across a double-barrier of Si buried in a Si$_{1-x}$Ge$_x$ layer has also been reported (6). Recently, growth of artificial crystals of Si$_m$Ge$_n$ made of alternating layers of pure Si and Ge has been demonstrated (7). The study of such materials is of much interest both from a basic physics point of view and for designing devices with unusual optical properties (8). In this paper, we report the growth of various Si/Si$_{1-x}$Ge$_x$ heterostructures including thick (~0.5 μm) periodic and Fibonacci strained-layer superlattices and ultra-thin (~10 nm) multilayers of Si$_m$Ge$_n$ ($m, n < 6$). The structural properties of these films are investigated by X-ray diffraction and Raman scattering spectroscopy.

2. EXPERIMENTAL

All the epitaxial layers discussed here were produced in a Vacuum Generators V80 MBE system. Both Si and Ge fluxes were obtained by-gun evaporators. Deposition rates were carefully controlled by Sentinel III (9) optical sensors and flux shutters were computer controlled. The epitaxial films were grown at 500°C on 100 mm (100) Si wafers. Details on the substrate preparation and growth methodology can be found elsewhere (10). Deposition rates of 0.2-0.5 nm/s were used for the growth of the thick Si$_{1-x}$Ge$_x$/Si strained-layer superlattices. Rates were reduced to about 0.02 nm/s for the synthesis of the thin Si$_m$Ge$_n$ structures. Also, for the latter, a portion of the wafers was capped by a 10-15 nm Si protective layer.

The structural properties of the various heterostructures were studied using cross-sectional transmission electron microscopy (XTEM), double-crystal X-ray diffraction (DCD) and Raman spectroscopy. In DCD, 400 rocking curves were recorded in a non-dispersive geometry using a (100) Si first crystal and Cu Kα radiation ($\lambda=0.154\text{nm}$). The Raman spectra were measured in a 90° geometry with the sample (100) surface inclined at an angle of 12.3° to the incident light. The large refractive index of these materials makes this effectively a backscattering experiment inside the crystal. The samples were placed in an helium atmosphere at 295 K and the spectra were excited with 500 mW of 457.9 nm argon laser light and analyzed with a Spex 14108 double monochromator. Further details on the Raman measurements can be found elsewhere (11).
3. THEORY

3.1. X-ray diffraction

In the present study, the kinematical theory of X-ray diffraction has been used to analyze the experimental results \((12-13)\). In the kinematical approximation, the diffracted amplitude is proportional to the Fourier transform of the spatial distribution of layered materials. The diffracted intensity \(I(\theta)\) (neglecting absorption) about the 400 Bragg reflexion \(\theta_B\) for a superlattice consisting of \(N\) periods of \(Si_{1-x}Ge_x\) Si of thickness \(t_{Si}\) and \(t_{Ge}\) is given by

\[
I(\theta) = \frac{\sin^2 NK(t_{Si}\Delta\theta_{Si} + t_{Ge}\Delta\theta_{Ge})}{\sin^2 K(t_{Si}\Delta\theta_{Si} + t_{Ge}\Delta\theta_{Ge})} \left[ \left( \frac{F_{Si} \sin K t_{Si} \Delta\theta_{Si}}{V_{Si} \Delta\theta_{Si}} \right)^2 + \left( \frac{F_{Ge} \sin K t_{Ge} \Delta\theta_{Ge}}{V_{Ge} \Delta\theta_{Ge}} \right)^2 \right] + \left( 2\cos K(t_{Si}\Delta\theta_{Si} + t_{Ge}\Delta\theta_{Ge}) \frac{F_{Si} F_{Ge}}{V_{Si} V_{Ge}} \sin K t_{Si} \Delta\theta_{Si} \sin K t_{Ge} \Delta\theta_{Ge} \right),
\]

where \(K = \pi \sin 2\theta_B / \lambda \sin \theta_B\), \(\Delta\theta_{Si,Ge} = \theta - \theta_B + \varepsilon_{Si,Ge} \tan \theta_B ; F_{Si}, V_{Si}, \varepsilon_{Si}\) and \(F_{Ge}, V_{Ge}, \varepsilon_{Ge}\) are the structure factor, volume of unit cell and perpendicular strain (with respect to bulk Si lattice constant) of the Si and \(Si_{1-x}Ge_x\) layers, respectively. From Eq. \([1]\), the diffracted spectrum from a superlattice consists of a series of satellite reflections of angular spacing \(\Delta\theta_n\) given by

\[
\Delta\theta_n = \frac{\lambda \sin \theta_B}{(t_{Si} + t_{Ge}) \sin 2\theta_B},
\]

with the zero order reflection centered at

\[
-\Delta\theta_0 = \frac{t_{Si} \varepsilon_{Si} + t_{Ge} \varepsilon_{Ge}}{t_{Si} + t_{Ge}} \tan \theta_B.
\]

Due to the limited thickness of the superlattice, the satellites have a finite width \(\Delta\theta_p = \Delta\theta_n / N\) and the spectrum also exhibits secondary peaks of spacing \(\Delta\theta_p\). Satellite intensity simulations using Eq. \([1]\) were performed to obtain the thickness and strain of individual layers \((13)\). Eqs \([1]\)-\([3]\) can also be applied in the case of thin multilayers. However, because of the very short period of these structures, the DCD spectrum will normally only display the zero order reflection which can be used to obtain the average composition and total thickness of the structure.

The \(j\)th generation Fibonacci superlattice is obtained from the recurrence relationship \(S_j = S_{j-2}S_{j-1}\) where \(S^1 = \{A\}\) and \(S^2 = \{BA\}\), with \(A\) and \(B\) two different layer units. For a \(Si/\)Si\(_{1-x}Ge_x\) Fibonacci superlattice, the position of the satellites is obtained from the following expression:

\[
\sin \theta_{m,n} = \frac{2\lambda}{\langle a \rangle} + \frac{\lambda(m\tau + n)}{2t_F},
\]

where \(\tau = (1 + \sqrt{5})/2, t_F = t_{Si} + t_{Ge}\) and \(\langle a \rangle\) is the average lattice constant. Eq. \([4]\) shows that a Fibonacci superlattice exhibits a complex spectrum consisting of an infinity of peaks that are subsequently labeled \((m,n)\).

3.2. Raman scattering

If light of wavelength \(\lambda\) and momentum \(k_i\) is incident on the superlattice surface, the large refractive index \(\eta\) of these materials ensures that the momentum \(k_i\) inside the crystal is essentially normal to the surface. If the scattered light has momentum \(k_s\) inside the crystal,