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Raman shifts and photoluminescence of the InSb nanocrystals ion beam-synthesized in buried SiO_2 layers

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ABSTRACT

The InSb nanocrystals embedded in buried SiO_2 layers were obtained by the In^+ and Sb^+ ion implantation into the SiO_2 layers thermally grown on Si wafers followed by the hydrogen transfer of a Si film and high-temperature annealing at 800–1100 °C. Transmission electron microscopy, Raman spectroscopy and photoluminescence were used to study the sample properties. The spherical-shaped InSb nanocrystals distributed close to the implanted atom profiles were obtained in buried SiO_2. The InSb TO- and LO-like phonon modes at 187 cm^{-1} and 195 cm^{-1} were observed in the Raman spectra. The effect of both phonon quantum confinement and stresses on the phonon frequency shift was calculated. The TO-LO splitting obtained from the ion-beam synthesized InSb nanocrystals was 3 cm^{-1} less than that from the unstressed bulk monocrystalline InSb. The obtained effect is discussed in the frames of decreasing the transverse effective charge, as well as that of the surface phonon influence. The photoluminescence peak at 1524 nm (0.81 eV) was seen in the low-temperature PL spectra from the samples annealed at 900–1000 °C. Its energy position corresponds to the localized charge carrier energy in the InSb nanocrystals.

1. Introduction

The incorporation of A_3B_5 semiconductor performance advantages in the mature complementary metal-oxide-semiconductor technology is a way to realize the silicon-based optical interconnection. From this point of view, InSb nanostructures attract particular interest because of their excellent electrical and optical properties resulting from their very small electron and hole effective masses (0.015 m_0 and 0.39 m_0, respectively) and very high permittivity (17.8). This provides the largest exciton Bohr radius (65.6 nm) among its quantities in all semiconductors. It allows observing the quantum-confinement effect in a wide range of nanocrystal sizes. Nanometer-sized InSb nanocrystals demonstrate their photoluminescence in the region 1.6–1.8 μm [1-3]. A large optical absorption edge blue shift from the infrared to ultraviolet spectral region was obtained from the InSb nanocrystals embedded in thin SiO_2 films [4]. The thin SiO_2 layers containing InSb nanocrystals also show an enhanced nonlinear optical coefficient [5]. InSb possess the narrow band gap (0.17 eV) and the high electron and hole mobilities at room temperature (78,000 and 750 $cm^2V^{-1}s^{-1}$, respectively) [6]. That makes it suitable for short-channel transistor applications, too.

The nanostructures properties depend on the preparation method. The InSb quantum dots produced by molecular-beam epitaxy are large in their dimensions and they possess inhomogeneous stresses [7-9]. The large lattice mismatch between Si and InSb is the base problem in preparing InSb/Si hetero-structures with epitaxial techniques. The radio-frequency sputtering [10,11,4] and ion-beam synthesis [12,13] methods were also used to fabricate the InSb nanocrystals in silicon dioxide films grown on silicon substrates. In the case of radio-frequency sputtering, InSb nanocrystals, deposited at a temperature of 300–400 °C, have their outer shell at the InSb/SiO_2 interface in which the In-Sb bond length differs from those in bulk InSb [11]. At that, the diffuse interface between an InSb nanocrystal and the surrounding SiO_2 matrix can result in a decrease in the nanocrystal real volume and the suppression of quantum-confinement effect [11].

The ion-beam synthesis is a method in which new phases formation occurs in ion-irradiated layers. The ion-beam synthesis consists of the ion implantation and subsequent high-temperature annealing of ion-implanted samples. These two steps are the standard manufacturing operations in the current silicon technology. Previously, we studied the

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InSb nanocrystal formation conditions, as well as the structural properties of the InSb nanocrystals synthesized within the buried SiO_2 layers and at the Si/SiO_2 bonding interface of silicon-on-insulator (SOI) structures [12,13]. The two-stage mechanism of InSb phase formation, including the antimony precipitation followed by the In and Sb atom diffusion, was obtained. Under these conditions, the InSb phase formation occurs during the ion-implanted In and Sb atom diffusion toward the nucleation centers at the temperature ≥ 800 °C that is higher than the InSb melting temperature. It means that InSb nanocrystals have to grow as a liquid phase followed by the crystallization under cooling. The difference of the atom density in liquid (6.43 g/cm^3) and solid (5.77 g/cm^3) InSb phases, as well as different thermal expansion coefficients of InSb nanocrystals and the surrounding matrix, can result in the respective In-Sb bond stresses. Along with the quantum confinement effect, the bond stresses in the nanocrystals can also result in modifying their optical properties. In the present paper, the optical properties of InSb nanocrystals ion-beam synthesized in buried SiO_2 layers, namely, optical phonon mode Raman spectra, as well as photoluminescence (PL) spectra, were experimentally studied. In order to separate the quantum-confinement effect from the In-Sb bond stress effect on the Raman frequency shift in InSb nanocrystals, the Raman spectra were calculated as a function of the nanocrystal size within the phonon confinement model [14].

2. Experimental

The InSb nanocrystals in the buried SiO_2 layers of SOI structures were formed by a technique described elsewhere [13]. In^+ and Sb^+ ions at the energy of 200 keV and doses 8.0×10^{15} cm^{-2} were embedded into the 300 nm thick SiO_2 films thermally-grown on p-type, (100)-oriented Si wafers by oxidation in a wet oxygen ambient at the temperature of 1100 °C. The ion implantation parameters produced Gauss-like In and Sb atom distributions with the peak concentrations $\sim 1.2 \times 10^{21}$ cm^{-3} at a depth of about 110 nm below the top surface. H_2^+ ions at the energy of 140 keV to the dose of 2.0×10^{16} cm^{-2} were incorporated into another Si wafer. Then, in a vacuum chamber, the wafers were joined with the implanted sides, simultaneously detaching the Si layer over the internal hydrophobic surface created by H_2^+ ions and transferring this silicon layer onto the first wafer. The transferred silicon film thickness was about 600 nm. Then the wafers were cleaned and cut in small pieces which were heat-treated at a temperature of 500 – 1100 °C for 0.5 h in the N_2 ambient. Raman spectroscopy, transmission electron microscopy (TEM) and PL were employed to study the produced layer properties.

The TEM analysis was carried out with a JEM-2200FS microscope at the accelerating voltage of 200 keV with the point resolution at 0.19 nm.

The Raman spectra were excited by an Ar laser line with a wavelength of ~514.5 nm at room temperature. The probing spot size was 10 µm. The laser beam power at the sample surface was about 2 mW. A Horiba Jobin Yvon system equipped with a T64000 triple monochromator spectrometer was used. The spectral resolution was not worse than 2 cm^{-1}. A light-sensitive silicon matrix cooled by liquid nitrogen was the detector. The measurements were performed in the backscattering geometry with the incident radiation polarization vector directed along the ⟨011⟩ silicon crystallographic direction. The scattered light was measured in the ⟨01$\bar{1}$⟩ polarization. The chosen geometry allowed the maximum suppression of the Raman signal from the silicon substrate.

The PL over the wavelength range 330–850 nm was excited by a He-Cd laser with the wavelength 325 nm and a laser beam power of about 20 mW at room temperature. The photomultiplier tube was used as a detector. A laser diode emitting at wavelength 488 nm was used to excite the PL in the infrared spectral region. The sample was cooled in liquid helium at 10K in a cryostat at a temperature stability of ± 0.5K. A monochromator equipped with a 600 lines/mm diffraction grating and a cooled InGaAs detector was used. The data were corrected for a spectral response of the detector.

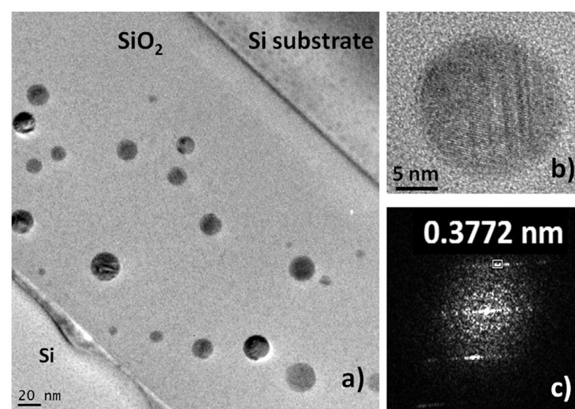

Fig. 1. (a) A cross-sectional TEM image, (b) high-resolution TEM image and (c) an electron-diffraction pattern of an individual nanocrystal obtained from the In^+ and Sb^+ ion-implanted sample after the annealing at 1000 °C.

3. Results

Beginning with the annealing temperature of 800 °C, the nanocrystal formation in the buried SiO_2 layer occurred. Both the nanocrystal distribution and their dimensions depended on the annealing temperature. It was described elsewhere in details [13]. As the annealing temperature grew from 800 to 1100 °C, the mean nanocrystal diameter increased from 5-15 to 13-25 nm respectively, and their distribution changed nonmonotonically [13]. The nanocrystals formed after the annealing at the temperature of 1000 °C are shown in Fig. 1a. The nanocrystals have a spherical shape, and their distribution is close to the ion implanted profiles. The nanocrystal size distribution is close to bimodal with a typical average nanocrystal diameter of about 13 and 25 nm, respectively. The largest nanocrystals are generally located close to the top Si/SiO_2 interface. That can be explained by the higher In and Sb atom concentration and their enhanced diffusion to the nanocrystal nucleus stimulated by excess defects generated in the top SiO_2 layer during ion implantation [15]. A high-resolution image obtained from a single nanocrystal in the SiO_2 layer is shown in Fig. 1b. The respective Fourier transform is presented in Fig. 1c. The interplanar spacing of the nanocrystal lattice is 0.377 nm and that corresponds to the (111) InSb lattice orientation.

The Raman and PL measurements were carried out both from the as-implanted and annealed samples. Before the measurements, the top silicon layer was removed by the 25% ammonia solution pickling at the temperature of 40 °C under gauging. The Raman spectra, measured from the silicon substrate, as well as from the as-implanted SiO_2 film and those subsequently annealed at 1000 °C, are presented in Fig. 2a. The main Raman peak position at 520.4 cm^{-1}, that corresponded to the vibrational mode in the monocrystalline silicon matrix, is invariable irrespective of the ion-implanted SiO_2 film presence on the silicon substrate. However, the SiO_2 covered samples show this peak intensity 1.8 times higher than that from the native silicon substrate. No changes in the intensity were obtained from the ion-implanted samples subsequently annealed at 1000 °C. The Raman line width is also invariable in all cases. It is about 4.5 cm^{-1}. The obtained results suggest that both the as-implanted and subsequently annealed SiO_2 films are equally transparent. An increase in the 520.4 cm^{-1} peak intensity of the SiO_2-covered samples is explained with a lower light reflection from the SiO_2 surface, as compared to that from the silicon surface. In other words, the SiO_2 layer acts as an anti-reflective covering. The 300 cm^{-1} peak defined by the scattering at the double acoustic phonon frequency in the silicon substrate is seen too. A peak around 190 cm^{-1} arises in the

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