

Observation of temperature-dependent heavy- and light-hole split direct bandgap and tensile strain from $\text{Ge}_{0.985}\text{Sn}_{0.015}$ using photoreflectance spectroscopy

Hyun-Jun Jo ^a, Geun Hyeong Kim ^a, Jong Su Kim ^{a,*}, Mee-Yi Ryu ^b, Yung Kee Yeo ^c, Thomas R. Harris ^{c,1}, John Kouvetakis ^d

^a Department of Physics, Yeungnam University, Gyeongsbuk, 712-749, Republic of Korea

^b Department of Physics, Kangwon National University, Kangwon-do, 200-701, Republic of Korea

^c Department of Engineering Physics, Air Force Institute of Technology, OH, 45433, USA

^d Department of Chemistry and Biochemistry, Arizona State University, AZ, 85287, USA

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ABSTRACT

Temperature- (T -) dependent photoreflectance (PR) measurements have been made for the tensile-strained, undoped $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film grown on n -Si substrate by ultra-high vacuum chemical vapor deposition method. The PR spectra at room temperature consist of two signals at around 0.739 and 1.022 eV, which are assigned to the direct transitions from conduction Γ valley to valence and spin-orbit split-off bands, respectively. The T -dependent PR measurements show tensile-strain split direct bandgap transitions from the Γ valley to the light-hole (E_{LH}) and heavy-hole (E_{HH}) bands at energies of 0.772 and 0.803 eV at 12 K, respectively, which are not usually observable from the photoluminescence measurements for relatively high Sn content $\text{Ge}_{1-y}\text{Sn}_y$ samples. The PR signals for both HH and LH bands are blue shifted and their intensities decrease with increasing temperature, but both LH and HH PR signals persist through 240 K and only one HH PR signal is observed at room temperature. It has been observed that the separation energy between the E_{HH} and E_{LH} increases as T decreases, which clearly indicates an increase in tensile strain as T decreases. From the analysis of the T -dependent separation energy between the E_{LH} and E_{HH} , the T -dependent tensile strain in the $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film was obtained, which might not be easily measured using the X-ray diffraction method.

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1. Introduction

Silicon has been used for its electronic device and integrated circuit applications for many decades, but it is a very poor light emitting source due to its indirect bandgap nature. Therefore, in recent decades, intensive research has been pursued to develop Si- and Ge-based direct bandgap semiconductors such as Ge, $\text{Ge}_{1-y}\text{Sn}_y$, and $\text{Ge}_{1-x-y}\text{Si}_x\text{Sn}_y$ films grown on Si or Ge substrates to expand its functionalities well beyond electronics to possible new optoelectronic devices [1–8]. Many research groups have reported the photoluminescence (PL) results of the tensile strained Ge and $\text{Ge}_{1-y}\text{Sn}_y$ alloys [9–13], and the electroluminescence (EL) emissions of

direct bandgap from various Ge- and $\text{Ge}_{1-y}\text{Sn}_y$ -based diode structures [6,11]. In spite of the great recent progress in the crystal growth methods of $\text{Ge}_{1-y}\text{Sn}_y$ alloys on Si substrate, the as-grown alloys often required a post-growth annealing treatment to improve its crystallinity. However, the annealed $\text{Ge}_{1-y}\text{Sn}_y/\text{Si}$ alloys could have a biaxial tensile strain upon cooling [14,15].

An enhancement in PL intensity has been observed for the tensile strained Ge film in the near-infrared regime compared to bulk Ge due to the shrinkage of the direct bandgap [16,17]. In addition, tensile strain causes the splitting of light-hole (LH) and heavy-hole (HH) valence bands of $\text{Ge}_{1-y}\text{Sn}_y$ [12,18]. Only a few researchers have reported the separation between LH and HH valence bands for the Ge and the $\text{Ge}_{1-y}\text{Sn}_y$ films at room temperature through photoreflectance (PR) spectroscopy so far [12,18,19]. Although the direct bandgap energies of strained $\text{Ge}_{1-y}\text{Sn}_y$ films grown on Si occurring between the conduction band Γ valley and LH- and HH-valence bands could be calculated as a function of in-

* Corresponding author.

E-mail address: jongsukim@ynu.ac.kr (J.S. Kim).

¹ Present address: Air Force Research Laboratory, Wright-Patterson AFB, OH 45433.

plane strain, the theoretical calculation of the tensile strain is difficult after the annealing process. In general, the strain is calculated by using rocking curves of a high resolution X-ray diffraction (HRXRD), but the measurements are difficult at low temperatures. Alternatively, Thomas et al. reported temperature dependent strain on a ZnSe/GaAs using PR measurements [20]. Although several PR papers for the strained $\text{Ge}_{1-y}\text{Sn}_y/\text{Si}$ alloys have been reported at room temperature, there have hardly been any reports on the temperature dependent tensile strain and separation energy between HH- and LH-valence bands for the $\text{Ge}_{1-y}\text{Sn}_y/\text{Si}$ alloys using the PR measurements. In this paper, the PR results of direct bandgap transition energies between Γ conduction valley and LH- and HH-valence bands in the biaxial tensile-strained $\text{Ge}_{0.985}\text{Sn}_{0.015}/\text{Si}$ are reported as a function of sample temperature, as well as the calculated results of the temperature-dependent tensile strain from the energy separation between LH and HH optical transitions.

2. Experiment

The undoped $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film was grown by using an ultra-high vacuum chemical vapor deposition method directly on n -type Si substrate with $\rho = 0.005 \text{ } \Omega\text{-cm}$ via deuterated stannane (SnD_4) assisted reactions of pure digermane (Ge_2H_6) at an optimum growth temperature of 375°C and at a pressure of 0.4 Torr. The content of 1.5% Sn and a film thickness of 620 nm were estimated from Rutherford backscattering (RBS) measurements. The as-grown $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film was annealed at 750°C for 30 min in a hydrogen environment, which resulted in the improved crystallinity of the sample greatly as observed from the narrowing of the XRD peak (not shown here). Apparently, the annealing process reduces the levels of threading defects. The strain in the as-grown $\text{Ge}_{0.985}\text{Sn}_{0.015}$ epitaxial layer was initially slightly compressive. However, the sample became fully relaxed after the annealing treatment, and then changed to slightly tensile strain upon cooling to room temperature due to the large difference in thermal expansion coefficients between $\text{Ge}_{0.985}\text{Sn}_{0.015}$ and Si [21,22]. The lattice constants were estimated using 224 reciprocal space maps measured by HRXRD, yielding $a = 5.6801 \text{ } \text{\AA}$ and $c = 5.6583 \text{ } \text{\AA}$. The latter is similar to the value $c = 5.6587 \text{ } \text{\AA}$ obtained from 004 on-axis peaks. The final residual tensile strain value was estimated to be 0.20% according to HRXRD measurement. More detailed descriptions of $\text{Ge}_{1-y}\text{Sn}_y$ crystal growth and annealing temperatures can be found from references [9] and [23], and those of RBS and HRXRD measurements can be obtained from references [23] and [24].

PR measurements were performed using a 637 nm laser diode as an excitation source with the excitation intensity of $400 \text{ mW}/\text{cm}^2$, and the PR signals were detected by an extended InGaAs detector (1000–2400 nm). The probe light was a monochromatic beam obtained from a tungsten-halogen lamp dispersed through a monochromator. PR spectra were measured at temperatures ranging from 12 to 300 K. A more detailed description of the PR setup can be found in Ref. [25].

3. Results and discussion

The PR spectra of undoped as-grown and annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}$ films grown on n -Si substrate are shown in Fig. 1. The inset of the figure shows the PR spectrum of a p -type bulk Ge, which was used as a reference sample. The measurements were performed at 300 K with a laser excitation intensity of $400 \text{ mW}/\text{cm}^2$. The PR spectrum for the as-grown undoped $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film exhibits a broad PR spectrum (black color), which may indicate slightly poor crystal quality for this film. On the other hand, the PR

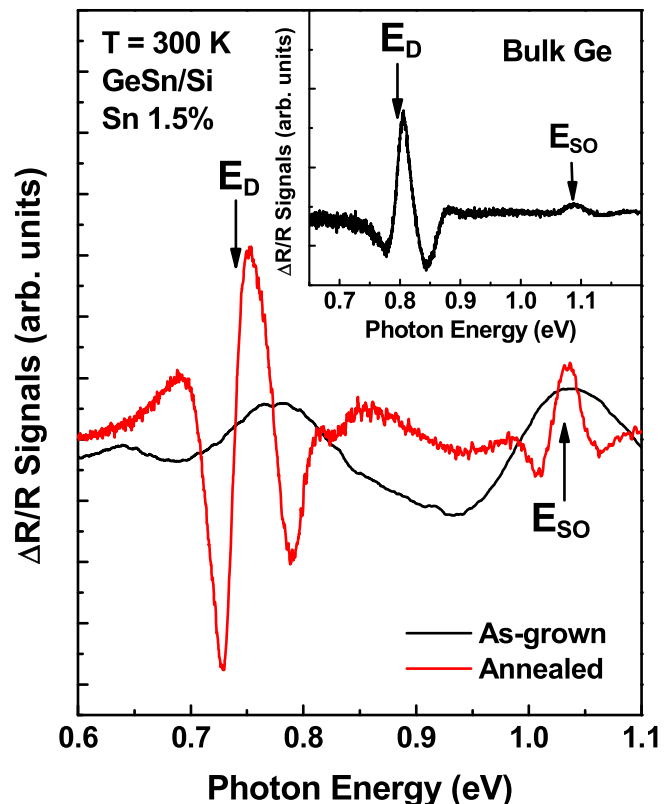


Fig. 1. Photoreflectance spectra of as-grown and annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}/\text{Si}$ thin films measured at room temperature. The inset is the PR spectrum of a bulk Ge.

spectrum of the thermally annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film (red color) shows very clear and specific features of spectrum. This result indicates that the crystal quality of the $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film improves greatly after thermal annealing. These PR observations agree with the fact that in general, the PR spectra of doped or poor quality semiconductors are broad, and the PR spectra of undoped, good quality semiconductors show sharp and specific features.

The PR spectrum of annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}/n\text{-Si}$ is mainly composed of two transition signals located at around 0.75 and 1.03 eV. For the bulk Ge sample, the direct bandgap transition PR signal was obtained at around 0.80 eV as shown in the inset of Fig. 1, which is in good agreement with the direct transition energy (E_D) of the Ge [26]. Also, the direct transition from the Γ valley in conduction band to the spin-orbit split-off band (E_{SO}) was observed approximately at 1.09 eV for this bulk Ge sample. In light of the positions of the E_D and E_{SO} transition signals for the bulk Ge sample, the PR signals at around 0.75 and 1.03 eV obtained from the undoped annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}$ film are attributed to the direct bandgap transition and the spin-orbit split-off band transition, respectively. The smaller direct bandgap transition energy of the undoped $\text{Ge}_{0.985}\text{Sn}_{0.015}$ sample ($E_D \approx 0.75 \text{ eV}$) compared to the bulk Ge sample ($E_D \approx 0.8 \text{ eV}$) is attributed to the 1.5% Sn concentration and 0.20% tensile strain induced by the thermal expansion mismatch between $\text{Ge}_{0.985}\text{Sn}_{0.015}$ and Si substrate during cooling to room temperature. These direct transitions for Ge and various $\text{Ge}_{1-y}\text{Sn}_y$ alloys have also been reported by several groups [14,15,27–31]. It is also worthwhile to point out that, the indirect bandgap related PR signal was not observed either from the bulk Ge or the annealed $\text{Ge}_{0.985}\text{Sn}_{0.015}/n\text{-Si}$ samples. In the case of indirect bandgap semiconductors, the PR signal is usually dominated by signals from the direct bandgap transition because of relatively inefficient indirect bandgap related re-emission. Therefore, it is often difficult to

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