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Effect of annealing treatment and nanomechanical properties for multilayer $Si_{0.8}Ge_{0.2}$–Si films

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ABSTRACT

Multilayered silicon–germanium (SiGe) films consisting of alternating sublayers with different mechanical properties have been epitaxially deposited by an ultra-high vacuum chemical vapor deposition (UHV/CVD) system. We report engineering of the mechanical properties of SiGe multilayer films by a commercial nanoindenter. From annealing treatment, it consists of an *ex situ* thermal treatments in furnace (600 °C) and rapid thermal annealing (800 °C) system. Subsequent roughness and microstructure of SiGe multilayer films were characterized by means of atomic force microscope (AFM) and transmission electron microscopy (TEM).

The annealing treatment not only produced misfit dislocations as a significant role in the critical pile-up event but also promoted hardness. The hardness of the films increased slightly and then gradually achieved a maximum value (from 12.6 ± 0.4 GPa to 14.2 ± 0.7 GPa) with increasing annealing temperature. This may be due to the relaxation effect from thermal annealing and is potential to provide the reliability behaviours to design periodical SiGe multilayer structure in further.

1. Introduction

Silicon–germanium (SiGe) is a very attractive semiconductor material which serves adjustable band gaps, enhanced carrier mobility, and higher dopant solubility than that of pure Si. Due to these outstanding properties, SiGe alloys is competitive with III–V alloys and its compatibility with Si processing makes it low cost to integrate with highspeed Si-based technologies applications [1–3]. However, there are several limiting factors for its applications such as the long growth time, high material consumption, rough surface, and partial strain relaxation [4,5]. Above all, the implication of the degree of Si/Ge interdiffusion in microstructure is difficult to decide because it was caused by many factors including Ge content, temperature, and so on [6,7]. Notice that, the lattice mismatch between SiGe and Si substrate can introduce crystal defects such as misfit dislocations and threading dislocations in the microstructure, which results in the degradation of device qualities, i.e., these misfit dislocations may exist at several interfaces and thread through the layers. In general, the abruptness of the interface misfit dislocations has the edge component in the active areas of device [8,9]. The high quality of the SiGe is required for devices application, that is to say, crystal defects and non-uniform composition are undesirable [10,11].

In recent, multilayer films with a large number of repeated layers have been intensively studied in the last decade. The enhancement of the films performance has been achieved by combining layers with different mechanical properties. The multilayers SiGe/Si is useful as model systems to improve the films hardness. It was found that the hardness of these multilayer films was significantly improved compared with that of one monolayer. In contrast, multilayer films in the SiGe/Si system not only has attracted widely interest but also plays a critical role in the monolithic integration of Si-based micro- and opto-electronic devices [12]. Therefore, the relation between hardness enhancement mechanism and the restriction of dislocation movement in multilayer SiGe/Si films is focused. In this case, indentation techniques can be used to delaminate films from the substrates and measure mechanical properties such as elastic modulus *E* and hardness *H* [13]. As ours knowledge, the annealing treated SiGe films is still not yet understood in the periodical structure to undergo a significant indentation process. The annealing treatment is potential to provide the reliability behaviours of periodical SiGe multilayered structure by using a quantitative method.

In this paper, the multilayer structure was deposited by an UHV/CVD system. This system has many benefits compared with

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other deposition technologies. For example, the low growth rates allow atomic layer precision in the growth of SiGe films. Steep doping as well as stable Ge profiles is required for high speed devices. In addition, UHVCVD avoids the segregation of Ge due to the low thermal budget. Therefore, the UHUCVD deposited SiGe shows high crystalline quality, smooth surfaces, and a low density of misfit dislocations [#2] and is different from sputtered one, which is in an amorphous form [#3]. We present experimental investigations during annealing under the influence of internal mechanical properties of period SiGe multilayer on Si substrate. It is demonstrated that the quantitative stress was inside the layer by means of nanoindentation technique. As a consequence, the annealing effect from the production of dislocations in the microstructure was evidenced.

2. Experimental procedure

The deposition process of SiGe multilayer films on the Si substrate was described as the following: (i) the *p*-type Si(1 0 0) wafers were prepared by a standard Radio Corporation of American (RCA) clean and a HF:H_2O (1:50) bath for 15 s. The samples were simultaneously introduced into load-lock chamber of ultra-high vacuum chemical vapor deposition (UHV/CVD) system. Afterwards, a 120-nm thick $Si_{0.8}Ge_{0.2}$ layer was deposited at 500 °C for ∼43 min from pure SiH_4 (in 85 sccm) and GeH_4 (in 15 sccm) mixing (Ge concentration $x = 20$), the rate of deposition is 2.8 nm/min and the vacuum is achieved at 10^{-7} mbar. (ii) A 10-nm-thick Si buffer layer was deposited on the Si substrate at 500 °C for 100 min from pure SiH_4 (in 85 sccm) gas and the rate of deposition is 0.1 nm/min and the vacuum is achieved at 10^{-7} mbar. Meanwhile, the SiGe and Si buffer layers were deposited following four period cycles, and the structures were terminated by a 10-nm-thick Si cap layer (ca. 530 nm in the total thickness). (iii) In the

Table 1
Variation of average surface roughness (R_a) and root-mean-square surface roughness (R_{ms}) at room temperature and various annealing temperatures.

Sample	Sample condition	Average roughness, R_a (nm)	Root mean square roughness, R_{ms} (nm)
(a)	Single-layer-SiGe RT	0.2	0.4
(b)	Multilayer-SiGe RT	3.1	3.6
(c)	Multilayer-SiGe (600 °C)	3.2	3.9
(d)	Multilayer-SiGe (800 °C)	3.5	4.1

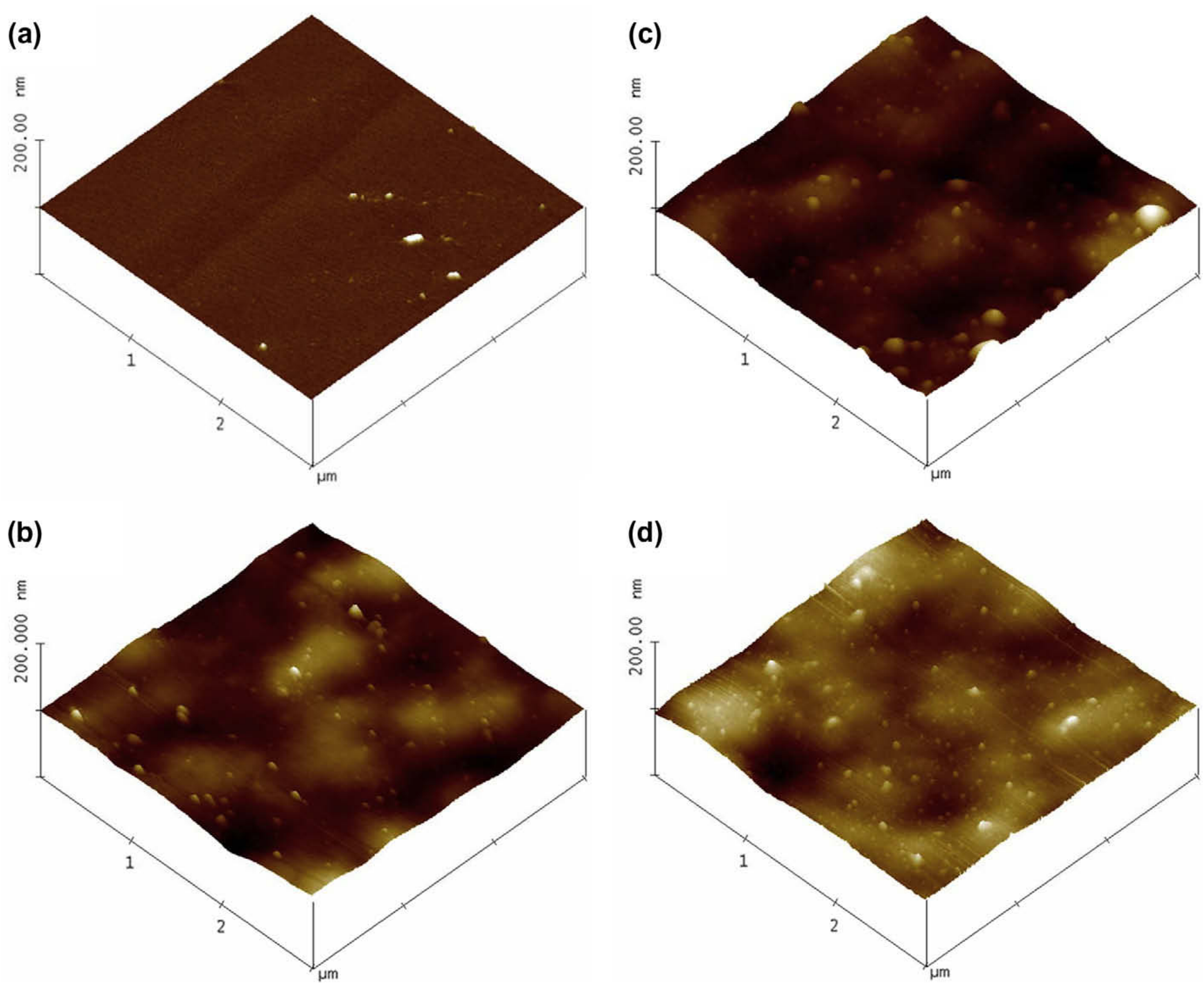

Fig. 1. AFM images of surface topography of samples: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s.

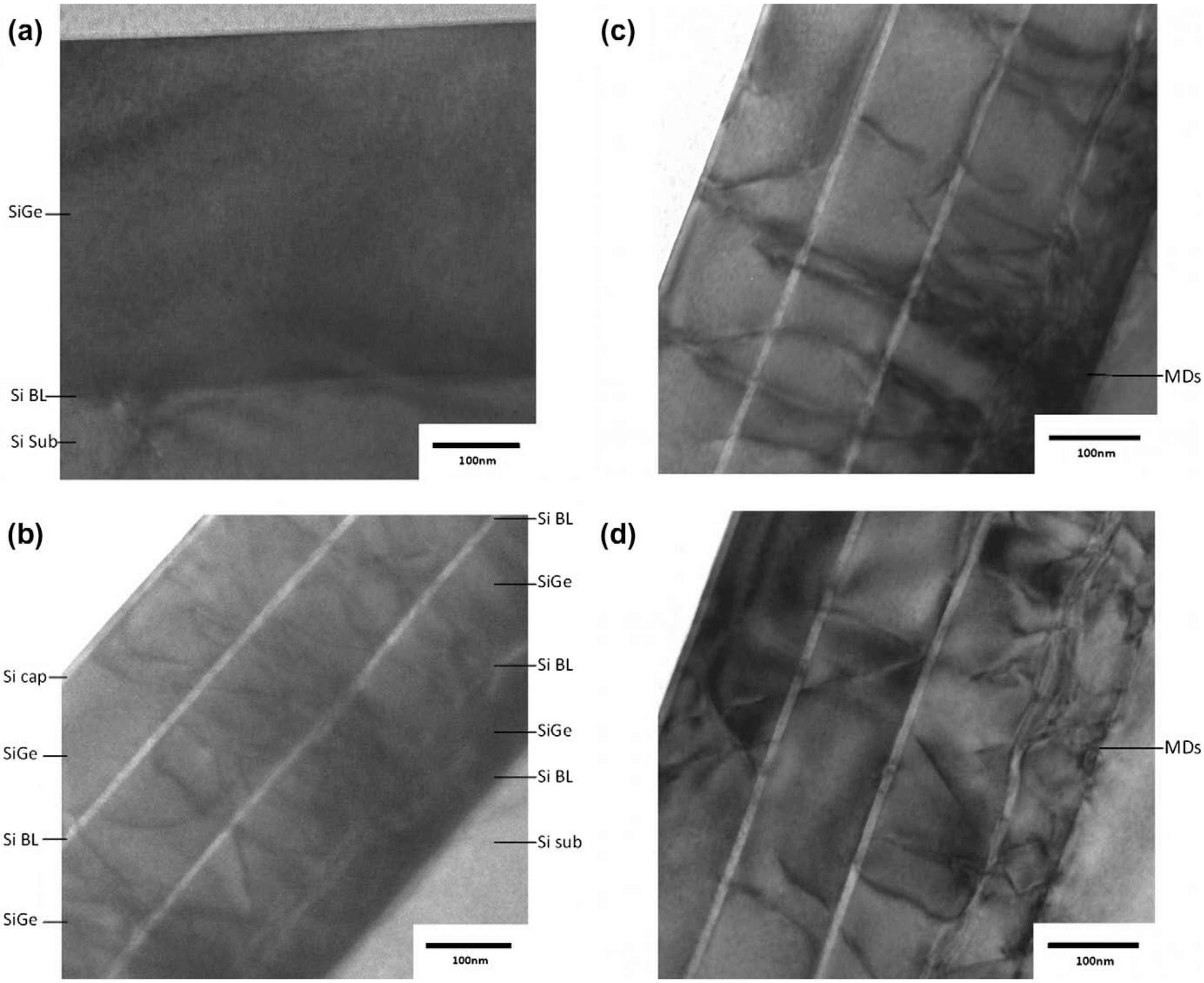

Fig. 2. The cross-sectional TEM image of the SiGe thin films samples: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s (Si cap: Si capping, Si BL: Si buffer layer, MDs: misfit dislocations).

annealing treatment, the SiGe multilayer films were *ex situ* endured thermal treatments in furnace on N_2 gas for 10 min (600 °C) and rapid thermal annealing for 20 s (800 °C).

Morphological character, roughness, and microstructure of the SiGe thin films were observed by means of scanning electron microscopy (SEM, Hitachi S-4000), atomic force microscope (AFM, Veeco D5000), and transmission electron microscopy (TEM, JEOL, JEM-2100F). From 3D patterns of AFM analysis, we mainly investigated two parameters: the height roughness parameters (R_a) and the root mean square (R_{ms}). In addition, TEM samples were prepared within mechanical polishing down to 20–30 μm, followed by Ar ion milling to electron transparency [14]. The observations were made at 200 kV.

Subsequent hardness and elastic modulus of the SiGe thin films were studied by using a Nano Indenter XP instrument (MTS Cooperation, Nano Instruments Innovation Center, TN, USA). The nanoindentation measurements used a diamond Berkovich indenter tip (tip radius ~50 nm), suggesting that plastic deformation can be generated at very small load. In addition, hardness data obtained with Berkovich indenter can be transformed to Vickers hardness because of the same shape of a three-sided pyramid, which means a similar area-to-depth function [13,15]. The continuous contact stiffness measurement (CSM) mode, which is executed by superimposing small oscillations on the force signal to measure displacement responses, offers a direct measure of dynamic contact stiffness during the loading process in the indentation test and is insensitive to thermal drift. Hence, the CSM mode can obtain hardness and Young's modulus relative to the indentation depth and is used in this experiment [16,17].

3. Results and discussion

The AFM system was employed to investigate the annealing process of the SiGe thin films in their surface and microstructure condition. Fig. 1 shows the sample of SiGe thin films before and after annealing treatments. Compared at Fig. 1a–b, we observe a smooth manner of the single-layer SiGe films and a comparatively rougher SiGe multilayer films. In Fig. 1c–d, it can be seen that the surface roughening of SiGe multilayer films is gradually increased at annealing treatment from 600 to 800 °C, as lists in Table 1. As well known, the dislocation nucleation may occur from surface roughening [18–22]. It is suggested that $Si_{1-x}Ge_x$ layers, grown on Si(0 0 1) substrates by UHV/CVD, relaxed either by dislocation multiplication or by dislocation nucleation based on surface roughening [23,24]. The surface roughness increases, which may be also

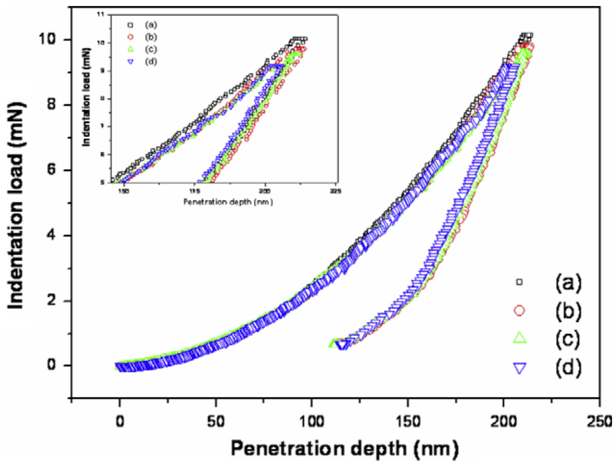

Fig. 3. The displacements of loading–unloading curves of the SiGe films: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s.

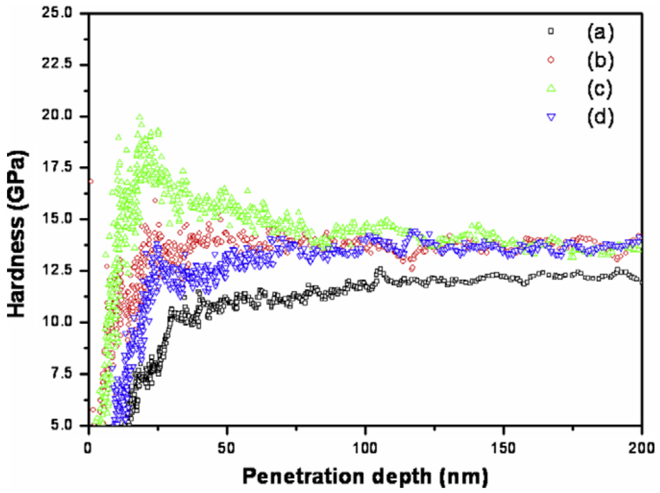

Fig. 4. The hardness of the SiGe thin films samples: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s.

attributed to the extra thermal budget and the relaxation of the strain [25–28]. Besides, the waved surfaces observed in multilayer films may be due to the 2D germanium segregation on surface during growing or annealing process at temperature below 600 °C [29]. We suggest that the SiGe multilayer films may be more complex than that of single films in the segregation mechanism. Therefore, more investigation in the microstructure of SiGe multilayer films is needed.

In order to examine the changes of SiGe films in the structural properties before and after annealing treatments, XTEM examination was employed in this work. Fig. 2a illustrates the profile of a single-layer SiGe films, and virtually no misfit dislocations as well as native dislocations have occurred. Fig. 2b shows the relatively stable in the interface of SiGe multilayer films. The amorphous Si buffer layer is embedded between SiGe layers which are totally crystalline. However, while the sample endured annealing treatment at 600 °C, we observe a low density of short misfit segments and some interdiffusion from the misfit dislocations (Fig. 2c). Meanwhile, the SiGe multilayer films allow a direct comparison of the dislocation formation while the annealing temperature rises to 800 °C (Fig. 2d). Both the conditions indicate a rapid increase in both misfit segments and the activated nuclei with increase in annealing temperature. This clearly demonstrates that the nucleation events and misfit segments are independent processes from thermally activated process in the SiGe multilayer films. Above all, TEM profile tends to display a serial nucleation seed at the each interface of SiGe multilayer films, which induces higher density of misfit dislocations. The similar phenomenon in the SiGe structures grown by means of UHV/CVD method occurred by a modified Frank–Reed mechanism. The dislocations are formed by the reproduction of corner dislocations [4]. In our experimental results, however, the high density of misfit dislocations can be aggregated between SiGe multilayer films. Thereby, the relative defects may induce the change in mechanical properties of the SiGe multilayer films.

The epilayer–substrate combination which corresponds to a softer epilayer (SiGe) on a harder substrate (Si) is investigated. The harder substrate, relatively difficult to deform, can impose a severe resistance to plastic deformation of the films, which could modify their response. Accordingly, the nanoindentation technique is powerful in probing the behaviours of the films. The samples belong to two categories: the materials (SiGe single-layer and multilayer) are investigated by CSM measurement; the penetration depth dependence of the *E* and *H* can be obtained. At the same time, the single-layer SiGe films is included for reference data. In Fig. 3, the same displacements of loading–unloading curves of the (a) single and (b–d) multilayer SiGe films are illustrated. Discrepancy in penetration depths of samples (a–d) are exhibited obviously (inset, Fig. 3). From this, we can observe that SiGe single-layer films is softer than that of all multilayer films due to the maximum penetration depth. Besides, we can also observe that the *H* of annealed SiGe multilayer films is higher than that of non-annealing one because of the less penetration depth. It is believed that the discrepancies among the multilayer SiGe films are mainly due to the increased activation energy, which is consistent with the kink model [30] for dislocation glide [31,32].

To further investigate this phenomenon, CSM nanoindentation technique is proposed. Fig. 4 shows typical CSM results of SiGe films for single-layer and multilayer structures on Si substrate. The effect of substrate is evident from the contact depth dependence of the *H*, as shown in Fig. 4. For indentation depths up to about 20 nm, the *H* increased as the indentation depth became deeper, which is normally attributed to the transition between purely elastic to elastoplastic contact whereby the *H* is actually the contact pressure. For indentation depths greater than about 20 nm, the H of SiGe films became constant. The *E* followed a trend similar to that of the *H*, which is converged at an indentation depth smaller than that for *H* (Fig. 5). Herein, *H* and *E* were therefore determined by averaging measurements at indentation depths from 100 to 200 nm, considering an adequate depth to achieve a fully developed plastic zone and meanwhile to avoid the more substrate effect [33]. However, in the present case the substrate probably affects the values of the elasto-plasticity coefficients since, for the experiments under a 10 mN peak load presented here, the penetration depth is ca. 200 nm, which does not exceed half of the film thickness; the influence of the substrate is expected [34]. For the measured results of each sample, the data at a contact depth of 100–200 nm are summarized in Table 2. It is interesting to observe that the *H* of annealing SiGe multilayer films is higher than non-annealing. In addition, the *E* decreases slightly with annealing treatment.

It is observed that the SiGe multilayer films can be more stable than that of SiGe single-layer. It is known that the films can relax by either the formation of misfit dislocations or interdiffusion. Misfit dislocations can enhance the *H* while significant interdiffusion exhibits the opposite tendency towards the *H* [#5]. Obviously, the enhanced *H* through annealing treatment in our study

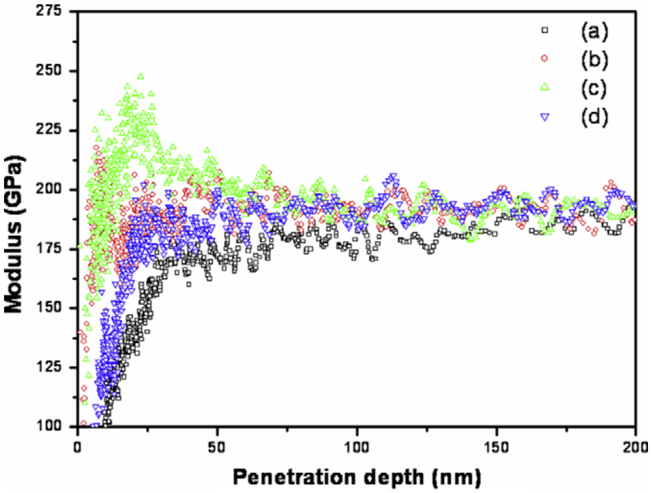

Fig. 5. The elastic moduli of the SiGe thin films samples: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s.

Table 2
The summary of hardness and elastic moduli of the SiGe thin films samples: (a) single-layer at RT, (b) multilayer at RT, (c) multilayer at 600 °C for 0.5 h and (d) multilayer at 800 °C for 20 s.

Sample	Sample condition	Elastic modulus (GPa)	Hardness (GPa)
(a)	Single-layer-SiGe RT	191.2 ± 6.5	12.6 ± 0.4
(b)	Multilayer-SiGe RT	194.1 ± 7.9	13.7 ± 0.6
(c)	Multilayer SiGe (600 °C)	184.1 ± 3.2	13.9 ± 0.6
(d)	Multilayer-SiGe (800 °C)	189.7 ± 5.3	14.2 ± 0.7

indicates that misfit dislocations dominate the relaxation mechanism. The annealing treatment not only leads nucleation seed but also slightly increased the *H* of the SiGe multilayer films. It is thus reasonable to conclude that slip and the orientation of the basal planes plays a key role in defining the mechanical properties of crystalline materials. The dislocations are formed by the reproduction of corner dislocations, which is based on the restriction of dislocation movement within and between SiGe multilayer films, as refer in the result of Fig. 4. However, the *E* of SiGe multilayer films with annealing treatments decreases slightly. The phenomenon may be due to the result of dislocation-induced additional strain, which would cause a decrease in stiffness [#1]. In other words, the *E* decreases because it is in proportion to stiffness. Nevertheless, the annealing treatment does not cause a significant decrease in the *E* in our experiment. When the contact depth exceeds 10–20% of the film thickness, substrate effect on the contact stiffness and *H* can be observed. The thickness of SiGe multilayers is approximately 530 nm, so the *H* and *E* data are obtained at a contact depth of 100 nm to measure these properties with certainty.

This result demonstrates that the SiGe multilayer films is more susceptible to plastic deformation and has the higher *H* in comparison to non-annealing samples. While annealing treatments are carried out, multilayer films will relax in order to reduce the strain energy; the restriction of dislocation movement therefore serves active role at the interfaces.

4. Conclusions

Our approach on an enhancement of mechanical ability of SiGe layer is based on a multi-heterostructure. High quality relaxed SiGe multilayer structures have been grown epitaxially by UHV/CVD methods. The agglomerated grains of the multilayer SiGe films were investigated by AFM analysis. It is obviously observed that the surface roughening of SiGe multilayer films is gradually increased, the dislocation multiplication or by dislocation nucleation may occur from the annealing treatment. The enhancement of the hardness at annealing temperature was achieved in the films by means of nanoindentation techniques. The *H* of the SiGe multilayer films within annealing treatment is higher due to the relaxed effect and, more susceptible to plastic deformation in comparison to the single-layer sample. Besides, the *E* decreases slightly through annealing treatment due to dislocation-induced additional strain. Dislocation glide is well understood and several mechanisms for dislocation nucleation have been studied.

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