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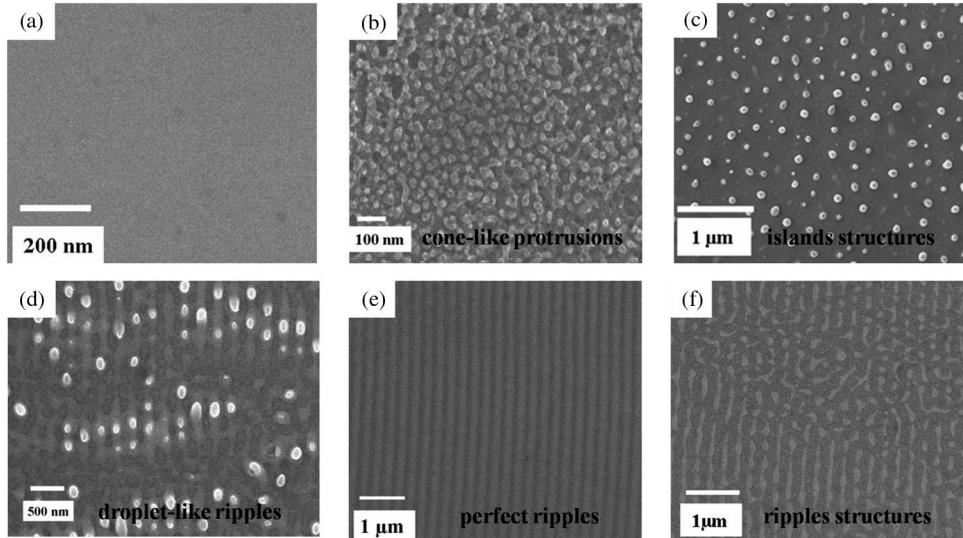
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Abstract: We have presented the mechanisms of nanosecond laser-induced specific nanostructures on a SiGe film by changing laser irradiation intensities. Our experimental results show that the types of the formed nanostructures were sensitive to the laser intensity because the structures such as cone-like protrusions, islands, and droplet-like ripples upon the irradiated SiGe film successively transform to ripple structures as the irradiation intensity decreases. Moreover, we obtain the threshold of the laser intensity for each nanostructure. Further studies give compelling evidence that the laser irradiation undergoes a transition from an initial thermal effect to an optical interference, which results in the evolution of SiGe nanostructures.

Index Terms: Laser processing, optical interference, microstructure, nanocrystalline materials.

1. Introduction

Surface nanostructures on semiconductors formed by laser irradiation have been studied in application of MEMS and optoelectronics [1]–[5]. The investigated laser wavelength varied from IR to UV, and the laser pulse duration is from nanoseconds to femtoseconds. In particular, pulse laser processing of semiconductor has attached great research interests in microelectronics and nanotechnology. By far, abundant types of nanostructures on semiconductors have been observed [6]–[9].

However, there is still lack of systematical study on the formation mechanisms of the laser-induced nanostructures. It is just different mechanism interactions between laser and material that result in the significantly morphology changes of the nanoscale structures under different laser irradiation conditions. In this condition, a deeper exploration of relevant mechanism is needed to achieve a direct and clear relationship between laser irradiation intensity and the induced nanostructures. As these relationship and process become more available, their use in material modification nanostructure applications is likely to increase.

In this paper, we obtain relationships between the nanostructures configuration and the laser irradiation intensities. A threshold of pulse energy (230 mJ/cm^2) is found, and thermal effect of laser

plays a major role with pulse energy greater than 230 mJ/cm^2 , ultimately forming cone-like protrusion and island structures. As laser energy decreases, the laser presents its wave character. The surface periodic structures become a dominant feature and completely replace disorder morphology when pulse energy is under 150 mJ/cm^2 .

2. Experimental

2.1. Materials Fabrication

The virtual SiGe substrate was epitaxially grown on a 4 inch n-type Si (100) wafer with resistivity of $0.1\text{--}1 \Omega\text{cm}$, using a cold-wall ultrahigh vacuum chemical vapor deposition (UHV/CVD) system under a base pressure of $5 \times 10^{-8} \text{ Pa}$ [9], [10]. Firstly, the wafers were cleaned by RCA method and dried by N_2 before loading into the growth chamber. The wafers were baked at 850°C for 30 min to de-oxide. Then a 32-nm-thick low temperature Ge (LT-Ge) film was deposited on n-type Si (100) substrate in the system from a solid Ge source. Ge was deposited by thermal evaporation of 99.999% elemental Ge in a Knudsen cell. The evaporation source used was an effusion cell of high temperature which was heated to 1100°C . The Si substrate temperature was maintained at 180°C and the sample was rotating during deposition to assure homogeneous coverage. Finally, pure Si_2H_6 and GeH_4 were used as precursors, and the pressure during the growth of material was about $10\text{--}2 \text{ Pa}$. About 170-nm-thick SiGe layer was deposited on the LT-Ge layer at $400\text{--}500^\circ\text{C}$ in the deposition system.

2.2. Laser Modification

A KrF excimer laser, which operates at wavelength of 248 nm with a 25-ns pulse width and 5-Hz repetition rate, was used in all experiments carried out herein. The SiGe samples were irradiated at different energy densities with 5 min irradiation time. The samples were protected in nitrogen ambient to avoid oxidation. The morphologies of the formed surface structures were characterized by scanning electron microscopy (SEM, LEO 1530, with an operating voltage of 20 kV). The crystal quality and the composition of the formed islands were characterized by transmission electron microscopy (TEM, FEI 200, with an operating voltage of 200 kV).

3. Results and Discussion

SEM images of various nanostructures on SiGe films are shown in Fig. 1(b)–(f) for various laser irradiation intensities from 70 to 300 mJ/cm^2 . When the irradiation energy density is 230 mJ/cm^2 and greater, the surface nanostructures are randomly distributed. The droplet-like ripples appear when the energy intensity reduces to 150 mJ/cm^2 . Perfect ripples emerged when the energy density is 100 mJ/cm^2 . However, as laser energy decreases, another threshold of pulse energy (70 mJ/cm^2) is found, periodic microstructure was observed in addition to the perfect ripples.

For further study, we investigate each formation mechanism of these nanostructures in the following papers. Fig. 2(a) shows SEM image of the SiGe surface morphology with energy of 300 mJ/cm^2 , and Fig. 2(b) is the close-up image. These figures precisely show cone-like protrusions can be achieved in our experiments. In comparison to as-grown samples, we can see there are some dark areas on the surface, and the surface becomes rough in these figures, which result from laser induced surface melting. We estimate that laser ablation drives the formation of cone-like protrusions morphology on the surface [10]–[12].

We can see in Fig. 1(c), island structures become a dominating morphology at the intensity of 230 mJ/cm^2 . Owing to its randomness, we conclude that the thermal effect induced the generation of disorder islands under this condition. It has been demonstrated that disorder SiGe islands appear on the surface, which are caused by obvious lattice distortion between the SiGe islands and Si film. We estimate that surface strain drives the formation of droplet-like morphology on the surface [13]–[15]. Fig. 3(b) and (c) are TEM images of a single island, in which we can see that there is a threading

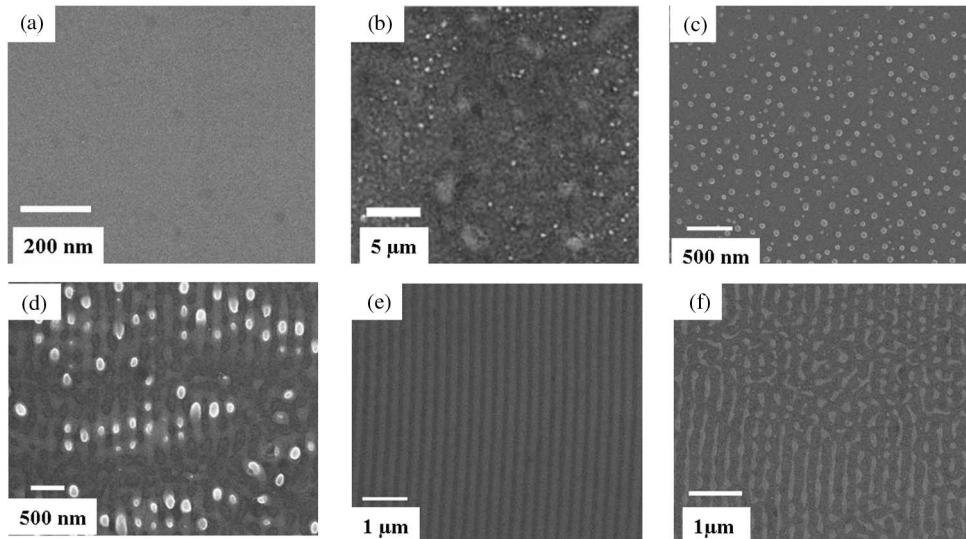


Fig. 1. SEM images of SiGe sample surfaces. (a) Surface without nanosecond laser irradiation. (b)–(f) SiGe surface nanostructures fabricated by KrF excimer laser with different incident energy densities, 300 mJ/cm^2 , 230 mJ/cm^2 , 150 mJ/cm^2 , 100 mJ/cm^2 and 70 mJ/cm^2 , respectively.

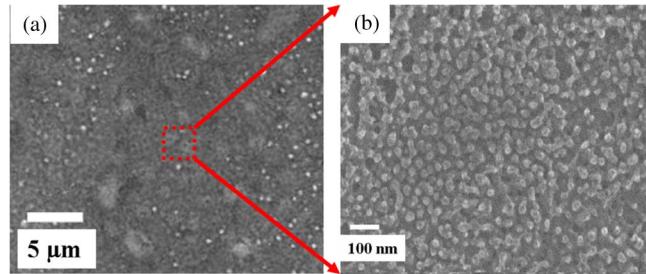


Fig. 2. SEM images of SiGe surface nano structures fabricated by KrF excimer laser with the energy density of 300 mJ/cm^2 . (b) Close-up image for (a).

dislocation in the island. Based on the former experiments we realize that the laser-induced island is fully relaxed, and thermal effect accelerates the relaxation of strain, which finally forms disorder surface islands [8], [16]. With the decrease of irradiation intensity, droplet-like ripples become the dominant feature, as shown in Fig. 1(d). The droplet-like ripples are actually a combination of the periodically array and disorder islands. In this condition, the droplet-like ripples are considered as the transition state from fully randomly distributed islands to periodic ripples. Co-existence of the thermal effect and the optical interference effect is proposed to the formation of the droplet-like ripples. As the laser energy further decreases, the perfect ripples become stronger, and islands finally disappear.

For further mechanism analysis of the perfect ripple structures, we changed the intensity of the beam in the following experiments. In our previous work [8], perfect annular nanostructures around scattering points on the SiGe film are obtained after the irradiation of a KrF excimer pulse laser beam (100 mJ/cm^2) at different incident angles. The different shapes of annular structures are related to different energy distributions due to the optical interference between the scattered light and the incident beam.

$$U(P) = A \exp[j \vec{k} \cdot \vec{r}] = A \exp[jk(x \cos\alpha + y \cos\alpha)] \quad (1)$$

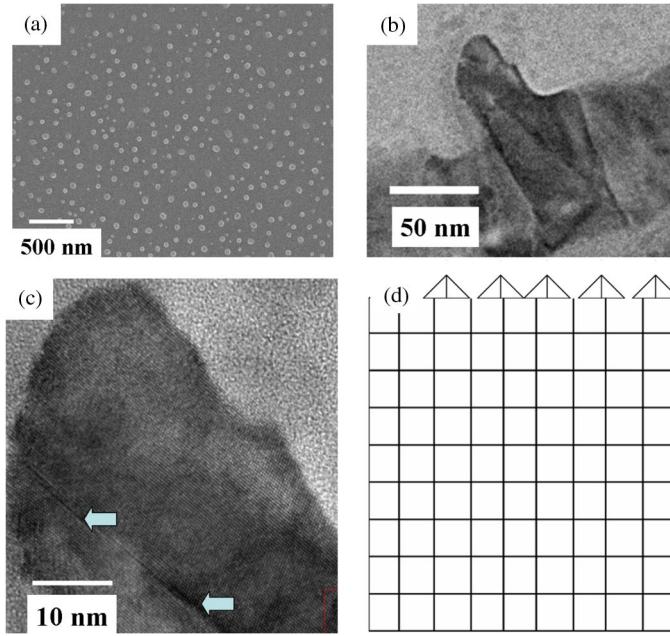


Fig. 3. (a) SEM images of SiGe surface island structures fabricated by KrF excimer laser with the energy density of 230 mJ/cm^2 . (b), (c) TEM images of single island, where arrows represent the threading dislocation in island, and (d) is schematic illustrations of the formation process of the SiGe islands.

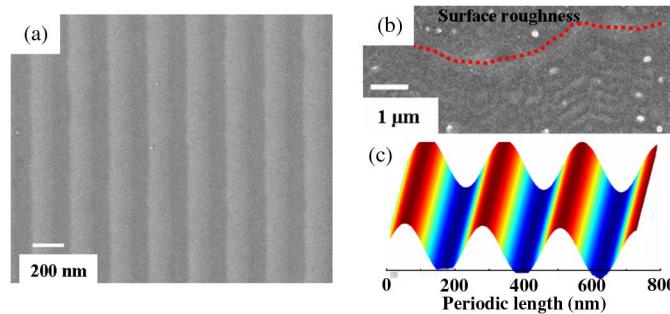


Fig. 4. (a), (b) SEM images of SiGe surface ripple structures fabricated by KrF excimer laser with the energy density of 100 mJ/cm^2 , the red line in (b) stands for the shape of surface roughness, and (c) is corresponding simulated result by optical interference.

where \vec{r} is the position vector of the plane wave, \vec{k} is the wave vector of the plane wave, and α in this Eq is the angle of the incident beam, respectively.

$$\begin{aligned} U(P) &= U_1(P) + U_2(P) = A_1 \exp[j \vec{k} \cdot \vec{r}_1] + A_2 \exp[j \vec{k} \cdot \vec{r}_2] \\ I &= U(P) * U^*(P). \end{aligned} \quad (2)$$

In this paper, there are no obvious scattering points on SiGe film, but uniform and perfect ripples with obvious interference can be observed in Fig. 4(a). And the period of the perfect ripples is similar with the laser wavelength (248 nm). Though the surface is not smooth [8], [18], obvious interference can also be found, just like Fig. 4(b), moreover, the distribution of ripples is in accord with the shape of surface roughness. In this condition, optical interference theory can be used to interpret these phenomena. Taking the optical interference into account, the intensity distribution and interference fringes will be determined as long as the wavelength and the angle of the incident laser are established [1], [8], [17], [18]. The complex amplitude of incident/scattered wave is given by Eq (1), here $\alpha = 0$. Moreover, we calculated the time-averaged interference intensity on SiGe

surface with the Eq. (2). Fig. 4(c) is the simulated result according to the former equation, and the periodic lengths of ripples correspond exactly to the experimental results, like Fig. 4(a).

However, ripples completely replaces the surface perfect ripples when the laser energy is low, as shown in Fig. 1(f). But the shapes of the fringe structures are consistent with the optical interference fringes. Only if the incident energy density is high enough can the interference phenomenon induce the redistribution of the laser energy on the surface, which fully results in the migration and agglomeration of atoms along the interference fringes. What we have obtained shows that the KrF excimer laser-patterned nanostructures on SiGe material are sensitive to pulse energy intensity, and appointed nanostructures can be caused via changing laser irradiation energy.

4. Conclusion

In conclusion, we have presented experimental results concerning the evolution of nanosecond laser-induced specific nanostructures on SiGe film. Various nanostructures can take shape with different laser irradiation intensities. The analyses of the structures reveal that these disorder islands are resulted from thermal effect of the incident laser when the pulse energy is above a threshold (230 mJ/cm^2). Droplet-like ripples appear after irradiated with the energy intensity of 150 mJ/cm^2 . Perfect ripples completely replace disorder structures as the energy intensity is 100 mJ/cm^2 . And further more, these ripple structures are caused by different energy distribution which are results of optical interference between the scattered light and the incident beam.

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