

Dislocation dynamics in SiGe alloys

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Abstract. The dislocation velocities and mechanical strength of bulk crystals of $\text{Si}_x\text{Ge}_{1-x}$ alloys grown by the Czochralski method have been investigated by the etch pit technique and compressive deformation tests, respectively. Velocity of dislocations in the SiGe alloys of the composition range $0.004 < x < 0.08$ decreases monotonically with an increase in Si content at temperature 450–700°C and under stress 3–24MPa. In contrast, velocity of dislocations in the composition range $0.92 < x < 1$ first increases, then decreases and again increases with a decrease in Si content at temperature 750–850°C and under stress 3–30MPa. The velocity of dislocations was quantitatively evaluated as functions of stress and temperature. Stress-strain behaviour in the yield region of the SiGe alloys of composition $0 < x < 0.4$ is similar to that of Ge at temperatures lower than about 600°C. However, the yield stress becomes temperature-insensitive at high temperatures and increases with increasing Si content. The stress-strain curves of the SiGe alloys of composition $0.95 < x < 1$ are similar to those of pure Si at temperatures 800–1000°C and the yield stress increases with decreasing Si content down to $x = 0.95$. The yield stress of the SiGe alloys is dependent on the composition, being proportional to $x(1-x)$, showing a maximum around $x \approx 0.5$. Built-in stress fields related to local fluctuation of the alloy composition and the dynamic development of a solute atmosphere around the dislocations, may suppress the activities of dislocations and lead to the hardening of SiGe alloys.

1. Introduction

Silicon-germanium ($\text{Si}_x\text{Ge}_{1-x}$ or Germanium-silicon $\text{Ge}_{1-x}\text{Si}_x$, where x indicates the mole fraction of silicon) is a complete solid-solution semiconductor having the diamond cubic structure. SiGe alloys have attracted great interests for both microelectronic and optoelectronic devices and various functional materials because of the potential for band-gap and strain/lattice parameter engineering they offer. That is, alloying leads to various unique effects on fundamental properties, absent in the component materials Si and Ge.

Usually these alloys are grown as thin films on Si substrates by various epitaxial techniques. The introduction of misfit dislocations is inevitable in such hetero-structures due to the stress caused by interfacial mismatch when the film thickness exceeds a critical value in the system of thin film/substrate. Although dislocations affect the electrical and optical properties of SiGe alloy and limit its application within various devices, only little is known about their dynamic properties such as the generation and motion of misfit dislocations at the film/substrate interface within stressed layers, mainly for compositions on the Si-rich side [1-5]. The large biaxial stress inherent to such hetero-structures hinders the quantitative study of the native properties of dislocations. The dynamic properties of dislocations in SiGe have often been assumed to be similar to those in Si or Ge and little



attention has been paid to the unique properties which have been found in several compound alloy semiconductors [6-8]. The present author reported that the flow stresses of GaAsP and InAsP ternary alloys have an athermal component that is absent in the binary GaAs, GaP, InAs and InP compounds [9,10]. Accordingly, it is of interest to investigate the dynamic activities of dislocations in SiGe alloys using bulk crystals and to deduce their unique properties brought about by alloying. SiGe alloys are quite suitable for such a basic study. The mechanical behaviour of the element materials Si and Ge has been under investigation since the 1960's pioneering studies by groups such as Patel and Chaudhuri [11] and Bell and Bonfield [12] and at present is well understood on the basis of kinetic properties of dislocations [13].

To clarify the dynamic properties of dislocations in SiGe alloys quantitatively, it is necessary to grow bulk crystals with low dislocation densities and measure the dislocation velocities and the mechanical properties. Growth of bulk single crystals of SiGe alloys is difficult because of a large miscibility gap and the differences in the densities, lattice parameters and melting temperatures of the constituent elements. However, this type of material is also suited for a basic study on the solidification of solid solution. Here, unique features that appeared in thermo-mechanical behaviour in SiGe grown by the Czochralski (CZ) method are reviewed, based on the results by the author's group [14-19].

2. Bulk crystals

Bulk crystals of $\text{Si}_x\text{Ge}_{1-x}$ alloys in the whole composition range $0 < x < 1$ were grown by the Czochralski technique at very low pulling rates ranging from 1 to 8 mm/h in an Ar gas flowing atmosphere [14, 19-22]. Seeds prepared from a Si or Ge crystal oriented parallel to [111] or [001] were used for the growth of the crystals.

Full single crystals of large size (i.e. larger than 15 mm in diameter and longer than 40 mm) were successfully grown for the composition ranges of $0 < x < 0.15$ and $0.9 < x < 1$. Small single crystal alloys of intermediate composition were obtained in the ingot near the seeds since the transition to polycrystallinity relates to the occurrence of constitutional supercooling [22].

Striations and dislocations were observed in the grown crystals. Dislocations were generated mainly at the seed/alloy interface. The density of grown-in dislocations in the alloys was in the range 10^3 – 10^5 cm^{-2} . The generation process may be controlled by the magnitude of the misfit strain between the alloy and seed, the temperature, the temperature gradient and the mobility of generated dislocations in the alloy [19].

This alloy shows a variety of unique properties, such as atomistic bonding structure, electron and hole mobilities, thermal conductivity, local-vibration of oxygen impurity, muonium, etc., which have been reported separately [23-31]. Here, it should be noted that SiGe alloy is a typical disorder material and that the bond lengths and bond angles are distorted with alloy composition in SiGe, classified to be quasi-Pauling type [23,24].

3. Dislocation velocities

Velocities of dislocations were measured in $\text{Si}_x\text{Ge}_{1-x}$ alloys of composition range $0 < x < 0.08$ and $0.922 < x < 1$ with low densities of grown-in dislocations of $\sim 10^3$ cm^{-2} . The specimen was stressed at elevated temperature by three-point bending in a vacuum. Dislocations were generated preferentially from a scratch drawn on the surface. Displacements of dislocations caused by stressing were measured by the etch pit technique [15,17,18].

Figure 1(a) shows the velocities of 60° dislocations at 550°C plotted against the resolved shear stress in CZ-SiGe alloys of the composition range $0 < x < 0.08$ together with that in pure Ge. As seen in the figure, the logarithm of the velocity of dislocations is linear with respect to the logarithm of the stress, with approximately the same slope for all the Si contents except the following feature: the velocity of 60° dislocations in the SiGe alloys of higher Si content shows a break at stress around 8 MPa, depending on the Si content. Figure 1(b) shows the velocities of 60° dislocations in Ge-rich SiGe

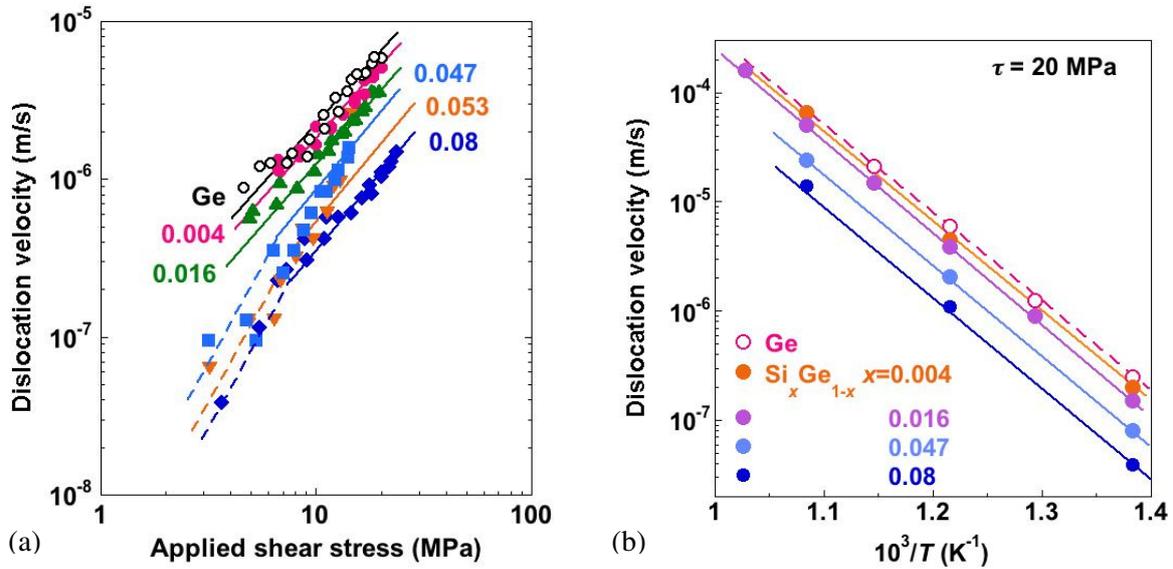


Figure 1. Velocities of 60° dislocations in the Ge-rich SiGe alloys. (a) Stress-dependence at 550°C, (b) temperature-dependence under a shear stress of 20 MPa. Numbers show the Si content.

alloys of various Si contents under a stress of 20 MPa, free from any threshold or kink, plotted against the reciprocal temperature together with that in pure Ge.

Figure 2(a) shows the velocities of 60° dislocations at 800°C plotted against the resolved shear stress up to 30 MPa in float-zone-grown (FZ-)SiGe alloys of the composition range 0.922 < x < 1 (Si-rich side) together with that in pure Si [32]. The logarithm of the velocity of dislocations in SiGe alloys is linear with respect to the logarithm of the stress at temperatures 750–850°C. The slope of the plot of the dislocation velocity versus the stress in the SiGe alloy with x = 0.996 is approximately the same as that in Si. On the other hand, in SiGe alloys of x = 0.922–0.979 the velocity of dislocations is

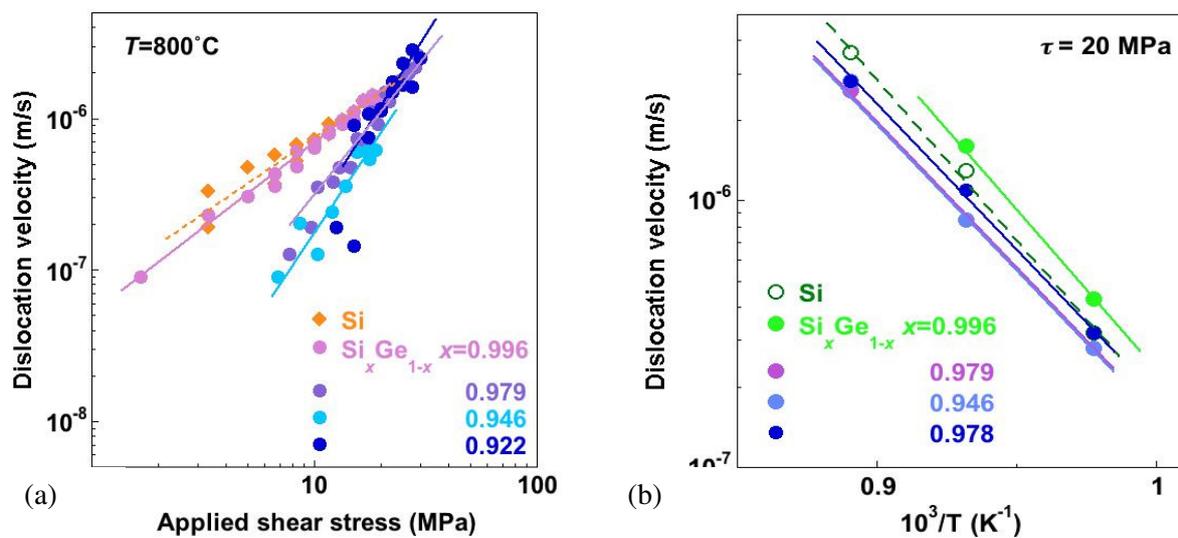


Figure 2. Velocities of 60° dislocations in the Si-rich SiGe alloys. (a) Stress-dependence at 800°C, (b) temperature-dependence under a shear stress of 20 MPa. Numbers show the Si content.

zero under a stress lower than the threshold one and then increases rapidly with an increase in stress beyond the threshold stress as reported by Iunin et al. [33]. The threshold stress for dislocation generation from a scratch increases with a decrease in the Si content. Figure 2(b) shows the velocities of 60° dislocations in Si-rich SiGe alloys of various Si contents under a stress of 20 MPa, free from an effect of the threshold, plotted against the reciprocal temperature together with that in pure Si.

In the Ge-rich SiGe alloys of composition range $0 < x < 0.08$ the dislocation velocity decreases monotonically with increasing Si content, reaching about one of seventh of that in pure Ge at $x = 0.08$ in the temperature range, as shown in figure 3. On the other hand, in the composition range $0.922 < x < 1$ (Si-rich SiGe) the dislocation velocity first increases, then decreases and again increases with increasing Ge content in the temperature range $750\text{--}850^\circ\text{C}$ and the stress range $3\text{--}30$ MPa, as shown in figure 3. The dislocation velocity for the Si content $x = 0.996$ is found to be higher than that in pure Si.

The velocities of dislocations in the SiGe alloys investigated in the present studies are expressed in a similar way to those in Ge, Si and other semiconductors as functions of the stress τ and temperature T by the following empirical equation [13, 34, 35]:

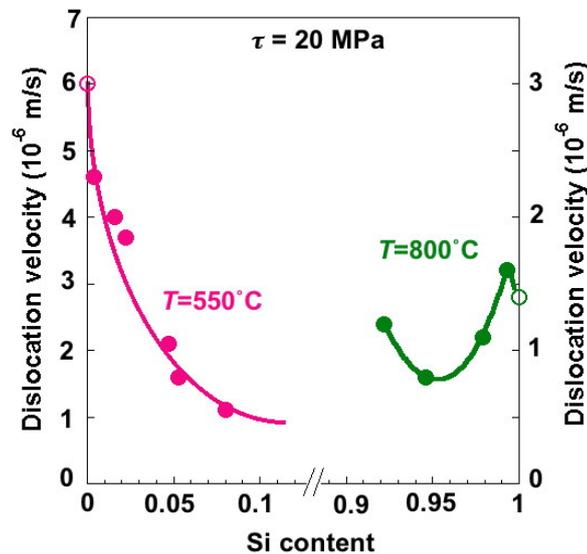


Figure 3. Velocities of 60 dislocations in the Ge-rich and Si-rich SiGe alloys at 550 and 800°C , respectively, under a shear stress 20MPa as dependent on the Si content.

$$v = v_0 (\tau/\tau_0)^m \exp(-Q/k_B T), \quad \tau_0=1 \text{ MPa}, \quad (1)$$

where k_B is the Boltzmann constant. The experimentally determined magnitudes of v_0 , m and Q in SiGe and pure Ge and Si are listed in Table 1.

Here, it should be noted that dislocations in SiGe alloys show a typical recombination enhanced dislocation motion under an irradiation by an electron beam [36,37].

4. Mechanical strength

The mechanical strength was investigated in $\text{Si}_x\text{Ge}_{1-x}$ alloys for the composition range $0 < x < 0.4$ and $0.95 < x < 1$ with grown-in dislocation densities of $10^3\text{--}10^5 \text{ cm}^{-2}$. Rectangular parallelepiped specimens were compressed under a constant strain rate using an Instron-type machine at elevated temperatures [16-18].

Table 1. Magnitudes of v_0 , m and Q for 60° dislocations in $\text{Si}_x\text{Ge}_{1-x}$ and pure Ge and Si.

Crystal	v_0 (m/s)	m	Q (eV)
Ge	2.9×10^2	1.7	1.62 ± 0.05
$\text{Si}_x\text{Ge}_{1-x}$	$x = 0.016$	4.6×10^2	1.68
	$x = 0.047$	2.8×10^2	1.68
	$x = 0.080$	2.3×10^2	1.6
	$x = 0.922$	1.2×10^1	3.3
	$x = 0.946$	9.4×10^1	2.1
	$x = 0.979$	2.1×10^2	1.9
	$x = 0.996$	1.4×10^4	1.0
Si	1.0×10^4	1.0	2.4

The stress-strain curves of the Si-rich SiGe alloys of the composition range $0.95 < x < 1$ are similar to those of pure Si at temperatures 800–1000°C, characterized by a stress drop followed by an increase in the stress with strain. Such a stress drop is commonly found in other semiconductors, such as Si, Ge, GaAs, and so forth, at relatively low temperatures [34,35]. The upper and lower yield stresses and flow stress increase with a decrease in Si content. Similarly, the stress-strain curves of the Ge-rich SiGe alloys with $x = 0.01, 0.10, 0.25$ and 0.40 at low temperatures were characterised by a stress drop followed by an increase in the stress with respect to strain, but at high temperatures those show no stress drop, differing from those of the Si-rich SiGe alloys. The highly enhanced mobility of dislocations at these temperatures may contribute to the feature. It is remarkable that the SiGe alloys with x larger than 0.10 exhibit much higher levels of the yield and flow stresses than pure Ge and Si [16-18].

Figure 4 shows the temperature dependence of the upper yield stresses of various SiGe alloys and also those of Si and Ge, for comparison, under a shear strain rate of $1.8 \times 10^{-4} \text{ s}^{-1}$. In a case where there is no stress drop after yielding, the yield stresses are plotted. The yield stresses in Si and Ge decrease with an increase in the temperature. Such a dependence can be described as a function of strain rate $\dot{\epsilon}$

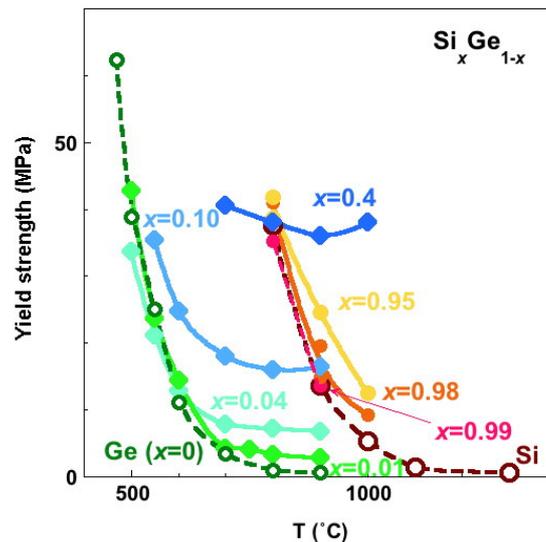


Figure 4. Yield stresses of the SiGe alloys plotted against the temperature for deformation under a shear strain rate of $1.8 \times 10^{-4} \text{ s}^{-1}$.

and temperature T by the following empirical equation:

$$\tau = A \dot{\epsilon}^{1/n} \exp(-U/k_B T), \quad (2)$$

where A , n and U are constants [34-38].

The Ge-rich SiGe alloys show similar reduction of the yield stresses with increasing the temperature in the low temperature regime. Their dependence on the temperature becomes weak in the high temperature region and finally nearly constant. The temperature-insensitive range expands towards the low temperature side with an increase of the Si content x . The magnitude of the yield stress of the alloys is higher in the high temperature region with increasing Si content up to 0.4. Typically, the yield stresses of the alloys $x = 0.4$ are temperature-insensitive in the range investigated. The magnitudes of the yield stress of the Si-rich SiGe alloy $x = 0.99$ are the same as, or slightly lower than, those of Si and the temperature dependence of the yield stress is similar to that of Si. With a decrease in the Si content to $x = 0.95$, the yield stress increases and the temperature dependence of the yield stress becomes weaker.

Figure 5 shows the composition dependence of the yield stress of the SiGe alloys under a shear strain rate of $1.8 \times 10^{-4} \text{ s}^{-1}$ at 900°C . The yield stress increases with increasing Si content in the composition range $x = 0-0.4$ and decreasing Si content down to $x = 0.95$ investigated. Typically, the yield stresses of the Ge-rich SiGe alloys with $x > 0.10$ are much higher than that of pure Si. Over the whole composition range of the SiGe alloys the yield stress seems to show a maximum around $x = 0.5$ and be dependent on the composition as proportional to $x(1-x)$ [18].

Hardness of the SiGe alloys obtained with a micro indenter with a 0.5 N load for 10 s at room temperature (RT), 600°C and 900°C is shown in figure 6. The hardness at RT and 600°C increases almost linearly with the Si content x from 0 to 1, while that at 900°C shows a maximum around $x = 0.5$, similar to the yield stress above-mentioned. The temperature dependence of the hardness well corresponds with that of the yield stress.

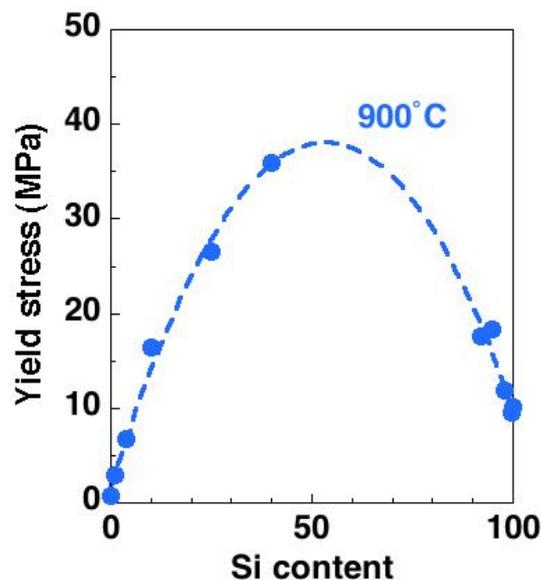


Figure 5. Yield stresses of SiGe alloys under a shear strain rate of $1.8 \times 10^{-4} \text{ s}^{-1}$ at 900°C .

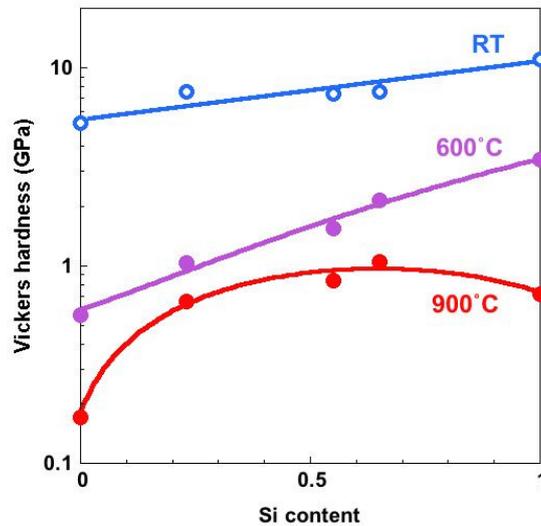


Figure 6. Micro-indentation hardness of the SiGe alloys plotted against the Si content at room temperature (RT), 600 and 900°C.

5. Origins of alloying effect

SiGe demonstrates a character as an alloy noticeably at high temperatures. This may also be understood from the fact that the hardness does not show alloying effects at room temperature but at 900°C as seen in figure 6. The velocity of isolated dislocations in SiGe alloys with compositions close to Si and Ge shows only small differences from that in Si and Ge. Thus, the difference in the dislocation mobility among various compositions of SiGe alloys may not lead to the drastic difference in the mechanical strength of the alloys observed at high temperatures. Here, it may be noted that the bulk modulus of SiGe alloys was evaluated to increase linearly with alloy composition [39]. Also, it was observed that dislocations induced by the plastic deformation are dissociated into Shockley partial dislocations bounding intrinsic stacking-faults. The intrinsic stacking-fault energy in the alloys decreases from 61 ± 10 to 55 ± 10 mJ/m² with increasing Si content, intermediate between those of Si and Ge [40].

There are two components of the flow stress of a crystal for deformation as seen in figure 7: One is the effective stress by which dislocations move at a certain velocity against the intrinsic resistance

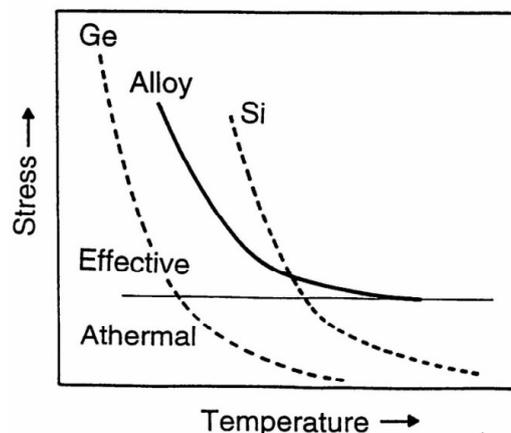


Figure 7. Schematic of the flow stress components and their dependence on temperature.

(Peierls potential) via thermal activation. The other is the athermal stress below which dislocations cannot move. The latter depends slightly on the temperature. If alloying results in a drastic increase in the Peierls potential and reduces the dislocation velocity, we may expect the stress drop at the yield point to be more remarkable in SiGe alloys than in Ge from the concept of dislocation dynamics of yielding in semiconductors. In addition, the strengthening effect caused by alloying should be less remarkable with an increase of temperature. As shown in figure 4, the yield stresses of the alloys are temperature-independent at elevated temperatures, remarkable in the alloys with the intermediate composition. Thus, the observed variation in the yield stress against the temperature in the SiGe alloys can be understood as a feature that the SiGe alloys have an athermal stress, that does not exist in other elemental and compound semiconductors. The athermal stress is in maximum in the SiGe alloy with a Si content $x \approx 0.5$. We can think reasonably that the athermal stress is related to the alloying effect.

As discussed in previous papers on GaAsP and InAsP alloys [9,10], several origins for athermal stress are plausible in alloying. First, short-range order of the $L1_1$ (CuPt-type) structure found in strained layer superlattice thin films prepared by molecular beam epitaxy [41] can lead to an extra stress of athermal nature since the motion of a dislocation destroys the short-range order along its slip plane [42]. However, there is no report detecting an ordered structure in bulk SiGe alloys [43] and also supported by XAFS study where the ordering parameter was evaluated to be around 0.22–0.29 from the coordination number [23].

Second, a long-range stress field may be developed by local fluctuation of the alloy composition in a crystal. Since the bond length of Ge is longer than that of Si by about 4%, local fluctuation of the alloy composition in the crystal, causing the development of Si or Ge enriched regions, may induce a long-range stress field that cannot be surmounted thermally by dislocations. Dislocations in SiGe alloys may move by a repeat bowing out process around the long-range stress fields.

Third, the dynamic development of a solute atmosphere around a dislocation during deformation at high temperatures leads to the additional stress for releasing the dislocation from the solute atmosphere. Indeed, many fine serrations on the stress-strain curve in the deformation under a strain rate as low as $1.8 \times 10^{-5} \text{ s}^{-1}$ at 900°C were observed [16]. Such a characteristic is known as the Portevin-LeChatelier phenomenon, being interpreted as repeated processes of locking and releasing of dislocations [44]. Indeed, though the width of dissociated dislocations formed by plastic deformation keeps constant, photoluminescence studies showed a variation of composition around deformation-induced dislocations by annealing [45,46]. Although the releasing process of a dislocation from its solute atmosphere is a thermally activated one, the development of a solute atmosphere around the dislocation is more enhanced at higher temperature. Thus, the contributions of these effects to the flow stress compensate each other and may give rise to a temperature-insensitive resistance to the dislocation motion, apparently looking like an athermal stress.

Either or both the local fluctuation of alloy composition and/or the dynamic development of a solute atmosphere around the dislocations are thought to suppress the dynamic activity of dislocations and result in the strengthening of bulk SiGe alloys at elevated temperatures.

6. Summary

The unique properties of $\text{Si}_x\text{Ge}_{1-x}$ crystals originating from alloying have been observed in an investigation of the mechanical strength and dislocation velocity by using bulk alloy crystals.

1. The dislocation velocity decreases monotonically with increasing Si content in the SiGe alloys of composition range $0.004 < x < 0.080$ in the temperature range $450\text{--}700^\circ\text{C}$ and the stress range 3–24 MPa, while the dislocation velocity first increases, then decreases and again increases with decreasing Si content in the composition range $0.92 < x < 1$ for the temperature range $750\text{--}850^\circ\text{C}$ and the stress range 3–30 MPa. The velocity of dislocations was determined as function of stress and temperature.

2. The stress-strain behaviour in the yield region of SiGe alloys of composition range $0 < x < 0.4$ is similar to that of Ge at temperatures lower than about 600°C . However, the yield stress becomes temperature-insensitive at high temperatures and increases with increasing Si content. The stress-strain behaviour of the SiGe alloys of composition range $0.95 < x < 1$ is similar to that of pure Si at

temperatures 800–1000°C and the yield stress increases with decreasing Si content down to $x = 0.95$. The composition dependence of the yield stress follows an $x(1 - x)$ relationship.

3. Built-in stress fields related to local fluctuation of the alloy composition, together with the dynamic development of a solute atmosphere around dislocations, seem to suppress the activities of dislocations and bring about a hardening of SiGe alloys.

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