

Flexible thermoelectric SiGe films formed by Al-induced layer exchange

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Abstract

An Al-induced layer exchange technique enabled us to form self-organically-p-doped SiGe on glass substrates at a low temperature (≤ 400 °C) in the whole SiGe composition range. Results indicated that a high power factor ($430 \mu\text{W}/\text{mK}^2$) and a low process temperature were simultaneously achieved for SiGe on glass. A SiGe layer fabricated on a plastic substrate exhibited a power factor of $190 \mu\text{W}/\text{mK}^2$, which is the highest among most thermoelectric films directly grown onto a plastic substrate.

1. Introduction

The SiGe alloy is the oldest, reliable and proven thermoelectric material. If a SiGe layer can be formed on plastic, it will be possible to create flexible thermoelectric devices with high performance. To achieve the SiGe on plastic structure, SiGe should be formed below the heat resistant temperature of plastic (e.g., 400 °C for polyimide). Recently, we developed the Al-induced layer exchange (ALILE) of SiGe and synthesized SiGe layers on glass at 400 °C or less in the whole composition range.^[1, 2] In this study, we clarified the thermoelectric properties of the ALILE-SiGe layers and developed this method on a plastic substrate.

2. Experiment procedures

Fig. 1 presents a schematic of the sample preparation process. Preparation of the Al and amorphous (a-) $\text{Si}_{1-x}\text{Ge}_x$ layers (each 50 nm thick) on a SiO_2 glass substrate and plastic substrate was performed at room temperature. The Ge composition x in the prepared a- $\text{Si}_{1-x}\text{Ge}_x$ layers was determined to be 0, 0.15, 0.3, 0.6, 0.8, and 1 by Rutherford backscattering spectrometry (RBS). Between the two deposition cycles, the Al film was exposed to air for 30 min to form a native Al oxide layer as a diffusion-limiting layer. The samples were then annealed at 340–450 °C in a N_2 ambient chamber until the layer exchange was finished. The Al and AlO_x layers were then etched away with HF solution (HF: 1.5%) for 1 minute.

3. Results and Discussion

As shown in Fig. 2(a), (b), the higher Ge fraction in SiGe, the temperature range where layer exchange occurs is lower. This reflects the activation energy required for crystallization.^[1] Fig. 2(a) shows that the hole concentration increases with increasing Ge fraction and growth temperature. This result is attributable to the fact that hole concentration in the SiGe layers are determined by the solubility limit of Al in SiGe. Thus, ALILE enables the self-organization of p-type SiGe layers at low temperature. Fig. 2(b) shows that the electrical conductivity increases with increasing Ge fraction reflecting the increase of hole concentration. These results mean that the hole concentration and electrical conductivity can be controlled by adjusting the Ge fraction and annealing temperature. Fig. 2(c) shows the Seebeck coefficient decreases with increasing hole concentration and Ge fraction. This is a well-known phenomenon in SiGe. Fig. 2(d) shows that the power factor improves with increasing hole concentration for high Ge fraction ($x \geq 0.8$), whereas the low Ge fraction ($x \leq 0.6$) shows the opposite trend. Fig. 2(e) shows the highest power factor in each Ge fraction. There was no clear Ge fraction dependence on the maximum power factor. The power factor is maximum ($435 \mu\text{W}/\text{mK}^2$) with pure-Ge. Fig. 2(f) shows that ZT becomes maximum (0.06) for $x = 0.6$. Therefore, for ALILE-SiGe for thermoelectric conversion applications, $x = 0.6$ is the optimum composition.

Fig. 3 shows that SiGe generally requires high process temperature to obtain high power factor.^[3-10] This is because a high temperature is required to activate impurities in SiGe. In layer exchange, impurities can be solid-solved (that is,

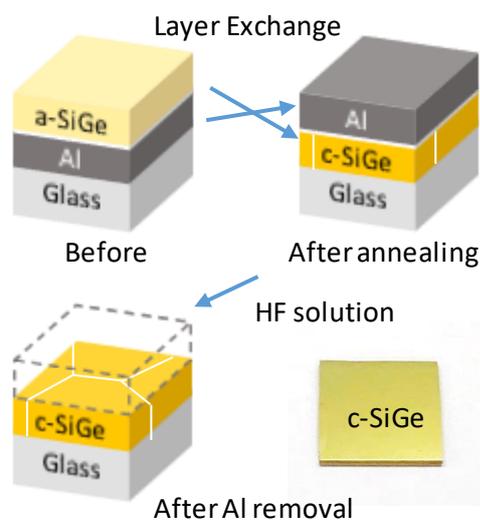


Fig. 1 Schematic of the sample preparation and photograph of a $\text{Si}_{0.7}\text{Ge}_{0.3}$ sample.

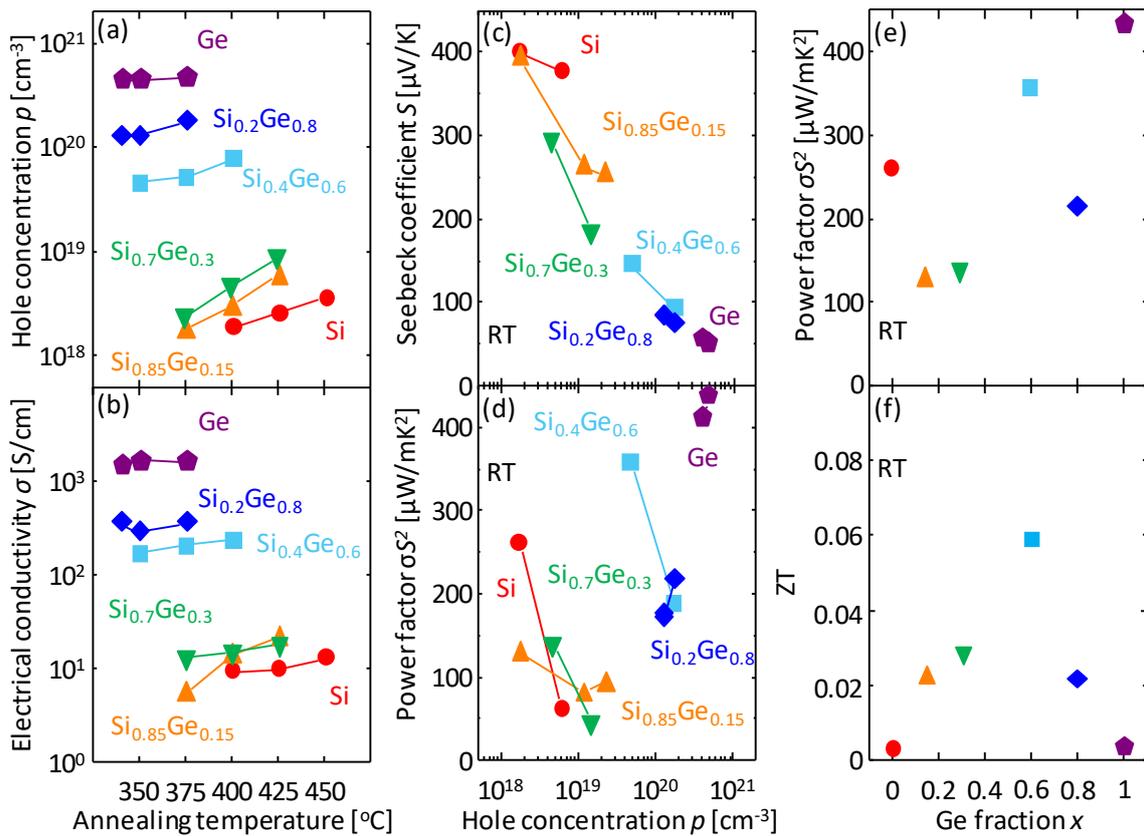


Fig. 2 Thermoelectric properties of $\text{Si}_{1-x}\text{Ge}_x$ (x : 0, 0.15, 0.3, 0.6, 0.8, and 1) layers at room temperature. (a) Hole concentration and (b) electrical conductivity as a fraction of annealing temperature. (c) Seebeck coefficient and (d) power factor as a function of hole concentration. (e) Power factor and (f) ZT as a function of Ge fraction.

doped) in a self-organizing manner during low temperature crystal growth, so that it is possible to achieve both high power factor and low temperature process. In particular, the power factor ($190 \mu\text{W}/\text{mK}^2$) of the sample with a plastic substrate is the highest among all thermoelectric thin films directly grown on plastics.

3. Conclusion

In conclusion, it was demonstrated that SiGe formed by Al induced layer exchange on the insulating substrate has excellent characteristics as thermoelectric thin film. We succeeded in forming $\text{Si}_{0.4}\text{Ge}_{0.6}$ thin film on plastic, showing the highest power factor ($190 \mu\text{W}/\text{mK}^2$) among all thermoelectric thin films directly grown on plastics. These achievements will accelerate the realization of thin film thermoelectric elements.

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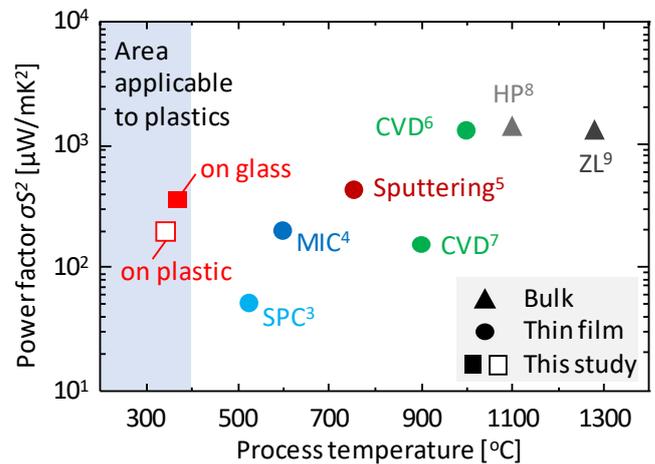


Fig. 3 Power factor of SiGe as a function of process temperature.

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