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Temperature dependence of electron and hole mobilities in heavily impurity-doped SiGe single crystals

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Heavily impurity-doped single crystals of Si₉Ge₁₋ₓ alloy with the composition 0.84 < x < 1 and large-grained polycrystals with x=0.80 were grown by the Czochralski technique. The Hall-coefficient measurements of the electron and hole mobilities in the grown crystals were carried out in the temperature range of 300–1000 K and compared with those in undoped SiGe. The Hall mobilities of electrons and holes in SiGe with a carrier concentration of 10¹⁹–10²⁰ cm⁻³ both show a Tⁿ, n = 1, temperature dependence up to elevated temperatures. This indicates that the carrier transport process is mainly rate controlled by charged impurity scattering. In single-crystal SiGe free from grain-boundary effects, the hole mobility increases with decreasing Si content at least up to 0.84 and the electron mobility is greater than in Si and polycrystalline SiGe. These results suggest that scattering processes in alloy semiconductors are more complicated than previously thought.


I. INTRODUCTION

Silicon-germanium alloys (SiₙGe₁₋ₓ or germanium-silicon Ge₁₋ₓSiₓ, where x is the mole fraction of Si) form a fully miscible solid solution with a diamond crystal structure. These alloys have attracted keen interest in view of their potential for band-gap and lattice-parameter engineering. SiGe thin films grown on Si substrates by various epitaxial techniques are being used for high-speed microelectronic devices. Also, bulk SiGe alloys have the potential for use as x-ray and neutron monochromators and solar cells, and might provide a lattice-matched substrate for SiGe epitaxial growth instead of Si. Highly conductive SiGe alloys are being employed in thermoelectric power generators operating at elevated temperatures up to about 1300 K. An electric power generation in SiGe alloys by conversion of a decay heat of Pt-238 has been employed in deep space probes such as Voyager, Galileo, and Cassini-Huygens. From the 1960s up to the present, extensive experimental and theoretical efforts have been aimed at enhancing the performance of SiGe alloys as thermoelectric devices.¹⁻³ For thermoelectric applications, however, in comparison with other basic factors such as the thermal conductivity and Seebeck coefficient, far less has been reported on the quantitative aspects of the carrier mobility, especially up to operating temperatures. One reason may be the general consensus that polycrystalline materials have a great cost advantage for these applications, although Slack and Hussain have suggested that a high-quality single-crystal SiGe might be a superior choice because grain-boundary scattering could be eliminated.⁴ Moreover, single crystals do not suffer the adverse effects of dopants segregating at grain boundaries. What has most

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range of 2–5 mm/h in a flowing argon gas atmosphere. Details of SiGe alloy growth have been described elsewhere. The alloy composition and homogeneity were determined by energy-dispersive x-ray spectroscopy. From the grown boules, square specimens with dimensions of 5 × 5 × 1 mm³ were prepared for Hall-effect measurements. The Hall coefficient was measured by the Van der Pauw method from room temperature (RT) to 1000 K to determine the mobility and concentration of the electrons or holes.

### III. RESULTS AND DISCUSSION

Table I summarizes the Si content, the Hall mobility (\(\mu_e\) and \(\mu_h\)), and carrier concentration at RT in the SiGe crystals investigated. Undoped Si\(_{0.93}\)Ge\(_{0.07}\) and Si\(_{0.9}\)Ge\(_{0.1}\) crystals were electrically \(p\) type at room temperature and became \(n\) type at temperatures in the range of 200–300 °C. The \(n\)- and \(p\)-type Si\(_{0.8}\)Ge\(_{0.2}\) specimens were polycrystalline with a grain size of 50–100 μm.

Figure 1 shows the carrier concentrations as a function of temperature for the SiGe crystals. The carrier concentrations of the undoped Si\(_{0.93}\)Ge\(_{0.07}\) and Si\(_{0.9}\)Ge\(_{0.1}\) crystals increase rapidly with increasing temperature after their \(p\) to \(n\) conversion, while the concentrations of the heavily impurity-doped crystals are almost independent of temperature up to 800 K. Some specimens show a slight increase of carrier concentrations at temperatures above 800 K due to the increase of intrinsic carriers.

Figure 2 shows the variations of the electron and hole Hall mobilities for the SiGe crystals against temperature. Temperature variations of electron and hole mobilities in SiGe polycrystals reported previously are superimposed. The electron mobilities \(\mu_e\) are somewhat higher than that in Si\(_{0.7}\)Ge\(_{0.3}\) doped with P at \(2.5 \times 10^{20}\) cm\(^{-3}\) by Vandersande,\(^\text{10}\) reproduced in the paper by Slack and Hussain,\(^\text{4}\) while the hole mobilities \(\mu_h\) are the same or a little lower than those in low-density, hot-pressed Si\(_{0.8}\)Ge\(_{0.2}\) doped with B at \(2.4 \times 10^{19}\) cm\(^{-3}\) by Rowe\(^\text{11}\) in the investigated temperature range. The \(\mu_e\) in SiGe with the composition 0.8 < \(x\) < 1 are higher than the \(\mu_h\) in the whole temperature range investigated, the difference originating from the difference of the effective masses. At RT, in undoped SiGe crystals with the composition 0.8 < \(x\) < 1, the electron mobility decreases from 1500 to ~ 500 cm\(^2\)/V s and the hole mobility from 450 to 250 cm\(^2\)/V s with increasing Ge content, and shows a somewhat flat bottom for Si content in the range of 0.5–0.8.\(^\text{12}\) This can be explained in terms of the alloy scattering produced by the disorder fluctuations of the lattice potentials at the lattice sites as noted below. In Fig. 2, the \(\mu_e\) and \(\mu_h\) shown for the impurity-doped SiGe alloys are seen to be much lower than those in undoped specimens in the low-temperature region. \(\mu_e\) and \(\mu_h\) exhibit a gradual decrease with increasing temperature. Apparently, the temperature dependence of \(\mu_e\) and \(\mu_h\) in the SiGe crystals, especially in undoped crystals, becomes larger at higher temperatures, although this feature is not as pronounced in some crystals.

### Table I. Various characteristics of impurity-doped SiGe alloys at room temperature.

<table>
<thead>
<tr>
<th>Si content</th>
<th>Dopant (type)</th>
<th>Single/Poly</th>
<th>(\mu_e)</th>
<th>(\mu_h)</th>
<th>Carrier concentration (10^{19}) cm(^{-3})</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.95</td>
<td>Ga((p))</td>
<td>Single</td>
<td>35</td>
<td>1.3</td>
<td></td>
</tr>
<tr>
<td>0.95</td>
<td>P((n))</td>
<td>Single</td>
<td>96</td>
<td>6.3</td>
<td></td>
</tr>
<tr>
<td>0.93</td>
<td>undoped ((p→n))</td>
<td>Single</td>
<td>150(^\text{a})</td>
<td>0.0002(^\text{a})</td>
<td>0.000006(^\text{a})</td>
</tr>
<tr>
<td>0.93</td>
<td>B((p))</td>
<td>Single</td>
<td>32</td>
<td>2.3</td>
<td></td>
</tr>
<tr>
<td>0.93</td>
<td>P((n))</td>
<td>Single</td>
<td>163</td>
<td>0.43</td>
<td></td>
</tr>
<tr>
<td>0.90</td>
<td>undoped ((p→n))</td>
<td>Single</td>
<td>266</td>
<td>0.0002(^\text{a})</td>
<td>0.000006(^\text{a})</td>
</tr>
<tr>
<td>0.90</td>
<td>B((p))</td>
<td>Single</td>
<td>30</td>
<td>8.5</td>
<td></td>
</tr>
<tr>
<td>0.90</td>
<td>P((n))</td>
<td>Single</td>
<td>64</td>
<td>4.8</td>
<td></td>
</tr>
<tr>
<td>0.84</td>
<td>B((p))</td>
<td>Single</td>
<td>34</td>
<td>11</td>
<td></td>
</tr>
<tr>
<td>0.84</td>
<td>P((n))</td>
<td>Single</td>
<td>81</td>
<td>3.4</td>
<td></td>
</tr>
<tr>
<td>0.80</td>
<td>In((p))</td>
<td>Poly</td>
<td>24</td>
<td>3.4</td>
<td></td>
</tr>
<tr>
<td>0.80</td>
<td>P((n))</td>
<td>Poly</td>
<td>53</td>
<td>9.7</td>
<td></td>
</tr>
</tbody>
</table>

\(^\text{a}\) The undoped specimens converted from \(p\) to \(n\) type at 200–300 °C.

\(^\text{b}\) At 150 °C.

![Graph showing carrier concentration vs. temperature for SiGe crystals.](image)
with a low hole mobility due to the heavy doping. The temperature at which the temperature dependence of \( \mu_e \) and \( \mu_h \) shifts from weak to large is higher in more heavily doped crystals.

The temperature dependence of the Hall mobilities (\( \mu_e \) and \( \mu_h \)) is described by the following equation:

\[
\mu_e \text{ and } \mu_h \sim T^n,
\]

where the exponent \( n = 1 \pm 0.1 \), within experimental error, in the low-temperature region from RT to about 500 K, irrespective of the \( \mu_e \) and \( \mu_h \) values. At high temperatures, \( n = 3 \) in the undoped \( \text{Si}_{0.95}\text{Ge}_{0.05} \) and \( \text{Si}_{0.9}\text{Ge}_{0.1} \) specimens and in \( n \)-type \( \text{Si}_{0.97}\text{Ge}_{0.03} \). The obtained magnitude of \( n \) at low temperatures is comparable to that of \( n \)-type polycrystalline \( \text{Si}_{0.97}\text{Ge}_{0.03} \) alloys reported by Vandersande, \( ^{10} \) and reproduced in the paper by Slack and Hussain. \( ^{4} \) Rowe reported that \( n = 0.8 \) for hot-pressed B-doped \( \text{Si}_{0.9}\text{Ge}_{0.1} \) alloys in the temperature range of 300–950 K. \( ^{11} \) In high-purity Si at room temperature \( n = 2.42 \) and 2.20 are known for \( \mu_e \) and \( \mu_h \), respectively. \( ^{13} \) Among the investigated SiGe crystals, \( \text{Si}_{0.84}\text{Ge}_{0.16} \) and \( \text{Si}_{0.8}\text{Ge}_{0.2} \) crystals show a weak variation of \( \mu_e \) and \( \mu_h \) with temperature up to the highest temperature. This may imply that carrier mobility is independent of temperature in the heavily impurity-doped SiGe alloys with such relatively high Ge contents.

Figures 3(a) and 3(b) show the relationship between the mobility and carrier concentration in the SiGe crystals at RT and 600 °C, respectively. In Fig. 3(a) the previous results in polycrystalline specimens of zone-level-grown \( \text{Si}_{0.7}\text{Ge}_{0.3} \) reported by Dismukes et al. \( ^{2} \) of hot-pressed \( \text{Si}_{0.8}\text{Ge}_{0.2} \) by Rowe, \( ^{11} \) and of arc-melted \( \text{Si}_{0.95}\text{Ge}_{0.05} \) by Yamashita and Sadatomi \( ^{14} \) are compared. The \( \mu_e \) and \( \mu_h \) versus carrier concentration relationships reported in Si by Masetti et al. \( ^{15} \) are also included. As seen in the figure, the measured \( \mu_e \) and \( \mu_h \) values decrease with increasing carrier concentration due to charged impurity scattering, as reported previously. The decrease of \( \mu_e \) with increasing carrier concentration is much larger than that of \( \mu_h \). The values of \( \mu_h \) in SiGe measured in the present work are close to those in polycrystalline SiGe reported by other groups \( ^{2,11,14} \) and are clearly lower than that in Si. \( ^{15} \) No systematic effect of different dopant species, such as Ga, In, or B, in \( p \)-type crystals can be detected explicitly. It is generally thought that \( \mu_e \) is smaller in SiGe than in Si. In the present work, some SiGe samples were found to have similar values of \( \mu_e \) as reported for polycrystalline SiGe by other groups, but it is seen that the single-crystalline \( \text{Si}_{0.95}\text{Ge}_{0.05} \) and \( \text{Si}_{0.9}\text{Ge}_{0.07} \) have greater values of \( \mu_e \) than those observed for Si and polycrystalline SiGe.

At present, far less is known about the carrier mobility versus concentration relation at elevated temperatures compared with such relation at RT. In Fig. 3(b), the concentration-dependent features of \( \mu_e \) and \( \mu_h \) seem to be similar at RT and 600 °C, with \( \mu_h \) having smaller magnitudes and weaker dependences than \( \mu_e \). Although the data in the present work are limited, a remarkable feature in single-crystalline alloys can be noted. The measured \( \mu_h \) for a carrier...
concentration of $\sim 10^{20}\text{cm}^{-3}$ seems to increase with decreasing Si content, i.e., in the order $\text{Si}_{0.95}\text{Ge}_{0.05}$, $\text{Si}_{0.9}\text{Ge}_{0.1}$, and $\text{Si}_{0.8}\text{Ge}_{0.16}$. Such enhancement effect was not observed for $\mu_e$.

Several scattering mechanisms contribute to the carrier transport process in SiGe alloys: dominantly, carrier-phonon scattering, alloy disorder scattering, charged (ionic) impurity scattering, and so on. Intervalley phonon scattering due to $L$ conduction-band transitions contributes to electron mobility when the electron concentration is higher than $10^{20}\text{cm}^{-3}$. Grain-boundary scattering can be excluded for single-crystal materials. Previously, based on the relation between the Seebeck coefficient and electrical conductivity, we reported that the carrier transport in heavily impurity-doped SiGe alloys is controlled by the alloy disorder and charged impurity scattering. The present results reveal that $\mu_e$ and $\mu_h$ in heavily impurity-doped SiGe alloys have similar characteristic temperature dependence, especially at temperatures around RT. In the undoped SiGe, the electron mobility decreases rapidly with increasing temperature, which originates around RT. The hole mobility increases rapidly with increasing temperature, which originates from RT to 1000 K. The Hall mobilities of electrons and holes in the SiGe with the carrier concentration of $10^{19} - 10^{20}\text{cm}^{-3}$ both show the temperature dependence as $T_n^\alpha$, $n \sim 1$, up to elevated temperatures, which suggests that carrier transport is controlled mainly by charged impurity scattering. Free from grain-boundary effects, the following two characteristics are detected in the SiGe single crystals: The hole mobility tends to increase with decreasing Si content to 0.84. The electron mobility in the Si-rich SiGe single crystals is greater than in Si and polycrystalline SiGe. These features, as well as recently acquired knowledge on the atomic structure, suggest the necessity of reevaluating the role of scattering processes in SiGe alloys, including further examination in a wide temperature range.

### IV. SUMMARY

This paper presents preliminary results of carrier mobility in the heavily impurity-doped $\text{Si}_x\text{Ge}_{1-x}$ single crystals with the composition $0.84 < x < 1$ in the temperature range from RT to 1000 K. The Hall mobilities of electrons and holes in the SiGe with the carrier concentration of $10^{19} - 10^{20}\text{cm}^{-3}$ both show the temperature dependence as $T_n^\alpha$, $n \sim 1$, up to elevated temperatures, which suggests that carrier transport is controlled mainly by charged impurity scattering. Free from grain-boundary effects, the following two characteristics are detected in the SiGe single crystals: The hole mobility tends to increase with decreasing Si content to 0.84. The electron mobility in the Si-rich SiGe single crystals is greater than in Si and polycrystalline SiGe. These features, as well as recently acquired knowledge on the atomic structure, suggest the necessity of reevaluating the role of scattering processes in SiGe alloys, including further examination in a wide temperature range.

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