Optimisation of the Figure of Merit of Modified Silicon-Germanium Alloys

Final Technical Report

D M Rowe

February 1989

United States Army
EUROPEAN RESEARCH OFFICE OF THE US ARMY
London, England
Contract Number: DAJA 45-87-C-0048

UNIVERSITY OF WALES COLLEGE OF CARDIFF

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Silicon–germanium alloys are established thermoelectric materials for high temperature applications and have been successfully used in the fabrication of the thermocouples of radioisotope thermoelectric generators which provided the onboard electrical power to several US space vehicles. Recently, material development scientists have focused attention on possible improvements in the figure of merit of alloys based on silicon–germanium through an increase in $zT$ (the so-called electrical figure of merit, or power factor). In the final report is embodied the results of a program of work to develop a realistic working theoretical model of the silicon germanium alloy systems and its use in identifying and investigating the parameters which determine the value of the electrical figure of merit. The analysis indicates that although the electrical figure of merit of silicon–germanium alloys in general decreases with increase in silicon content, the relative independence on alloy composition at high carrier concentrations ($5 \times 10^{19} \text{ cm}^{-3}$) tilts the final choice of composition towards an alloy which possesses (1) a large energy...
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Since this present program of work started, it has been reported that the electrical figure of merit of silicon germanium alloys to which small amounts of gallium phosphide additive have been added can be significantly increased by high temperature thermal anneals. Once conclusion of the work reported here is that increasing the carrier concentration by enhancing the dopant solubility could result in an improved EFOM. It could well be that the presence of gallium phosphide does just that, although it is very unlikely that the magnitude of the reported increase (approaching 40% at 500°C) can be solely attributed to an increase in carrier concentration. In addition, an increase in carrier concentration will result in an unwanted increase in the electronic contribution to the thermal conductivity, although this may well be offset by the reduction in the lattice component which is reported to accompany the addition of III-V additives to these alloys.
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ABSTRACT

Silicon-germanium alloys are established thermoelectric materials for high temperature applications and have been successfully used in the fabrication of the thermocouples of radioisotope thermoelectric generators which provided the onboard electrical power to several United States space vehicles.

Recently, material development scientists have focussed attention on possible improvements in the figure of merit of alloys based on silicon-germanium through an increase in $\alpha^2\sigma$ (the so-called electrical figure of merit, or power factor).

In this report is embodied the results of a programme of work to develop a realistic working theoretical model of the silicon germanium alloy system and its use in identifying and investigating the parameters which determine the value of the electrical figure of merit.

The analysis indicates that although the electrical figure of merit of silicon-germanium alloys in general decreases with increase in silicon content, the relative independence on alloy composition at high carrier concentrations ($> 5 \times 10^{19} \text{ cm}^{-3}$) tilts the final choice of composition towards an alloy which possesses (i) a large energy band gap to decrease unwanted minority effects, and (ii) a low lattice thermal conductivity - which favours a higher silicon content.

Having optimised the alloy composition bearing in mind the factors identified above, it should be noted that a high electrical figure of merit favours a large number of equi-energetic valleys and a high carrier concentration to take advantage of the multivalleyed band structure. However, in order to approach the optimum value of the electrical figure of merit of silicon germanium alloys, it is found necessary in practice to dope silicon germanium alloys to concentrations at the solubility limits of the dopants in these alloys. Since this present programme of work started, it has been reported that the electrical figure of merit of silicon germanium alloys to which small amounts of gallium phosphide additive have been added can be significantly increased by high temperature thermal anneals. One conclusion of the work reported here is that increasing the carrier concentration by enhancing the dopant solubility could result in an improved EFOM. It could well be that the presence of gallium phosphide does just that, although it is very unlikely that the magnitude of the reported increase (approaching 40% at 500 C) can be solely attributed to an increase in carrier concentration. In addition, an increase in carrier concentration will result in an unwanted increase in the electronic contribution to the thermal conductivity although this may well be offset by the reduction in the lattice component which is reported to accompany the addition of III-V additives to these alloys.
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II. General Introduction

Silicon germanium alloys are established thermoelectric materials for high temperature applications and they have been successfully used in the fabrication of the thermocouples of radioisotope thermoelectric generators (RTG) which provided onboard electric power to space vehicles such as the LES 8/9 communication satellite and the Voyager spacecrafts (1). Once the operating conditions of a thermoelectric generator have been decided, the performance of the conversion system can be expressed in terms of the thermocouple material's so-called figure of merit $z = \alpha^2 \sigma / \lambda$, where $\alpha$ is the Seebeck Coefficient and $\sigma$ is the electrical conductivity. The thermal conductivity $\lambda$ consists mainly of two parts, an electronic component $\lambda_e$ and a lattice component $\lambda_L$. The lattice is the major component, even in the heavily doped materials used in thermoelectric applications and typically accounts for around 75% of the total thermal conductivity. In space applications, savings in weight and isotopic fuel inventory is an important consideration and over the past ten years or so a sustained effort has been maintained, principally in the United States to improve the figure of merit of high temperature thermoelectric materials. Emphasis has concentrated on reducing the lattice thermal conductivity and substantial reductions have been obtained by (a) employing very small grain size materials (2) and (b) the addition of small amounts of III-V materials such as Ga-P (3). The reduction in thermal conductivity which accompanies the use of these two methods is displayed in Figures 1 and 2 (4). However, little data has been published on the electrical properties of these modified materials and what there is indicates that a reduction in thermal conductivity is usually accompanied by a reduction in electrical conductivity (5). In addition, there is a general awareness that having introduced these additional phonon scattering mechanisms into the material's structure we may be approaching the minimum of the lattice thermal conductivity in these alloys (6).

During the proposal stage of this programme of work the investigator was unaware of the substantial improvements in the electrical figure of merit (EFOM) being obtained by researchers at JPL (7). In view of these developments and following discussion with Dr. Guido Guazzoni, the scientific representative for the contract sponsors (US Army - Fort Monmouth) it was decided to modify the investigation and concentrate the effort on the behaviour of the electrical figure of merit of silicon germanium alloys and not the overall figure of merit as had been originally envisaged.

Adopting this approach would provide useful information on silicon germanium alloys and enable a solid foundation to be laid for any subsequent
programme of work into the behaviour of silicon germanium-gallium phosphide material and in particular into the conditions which have been reported to enhance their thermoelectric performance.

The development of a comprehensive and realistic theoretical model which provides a description of the electrical properties of the silicon germanium alloy system is difficult and is best approached in stages, commencing with a working model and progressing to more realistic models of increased complexity. Based upon these considerations, a programme of work was proposed which had the following objectives.

III. Objectives
1. Study the variation of $\alpha^2\sigma$ with reduced Fermi energy and carrier concentration for a range of alloy compositions.
2. Undertake a literature survey with the view to exploring changes in the band structure of silicon germanium alloys with change in composition from germanium to pure silicon; identifying the effect of these changes on $\alpha$ and $\sigma$.
3. As stage 1, but including the effect of non-parabolic energy bands on the theoretical model.
4. Develop an idealised model and explore the variation of $\alpha^2\sigma$ with number of valleys.
5. Identify the parameters and conditions which optimise the electrical figure of merit $\alpha^2\sigma$.

IV. General Considerations
1. Literature Survey
A survey of the literature revealed that a detailed investigation of the band structure of these alloys had been carried out by a number of workers (8, 9, 10, 11, 12, 13, 14, 15, 16). Papers by Amith and Rosi (17, 18) provide a good description of the subject matter as it appeared two decades ago. The alloys are substitutional and maintain the diamond structure characteristic of both silicon and germanium. Although the periodicity of the lattice is disturbed in an alloy, a distinct energy band structure does appear to exist. Following Herman (19), it is possible to speculate on the manner in which the band structure of germanium deforms to become the band structure of silicon as the composition of the alloys is changed from 0% silicon to 100% silicon. In developing the theoretical model, account is taken of the basic band structure parameters that have a strong bearing on the electrical properties - the EFOM can then be optimised with respect to variations in these parameters and level of doping.
2. Energy Band Gap and Effective Mass

The energy band gap is an important band structure parameter - among other things it determines the highest utility temperature of a material in thermoelectric applications (20). The variation of energy gap with composition exhibits the following features - in the region 0 to 15 mol % silicon the energy gap increases linearly from 0.72 to 0.94 electron volts; a break then occurs and the gap continues to increase, reaching 1.2 electron volts at pure silicon.

As the silicon content increases, the conduction band of germanium having (111) minima is raised relative to the conduction band (100) minima of silicon. Both of the conduction bands move away from the valence bands with the top of the valence band structure remaining at the central point of the reduced zone over the entire range of alloy compositions. Measurements of magneto-resistance and Hall effect on n-type alloys showed that in alloys with a silicon content greater than 23%, the electronic conduction takes place in minima which are spheroids orientated along the (100) axes in the reduced zone. It is concluded that the electrons in these minima have about the same effective mass in the alloys as they have in silicon.

3. Seebeck Coefficient

Measured values of the Seebeck Coefficient exhibit a peak in alloys having 15% silicon. This corresponds to the composition where there is a break in the energy gap versus composition curve. Amith's theoretical analysis of the composition dependence of the Seebeck Coefficient, which is in good agreement with the experimental data, shows that the behaviour of \( \alpha \) near this composition reflects the fact that there are ten equi-energetic valleys, four along the (111) axis and six along the (100) axis, resulting in a very large density of states.

4. Theoretical Options

The presence of a maxima in the Seebeck Coefficient versus alloy composition curve is not sufficient reason to choose an alloy of this composition as being the most suitable for use in thermoelectric applications. A number of factors must be taken into account, one the most important of which is the thermal conductivity. The lattice thermal conductivity exhibits a much stronger dependence on alloy composition than the electrical properties including the Seebeck Coefficient. However, our concern in this programme of work is the behaviour of the electrical figure of merit and the parameters which have a strong influence on it, namely the energy gap and the carrier density of states.
5. Compositional Dependence

It is not easy to incorporate the compositional dependence of the electrical properties in the theoretical model - unlike their dependence on carrier concentration. All theoretical expressions for the electrical properties can be conveniently expressed in terms of the Fermi level which in turn depends upon the level of doping and temperature. An equivalent situation does not cover the dependence on composition. Two options are available: (i) to examine each composition separately and independently, (ii) identify those parameters which are effective in influencing the Seebeck Coefficient and electrical conductivity.

6. Equivalent Valleys

The carrier density of states has a profound influence on the electrical figure of merit.

The density of states corresponding to a single valley is given by

\[ g_s(E) = 4\pi \left( \frac{2m^* ds}{\hbar^2} \right)^{1/2} E^{3/2} dE \]

where \( E \) is the carrier energy measured with respect to the band edge and \( m^* ds \) is the density of states effective mass. If there are \( N_v \) equivalent valleys, each will contribute \( g_s(E) \) to the total density of states \( g_t(E) \) and is given by

\[ g_t(E) = N_v g_s(E) \]

This situation may then be taken into account by defining a total density of states \( m^* dt \) where

\[ g_t(E) = N_v 4\pi \left( \frac{2m^* dt}{\hbar^2} \right)^{1/2} E^{3/2} dE \]

This gives

\[ m^* dt = N_v^{2/3} m^* ds = N_v (m_1 m_2 m_3)^{1/3} \]

where \( m_1, m_2 \) and \( m_3 \) are the components of the mass tensor along the principal axes.

It should be noted that it may be possible to increase the total density of states which changes the total density of states effective mass without affecting the conductivity effective mass defined by

\[ \frac{1}{m^*} = \frac{1}{3} (m_1^{-1} + m_2^{-1} + m_3^{-1}) \]

The relevant parameter which must be varied is the number of equivalent valleys.
7. Variation of \( N \) for a Fixed Composition

It is clearly relevant to investigate the variation of the electrical properties with a change in the number of equivalent valleys (avoiding the complications associated with the difficult problem of the composition dependence of the various parameters). As a first step, the changes in parameters closely linked with the band structure are investigated in an indirect way to provide an insight of the magnitude of the possible changes in the system which tend to optimise the electrical figure of merit.

8. Non-Parabolicity of Bands

Although silicon germanium alloys cannot be described as narrow band semiconductors, the high level of doping usually employed in thermoelectric materials necessitates the inclusion of a deviation from the usually assumed parabolic band. The non-parabolicity of the bands is interpreted in terms of a parameter \( \beta = \frac{k_B T}{E_g} \) where \( k_B \) is the Boltzmann constant and \( T \) is the absolute temperature; \( E_g \) is the energy band gap. Non-parabolicity has a strong influence on the electrical properties at high carrier concentrations. Moreover, these effects are quantitatively dependent on the scattering mechanisms.

V. Theoretical Model

At room temperature and a little above, acoustic phonon scattering is the dominant charge carrier scattering mechanism. In the model adopted we have considered a multivalled, non parabolic band structure but excluded the effect of intervalley scattering. Intervalley scattering requires the presence of high energy phonons which are not excited at around room temperature.

It is convenient to work in terms of reduced electrical conductivity \( \sigma' \) and reduced Seebeck coefficient \( \alpha' \) where

\[
\sigma' = \left[ \frac{k_B}{\theta} \right]^2 \frac{T}{\lambda_L} \sigma \quad \text{and} \quad \alpha' = \frac{\theta}{k_B} \alpha
\]

\[
\sigma' = \left( \frac{k_B^2 \hbar c \lambda_1}{3 n^2 e_1^2} \right) \frac{N_{\lambda_1} O_{\lambda_1} T}{(2 e_1^2 \lambda_1)}
\]

and \( \alpha' = (5 - \xi) \) with \( \xi = \left( \frac{O_{\lambda_1}}{O_{\lambda}} \right) \)

The electrical figure of merit is given by

\[
\alpha'^2 \sigma = \frac{\lambda_L}{T} \sigma'^2 \sigma' = \left( \frac{k_B^2 \hbar c \lambda_1}{3 n^2 e_1^2} \right) \frac{N_{\lambda_1} O_{\lambda_1}}{k_B^2} (5 - \xi)^2
\]
with $\epsilon_1$ the acoustic deformation potential, $C_{11}$ the longitudinal elastic constant, $N_V$ the number of equivalent energy valleys, $m_{c*}$ the conductivity (inertial effective mass), and $\lambda_L$ the lattice thermal conductivity. The generalised Fermi integrals $n_{L}^{\pm}$ which appears in the electronic transport coefficient expressions are given by

$$n_{L}^{\pm} (\tau) = \int_{0}^{\infty} \left[ \frac{\partial f}{\partial \eta} \right] n^\eta (\eta(1 + Bn)) \eta (1 + 2Bn)^\eta d\eta$$

$f$ is the Fermi distribution function, $\eta = E/kT$, $B = k_B T/E_g$

The carrier concentration $n_c$ is related to the Fermi level by

$$n_c = N_V \frac{1}{3n^2} (2m_{d*}^n k_B T)^{3/2} \sigma_{L0}^{3/2}$$

The effective mass values for the electronic states in the alloys are about the same as for corresponding states in pure crystals; consequently, we initially employed a simple linear compositional dependence of effective mass using values given in Table 1, making no distinction between the density of states and conductivity effective masses.

<table>
<thead>
<tr>
<th></th>
<th>Ge</th>
<th>Si</th>
</tr>
</thead>
<tbody>
<tr>
<td>$m_{L*}/m_0$</td>
<td>0.08</td>
<td>0.19</td>
</tr>
<tr>
<td>$m_{L*}/m_0$</td>
<td>1.58</td>
<td>0.92</td>
</tr>
<tr>
<td>$m_{d*}/m_0$</td>
<td>0.216</td>
<td>0.32</td>
</tr>
<tr>
<td>$m_{c*}/m_0$</td>
<td>0.117</td>
<td>0.26</td>
</tr>
</tbody>
</table>

Table 1. Effective mass values for germanium and silicon

Values for $C_{11}$, $\epsilon_1$ and $\lambda_L$ are taken to be $1.7 \times 10^{11} \text{N m}^{-2}$, 6.2 eV and 5.00 W m$^{-1}$ deg$^{-1}$ respectively.

In the calculations the atomic percent of silicon has been varied from 30% to 90% in view of the expected usefulness of silicon rich alloys in device applications. The energy gap varies linearly over the range of composition.

A slight improvement in the model can be made by considering the density of states and conductivity effective masses separately for silicon and germanium and obtaining the corresponding values for the alloy compositions by linear interpolation.
Results

1. Carrier Concentration Dependence

The dependence of the Seebeck Coefficient electrical conductivity and EFOM with carrier concentration and parameter N has been obtained from the equations described previously. Other relevant parameters - energy gap, effective mass elastic constant, deformation potential and lattice thermal conductivity correspond to a Si70Ge30 alloy composition. However, \( N_v \) has been varied in the range 1 to 10 which makes the analysis a study of the band structure dependence rather than that of a single valley composition. As indicated previously, a variation of \( N_v \) incorporates into the model an important feature of the band structure and this can be treated as an essential first step towards the study of the thermoelectric behaviour of these alloys with respect to composition variation.

The results of the calculations are presented in Figures 3, 4 and 5. It is observed that an increase in the number of valleys in the absence of intervalley scattering has two important effects: (i) increases the value of \( \alpha \) and \( \sigma \) and hence \( \alpha^2\sigma \), (ii) shifts the maximum in the \( \sigma^2\sigma \) versus carrier concentration curve towards higher concentration. The maximum value of \( \sigma^2\sigma \) also increases as \( N_v \) increases as also does \( n_{opt} \) where \( n_{opt} \) is the carrier concentration which optimises \( \sigma^2\sigma \). In Table 2 is presented \( \alpha^2\sigma_{\text{max}} \) and \( n_{opt} \) for a fixed carrier concentration and \( N_v \) values. Figures 6 and 7 display the variation of \( \sigma^2 \) and \( \sigma^2n_{\text{opt}} \) with number of valleys \( N_v \).

<table>
<thead>
<tr>
<th>( N_v )</th>
<th>( (\alpha^2\sigma)_{\text{max}} ) (W/mdeg²)</th>
<th>( n_{opt} ) (cm⁻³)</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>7.5</td>
<td>4 x 10¹⁸</td>
</tr>
<tr>
<td>2</td>
<td>13.5</td>
<td>9 x 10¹⁸</td>
</tr>
<tr>
<td>4</td>
<td>26.0</td>
<td>1.7 x 10¹⁹</td>
</tr>
<tr>
<td>6</td>
<td>38.0</td>
<td>2.5 x 10¹⁹</td>
</tr>
<tr>
<td>8</td>
<td>51.5</td>
<td>3.4 x 10¹⁹</td>
</tr>
<tr>
<td>10</td>
<td>64.5</td>
<td>4.5 x 10¹⁹</td>
</tr>
</tbody>
</table>

Table 2. Optimum Values of \( \alpha^2\sigma \) and \( n \)

2. Composition Dependences

The energy gap is an important parameter which changes with alloy composition and has an important bearing on the thermoelectric behaviour of the material. It is the parameter which determines the highest utility temperature of a semiconductor when used in thermoelectric applications. This is manifested through the effect of impurity carriers across the energy band gap which in turn
influences the electrical conductivity, Seebeck Coefficient and the thermal conductivity. This requires that account be taken of the bipolar thermal conductivity. Initial calculations employed a relatively simple model in which no distinction is made between the density of states and the conductivity effective masses. In Figure 8 is displayed the variation of $\alpha^2 \sigma$ with reduced Fermi potential and for four different alloy compositions. Figure 9 shows $\alpha^2 \sigma$ plotted against electron concentration while in Figure 10 is plotted $\alpha^2 \sigma$ against silicon content for four different carrier concentrations; the calculations corresponding to a temperature of 300 K.

A slight improvement in the model is possible if the density of states effective masses and conductivity effective masses are taken separately for silicon and germanium and the appropriate values obtained for the alloy composition by assuming a linear interpolation (Figures 11, 12 and 13).

VII. Conclusions and Concluding Remarks

1. Conclusions

Although the model adopted is relatively simplistic, a number of general conclusions can be drawn regarding the electrical figure of merit.

(a) At relatively low temperatures when intervalley scattering can be taken as negligibly small, a high electrical figure of merit favours a large number of equienergetic valleys.

(b) The maximum in the $\alpha^2 \sigma$ versus $n$ shifts towards higher carrier concentrations for alloy compositions with larger silicon content.

(c) Above a critical carrier concentration (about $5 \times 10^{19}$ cm$^{-3}$), the electrical figure of merit becomes independent of alloy composition.

(d) The absolute values of $\alpha^2 \sigma$ are sensitive to changes in the value of the effective mass. However, it is apparent from the results that an inappropriate choice of effective mass does not change its qualitative behaviour.

(e) The electrical figure of merit decreases with an increase in the silicon content. Other parameters such as the thermal conductivity, which is strongly dependent on alloy composition, must be taken into account when considering the overall figure of merit.

(f) Although the electrical figure of merit in general decreases with increase in silicon content, its relative independence on alloy composition at high carrier concentrations tilts the final choice of composition towards an alloy which possesses (i) a large energy band gap to decrease unwanted minority effects, and (ii) a low lattice thermal conductivity - which favours a somewhat higher silicon content.
2. Concluding Remarks

Having optimised the alloy composition bearing in mind the factors identified above, it should be noted that a high electrical figure of merit favours a large number of equi-energetic valleys and a high carrier concentration to take advantage of the multivallied band structure. However, in order to approach the optimum value of the electrical figure of merit of silicon germanium alloys, it is found necessary in practice to dope silicon germanium alloys to concentrations at the solubility limits of the dopants in these alloys. Since this present programme of work started, it has been reported that the electrical figure of merit of silicon germanium alloys to which small amounts of gallium phosphide additive have been added can be significantly increased by high temperature thermal anneals. One conclusion of the work reported here is that increasing the carrier concentration by enhancing the dopant solubility could result in an improved EFOM. It could well be that the presence of gallium phosphide does just that, although it is very unlikely that the magnitude of the reported increase (approaching 40% at 500 C) can be solely attributed to an increase in carrier concentration. In addition, an increase in carrier concentration will result in an unwanted increase in the electronic contribution to the thermal conductivity although this may well be offset by the reduction in the lattice component which is reported to accompany the addition of III-V additives to these alloys.

VIII. Acknowledgements

Dr. C. M. Bhandari is thanked for acting as consultant to the project and his valuable contributions to the work embodied in this report are appreciated. The United States Army is acknowledged for supporting this work under Contract No., DAJA 45-87-C-0048. Mrs. M. Price is thanked for typing the report.
IX. References


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Fig. 4. Variation of $\alpha'$ with carrier concentration and number of valleys
Fig. 5. Variation of $a^4\sigma$ with carrier concentration and number of valleys.

Fig. 6. Variation of $a^4$ with number of valleys.
Fig. 7. Variation of $\alpha^2 \sigma$ and $\alpha$ with number of valleys

Fig. 8. Variation of $\alpha^2 \sigma$ with reduced Fermi potential for four different carrier concentrations
300 K
At fraction Si
1. 0.3
2. 0.5
3. 0.7
4. 0.9

Fig. 9. Variation of $a^2\sigma$ with electron concentration for four different alloy compositions.

300 K
A. $10^{18}$ cm$^3$
B. $2 \times 10^{19}$ cm$^3$
C. $6 \times 10^{19}$ cm$^3$
D. $10^{20}$ cm$^3$

Fig. 10. Variation of $a^2\sigma$ with silicon content for four different carrier concentrations.
Fig. 11. Variation of $\alpha^2 \sigma$ with reduced Fermi potential for four different carrier concentrations (improved model)

Fig. 12. Variation of $\alpha^2 \sigma$ with electron concentration for four different alloy compositions (improved model)
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