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Thermal conductivity of thermoelectric Si_{0.8}-Ge_{0.2} alloys

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The thermal conductivity of heavily doped, n-type Si-Ge alloys has been studied from 300 to 1200 K. The scattering rate of several phonon scattering mechanisms has been calculated, including intrinsic scattering, mass defect and distortion scattering, phonon-electron scattering, and scattering by inclusions. These rates were then used to calculate the lattice thermal conductivity. The electronic component of the thermal conductivity was calculated from the calculated Lorenz ratio and measured values of the electrical conductivity. The total thermal conductivity was then compared to measured values for a specimen studied by Vining *et al.* [J. Appl. Phys. **69**, 15 (1991)].

I. INTRODUCTION

The thermal conductivity of thermoelectric materials is of interest because the efficiency of thermoelectric power generators is directly related to the dimensionless figure of merit,

$$ZT = S^2 T / \rho k = (S^2 / L) [k_e / (k_e + k_l)]$$

where S is the thermoelectric power, ρ is the electrical resistivity, k is the thermal conductivity, k_e and k_l are the electronic and lattice components of the thermal conductivity, L is the Lorenz ratio, and T is the temperature. To effect improvements it is important to characterize the mechanisms that determine their lattice conductivity.

In this paper the phonon relaxation rates for the relevant interaction processes are presented and the thermal conductivity is calculated from these relaxation times using standard theory.¹⁻³ Different processes are important in different frequency regimes, and are needed in combination to reduce k_k . The interaction of phonons with electrons is shown to be sufficiently strong in the materials doped for optimal performance to overshadow the phonon scattering by grain boundaries in the relevent frequency range. It is also shown that the scattering of phonons by unionized donors, a mechanism first proposed by Keyes,⁴ is a significant source of phonon scattering in these materials at 300 K but becomes less significant at higher temperatures. The frequency regime of the phonon spectrum that carries most of the thermal current in these materials is identified, so that further work on reducing the lattice thermal conductivity may concentrate on that frequency regime.

II. INTRINSIC SCATTERING

It can be shown, e.g., Klemens,² that the scattering of phonons by three-phonon umklapp processes may be described by a relaxation time of the form

$$\frac{1}{\tau_i} = \frac{1}{\tau_0} \frac{\omega^2}{\omega_D^2} \frac{T}{\Theta},\tag{1}$$

where τ_0 is a constant of the dimension of time, ω is the phonon frequency, ω_D is the Debye frequency, T is the temperature, and Θ is the Debye temperature. The intrinsic thermal conductivity is given by the integral

$$\lambda_i = \frac{1}{3} \int_0^{\omega_D} \tau_i(\omega) v^2 C(\omega) d\omega, \qquad (2)$$

where $C(\omega)$ is the contribution to the specific heat and v is the phonon velocity. Substituting τ_i from Eq. (1) into Eq. (2) and using the high-temperature limit for the spectral specific heat, the intrinsic relaxation time can be expressed in terms of the intrinsic thermal conductivity by

$$\frac{1}{\tau_i} = \frac{k_B \omega_D}{2\pi^2 v} \frac{1}{\lambda_i} \omega^2, \tag{3}$$

where k_B is the Boltzmann constant. The intrinsic thermal conductivity of the alloy is calculated as follows. The intrinsic conductivity obtained by Leibfried and Schloemann⁵ is of the form

$$\lambda_i = C \frac{1}{\gamma^2} \left(\frac{k_B}{h}\right)^3 M a \frac{\Theta^3}{T}.$$
 (4)

Here C is a constant, M is the mass per atom, γ is the Grueneisen constant, a^3 is the volume per atom, and h is Planck's constant. This was done by linearly interpolating the values of C, M, a, and Θ , for an 80% Si 20% Ge alloy between Si and Ge, using the conductivities of Si and Ge measured by Glassbrenner and Slack.⁶

III. POINT DEFECT SCATTERING

It can be shown^{7,8} that the relaxation rate for mass defect and distortion scattering is given by

$$\frac{1}{\tau} = \frac{a^3}{4\pi v^3} \,\epsilon \omega^4,\tag{5}$$

where

$$\epsilon = \sum_{i} c_{i} [(1 - M/M_{i}) + (9\gamma/2)(1 - a_{i}/a) \\ \times (1 - c_{i})\kappa/c_{i}\mu]^{2}.$$
(6)

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The sum is over the components of the alloy, c_i is the concentration of species *i*, κ is the bulk modulus of the matrix, μ is the shear modulus of the matrix, a_i is the lattice spacing of species *i*, $M = \sum_i c_i M_i$, $a = \sum_i c_i a_i$, $\kappa = \sum_i c_i \kappa_i$, and $\mu_i = \sum_i c_i \mu_i$.

IV. GRAIN-BOUNDARY SCATTERING

The form assumed for the relaxation rate due to boundary scattering was

$$\frac{1}{\tau} = \frac{v}{l},\tag{7}$$

where l is the average diameter of the grains that make up the material.

While the actual mechanism of grain-boundary scattering is not yet understood in detail, experimental evidence suggests that this is of the right magnitude,⁹ particularly as the average grain diameter is not easy to determine in polycrystalline specimens.

V. PHONON-ELECTRON SCATTERING

The phonon relaxation time due to the interaction with free electrons was calculated using a momentum balance argument. This procedure necessitates describing the departure from equilibrium of the phonon and electron distribution functions and relates the phonon relaxation time to the electron relaxation time.¹⁰ The departure from equilibrium of an electron distribution in the presence of an electric field can be written in the form

$$g = -\lambda \cdot \mathbf{k} \left(\frac{\partial f^0}{\partial E} \right), \tag{8}$$

where $\lambda = \hbar \mathbf{v}_d$, v_d is the electron drift velocity, \hbar is $h/2\pi$, **k** is the electron wave vector, and f^0 is the equilibrium electron distribution function. Similarly the departure from equilibrium of a phonon distribution in the presence of a temperature gradient is given by

$$n = (\lambda \cdot \mathbf{q}) \frac{\partial N^0}{\partial E}, \qquad (9)$$

where $E = \hbar \omega$, **q** is the phonon wave vector, N^0 is the equilibrium distribution, and λ is at this point an arbitrary vector.

The rate integral for the phonon-electron interaction contains the term

$$f(\mathbf{k}')[1-f(\mathbf{k})][N(\mathbf{q})+1] - f(\mathbf{k})[1-f(\mathbf{k}')]N(\mathbf{q}),$$
(10)

where $f = f^0 + g$, and $N = N^0 + n$. Substituting in Eqs. (8) and (9) into this expression it is found that it is stationary only if λ is common to both the electron and phonon distributions. If the λ 's differ the rate integral is proportional to the difference. Thus if the time rate of change of the phonon gas momentum is equated to the rate of change of the electron gas momentum a momentum balance condition can be described,

$$\left|\frac{dP_e}{dt}\right| = \left|\frac{dP_p}{dt}\right|,\tag{11}$$

where P_e is the electron momentum and P_p is the phonon momentum. Assuming relaxation times τ_e and τ_p for the electrons and phonons, respectively, this condition becomes

$$\frac{\partial P_e}{\partial \lambda} \frac{1}{\tau_e} = \frac{\partial P_p}{\partial \lambda} \frac{1}{\tau_p},\tag{12}$$

thus the "momentum capacities" $\partial P_e / \partial \lambda$ and $\partial P_p / \partial \lambda$ and the electron relaxation time must be determined in order to determine the phonon relaxation time. The phonon momentum capacity may be expressed in terms of the phonon heat capacity as

$$\frac{\partial P_p}{\partial \lambda} = \frac{1}{3} \frac{C_p T}{v^2 \hbar}.$$
(13)

This argument applies to metals as well as to semiconductors. In the latter case, since there is a maximum value for the electron energy, wave-vector conservation dictates a maximum interacting phonon wave vector and in the evaluation of C_p it is necessary to consider only those phonons with wave number less than this maximum, q_0 . For Maxwell-Boltzmann statistics one can approximate the maximum electron energy to be k_BT and thus

$$q_0 = \frac{4\pi}{h} \sqrt{2mk_B T},\tag{14}$$

where m is the electron effective mass. In the high-temperature limit

$$C_p = 3\mathcal{N}k_B \left(\frac{q_0}{q_D}\right)^3 = 3\mathcal{N}k_B \alpha \left(\frac{T}{\Theta}\right)^{3/2},\tag{15}$$

where \mathcal{N} is the number of atoms per unit volume, q_D is the Debye wave number, and $\alpha = 4^3 (\pi/6)^{1/2} (vma/h)^{3/2}$. The Maxwell-Boltzmann electron distribution is $f^0 = \exp[-(E - \zeta)/kT]$, where E is the electron energy and ζ is the chemical potential. The momentum capacity of the electrons may be expressed in terms of the total electron energy \mathscr{E} as

$$\frac{\partial P_e}{\partial \lambda} = \frac{2m\mathscr{C}}{3\hbar kT}.$$
(16)

Since $\mathscr{C} = \frac{3}{2}n_c kT$ this becomes

$$\frac{\partial P_e}{\partial \lambda} = \frac{n_c m}{\hbar},\tag{17}$$

where n_c is the electron density.

It is now left to obtain an expression for τ_e . Assuming the electronic conductivity to be given by $\sigma = (n_c e^2 \tau_e)/(m)$, where *e* is the charge of the electron, τ_e may be determined from measured values of σ . However this expression takes account of intravalley and intervalley scattering of electrons. Phonons that promote intervalley scattering have a wave vector much greater than the greatest interacting phonon wave vector q_0 and were excluded from the phonon momentum because they are strongly

TABLE I. Calculated values of n_c , η , S, N_d and N_{dn} .

Т (К)	σ 10 ⁴ (Ω m) ⁻¹	n_c (10 ²⁵ m ⁻³)	η	<i>S</i> (μV/K)	$\frac{N_d}{(10^{26} \text{ m}^{-3})}$	N_{dn} (10 ²⁶ m ⁻³)
300	10	6.61	1.90	- 115	2.61	1.95
500	6.7	8.69	1.06	- 148	2.91	2.04
700	5.3	9.40	0.43	- 178	2.43	1.49
900	4.3	9.74	- 0.01	- 205	2.14	1.16
1000	4.0	10.2	- 0.15	- 213	2.14	1.12
1100	4.7	13.3	- 0.09	- 210	2.94	1.61
1200	5.0	15.5	- 0.06	- 208	3.55	2.00

scattered by point defects. Thus in this momentum balance it is only valid to include contributions to the relaxation time that are due to intravalley scattering. The relaxation time for intravalley scattering of electrons by phonons will be estimated as $\tau_{e,intra} = \beta \tau_e$, where β is the number of equivalent valleys in the conduction band, which is six in this case. Thus

$$\frac{1}{\tau_{e,\text{intra}}} = \frac{n_c e^2}{\beta \sigma m},$$
(18)

and thus

$$\frac{1}{\tau_p} = \frac{n_c^2 e^2 v^2}{\beta \mathcal{N} k T \sigma} \frac{1}{\alpha} \left(\frac{\Theta}{T}\right)^{3/2}.$$
(19)

It is necessary to determine n_c in order to evaluate τ_p . This is done by analyzing the temperature dependence of the electrical resistivity. If n_c were constant then ρ would be proportional to T and a slower increase is attributed to an increase in n_c . Thus, knowing n_c at one temperature one can determine it at other temperatures. In our analysis we have used the electrical resistivity curve and the value of n_c at 300 K as determined from Hall measurements of Vining *et al.*¹¹ in order to determine n_c at all temperatures.

VI. NEUTRAL DONOR SCATTERING

Neutral donor scattering was first seen by Goff and Pearlman¹² at low temperatures in Ge, and a theory was given by Keyes.⁴ While this process is particularly strong at low temperatures, it is by no means negligible at higher temperatures in the heavily doped materials of interest in thermoelectric conversion, which have a large concentration of neutral donors. The theory has been adapted to the Si symmetry¹⁰ and will also be reported at a later date.¹³ The form for the relaxation time due to neutral donor scattering was found to be

$$\frac{1}{\tau} = N_{\rm dn} \left(\frac{4}{3}\right)^2 \left(\frac{\Xi^2}{\Delta}\right)^2 \frac{a^6}{M^2 \pi} \frac{3}{v^7} \left(1 + \frac{a_0^2 q^2}{4}\right)^{-8} \omega^4, \qquad (20)$$

where N_{dn} is the neutral donor density, Ξ is the deformation potential, Δ is the splitting of the degenerate donor ground state by the impurity potential, and a_0 is the Bohr radius of the donor wave function.

It is necessary to determine the number of neutral donors at different temperatures which is done as follows. The number of electrons in the conduction band n_c is given by

$$n_c = 12 \left(\frac{2\pi m kT}{h^2}\right)^{3/2} \mathscr{F}_{1/2}(\eta) \tag{21}$$

and

$$\mathscr{F}_{j}(\eta) = \frac{1}{\Gamma(j+1)} \int_{0}^{\infty} \frac{e^{j}}{1+e^{\epsilon-\eta}} d\epsilon, \qquad (22)$$

and the number of unionized donors is given by

$$N_{\rm dn} = N_d \frac{1}{\frac{1}{2} \exp[(E_1 - \zeta)/kT] + 1},$$
 (23)

where N_d is the donor concentration and E_d is the donor energy. Combining these equations it is found

$$N_{d} = 6N_{c}\mathcal{F}_{1/2}(\eta) \left[1 + 2 \exp\left(\eta - \frac{(E_{d} - E_{c})}{kT}\right) \right],$$
(24)

where

$$N_c = 2 \left(\frac{2\pi m kT}{h^2}\right)^{3/2},$$
 (25)

Using an expression for the thermoelectric power

$$S = \frac{k}{e} \left(2 \frac{\mathcal{F}_1(\eta)}{\mathcal{F}_0(\eta)} - \eta \right), \tag{26}$$

the Fermi level may be determined from measured values of S. Using these values of η and N_d for a specific sample Eq. (24) can be solved for $E_d - E_c$ and $N_{dn} = N_d - n_c$. This was done at 300 K where a value of n_c was known from Hall measurements. At higher temperatures the values of n_c at those temperatures determined from the temperature dependence of the electrical resistivity as described in the previous section were used to determine η from Eq. (21) and these values of η were then used to calculate N_d from Eq. (24) and then $N_{dn} = N_d - n_c$.

VII. SAMPLE CHARACTERISTICS

Calculations are based on the results of a specific $Si_{0.8}Ge_{0.2}$ sample (no. 93) of Vining *et al.*¹¹ The calculated values of n_c , N_d , N_{dn} , S, and η are shown in Table I. The other material constants used were $\Delta = 4.3$ MeV and $\Xi = 7$ eV.^{14,15} These are the values for P in Si; It is not known how alloying affects these values. The values of a_0 , shear modulus, bulk modulus, lattice spacing, velocity, and Debye temperatures for Si and Ge are shown in Table

TABLE II. Material constants.

Constant	Si	Ge	Si _{0.8} Ge _{0.2}	
Lattice Spacing [*] (Å)	2.715	2.830	2.738	
Mass (10^{-26} kg)	4.668	12.06	6.15	
Debye temperature ^b (K)	645	374	591	
Sound velocity ^c (10 ³ m/s)	6.6	3.98	6.08	
Bulk modulus ^c $(10^{12} \text{ dyn cm}^{-3})$	0.979	0.732	0.930	
Shear modulus ^c	0.666	0.530	0.638	
Radius ^d a_0 (Å)	20.7	45.5	25.7	

*See Ref. 16.

^bSee Ref. 17. ^cSee Ref. 18.

^dSee Ref. 19.

II and were linearly interpolated for a 80%Si 20% Ge alloy. A value of 2 was also assumed for the Grueneisen constant in all cases.

VIII. BOUNDARY SCATTERING VERSUS PHONON-ELECTRON SCATTERING

For the sample for which the thermal conductivity was calculated the average grain diameter was $l = 1.8 \times 10^{-6}$ m and thus

$$\frac{1}{\tau_b} = 3.38 \times 10^9 \text{ s}^{-1}.$$
 (27)

This compares to the phonon-electron relaxation times shown in Table III. Thus the phonon-electron scattering is stronger than the boundary scattering at all the high temperatures.

IX. THERMAL CONDUCTIVITY CALCULATIONS

Using the above relaxation times, the thermal conductivity was calculated using the theory of Callaway:³

$$K_{I} = \frac{(6\pi^{2})^{1/3}}{2\pi^{2}} \frac{k^{3}}{\hbar^{2}} \frac{T^{3}}{\Theta a} \left(I_{1} + \frac{I_{2}^{2}}{I_{3}} \right), \qquad (28)$$

where

$$I_{1} = \int_{0}^{\Theta/T} \tau_{c} \frac{x^{4} e^{x}}{(e^{x} - 1)^{2}} dx,$$
$$I_{2} = \int_{0}^{\Theta/T} \frac{\tau_{c}}{\tau_{n}} \frac{x^{4} e^{x}}{(e^{x} - 1)^{2}} dx,$$

Temperature (K)	$\frac{1/\tau_{p}}{(10^{10} \text{ s}^{-1})}$		
300	10.5		
500	7.57		
700	4.83		
900	3.41		
1000	3.09		
1100	3.52		
1200	3.62		

TABLE IV. Calculated lattice thermal conductivity.

Temperature	(W m	Difference	
(K)	Neutral scattering	No neutral scattering	(%)
300	3.81	4.71	19
500	3.43	3.76	9
700	3.03	3.31	8
900	2.79	3.01	7
1000	2.67	2.89	8
1100	2.58	2.79	7
1200	2.52	2.74	8

$$I_3 = \int_0^{\Theta/T} \frac{\tau_c}{\tau_n \tau_p} \frac{x^4 e^x}{(e^x - 1)^2} dx,$$

where $x = \hbar \omega/kT$, τ_n is the relaxation time for normal processes, $\tau_c^{-1} = \tau_n^{-1} + \tau_p^{-1}$, $\tau_p^{-1} = \sum_j \tau_j^{-1}$, and the τ_j are the relaxation times for the various resistive scattering processes. τ_n has been estimated by Abeles²⁰ as τ_n^{-1} = $2.5\tau_i^{-1}$, where τ_i is the intrinsic relaxation time. The calculated lattice thermal conductivity is shown in Table IV with the neutral donor scattering included, and also with this mechanism excluded to show the effect of neutral donor scattering.

X. SPECTRAL CONDUCTIVITY

The spectral conductivity is the integrand of the thermal conductivity integral $K = \int K(\omega) d\omega$. Figure 1 gives an example of the spectral conductivity. At the highest frequencies the spectral conductivity is low because of strong mass defect scattering, at low frequencies the spectral conductivity is low due to strong phonon-electron scattering. At intermediate frequencies there is a peak in the spectral conductivity and this peak occurs around the frequency at which mass defect scattering has the same strength as in-

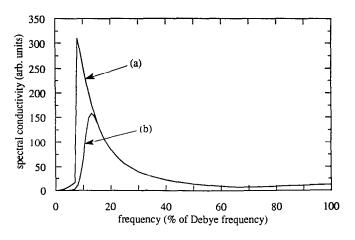


FIG. 1. Spectral conductivity at 300 K. Frequencies are given as percentages of the Debye frequency ($\omega_D = 7.73 \times 10^{13} \text{ s}^{-1}$). Values of ω_0 and ω_c as percentages of the Debye frequency in this case are 6.6% and 7.3%, respectively. (a) Spectral conductivity without neutral donor scattering included, and (b) spectral conductivity with neutral donor scattering included. These are plotted to show that part of the spectrum affected by neutral donor scattering.

TABLE V. Calculated values of ω_0 and ω_c .

Т (К)	(10^{12} s^{-1})	$(10^{12} s^{-1})$		
300	4.39	5.63		
500	6.08	7.26		
700	7.54	8.59		
900	8.98	9.75		
1000	9.53	10.3		
1100	9.92	10.8		
1200	10.2	11.3		

trinsic scattering ω_0 and the cutoff frequency for the phonon-electron interaction ω_c , where

$$\omega_0 = \left(\frac{2k_B v^2 \omega_D}{\pi \lambda a^3 \epsilon}\right)^{1/2} \tag{29}$$

and

$$\omega_c = \frac{4\pi v}{h} \sqrt{2mk_B T}.$$
(30)

Both ω_0 and ω_c increase as \sqrt{T} and this peak which carries most of the heat current moves up in frequency as temperature increases. Because the neutral donor scattering has a sharp cutoff it affects only phonons in this peak at the lower temperatures studied, which explains the larger effect of neutral donor scattering at 300 K. Table V gives ω_0 and ω_c , and Fig. 1 is an actual plot of the spectral conductivity.

XI. ELECTRONIC THERMAL CONDUCTIVITY

The Lorenz ratio is given by

$$L = \left(\frac{k_B}{e}\right)^2 \left(\frac{6\mathscr{F}_2(\eta)}{\mathscr{F}_0(\eta)} - 4\eta \frac{\mathscr{F}_1(\eta)}{\mathscr{F}_0(\eta)} + \eta^2\right) - S^2, \quad (31)$$

where S is the thermoelectric power. Using the values of η calculated previously, k_e , the electronic component at the thermal conductivity, was calculated and the total conductivity $k_e + k_l$ was then compared to the measured values for this sample¹¹ in Fig. 2.

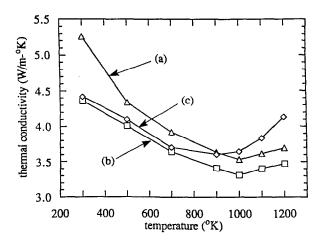


FIG. 2. Thermal conductivity vs temperature. (a) Calculated thermal conductivity without neutral donor scattering included; (b) calculated thermal conductivity with neutral donor scattering included, and (c) measured values of the thermal conductivity.

XII. INCLUSION SCATTERING

The scattering of phonons by small insulating inclusions has been proposed as a possible mechanism for reducing the lattice thermal conductivity in Si-Ge alloys. In this treatment the inclusions are assumed to be spheres. Inclusions with a diameter greater than the phonon wavelength scatter as shadow scattering and the phonon relaxation time is given by

$$\frac{1}{\tau_{ni}} = \frac{1}{NAv}$$
(32)

where v is the phonon velocity, N is the number of inclusions per unit volume, and A is the cross-sectional area of the inclusions. For inclusions of 40 Å diameter the lattice conductivity has been calculated for 3%, 6%, 9%, and 12% volume concentrations and the results are presented in Table VI. These results suggest that if neutral inclusions could be incorporated into these materials a substantial reduction in the lattice conductivity could be effected.

TABLE VI. Lattice thermal conductivity ($Wm^{-1}K^{-1}$) calculated with various values of the volume concentration c of inclusions with diameter of 40 Å. Columns labelled w/o are values calculated without considering neutral donor scattering; columns labeled w are values calculated with neutral donor scattering.

Temperature (K)	c = 0.00		c = 0.03		c = 0.06		c = 0.09		c = 0.12	
	w/o	w								
300	4.71	3.81	3.12	2.97	2.67	2.61	2.42	2.38	2.25	2.22
500	3.76	3.43	2.96	2.85	2.62	2.56	2.41	2.38	2.27	2.24
700	3.31	3.03	2.76	2.67	2.50	2.45	2.32	2.29	2.20	2.17
900	3.01	2.79	2.58	2.50	2.36	2.31	2.22	2.18	2.11	2.08
1000	2.89	2.67	2.51	2.43	2.31	2.26	2.17	2.14	2.07	2.05
1100	2.79	2.58	2.45	2.36	2.27	2.21	2.14	2.10	2.04	2.02
1200	2.74	2.52	2.42	2.32	2.24	2.19	2.12	2.08	2.02	2.00

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