

Low-temperature heat transport by charge carriers in doped semiconductors

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The electronic contribution to the thermal conductivity in degenerate semiconductors has been measured at low temperature in fine-grained ceramic samples in which the lattice thermal conductivity is reduced by grain-boundary scattering of the phonons. The validity of the Wiedemann-Franz-Lorenz law has been demonstrated; the Lorenz number was found to be close to the Sommerfeld value L_0 .

I. INTRODUCTION

In dielectric crystals, heat is carried exclusively by lattice vibrations which are scattered by each other or by a variety of lattice defects. Although this defect scattering can lead to a wide range of thermal conductivities, and widely differing temperature dependences, the heat transport in these solids is, in principle, far easier to understand than in solids which also contain electric charge carriers, which by themselves can carry heat, but can also scatter phonons.¹ Because of this added complexity, the analysis of the heat flow in electrical conductors is feasible only in certain simple cases, and usually requires a good deal of effort and adjustable parameters.

It is the purpose of this paper to report on a particularly simple case of heat transport in an electrical conductor in which all of the contributions to the heat flow are readily measurable. This investigation has also allowed us, for the first time, to test the Wiedemann-Franz-Lorenz law for a doped semiconductor in the low-temperature limit in which the electrons are expected to be scattered solely by elastic processes and hence the Lorenz number should approach its Sommerfeld value.

Determinations of the Lorenz number at low temperatures have been reported in a large number of metals and semimetals, too many in fact, to be mentioned here. The interested reader is referred to a recent review.² The range of residual electrical conductivities σ_{res} spanned in these materials extends over five orders of magnitude, with gallium,³ $\sigma_{\text{res}} = 7 \times 10^9 \Omega^{-1} \text{cm}^{-1}$, having the highest, and ytterbium,⁴ $\sigma_{\text{res}} = 6.5 \times 10^4 \Omega^{-1} \text{cm}^{-1}$, having the lowest conductivity. In semiconductors, however, investigations of the Wiedemann-Franz-Lorenz law have emphasized higher temperatures, where the lattice part of the thermal conductivity decreases with increasing temperature, while the electronic part increases. Separation of the two parts of the thermal conductivity has been achieved through high magnetic fields; this technique is restricted to high carrier mobilities.⁵ The observed deviations of the Lorenz numbers from their theoretically expected value have been explained through inelastic electron scattering.^{5,6}

In the present investigation, the problem of distinguish-

ing between the two parts of the thermal conductivity was achieved by intentionally decreasing the lattice part through grain-boundary scattering, as first done by Eucken and co-workers⁷ (because of low carrier mobility, a magnetic separation was not feasible). In small-grained ceramic samples, the lattice thermal conductivity at low temperatures ($< 1 \text{ K}$) was limited entirely by grain-boundary scattering, and hence decreased as T^3 . Thus, the electronic part, which decreased as T , became the dominant contribution to the total thermal conductivity.

For reasons that play no role in connection with the present investigation, the semiconducting material used was an alloy of 78 mol % Si and 22 mol % Ge. The only consequence this alloying had for our work was that we had to measure the specific heat of our samples, in order to determine their Debye speed of sound needed for the evaluation of our data.

II. EXPERIMENTAL MATTERS

The polycrystalline samples, $5 \times 5 \times 40 \text{ mm}^3$, were pressed (1.8 kbar, 1345 °C, 0.67 h) from appropriately ground powders obtained from ingots which were either undoped (i.e., not doped intentionally), or which were doped with $\sim 2 \text{ mol } \%$ boron (p) or phosphorous (n). One sample, labeled TECO, also contained about 5 mol % GaP which had, however, no influence on our results, as we will see. One undoped silicon single crystal (produced by Wacker Chemie) of the same size, with roughened faces, heat flow parallel to $\langle 111 \rangle$, was kindly provided by Dr. V. Narayanamurti from Bell Telephone Laboratories.

Mass density of the undoped ceramic samples was measured by Archimedes's principle, and compared with the single-crystal density determined by Dismukes *et al.*⁸ Grain sizes were determined on polished surfaces which had been etched with a copper-doped acid mixture,⁹ using a counting technique as described by Hilliard.¹⁰ Charge-carrier densities of the undoped samples were determined through measurement of the Hall effect and, for the doped samples in which the Hall voltages were inconveniently small, by comparison of the electrical conductivity at room temperature with those measured by Morin

and Maita¹¹ on doped Si of known carrier concentration. Electrical conductivity was measured with a four-probe technique on the same samples, and using the same clamp positions as used for the thermal conductivity measurements. Thermal conductivity was measured with the steady-state, two-heater, one-thermometer method in ⁴He, ³He, and dilution cryostats which have been described previously.¹²⁻¹⁴ Specific heat was measured with transient heat pulses in a cryostat small enough that it could be inserted into ⁴He storage Dewars with a neck size of 5 cm.¹⁵

III. EXPERIMENTAL RESULTS

In Table I the specific heat data obtained on an undoped Si₇₈Ge₂₂ sample are listed. Figure 1 shows a comparison of the temperature-dependent effective Debye temperature $\Theta_D(T)$ with those of pure Si and Ge published previously.¹⁶ From its low-temperature limit, the Debye limiting velocity and specific heat of Si₇₈Ge₂₂ can be computed; see Table II.

The influence of the grain size on the thermal conductivity of the undoped samples is shown in Fig. 2. The data on single-crystal pure Si are from Glassbrenner and Slack¹⁷ above 200 K, and from Holland and Neuringer¹⁸ from 2 to 200 K. Below 2 K, the data were obtained in the present work. Previously unpublished data obtained on a single-crystal Si containing 1 mol% Ge (Ref. 19) demonstrate the vanishingly small influence this alloying has on the scattering processes at low temperatures. This is expected since the point-defect phonon scattering diminishes as ω^4 (or T^4), where ω is the phonon angular frequency. Both the large and the small grained, undoped Si₇₈Ge₂₂ ceramic samples approach a T^3 thermal conductivity at low temperature, which shows that the point-defect scattering vanishes below 1 K even for these high Si-Ge alloying ratios. Note that the data of the large-

TABLE I. Specific heat of Si₇₈Ge₂₂ (large-grain, undoped, as described in Table III).

Temperature (K)	Specific heat (J g ⁻¹ K ⁻¹)
2.83	8.60×10^{-6}
3.44	1.42×10^{-5}
3.86	1.97×10^{-5}
4.14	2.28×10^{-5}
4.84	3.52×10^{-5}
5.90	6.51×10^{-5}
6.74	1.01×10^{-4}
7.96	1.74×10^{-4}
9.85	3.50×10^{-4}
13.2	9.38×10^{-4}
17.9	3.55×10^{-3}
21.2	6.84×10^{-3}
26.5	1.54×10^{-2}
33.3	3.10×10^{-2}
46.0	6.67×10^{-2}
62.3	1.15×10^{-1}
76.4	1.53×10^{-1}
94.5	1.99×10^{-1}
112.0	2.38×10^{-1}

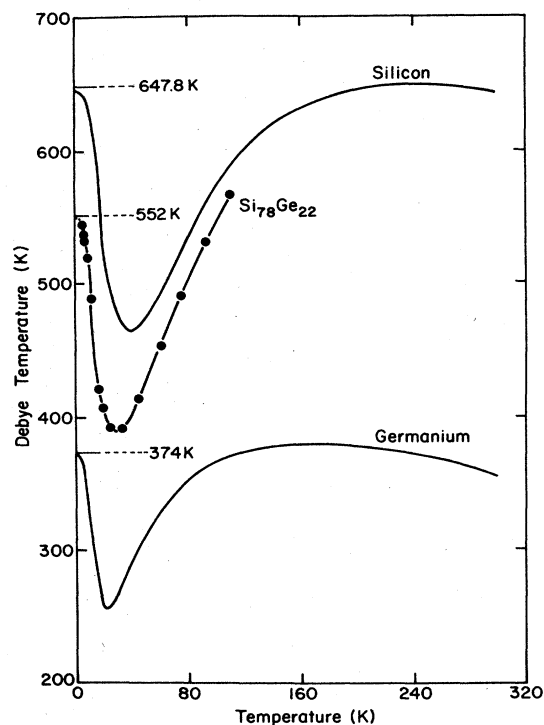


FIG. 1. Effective Debye temperatures $\Theta_D(T)$ for sample listed in Table I, compared with those of pure Si and Ge (Ref. 16).

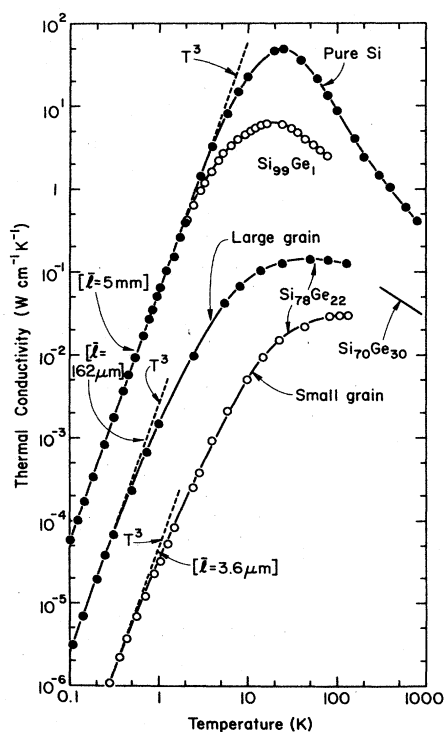


FIG. 2. Thermal conductivity of undoped Si and Si-Ge alloys. For pure Si, single-crystal data above 2 K after Refs. 17 and 18. Heat flow along $\langle 111 \rangle$. Si₉₉Ge₁, unspecified single crystal, after Ref. 19. Si₇₀Ge₃₀, single crystal, after Ref. 20.

TABLE II. Low-temperature asymptotic values of the specific heat C_v , Debye speed of sound v_D , and Debye temperature $\Theta_0 = \Theta_D(T=0)$, for Si and Ge (Ref. 16), and for $\text{Si}_{78}\text{Ge}_{22}$ (present work). The specific heat per unit mass, $C_{v,m}$, which is the quantity usually measured, is connected with C_v through ρ , the mass density: $C_{v,m} = C_v / \rho$. The mass densities ρ and number densities N , also listed in this table, are room-temperature values after Ref. 8, and refer to the fully dense material.

	N (cm^{-3})	ρ (g cm^{-3})	C_v / ρ ($\text{J g}^{-1} \text{K}^{-1}$)	C_v ($\text{J cm}^{-3} \text{K}^{-1}$)	v_D (cm s^{-1})	Θ_0 (K)
Si	5×10^{22}	2.33	$2.53 \times 10^{-7} T^3$	$5.90 \times 10^{-7} T^3$	5.92×10^5	648
Ge	4.42×10^{22}	5.32	$5.10 \times 10^{-7} T^3$	$2.71 \times 10^{-6} T^3$	3.56×10^5	374
$\text{Si}_{78}\text{Ge}_{22}$	4.89×10^{22}	3.07	$3.04 \times 10^{-7} T^3$	$9.33 \times 10^{-7} T^3$	5.17×10^5	552

grain sample approach the single-crystal $\text{Si}_{70}\text{Ge}_{30}$ data obtained by Abeles *et al.* at high temperatures.²⁰

For the four (B- or P-) doped samples, the thermal conductivities do not vary as T^3 at the lowest temperatures; see Fig. 3. In the doped, small-grain samples, the conductivities, instead, are larger than in the undoped sample, and approach a linear temperature dependence:

$$\Lambda = aT + bT^3. \quad (1)$$

The temperature dependence expressed in this equation is shown in Fig. 4, where Λ/T versus T^2 is plotted on linear scales. The straight lines have almost identical slopes, corresponding to equal values of b ; their intercepts determine the linear terms a in Eq. (1). Also shown are the data for the undoped small grain sample. A similar plot of the large-grain-doped sample shown in Fig. 5 also yields a straight (dashed) line, although only below $T \sim 0.15$ K. The values of a and b for all five samples are listed in Table III. (For the undoped samples, a , of course, is 0.)

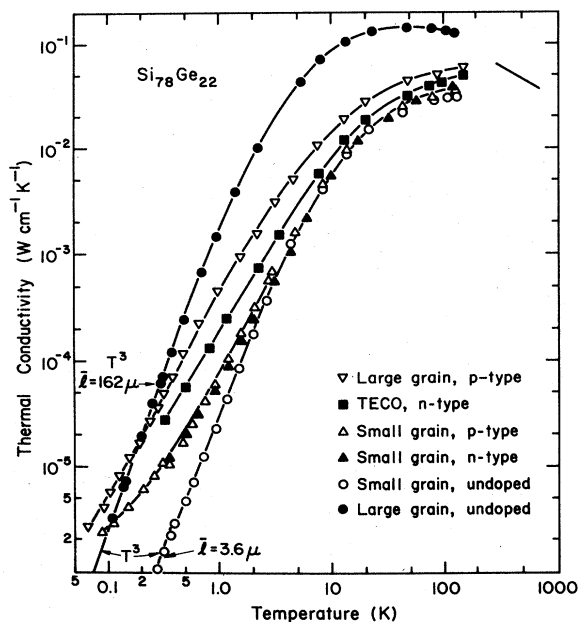


FIG. 3. Thermal conductivity of six $\text{Si}_{78}\text{Ge}_{22}$ ceramic samples, two undoped ones (same as the ones shown in Fig. 2) and four doped ones (see Table III for details).

The electrical conductivity of the four doped samples and of one undoped sample are compared in Fig. 6 with several silicon samples measured by Morin and Maita.¹¹ The electrical conductivities of the doped samples vary only very little below room temperature, which shows that they are degenerate. Their values at 4.2 K are listed in Table III. The carrier concentrations are also listed in Table III; their values are probably uncertain by as much as 15%, but this uncertainty is of little importance in the following discussion.

IV. DISCUSSION

In the low-temperature limit, when the lattice specific heat obeys the Debye T^3 law, the phonon thermal conductivity is given by

$$\Lambda = \frac{1}{3} C_v v_D \bar{l} = \frac{2\pi^2}{5} \frac{k_B^4}{\hbar^3 v_D^3} v_D T^3 \bar{l}, \quad (2)$$

where C_v is the specific heat (per unit volume at constant volume), v_D the Debye speed of sound, \bar{l} is the average phonon mean free path, and k_B and \hbar are Boltzmann's constant and Planck's constant (divided by 2π), respectively. In a pure single crystal with roughened surfaces, \bar{l} should be approximately equal to the sample diameter. In a polycrystalline ceramic \bar{l} should be of the order of d , the average grain size. Porosity is important in determining \bar{l} , though; in ceramic samples with mass densities close to

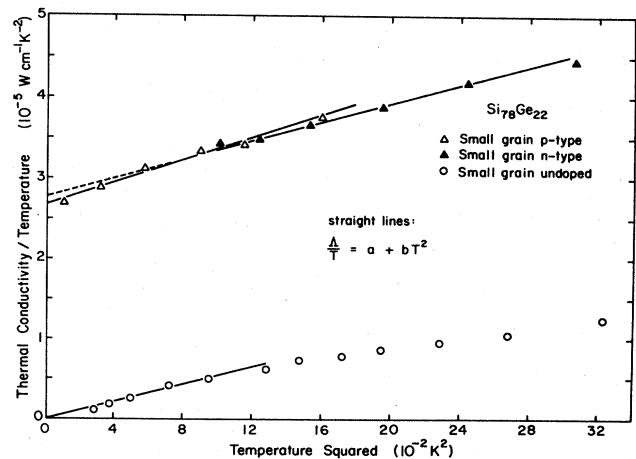


FIG. 4. $\Lambda(T)/T$ vs T^2 [see Eq. (1)] for the small-grain $\text{Si}_{78}\text{Ge}_{22}$ undoped and doped samples.

TABLE III. $\text{Si}_{78}\text{Ge}_{22}$ alloy ceramics. a and b were determined from Figs. 4 and 5, as best fits to a temperature-dependent thermal conductivity given by Eq. (1). The electrical conductivities $\sigma_{4.2\text{K}}$ listed are those measured at 4.2 K; see Fig. 6. The Lorenz numbers L were determined from $\sigma_{4.2\text{K}}$ and the linear terms of the thermal conductivity given in Eq. (1). Grain sizes d as determined microscopically; phonon mean free path \bar{l} from the T^3 term in Eq. (1), using the quantities required as listed in Table II; for the specific heat, a correction for the porosity was applied. n is the number density of free charge carriers.

Sample	ρ (g cm^{-3})	d (μm)	a ($\text{W cm}^{-1}\text{K}^{-2}$)	b ($\text{W cm}^{-1}\text{K}^{-4}$)	\bar{l} (μm)	$\sigma_{4.2\text{K}}$ ($\Omega^{-1}\text{cm}^{-1}$)	n (cm^{-3})	L ($10^{-8}\text{W}\Omega\text{K}^{-2}$)
Large-grain, undoped	3.01	29	0	2.48×10^{-3}	162	4.6×10^{-6}	1.2×10^{17a}	
Large-grain, p -type TECO,		12.8	3.15×10^{-5}	1.66×10^{-3}	108	1.42×10^3	9.1×10^{19}	2.22
n -type		11.2				2.04×10^3	1.6×10^{20}	
Small-grain, undoped	2.966	3.9	0	5.44×10^{-5}	3.6	$< 10^{-7}$	2.8×10^{15}	
Small-grain, p -type		2.6	2.67×10^{-5}	6.96×10^{-5}	4.5	8.77×10^2	5.6×10^{19}	3.04
Small-grain, n -type		1.5	2.76×10^{-5}	5.74×10^{-5}	3.8	1.01×10^3	7.1×10^{19}	2.74

^aObtained by comparison with the conductivities given in Ref. 11 (from Hall coefficient measurements, $n = 1.4 \times 10^{17}\text{ cm}^{-3}$ was obtained).

the crystalline densities ($\rho > 0.99\rho_{\text{cryst}}$), $\bar{l} \sim 10d$ has been observed, while in porous samples ($\rho \sim 0.95\rho_{\text{cryst}}$), $\bar{l} \sim d$ is common.²¹⁻²³ Details of the thermal conductivity in granular materials are still poorly understood.²⁴

In the single-crystal silicon, \bar{l} was indeed observed to be approximately equal to the sample diameter, 5 mm. In the two undoped ceramic $\text{Si}_{78}\text{Ge}_{22}$ samples, the thermal conductivity also follows a T^3 temperature dependence; see Figs. 2 and 3; the phonon mean free paths \bar{l} , calculated with the help of Eq. (2), and listed in Table III, agree well with the average grain size for the small grain sample; its mass density was determined to $\rho = 0.966\rho_{\text{cryst}}$. For the large grain sample, $\bar{l} \sim 5d$; in this case, $\rho = 0.98\rho_{\text{cryst}}$. Conceivably, the lower porosity resulted in a relatively larger \bar{l} .

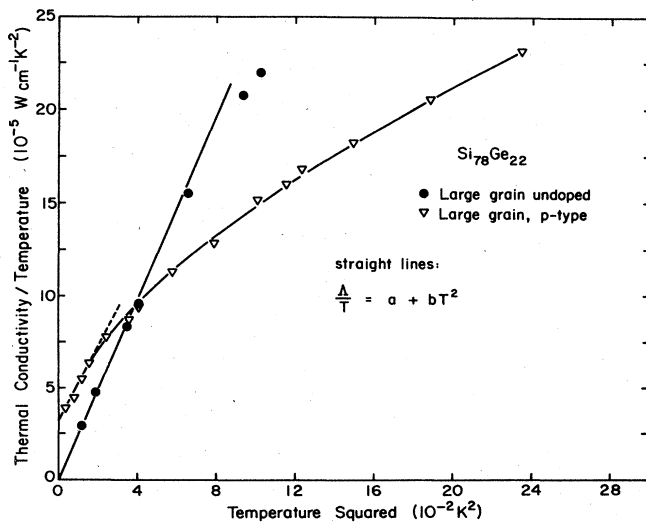


FIG. 5. $\lambda(T)/T$ vs T^2 [see Eq. (1)] for the large-grain, undoped and for the large-grain, p -type $\text{Si}_{78}\text{Ge}_{22}$ ceramic samples.

In the doped ceramic samples, we also ascribe the T^3 terms of the thermal conductivities to phonon heat transport, with the phonon mean free path \bar{l} , limited by the grain boundaries; see Table III. In the large-grain, p -type sample, the T^3 term extends only to $T \sim 0.15\text{ K}$; above

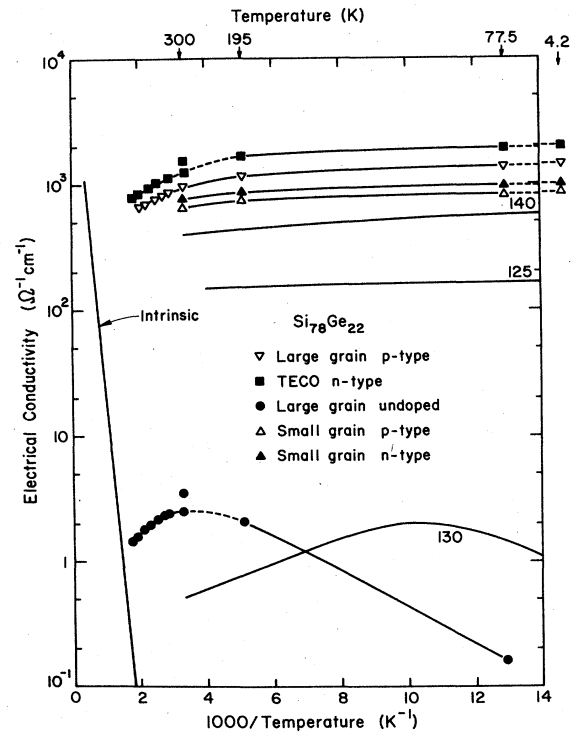


FIG. 6. Variation of electrical conductivity with temperature for different $\text{Si}_{78}\text{Ge}_{22}$ ceramic samples, and for doped single-crystal silicon samples by Morin and Maita, Ref. 11, listed as follows with the carrier concentration for each in cm^{-3} : sample no. 140, 2.7×10^{19} (arsenic doped); sample no. 125, 1.5×10^{19} (boron doped); sample no. 130, 2.1×10^{15} (arsenic doped).

this temperature, the conductivity rises less rapidly. This deviation is probably the result of phonon scattering by point defects within the grains. The mean free path connected with this scattering can be estimated to $\sim 100 \mu\text{m}$ at 0.5 K. This value is close to the mean free path determined from thermal conductivity measurements on *single crystals* containing boron or phosphorous in concentrations similar to those used in our samples: Measurements by Slack²⁵ above 2 K can be extrapolated to 0.5 K to yield a mean free path of $\sim 50 \mu\text{m}$. Note also that impurity scattering of the same magnitude should be totally invisible in the small-grain-doped samples, in agreement with our findings (see Fig. 4): In these samples, phonon mean free paths resulting from grain-boundary scattering were determined to be 4.5 and 3.8 μm , much smaller than the 100 μm mean free path resulting from the point-defect scattering.

Next, we will show that the low-temperature contribution to the thermal conductivity, which is linear in T , has the magnitude expected for heat transport by charge carriers. According to the Wiedemann-Franz-Lorenz law (see Appendix), the electronic part Λ_e of the thermal conductivity is proportional to the electrical conductivity and to the temperature T :

$$\Lambda_e = L\sigma T. \quad (3)$$

For a degenerate gas of electrons (or holes) undergoing elastic collisions only, as should be expected for scattering by point defects, the Lorenz number L should be equal to the Sommerfeld value,

$$L_0 = \pi^2 k_B^2 / 3e^2 = 2.44 \times 10^{-8} \text{ W } \Omega \text{ K}^{-2}.$$

Where e is the electronic charge. From the comparison of the linear term of the measured thermal conductivity, and the temperature-independent electrical conductivity ($=\sigma_{4.2\text{K}}$) we can determine L :

$$\Lambda_e = L\sigma_{4.2\text{K}}T = aT. \quad (4)$$

$L = a/\sigma_{4.2\text{K}}$ is listed in Table III. For all three doped samples, L is close enough to L_0 to serve as proof that the linear term of the thermal conductivity in these doped semiconductors is indeed caused by the mobile charge carriers. We believe that the discrepancy between the experimental L and the theoretical L_0 is most likely caused by experimental uncertainties, mainly the uncertainty in determining the low-temperature thermal conductivity ($=aT$).

While the grain boundaries have a pronounced influence on the heat transport by the phonons, their impact on the electrical conductivity, and thus on the heat transport by charge carriers is likely to be negligible. In order to prove this point, we calculated l_e , the electron (hole) mean free path by the well-known expression of σ for a degenerate electron gas²⁶

$$\sigma = \frac{ne^2 l_e}{\hbar(3\pi^2 n)^{1/3}}, \quad (5)$$

where n , the number density of the charge carriers, and $\sigma = \sigma_{4.2\text{K}}$ are given in Table III. From these values, we obtained $l_e = 130 \text{ \AA}$ in the small-grain n - and p -type samples, and $l_e = 95 \text{ \AA}$ in the large-grain, p -type sample.

These values are far smaller than the grain sizes and suggest a scattering within the grains, probably by point defects. From the temperature independence of the electrical conductivity shown in Fig. 6, we conclude that electron-phonon scattering is unimportant.

We may thus conclude that these doped ceramic samples present a particularly simple case of both phonons and charge carriers contributing independently to the heat transport, according to the simple formula

$$\Lambda_{\text{total}} = \Lambda_e + \Lambda_{\text{lattice}}, \quad (6)$$

where the electronic part of the thermal conductivity Λ_e is limited by scattering of the charge carriers by lattice defects within the grains, and the lattice part of the thermal conductivity, Λ_{lattice} , is limited by phonon scattering at the grain boundaries.

The successful low-temperature test of the Wiedemann-Franz-Lorenz law presented here for doped semiconductors has been performed on a substance of very low electrical conductivity ($\sim 2 \times 10^3 \Omega^{-1} \text{ cm}^{-1}$): The metal with the largest residual electrical conductivity on which this law has been tested previously is gallium, with $\sigma_{\text{res}} = 7 \times 10^9 \Omega^{-1} \text{ cm}^{-1}$; the only semiconductor on which measurements have been extended to 4 K, PbTe, had an electrical conductivity of $4 \times 10^5 \Omega^{-1} \text{ cm}^{-1}$ at that temperature.⁵ While there is no reason why for a quasi-free-electron gas the Wiedemann-Franz-Lorenz law with $L = L_0$ should be limited to a certain range of electrical conductivity, the observation that it indeed holds over more than six orders of magnitude is, nevertheless, gratifying.

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APPENDIX: A HISTORICAL REMARK

Equation (3) is usually referred to as the Wiedemann-Franz law. In their pioneering work, Wiedemann and Franz²⁷ noted that the thermal conductivities of several metals they had measured at room temperature (in relative units) appeared to be proportional to the electrical conductivities of these metals (measured by earlier investigators). They concluded that "the conductivities of metals for electricity and heat are very closely related to each other, and are probably both functions of the same quantity."

The first study of the temperature dependence in Eq. (3) was done by Lorenz.²⁸ He compared earlier work by Matthiessen and von Bose on the electrical conductivity of metals, and by Ångström and other, unreferenced investigators on the thermal conductivity of metals. He concluded that "the ratio of the conductivities for heat and electricity of pure metals is proportional to the absolute

temperature." Lorenz provided the experimental proof in 1881;²⁹ refined measurements did not follow until 1899.³⁰

It thus appears that the recognition for the discovery of

the relationship expressed in Eq. (3) ought to be shared by Wiedemann, Franz, and Lorenz, and hence we prefer to call it the Wiedemann-Franz-Lorenz law.

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