Quenched Phonon Drag in Silicon Microcontacts

R. Trzcinski, E. Gmelin, and H. J. Queisser
Max-Planck-Institut für Festkörperforschung, D-7000 Stuttgart 80, Federal Republic of Germany
(Received 19 December 1985)

The thermoelectric power of silicon decreases precipitously when the dimensions of a microcontact are reduced below a critical value. The critical dimension L decreases with rising temperature T and is a measure for an interaction length of electron-phonon coupling. The dimensional reduction inhibits the phonon-drag contribution to the thermoelectric power; the predicted scaling with $L^{-1}T^{-4}$ is verified for the first time.

PACS numbers: 72.20.Pa, 63.20.Kr, 73.40.Lq

Electronic transport in solids is strongly afflicted by reductions of the sample geometry toward dimensions comparable to the mean free paths of the carriers and the phonons.[1,2] In semiconductors, ballistic transport of electrons within structures of small sizes has become a topic of great interest not only for the basic understanding of conduction mechanisms but also for applications in novel high-speed devices.[2-4] Ballistic transport, also named "Knudsen flow," of electrons has recently been demonstrated in semiconductor microcontacts by the measurement of an asymmetric generation of heat by electronic currents flowing through a symmetrical arrangement.[5,6]

In this paper, we report studies of the thermoelectric power of thermocouples of silicon/copper, in which a microcontact of variable contact area is inserted into the silicon leg of the couple by means of highly controlled techniques in ultrahigh vacuum.[6] From the value of the contact resistance we deduce its geometrical dimension. We find dramatic decreases of the thermoelectric power when the dimensions are reduced below a critical value, which depends upon temperature. This decrease is seen clearly at temperatures as high as room temperature. We interpret this drop of thermoelectric power by a quenching of the phonon-drag contribution upon size reduction.

The samples used in our experiments reported here consist of n-type silicon of wedge shape, similar to the samples described earlier.[5] We chose phosphorus doping yielding of the order of $n = 8\times10^{17}$ electrons/cm^3 at 300 K. The two wedges face each other crosswise. The microcontact is established by pressing them against each other by means of a sensitively controllable piezoelectric drive. The samples are mounted on copper base plates, which are placed onto cryostats operated by continuous flow of liquid nitrogen, which enables us to maintain controlled temperature differences within the range from 120 to about 500 K. The entire arrangement is mounted in ultrahigh vacuum to control surface conditions and avoid contamination. Prior to the measurements, the wedges are cleaned by ion bombardment and analyzed by Auger spectroscopy. We always investigate first the current-voltage relation to check for Ohmic conduction and to obtain reliable values for the resistance R of our double-wedge arrangement.

Our results are exemplified in Fig. 1. The thermoelectric power Q is measured with a voltmeter having an impedance exceeding 100 MΩ. We plot Q as a function of the resistance R, which can be split into a Maxwellian part R_M and a Knudsen part R_K: $R = R_M + R_K = (2\sigma L)^{-1} + K/2\sigma L$, where σ is the electrical conductance and L the contact radius, as discussed in detail earlier.[5] Measurements of R thus enable us to derive the contact dimension L, which is plotted as the upper abscissa in Fig. 1. We select for Fig. 1 merely a family of four typical curves from our data, with temperatures T_1 and T_2 indicated in the inset for the schematical setup.

The thermoelectric power Q is seen to be initially fairly constant for small R, i.e., for contacts large enough to resemble bulk conditions. Within a rather narrow range of R, a sharp drop occurs, after which

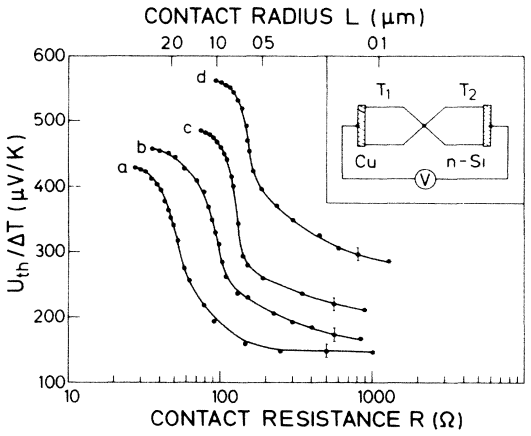

FIG. 1. Measured thermoelectric voltage U_{th}, divided by temperature difference $T_2 - T_1 = \Delta T$, plotted vs contact resistance R of silicon microcontacts, as shown schematically in the inset. The contact radius L is derived from R. Curves are for fixed $T_1 = 300$ K; a, $T_2 = 342$ K; b, $T_2 = 388$ K; c, $T_2 = 406$ K; and d, $T_2 = 450$ K.

the thermoelectric power varies only slightly. Figure 1 indicates clearly that the drop of Q arises for smaller values of the contact area (i.e., for larger R) when the temperature is raised. We have observed this qualitative behavior for a sizable number of samples and for wide temperature ranges (145 to 480 K).

We first consider the magnitudes of Q. The thermoelectric power is conventionally split into two parts,

$$Q = Q_e + Q_p, \tag{1}$$

where Q_e is the electronic part, while Q_p is the additional contribution which arises from a drag on the electronic system by the disequilibrated phonon system.[7-10] (We use the notation of Herring[8] in this Letter.) The electronic contribution is dominated by the first term in the equation[8]

$$\mp eQ_e = (\epsilon_F - \epsilon_b)/T - |\Delta\epsilon_T|/T, \tag{2}$$

where e is the electronic charge. The difference of the Fermi energy ϵ_F and the band-edge energy ϵ_b usually exceeds[8] the term $\Delta\epsilon_T$, being the average carrier energy. From our known doping level we know that ϵ_F lies about 100 meV below ϵ_b,[11] and hence we can calculate the electronic part to be $Q_e \cong 300$ μV/K typically for our conditions of doping and temperature, which is of the order of magnitude of our data, but typically larger by about 20% than our measured values of Q at the largest resistances R. The phononic part Q_p is a rather complicated function of both the electronic system and the lattice dynamics. For an infinite specimen, Herring[8] gave an asymptotic expression

$$\mp Q_p = m^* c^2 f e^{-1} T^{-1} (\bar{\tau}/\bar{\tau}_e), \tag{3}$$

where m^* is the effective mass, c the sound velocity, and f the fraction of interacting carriers, and $\bar{\tau}$ and $\bar{\tau}_e$ are average relaxation times for the phonons and electrons.[8] As a rule, Q_p must thus be derived from experiment by subtracting from the total, measured Q a calculated electronic part Q_e.[9,10] Previous measurements have indeed demonstrated the existence of the phonon-drag contribution, but all experiments lacked a systematic size variation and were restricted to large samples of the order of square millimeters (thus a factor of 10^8 larger than reported here), and therefore could prove phonon-drag effects only at temperatures well below room temperature, where the phonon mean free path approaches sample dimensions.[8-10] For our conditions and with the rough estimate of $f = 1$, we get

$$Q_p = (10\ \mu\text{V/K})(l_p/l_e), \tag{4}$$

where we express the mean free paths of phonons and electrons by $l_p = c\bar{\tau}$ and $l_e = \tau_e v_{th}$, where v_{th} is the thermal electron velocity. From the data of Geballe and Hull[10] for Si, we take $Q_p' \cong 350$ μV/K for an infinite specimen of similar doping and comparable electron mobility μ. The mean free path $l_e = v_{th} m^* \mu/e$ can be estimated with $v_{th} = 10^7$ cm/s, m^* approximately the free-electron mass, and $\mu = 300$ cm^2/V·s. We can therefore obtain from Eq. (4) with Q_p' and l_e an approximation for the mean free path of the phonons which are relevant for the drag on the electrons: $l_p = 5 \times 10^{-5}$ cm.

The data of Fig. 1 indicate that indeed the thermoelectric power is quenched as soon as dimensions are reached which are of the order of this mean free path l_p of the relevant phonons, as estimated above. Conversely, experiments such as the ones described here, with controlled, variable small dimensions, can be utilized to measure mean free paths and relaxation times. Intricate details, such as the relative contributions of phonon-phonon scattering or phonon-boundary scattering to the mean free path l_p, or the relative weights of the various phonon branches, seem now much more directly accessible to experimental scrutiny than with the previous techniques limited to macroscopic samples.

Measurements of the thermoelectric Seebeck coefficient for semiconducting samples with extreme geometrical constrictions are thus very useful to extract information concerning microscopic transport parameters. Such an experimental arrangement, as schematized in the inset of Fig. 1, must, however, be carefully analyzed. Evaluations of the thermal resistance of such a structure show[12] that almost the total temperature difference $\Delta T = T_1 - T_2$ is subtended by regions very close to the constriction; there is very little temperature gradient ∇T across the wedge bulk. Thus, the measured thermoelectric voltage U_{th} originates almost exclusively from the constricting orifice and its immediate surroundings, when we consider the usual summation to obtain the thermovoltage: $U_{th} = \int Q \nabla T\, dx$, to be integrated along the coordinate x throughout the thermocouple. The contributions to U_{th} from leads and interfaces at the metallic plates amount at most to 1%.[8-10] A thermovoltage measurement therefore concerns directly the portions of the samples with the reduced geometry.

Next, we concern ourselves with the observed dependence of the critical dimension upon temperature. Qualitative interpretation is straightforward. As the lattice temperature rises, more phonons are present, especially at the high-energy, short-wavelength end of the distribution of the relevant phonons. These phonons are less affected by size reduction than their companions of longer wavelengths. Therefore, the quenching of the phonon drag arises for smaller dimensions. Quantitative arguments for this situation have been given in considerable detail by Herring.[8]

The effects of finite size were treated with various simplifying assumptions.[13] When longitudinal pho-

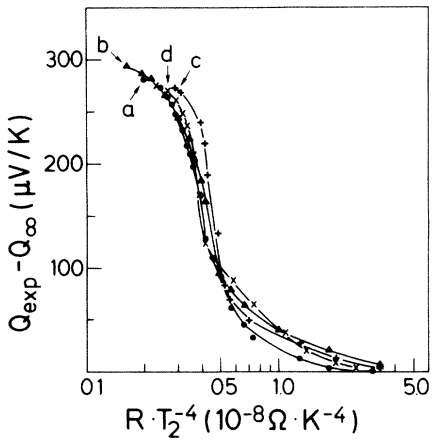

FIG. 2. Experimental thermoelectric power $Q_{exp} = U_{th}/T$, reduced by Q_∞ (which is asymptotic thermopower at maximal measured R), plotted vs RT_2^{-4} to test the scaling law predicted by Herring (Ref. 13). Curves a–d measured at various temperatures T_2, as indicated for Fig. 1.

nons are considered to cause the drag effect, low temperatures are assumed to allow resort to the Debye approximation, and effects of impurities to scatter the electrons are neglected, a simple scaling law can be derived in order to link dimension L and temperature T. Herring introduced a dimensionless variable ξ,[13] which for the longitudinal phonon modes is

$$\xi_l \propto T^{-4} L^{-1}. \quad (5)$$

We have examined whether this variable will indeed provide a universal scaling description by providing one common plot for varieties of L, T. Figure 2 gives the result for a number of experimental runs. The universality is indeed remarkable; the curves coincide. The assumption of being in the Debye regime to describe the phonon distribution is not unjustified, although we work up to room temperature, because the Debye temperature of Si is reported[14] to be $\theta_D(\mathrm{Si}) = 636$ K.

More detail can be extracted from universal curves, such as Fig. 2, when the degree of abruptness of the quenching is analyzed.[13] Our curves always show a more abrupt decrease of the thermoelectric power than originally anticipated theoretically. There are several experimental facts which may cause this more abrupt quenching effect. First, the dimensional decrease will enhance and concentrate the temperature gradient ∇T, which favors the contributions to the thermoelectric voltage from the more constricted regions and amplifies the abruptness. Second, we have not considered the increasing one-dimensionality of transport through the orifice as dimensions are decreased, and thus we ought to introduce a selection in k space. We can, at present, not yet decide which of these two—or other—phenomena is most strongly responsible for the abruptness experimentally observed; nevertheless, we verify Herring's prophecy[13] concerning the important inferences to be drawn from such analysis.[15] Similar arguments apply to our observation of generally lower electronic contributions Q_e as compared to large bulk samples. Work is in progress to elucidate these points.

In conclusion, we have shown that thermoelectric studies at well-defined semiconductor microcontacts provide much insight into transport mechanisms of electrons and phonons and the electron-phonon interaction. A much more direct experimental access to characteristic lengths describing microscopically these phenomena in semiconductors seems now feasible. As one example, we have here verified a long-standing early prediction of a universal scaling, which combines the effects of temperature and sample size. The severe reduction of thermoelectric power at microcontacts dashes some speculative hopes to achieve superior Seebeck or Peltier devices[16] by size reductions, yet our improved understanding of thermoelectricity in small dimensions might eventually also prove to become a useful guideline for novel applications.

This work was sponsored by a grant from the Stiftung Volkswagenwerk, which supported one of us (R.T.) and is gratefully acknowledged. We thank Elisabeth Bauser for sample materials and preparation. Ursula Nöcker deserves our thanks for her careful and patient participation in the often delicate and difficult measurements. Lutz Weber was a most stimulating discussion partner, as was Ursula Gerlach-Meyer, who constructed most of our equipment in its earlier configuration.

[1]See, for example, *Localization, Interaction, and Transport Phenomena,* edited by B. Kramer, G. Bergmann, and Y. Bruynseraede (Springer, New York, 1985).

[2]J. R. Hayes, A. F. Levi, and W. Wiegmann, Phys. Rev. Lett. **54**, 1570 (1985); M. Heiblum, M. I. Nathan, D. C. Thomas, and C. M. Knoedler, Phys. Rev. Lett. **55**, 2200 (1985).

[3]L. F. Eastman, in *Festkörperprobleme: Advances in Solid State Physics,* edited by P. Grosse (Vieweg, Braunschweig, 1982), Vol. 22, p. 173.

[4]D. K. Ferry, in *Handbook of Semiconductors,* edited by W. Paul and T. S. Moss (North-Holland, New York, 1982), Chap. 11A.

[5]U. Gerlach-Meyer and H. J. Queisser, Phys. Rev. Lett. **51**, 1904 (1983).

[6]U. Gerlach-Meyer, Appl. Phys. **A33**, 161 (1984); U. Gerlach-Meyer, E. Gmelin, and H. J. Queisser, in *Proceedings of the Seventeenth International Conference on the Physics of Semiconductors, San Francisco, California, 1984,* edited by J. D. Chadi and W. A. Harrison (Springer, New York, 1985), p. 1309.

[7]J. Tauc, *Photo and Thermoelectric Effects in Semiconductors* (Pergamon, Oxford, London, 1962).

[8]C. Herring, Phys. Rev. **96**, 1163 (1954).

[9]H. P. R. Frederikse, Phys. Rev. **92**, 248 (1953).

[10]T. H. Geballe and G. W. Hull, Phys. Rev. **98**, 940 (1955).

[11]See, e.g., S. Sze, *Physics of Semiconductor Devices* (Wiley, New York, 1981), 2nd ed., p. 26.

[12]E. Gmelin, unpublished.

[13]Ref. 8, Chap. 6 and Fig. 6.

[14]P. H. Keesom and G. Seidel, Phys. Rev. **113**, 33 (1958).

[15]For a recent consideration of drag effects in thermoelectricity in miniaturized metallic systems, see E. N. Bogachek, I. Q. Kulik, A. N. Omel'yanchuk, and A. G. Shkorbatov, Pis'ma Zh. Eksp. Teor. Fiz. **41**, 519 (1985) [JETP Lett. **41**, 633 (1985)].

[16]R. Dahlberg, German Patent No. DT2547262, and private communication.