Thermoelectric Voltage between Identical Metals in a Point-Contact Configuration

O. I. Shklyarevskii,[a] A. G. M. Jansen,[b] J. G. H. Hermsen, and P. Wyder[b]
Research Institute for Materials, University of Nijmegen, 6525 ED Nijmegen, The Netherlands

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In an electric circuit made from only one metal with a thermal gradient over a point contact in the chain, thermoelectric voltages emerge as a result of the finite size of the contact. The phenomenon is described in terms of the difference in the transport properties of a point contact and the bulk, yielding the phonon drag as a major contribution to the measured thermopower at temperatures $T < \theta_D/2$.

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The resistance of metallic point contacts has been studied extensively as a function of the applied voltage. The nonlinearities in the current-voltage characteristics can be used for an experimental study of the energy dependence of the scattering of the conduction electrons with elementary excitations (i.e., phonons).[1,2] In this paper we report on measurements where we applied a temperature gradient over the contact. Now thermoelectric effects come into play as well. Usually, no thermal voltage is observed in a homogeneous chain with temperature gradients somewhere within the chain; however, by inserting a metallic constriction in the chain and applying a temperature gradient over the contact, we observed a thermal emf. The evaluated thermopower of this point-contact configuration measures the phonon-drag contribution of the thermopower of the bulk metal. A first step has been made for a theoretical understanding of this phenomenon.[3] Phonon-drag quenching has recently been studied in the thermopower of semiconducting Si microcontacts.[4]

The experimental setup is shown in Fig. 1. The thermal emf is measured over the homogeneous chain ABC which includes a metallic constriction without an externally applied voltage. One part of the contact (A) is electrolytically etched to a sharp needle and kept at liquid He temperature. The other part (B) is thermally attached to the heater. The wires BC (diameter 50–100 μm) are made from the same starting material as the point contact. The materials investigated were the noble metals Ag, Cu, and Au, and some transition metals with at least a purity of 99.99+%. Here, explicit results will be presented for Ag only. With the heater at one side of the contact, the "anvil" could be heated up to room temperature. The temperature was measured with a AuFe-Chromel differential thermocouple with an error less than 0.2 K. The whole system is in a vacuum can, immersed in liquid helium.

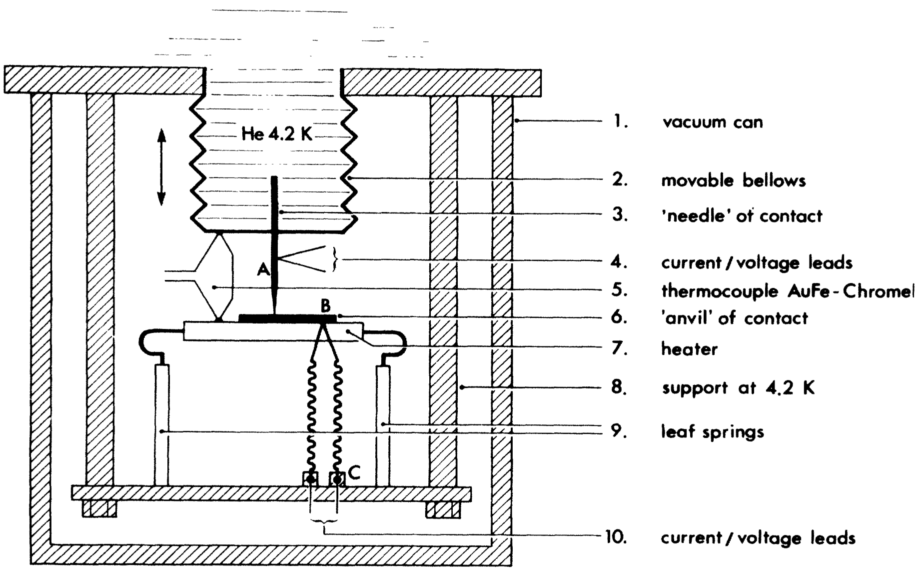

FIG. 1. Schematic view of the experimental arrangement for the measurement of the Seebeck voltage between the isothermals A and C in a circuit with a point contact. The chain A-B-C with the constriction is made from the same metal.

With a differential screw mechanism the spear-anvil contacts were adjusted. The investigated electrical contact resistances were measured in a four-probe arrangement and ranged from 0.001 to 1 Ω. For Ag this gives contact dimensions from 300 to 10 nm; this value was calculated from the expression $R = ql/a^2$ for the resistance of a clean contact[5] (mean free path l long compared to the contact radius a), where q is the specific resistivity of the metal.

For the homogeneous thermocouple with a point contact the thermoelectric emf was measured between the isothermals A and C (see Fig. 1) as a function of the temperature of the anvil part. Spurious emf effects did not exceed $\sim 1\ \mu$V. In our experimental arrangement the measured Seebeck voltage V_{exp} is equal to the difference between a voltage over the bulk wire (V_b) and over the point contact (V_{pc}) such that

$$\begin{aligned} V_{\mathrm{exp}}(T) &= V_b - V_{pc} \\ &= \int_{4.2}^{T} [S_b(t) - S_{pc}(t)]\,dt, \end{aligned} \tag{1}$$

where S is the thermopower. Numerical differentiation with respect to the temperature T gives the difference in the thermopower $S(T)$ between the two parts of the thermocouple chain.

In Fig. 2 we show the measured thermopower for a Ag chain with a point contact. The data are given for several contact resistances; the two curves with the same contact resistance give an indication of the reproducibility. In the same figure we have also plotted the thermopower for pure Ag as given by Pearson.[6] We see that especially for low temperatures the thermopower for the homogeneous chain resembles the bulk behavior and is independent of the point-contact. For temperatures higher than indicated in the figure ($T > 200$ K) the data are not reliable as the contact resistance is no longer stable, but there is a strong indication that the measured thermopower stays roughly constant up to room temperature. In view of Eq. (1), we have to compare the thermopower of the bulk with that of a metallic constriction. The thermopower of a metal has two important contributions,[7] such that $S = S^e + S^{\mathrm{ph}}$. The term S^e has its origin in the diffusion of the electrons in the presence of a temperature gradient. The other term, S^{ph}, is due to the nonequilibrium distribution of the phonons, which drags the electrons.

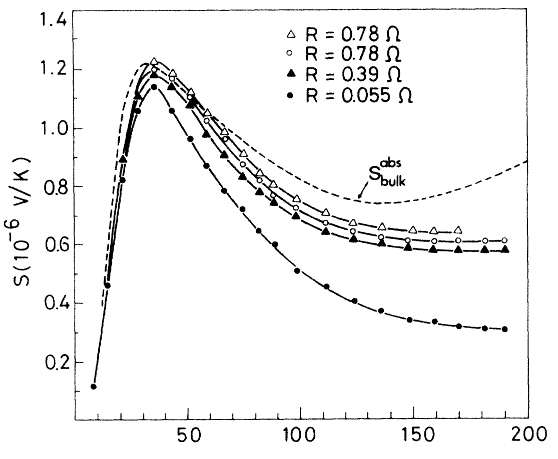

FIG. 2. Measured thermopower $S = S_b - S_{pc}$ as a function of the temperature, obtained in the point-contact configuration for Ag with the indicated contact resistances. For comparison, the dashed curve shows the absolute thermopower S_b^{abs} for Ag (Ref. 6).

For a contact with a long mean free path of the electrons compared to the contact dimension, the current density through the contact can be calculated from the Sharvin distribution[1,2,5] of the electrons, at the orifice consisting of two half Fermi spheres with different electrochemical potentials. In the presence of a voltage V and a temperature gradient ΔT across such a clean contact the current density J can be written in this model as

$$\begin{aligned} J = -2e \textstyle\sum_k |v_{kz}| [\, & f_0^T(\epsilon_k - \mu^T - eV) \\ & - f_0^{T+\Delta T}(\epsilon_k - \mu^{T+\Delta T})], \end{aligned} \tag{2}$$

where the summation over the k vectors is only taken over half the Fermi volume. v_{kz} is the velocity component perpendicular to the contact orifice. f_0^T is the Fermi distribution function at a temperature T; the argument of this function involves the electron energy ϵ_k and the temperature-dependent chemical potential μ^T. For $\Delta T = 0$ this expression yields the Sharvin resistance[5] for a clean contact. Using Eq. (2) at zero current density gives the diffusion term in the thermopower of a point contact:

$$S_{pc}^e \equiv \lim_{\Delta T \to 0} \left. \frac{\Delta\mu - eV}{e\Delta T} \right|_{J=0} = \frac{1}{-eT} \left(\frac{F_2}{F_1} - \mu^T \right), \tag{3}$$

for a free-electron model with a spherical Fermi surface with $F_n = \int \epsilon^n (\partial f_0 / \partial \epsilon)\, d\epsilon$. In the definition of S_{pc}^e, $\Delta\mu = \mu^{T=\Delta T} - \mu^T$ is the difference in chemical potential across the contact. Equation (3) yields the well-known linear term in the thermopower of a metal. For the bulk one obtains a similar expression assuming that the scattering time τ of the electrons behaves like $\tau(\epsilon) = l/v_{\mathrm{F}} \sim \epsilon^{-1/2}$ (constant mean free path l).[7] Therefore, we expect that the diffusional contribution to the thermopower is in balance between a point contact and the bulk.

In a simple model the phonon-drag component in the thermopower is proportional to the lattice specific heat and a reduction factor for the effectiveness of momentum transfer from the phonons to the electrons. The reduction factor α is a quotient between the relevant scattering times, such that

$$\alpha = \tau_{p,x}/(\tau_{p,x}+\tau_{p,e}), \tag{4}$$

where $\tau_{p,e}$ is the scattering time of a phonon with the electrons and $\tau_{p,x}$ with all other scatterers. At temperatures low compared with the Debye temperature θ_D, we have $\tau_{p,x} >> \tau_{p,e}$ yielding $\alpha \rightarrow 1$, and at high temperatures $\tau_{p,x}$ is reduced by the scattering with phonons ($\tau_{p,x}=\tau_{p,p} << \tau_{p,e}$), which gives $\alpha \rightarrow 0$. Together with the temperature dependence for the specific heat this behavior for α results in the well-known maximum in the phonon-drag thermopower at about $0.2\theta_D$. However, for a point contact, at low temperatures $\tau_{p,x}$ will be dominated by the contact dimensions such that $\alpha = a/l_{p,e} << 1$ and the phonon-drag term in the thermopower of a point contact will be suppressed.[3]

Therefore, by combination of the diffusive and phonon-drag contributions in Eq. (1) the experimentally determined thermopower of a homogeneous chain with a point contact will resemble the phonon-drag term of the bulk. In Fig. 2 we clearly see this phonon-drag peak in our measured signal for Ag. For temperatures $T > \theta_D/2$ (for Ag $\theta_D = 225$ K) still a constant signal remains, whereas the phonon drag is expected to go to zero. The constant unexplained signal depends on the contact resistance, being smaller for larger contact dimensions.

The origin of this constant signal in the measured thermopower at high temperatures has to be found in the description of the diffusional thermopower for a point contact or in a contribution of the phonon drag from the nonequilibrium distribution of the phonons in a point contact. In the point-contact model described by Eqs. (2) and (3), the diffusional thermopower gives the same functional dependence as for the bulk sample, independent of the contact dimensions. At high temperatures the diffusional thermopower depends linearly on the temperature and, in the difference between bulk and point-contact diffusional thermopower, one would not expect a constant contribution, which also depends on the area of the contact. If the characteristic length for bringing into equilibrium with each other the phonon systems at the hot and the cold parts of the contact is larger than the contact dimension, phonon drag in the point-contact region can still be important. Note that the distribution of the phonons is very anisotropic near the contact area, consisting of a hot and a cold part. The effectiveness for the phonon-drag process in a point contact should saturate to a constant value as a function of the temperature in order to explain the observed constant contribution to the thermopower. The sign of this contribution to the thermopower of the point contact is opposite to the normally positive sign for both the diffusional and the phonon-drag term in the thermopower of the noble metals. Umklapp processes are believed to be responsible for the positive thermopower in the noble metals, where a simple free-electron model would yield a negative contribution. Of course, this type of analysis for the phonon drag in a point-contact geometry is too simple and one has to solve the full nonequilibrium transport problem for thermal-electric effects in the contact region. A detailed investigation of this unexplained phenomenon is interesting, also in view of the electron transport efficiency in a point contact.[1,2] Because of the dependence on contact resistance a critical length scale for the occurrence of this effect can be given: For $R = 0.01$ Ω this would give a contact dimension $a = 100$ nm, where this effect becomes important.

We have studied other metals in the configuration depicted in Fig. 1. For the noble metals Cu and Au we see qualitatively the same behavior. However, at low temperatures the reproducibility of the phonon-drag peak seems to be obscured by the contribution of paramagnetic impurities. In the point-contact area the number of impurities depends on the specific contact, giving a variable point-contact thermopower compared to the averaged bulk thermopower. For these noble metals again we observe the constant signal at high temperatures. Also the homogeneous chains of Pt and Pd with a point contact show phonon-drag structure in the measured thermopower.

We conclude that a thermoelectric chain of the same material which includes a point contact shows a thermoelectric emf. The obtained thermopower yields the phonon drag of the bulk metal because of phonon-drag suppression in the constriction.

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[a] Permanent address: Institute for Low Temperature Physics and Engineering, Ukrainian S.S.R. Academy of Sciences, 47 Lenin Avenue, Kharkov, USSR.

[b] Present address: Max-Planck-Institut für Festkörperforschung, Hochfeld-Magnetlabor, 25 Avenue des Martyrs, F-38042 Grenoble Cedex, France.

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