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MODELLING OF CHARGE CARRIER NON-ISOTHERMAL TRANSPORT IN SILICON AND SILICON CARBIDE

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Abstract. Extending our previously published model of charge carrier transport and mobility, a procedure for calculation of the semiconductor thermoelectric coefficients is developed. It is shown that the phonon drag strongly influences the Seebeck and Peltier coefficients even at high temperatures, up to 400 K for Si and 600 K for SiC. As a new feature, the presented model gives a possibility to investigate the influence of the electron-hole scattering on thermoelectric coefficients. A strong underestimation of non-isothermal ∇T-proportional current density terms in most of the semiconductor device simulators is pointed out.

Key words: Seebeck coefficient, Peltier coefficient, Soret coefficient, phonon drag, electron-hole scattering.

1. INTRODUCTION

It is well known that electron $(\alpha = e)$ and hole $(\alpha = h)$ current densities $\mathbf{j}_\alpha$, deduced from irreversible thermodynamics, can be expressed as follows:

$$\mathbf{j}_\alpha = L_{1\alpha}(-\nabla\phi_\alpha) + L_{2\alpha}(-\nabla T), \tag{1}$$

where ϕ_α is the quasi-Fermi potential and T is the crystal temperature, common for lattice, electron, and hole subsystems. The transport coefficients $L_{1\alpha}$ and $L_{2\alpha}$ are to be determined either empirically from experiments or theoretically from the kinetic transport theory based on the Boltzmann transport equation (BTE). In terms of electrical conductivity σ_α and absolute thermoelectric power (the Seebeck coefficient) S_α, Eq. (1) can be rewritten as

$$\mathbf{j}_\alpha = \sigma_\alpha(-\nabla\phi_\alpha) + \sigma_\alpha S_\alpha(-\nabla T). \tag{2}$$

Stratton [1,2] was probably the first who examined the non-isothermal current equations from the viewpoint of the theory of the semiconductor devices. Later on, Nakagawa and Navon [3], referring to Stratton [2], used the non-isothermal current term in their gate-turn-off thyristor simulations. However, the constant value of the Seebeck coefficient $|S_\alpha| = k_B/2q$, used in [3] and later in several other publications, strongly underestimates the thermoelectric effect in semiconductors (k_B is the Boltzmann constant and $q = |e|$ is the unit charge).

A detailed treatment of various aspects of non-isothermal modelling of the semiconductor problems can be found in recent papers by Lindefelt [4–6].

In this paper we report some new results on the calculation of the thermoelectric coefficients obtained by extending our earlier model of charge carrier transport [7] based on the well-known Kohler's variational principle.

2. BASIC THEORY

For the isothermal case, it was shown in [7,8] that correct accounting for electron-hole scattering (EHS), i.e., regarding both electrons and holes as moving scattering centres, converts current equations into so-called cross-term form

$$\begin{aligned} \mathbf{j}_\alpha &= qn_\alpha\mu_{\alpha 2}(-\nabla\phi_\alpha) - qn_\beta\mu_{\alpha 3}(-\nabla\phi_\beta) \\ &= qn_\alpha\mu_{\alpha 1}\mathbf{E} - q_\alpha k_B T\mu_{\alpha 2}\nabla n_\alpha + q_\beta k_B T\mu_{\alpha 3}\nabla n_\beta , \end{aligned} \tag{3}$$

with two (three) mobility-like transport coefficients $\mu_{\alpha 1} = \mu_{\alpha 2} - \mu_{\beta 3}$. Here subscript β marks a carrier type opposite to the α-type carrier, n_α is the density of the α-type charge carriers, $\mathbf{E}$ is the electric field vector, and q_α denotes α-type particle charge sign (i.e. $q_e = -1$ for electrons and $q_h = 1$ for holes). According to Onsager's reciprocity relationship, $n_\alpha\mu_{\beta 3} = n_\beta\mu_{\alpha 3}$.

It should be emphasized that a more detailed treatment of the thermoelectric power in case of non-negligible EHS causes similar splitting of the Seebeck coefficient and, consequently, the splitting of the term proportional to ∇T in Eq. (2) as well. However, throughout this paper we shall follow the traditional interpretation of S_α in accordance with Eq. (2). Nevertheless, in the BTE solution using the variational principle, we actually account for the influence of EHS and all three driving forces, namely the gradients $\nabla\phi_\alpha$ and ∇T directly and gradient $\nabla\phi_\beta$ for each carrier type through the ratios $R_{\alpha\beta}$ specified below by Eq. (16).

The calculation procedure of the non-isothermal transport parameters implemented in this work is based on Kohler's variational method for the solution of the BTE for the electron and hole, coupled through the electron-hole collision integral. As in [7], we assume isotropic parabolic bands for both

electrons and holes. The quasi-Fermi potential gradients $\nabla\phi_\alpha$ and the temperature gradient ∇T are assumed to be weak and collinear, and the magnetic field to be zero. The referred isothermal transport and mobility model [7] and a relevant computer code constitute the mathematical basis of the present study. Here we will describe only some new extensions needed for the calculation of the thermoelectric coefficients.

The central position among the charge carrier non-isothermal transport parameters occupies the Seebeck coefficient S_α. Sometimes, especially for convenient scaling of the dependence on the temperature, the Peltier coefficients Π_α may be considered instead of S_α. From Kelvin's formula we have

$$\Pi_\alpha = TS_\alpha. \tag{4}$$

The Soret[1] coefficient D_α^T, as used by Stratton [1,2] for the description of the thermal diffusion contribution to the non-isothermal current flow in semiconductors, is straightforwardly related to the Seebeck coefficient

$$D_\alpha^T = q_\alpha \frac{\sigma_\alpha S_\alpha}{qn_\alpha}. \tag{5}$$

In case of conventional non-cross-term formulation we obtain $D_\alpha^T = q_\alpha \mu_\alpha S_\alpha$. In terms of variational parameters, the transport coefficients in Eq. (2) may be written as follows:

$$\sigma_\alpha = q^2 \sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_{\alpha r}^{(E)}, \tag{6}$$

$$\sigma_\alpha S_\alpha = \frac{q_\alpha q}{T} \sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_{\alpha r}^{(T)}, \tag{7}$$

which enable us to define the Seebeck coefficient as

$$S_\alpha = \frac{q_\alpha}{qT} \frac{\sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_{\alpha r}^{(T)}}{\sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_{\alpha r}^{(E)}}, \tag{8}$$

where N is the order of approximation of the solution.

The variational parameters $c_{\alpha r}^{(E)}$ are coefficients of the polynomial function of the reduced carrier energy ε_α

[1] Sometimes the Soret coefficient is defined as $n_\alpha D_\alpha^T$, where D_α^T is called the thermal diffusion coefficient.

$$\varphi_\alpha^{(E)} = \sum_{r=0}^{N} c_{\alpha r}^{(E)} \varepsilon_\alpha^r, \quad \varepsilon_\alpha = \frac{E_\alpha}{k_B T}. \tag{9}$$

This function was used as a trial function for Kohler's variational solution of the BTE. The limit $N \to \infty$ corresponds to the exact solution of the BTE. Fortunately, successive approximations reveal a rapid convergence after the first order correction and, therefore, often the first and in some cases even the zero order approximation provide practically acceptable accuracy of the examined transport parameters.

Instead of explicit calculation of the coefficients $c_{\alpha r}^{(E)}$, the sums in Eqs. (6)–(8) can be evaluated making use of the technique of the bordered determinants introduced by Enskog [9]. In this case

$$\sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_\alpha^{(E)} = \frac{1}{G} D_\alpha^{(E)}, \tag{10}$$

$$\sum_{r=0}^{N} c_{\alpha r}^{(E)} \beta_{\alpha r}^{(T)} = \frac{1}{G} D_\alpha^{(T)}, \tag{11}$$

and in accordance with Eq. (7),

$$S_\alpha = \frac{q_\alpha}{qT} \frac{D_\alpha^{(T)}}{D_\alpha^{(E)}}. \tag{12}$$

For the first order approximation $(N = 1)$ the "electrical" and "thermal" 5×5 bordered determinants for electrons and holes and the "scattering" 4×4 G-determinant become:

$$D_e^{(E,T)} = \begin{vmatrix} 0 & \beta_{e0}^{(E,T)} & 0 & \beta_{e1}^{(E,T)} & 0 \\ +R_{ee}\beta_{e0}^{(E)} & \dots & \dots & \dots & \dots \\ -R_{eh}\beta_{h0}^{(E)} & \dots & G & \dots & \dots \\ +R_{ee}\beta_{e1}^{(E)} & \dots & \dots & \dots & \dots \\ -R_{eh}\beta_{h1}^{(E)} & \dots & \dots & \dots & \dots \end{vmatrix}, \tag{13}$$

$$D_h^{(E,T)} = \begin{vmatrix} 0 & 0 & \beta_{h0}^{(E,T)} & 0 & \beta_{h1}^{(E,T)} \\ -R_{he}\beta_{e0}^{(E)} & \dots & \dots & \dots & \dots \\ +R_{hh}\beta_{h0}^{(E)} & \dots & G & \dots & \dots \\ -R_{he}\beta_{e1}^{(E)} & \dots & \dots & \dots & \dots \\ +R_{hh}\beta_{h1}^{(E)} & \dots & \dots & \dots & \dots \end{vmatrix}, \tag{14}$$

$$G = \begin{vmatrix} \Gamma_{00}^{ee} & \Gamma_{00}^{eh} & \Gamma_{01}^{ee} & \Gamma_{01}^{eh} \\ \Gamma_{00}^{he} & \Gamma_{00}^{hh} & \Gamma_{01}^{he} & \Gamma_{01}^{hh} \\ \Gamma_{10}^{ee} & \Gamma_{10}^{eh} & \Gamma_{11}^{ee} & \Gamma_{11}^{eh} \\ \Gamma_{10}^{he} & \Gamma_{10}^{hh} & \Gamma_{11}^{he} & \Gamma_{11}^{hh} \end{vmatrix}. \tag{15}$$

The evaluation of the Γ-elements of the scattering determinant has been described in [7]. The coefficients $R_{\alpha\beta}$, introduced but not used in [7] for mobilities calculation, represent the ratio of the quasi-Fermi potential gradients of the electron and hole

$$R_{\alpha\beta} = -\frac{\partial\phi_\beta / \partial z}{\partial\phi_\alpha / \partial z}. \tag{16}$$

Semiconductor device simulation shows that $R_{\alpha\beta} \simeq -1$ for the structure of homogeneous quasineutral regions and, in average, for all the contact-to-contact interval as well. Therefore in the present calculations we have assumed $R_{\alpha\beta} = -1$. The carrier concentration-related terms $\beta_{\alpha r}^{(E)}$ are calculated similarly to the isothermal case [7] as

$$\beta_{ar}^{(E)} = \frac{n_\alpha}{m_\alpha} \frac{(r+3/2)!}{(3/2)!} \frac{\mathcal{F}_{r+1/2}(\eta_\alpha)}{\mathcal{F}_{1/2}(\eta_\alpha)}, \tag{17}$$

assuming that the effective mass m_α is isotropic and using Fermi integrals

$$\mathcal{F}_j(\eta_\alpha) = \frac{1}{j!} \int_0^\infty \frac{\varepsilon^j}{\exp(\varepsilon - \eta_\alpha) + 1} \, \mathrm{d}\varepsilon, \tag{18}$$

with reduced quasi-Fermi energy η_α as a parameter.

The new "thermal" $\beta_{\alpha r}^{(T)}$-terms are to be calculated as follows:

$$\beta_{\alpha r}^{(T)} = k_B T \cdot \beta_{\alpha r}^{(E)} \left\{ \left(r + \frac{5}{2} \right) \frac{\mathcal{F}_{r+3/2}(\eta_\alpha)}{\mathcal{F}_{r+1/2}(\eta_\alpha)} - \eta_\alpha \right\}. \tag{19}$$

Note that in case of $N = 0$, Eqs. (17) and (19), inserted into Eq. (8), yield for Maxwell–Boltzmann statistics

$$S_\alpha = \frac{q_\alpha}{qT} \frac{\beta_{\alpha 0}^{(T)}}{\beta_{\alpha 0}^{(E)}} = q_\alpha \frac{k_B}{q} \left(\frac{5}{2} - \eta_\alpha \right). \tag{20}$$

Equation (20) reveals that in zero order approximation the Seebeck coefficients are EHS independent and the calculation of determinants in Eq. (12) is actually not needed.

Interrelating the reduced quasi-Fermi energies to the conduction and valence band effective densities of state, N_C and N_V, two useful estimations may be deduced from Eq. (20):

$$S_e = -\frac{k_B}{q}\left(\frac{5}{2} + \ln\frac{N_C}{n_e}\right), \quad n_e < N_C, \tag{21}$$

$$S_h = \frac{k_B}{q}\left(\frac{5}{2} + \ln\frac{N_V}{n_h}\right), \quad n_h < N_V. \tag{22}$$

Further, by introducing well-known functions of the temperature $N_{C,V}(T) = N_{C300,V300}\left(\frac{T}{300\text{K}}\right)^{3/2}$, a rather weak temperature dependence of the Seebeck coefficients follows:

$$S_e = -\frac{k_B}{q}\left(\frac{5}{2} + \ln\frac{N_{C300}}{n_e} + \frac{3}{2}\ln\frac{T}{300\text{K}}\right), \tag{23}$$

$$S_h = \frac{k_B}{q}\left(\frac{5}{2} + \ln\frac{N_{V300}}{n_h} + \frac{3}{2}\ln\frac{T}{300\text{K}}\right). \tag{24}$$

However, measurements by Frederikse [10] and Geballe and Hull [11] have shown a remarkable rise of the thermoelectric power in Ge and Si single crystal specimens at low lattice temperatures, typically below 100 to 200 K. The explanation, suggested independently by Frederikse [10] and Herring [12], is based on the effect of drag of the electrons and holes caused by non-equilibrium phonons in the crystal. The strength of the phonon drag mechanism is proportional to the ratio of phonon and electron or hole mean free paths [10] which rises rapidly at low temperatures due to the predominating increase of the phonon mean free path. At very low temperatures, below 20 to 40 K, the sample boundary scattering will limit phonon mean free path to the order of the crystal size while the charge carrier mean free path continues to rise. Consequently, the influence of the phonon drag on the thermopower will decrease.

The phonon drag has been modelled by adding an extra term $\beta^{(T)}_{\alpha r.ph}$ to the regular term $\beta^{(T)}_{\alpha r}$ given by Eq. (19). Following Frederikse [10] and Herring [12,13], we obtain

$$\beta^{(T)}_{\alpha r.ph} = k_B T \beta^{(E)}_{\alpha r} A^H_{\alpha.ph} \frac{(r+1)!}{(r+3/2)!} \frac{\mathcal{F}_r(\eta_\alpha)}{\mathcal{F}_{r+1/2}(\eta_\alpha)}, \tag{25}$$

where

$$\left(\frac{n}{2}\right)! = \frac{1}{2}\cdot\frac{3}{2}\cdot\ldots\cdot\frac{n}{2}\sqrt{\pi}, \quad \text{if } n = 1,\ 3,\ 5,\ldots$$

Dimensionless factor $A^H_{\alpha.ph}$ in Eq. (25) characterizes the phonon drag force $k_B A^H_{\alpha.ph} \varepsilon_\alpha^{-1/2} (-\nabla T)$ in accordance with Herring's phonon-phonon scattering theory [13]. In case of large crystals of cubic symmetry and negligible phonon boundary scattering, according to [13,14], the factor $A^H_{\alpha.ph}$ can be written as follows:

$$A^H_{\alpha.ph} = \frac{E_{1\alpha}^2 u_l^3 m_\alpha^{3/2}}{\pi 2^{3/2} (k_B \cdot 300\text{K})^{7/2}} \left(\frac{T}{300\text{K}} \right)^{-7/2}, \tag{26}$$

where $E_{1\alpha}$ is the deformation potential constant for intravalley acoustic scattering and u_l is the longitudinal sound velocity.

In case of non-negligible phonon drag effect, Eq. (20) should be modified:

$$S_\alpha = \frac{q_\alpha}{qT} \frac{\beta^{(T)}_{\alpha 0} + \beta^{(T)}_{\alpha 0.ph}}{\beta^{(E)}_{\alpha 0}} = q_\alpha \frac{k_B}{q} \left(\frac{5}{2} - \eta_\alpha + \frac{4}{3\sqrt{\pi}} A^H_{\alpha.ph} \right). \tag{27}$$

For use in device simulators, a more practical formula can be proposed:

$$S_\alpha = q_\alpha \frac{k_B}{q} \left(\frac{5}{2} + \ln \frac{N_{C,V}(300\text{K})}{n_\alpha} + \frac{3}{2} \ln \frac{T}{300\text{K}} + C_\alpha \left(\frac{T}{300\text{K}} \right)^{-7/2} \right), \tag{28}$$

where

$$C_\alpha = \frac{\sqrt{2}}{3\pi^{3/2}} \frac{E_{1\alpha}^2 u_l^3 m_\alpha^{3/2}}{(k_B \cdot 300\text{K})^{7/2}}. \tag{29}$$

Our calculations provide $C_e = 4.29$, $C_h = 5.46$ for Si, and $C_e = 22.5$, $C_h = 82.6$ for 6H–SiC in the crystallographic direction perpendicular to the *c*-axis. By that we used the following sets of input data: 1) $m_e = 1.185 m_0$, $m_h = 1.153 m_0$, $E_{1e} = 3.3$ eV, $E_{1h} = 3.8$ eV, and $u_l = 9.04 \times 10^5$ cm/s for Si, 2) $m_e = 0.42 m_0$, $m_h = 1.0 m_0$, $E_{1e} = E_{1h} = 10$ eV, and $u_l = 12.6 \times 10^5$ cm/s for 6H–SiC.

3. RESULTS AND DISCUSSION

Generally, the Seebeck coefficient dependence on the carrier concentration, doping, and temperature is of interest. Figure 1 shows calculated S_h versus hole concentration in order to demonstrate the applicability of Eqs. (21)–(24) and to check the possible first order corrections due to the influence of the carrier-carrier and carrier-impurity scattering mechanisms through the determinants in Eq. (12).

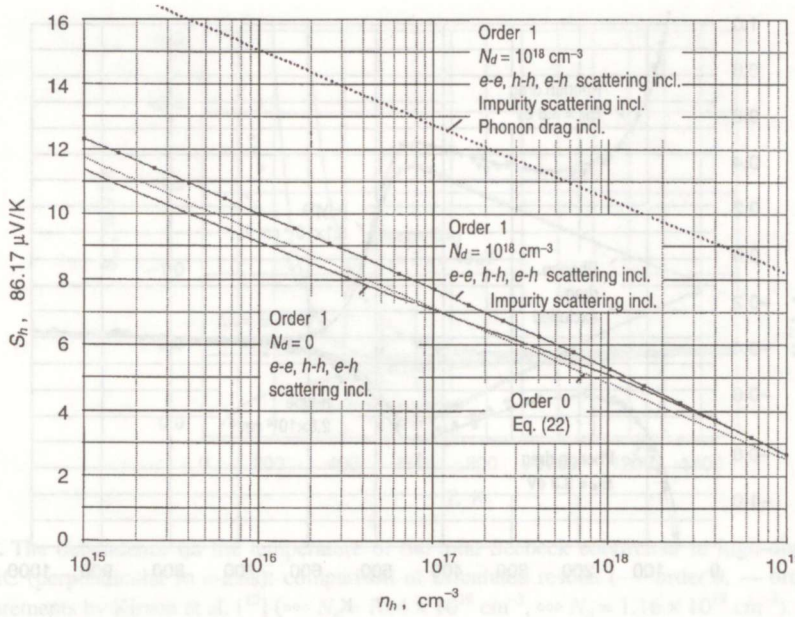

Fig. 1. Calculated Seebeck coefficient for holes in units of $k_B/q = 86.17\,\mu\text{V/K}$ versus hole concentration in intrinsic (donor concentration $N_d = 0$) and heavily doped ($N_d = 10^{18}$ cm^{-3}) n-type Si.

In order to check the overall accuracy of the present model, especially the phonon drag term, Fig. 2 compares the temperature dependence of the calculated total Peltier coefficient (for mixed electron and hole conductivity) with measurements in silicon by Geballe and Hull [11].

The total Peltier (or Seebeck) coefficient is expressed as

$$\Pi_{\text{tot}} = TS_{\text{tot}} = T\frac{\sigma_e S_e + \sigma_h S_h}{\sigma_e + \sigma_h}. \tag{30}$$

To obtain a reasonable fit to the measured results we used in Eq. (26), instead of Herring's theoretical limit $T^{-3.5}$, a weaker temperature dependence $T^{-2.3}$ as suggested by Geballe and Hull [11]. Additionally, we selected proper values of the deformation potential constants E_{1e} and E_{1h} to fit experimental curves at low temperatures. In order to approach experimental points at the high temperature intrinsic region where $n_e \simeq n_h \to n_i(T)$, we had to use gradually diminishing ratio of mobilities $b = \mu_e/\mu_h$ at higher temperatures ($b = 2.9$ at 300 K and $b \to 1.6$ at 1000 K).

To our knowledge, only one paper considers the Seebeck coefficient measurements in SiC [15]. In this work, the Seebeck coefficient of the nitrogen doped n-type 6H–SiC has been measured in the hexagonal plane, applying temperature gradient in perpendicular direction to the *c*-axis.

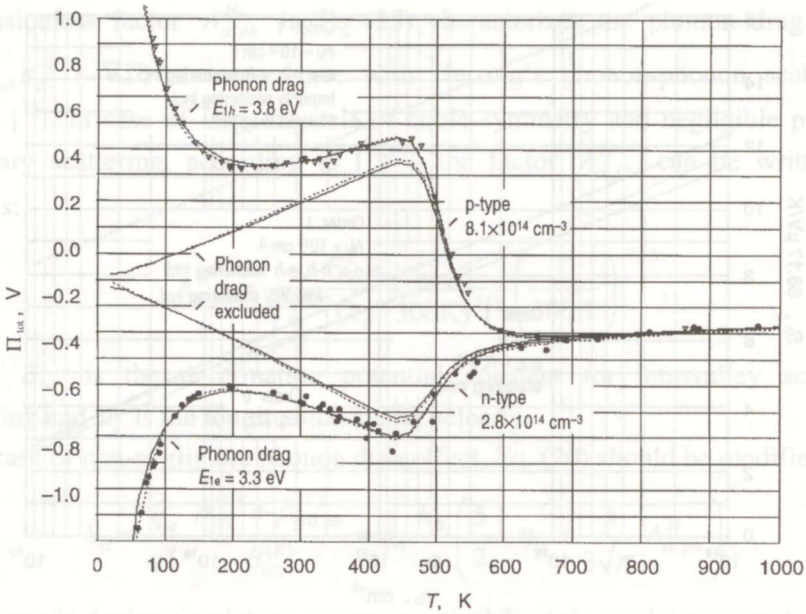

Fig. 2. Dependence on the temperature of the total Peltier coefficient for mixed electron and hole conductivity for low-doped n-Si and p-Si: calculated results (--- order 0, — order 1) are compared to Geballe and Hull [10] measurements for donor concentration 2.8×10^{14} cm^{-3} and acceptor concentration 8.1×10^{14} cm^{-3}.

In our calculations for 6H–SiC, the effective masses $m_e = 0.42m_0$ and $m_h = 1.0m_0$, the longitudinal sound velocity $u_l = 12.6 \times 10^5$ cm/s, and the deformation potential constants $E_{1e} = E_{1h} = 10$ eV were taken according to [16]. In this work, the value of 10 eV was established as a value providing the best fit of calculated mobilities and their anisotropy ratios to the measured ones of both 6H–SiC and 4H–SiC.

Figure 3 compares the temperature dependence of the experimental and calculated Seebeck coefficients for the n-type 6H–SiC with $n_e \simeq 10^{19}$ cm^{-3}.

The measured and calculated Seebeck coefficients for the n-type 6H–SiC agree well at higher temperatures $T > 600$ K, but at lower temperatures, where the phonon drag component is dominating, our theoretical model with $E_{1e} = E_{1h} = 10$ eV strongly overestimates the phonon-drag-induced Seebeck effect. A somewhat better fit in the range $T < 300$ K was obtained by using the reduced deformation potential coefficients $E_{1e} = E_{1h} = 5$ eV, but presumably the real reason of the above discrepancy is hidden in the incompleteness of the low energy phonon-phonon scattering model resulting in the factor $A^H_{\alpha \cdot ph}$ specified by Eq. (26).

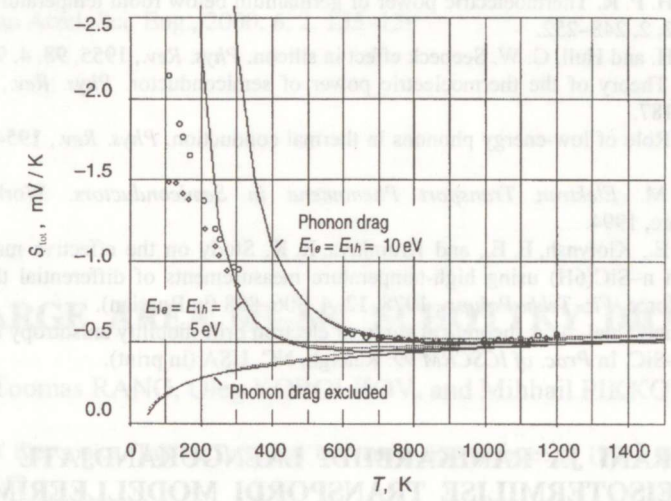

Fig. 3. The dependence on the temperature of the total Seebeck coefficient in high-doped n-type 6H–SiC (perpendicular to *c*-axis): comparison of calculated results (····· order 0, — order 1) with measurements by Kirson et al. [15] (○○○ $N_d = 1.04 \times 10^{19}$ cm^{-3}, ◇◇◇ $N_d = 1.16 \times 10^{19}$ cm^{-3}).

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RÄNI JA RÄNIKARBIIDI LAENGUKANDJATE MITTEISOTERMILISE TRANSPORDI MODELLEERIMINE

Enn VELMRE ja Andres UDAL

Samade autorite varem avaldatud isotermilise transpordimudeli üldistamise tulemusel on esitatud pooljuhi termoelektriliste nähtuste teoreetiline mudel ja vastavate koefitsientide arvutusprotseduur. Seebecki ja Peltier' tegurite modelleerimisel madalamate temperatuuride piirkonnas on olulise mõjurina arvesse võetud elektronide foononkanne. Esitatud mudel võimaldab arvestada ka elektron–aukhajumise mõju termoelektrilistele nähtustele. On näidatud, et enamikus pooljuhtseadiste simulaatorites rakendatud transpordimudelites on mitteisotermilise, s.t. temperatuurigradiendiga võrdelise voolukomponendi osa tugevasti alla hinnatud.