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Thermoelectric power of hot carriers in the nonequilibrium-statistical-operator approach

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The thermoelectric power of charge carriers heated under a strong applied electric field in semiconductors is obtained by use of the nonequilibrium-statistical-operator (NSO) method. The balance equations are derived in terms of the NSO density matrix and the force-force correlation functions which can easily be calculated for a system with electron-impurity and electron-phonon interactions. A numerical study has been performed for hole-doped Ge. It is shown that the hot-electron thermoelectric power is sensitively affected by the applied electric field and that its sign is reversed at higher electric fields.

I. INTRODUCTION

Recently there has been a revival of research activities\textsuperscript{1-3} on the thermoelectric properties of materials, particularly due to their relevance to the thermoelectric cooling technology.\textsuperscript{3} For a material to be a good thermoelectric refrigerator, it must have a high Seebeck coefficient or thermoelectric power \textit{S}.\textsuperscript{1-3} Therefore, it would be interesting to derive an effective formula to calculate the thermoelectric power and examine the hot-electron effect that often exists in thermoelectric measurements.

The purpose of this paper is to study \textit{S} of hot carriers in semiconductors under a uniform electric field \textit{E} and calculate the variation of \textit{S} with \textit{E}. The physical process under consideration is the thermoelectric Seebeck effect in the presence of a crossed electric field \textit{E} and a lattice temperature gradient \textit{\nabla} \textit{T}. With strong electron-electron interaction, electrons heated by the strong electric field equilibrate among themselves at an electron temperature \textit{T}_e, which is usually higher than the lattice temperature \textit{T}. On the other hand, the applied \textit{\nabla} \textit{T} causes an electronic temperature gradient \textit{\nabla} \textit{T}_e via electron-phonon interactions so as to give rise to a nonuniform electronic distribution or a chemical potential gradient \textit{\nabla} \textit{\mu} under the open circuit conditions. Since \textit{\nabla} \textit{T} is the driving force responsible for the Seebeck effect, we define a hot-electron thermoelectric power as \textit{S} = \textit{e}^{-1}(\textit{\nabla} \textit{T}_e/\textit{\nabla} \textit{T}) with \textit{e} the electron charge. It is assumed that the phonon-drag contribution to \textit{S} may be neglected in the range of electron temperatures of interest. The present definition for \textit{S} differs from that in Ref. 4, where the crystal lattice is assumed to remain at uniform temperature (\nabla \textit{T} = 0), and so the only driving force for the Seebeck effect is \textit{\nabla} \textit{T}_e caused by a nonuniform microwave heating.

Although there exist some investigations of the thermoelectric power for charge carriers heated by a strong microwave field in some literatures,\textsuperscript{4,5} to our knowledge, the topic proposed here has not yet been studied. Furthermore, the previous interpretation of the experimental results\textsuperscript{4} was based upon the Boltzmann equation in the relaxation-time approximation.\textsuperscript{5} It is significant to develop a fundamental theory for \textit{S} in terms of the nonequilibrium-statistical-operator (NSO) method\textsuperscript{6} and the balance equation approach.\textsuperscript{7-9} In the next section, we will extend the NSO method of hot-electron transport\textsuperscript{8} to weakly nonuniform systems in the presence of both a strong \textit{E} and a small \textit{\nabla} \textit{T}, yielding its statistical density matrix. Starting from this, in Sec. III, we derive an analytic formula for \textit{S} as a function of \textit{T}_e, the latter being determined by force and energy balance equations. In Sec. IV, a similar formula for \textit{S} is obtained by solving the Boltzmann equation in the relaxation-time approximation. Finally, numerical results for hole-doped Ge are presented in Sec. V. It is shown that the hot-electron thermoelectric power is sensitively affected by \textit{E} and that its sign changes at higher electric fields.

II. NSO DENSITY MATRIX

Let us consider a system involving \textit{N} interacting electrons under the influence of a uniform electric field \textit{E} along the \textit{x} axis and a small lattice temperature gradient \textit{\nabla} \textit{T} along the \textit{y} axis, subject to impurity and phonon scattering. The total Hamiltonian of the system is 

\[ H = \int d\textit{r}[\textit{H}_e(\textit{r}) + \textit{H}_e(\textit{r}) + \textit{H}_{\text{ph}}(\textit{r}) + \textit{H}_{\text{el}}(\textit{r})] \]

where
we have separated the electron Hamiltonian into the center-of-mass part $H_e$ and the relative part $H_r$. $H_{\text{ph}}$ is the phonon Hamiltonian, and $H_{\text{el}}$ includes the electron-impurity and electron-phonon interactions. Thermal perturbations due to the applied temperature gradient are not included in the Hamiltonian, but described by adding an extra term to the following entropy production operator. The statistical density matrix in the NSO approach has the form:

$$\rho(t) = \exp\left[-S(t,0) + \int_{-\infty}^{0} dt' e^{st'} \tilde{S}(t + t', t')\right].$$

Here $S(t,0) = \Psi(t) + \sum_m \int dx F_m(x, t) P_m(x, t)$ is the entropy operator of the system where $F_m$ are the basis dynamic operators, and $P_m$ are the thermodynamic parameters conjugate to the average values of $F_m$. $\Psi(t) = \ln \text{Tr}[\exp[-\sum_m \int dx F_m(x, t) P_m(x, t)]]$, is the normalization coefficient such that $\text{Tr}\rho_t = 1$ with $\rho_t = \exp[-S(t,0)]$ corresponding to the quasiequilibrium statistical operator in the absence of dissipative processes. The entropy production operator is the time derivative of $S(t,0)$. For the steady-state transport under consideration, $\hat{F}_m(t) = 0$, then we have

$$\tilde{S}(t,0) = \sum_m \int dx F_m(x, t) [\hat{P}_m(x, t) - (\hat{P}_m(x, t))_t],$$

where $(\cdots)_t = \text{Tr}(\cdots \rho_t)$, and

$$\tilde{S}(t,t') = \text{exp}(iHt') \tilde{S}(t,0) \text{exp}(-iHt'),$$

which second argument denotes the Heisenberg time dependence. The success of the application of the NSO method often depends on the appropriate choice of $F_m$ and $P_m$. In order to describe correctly hot-electron transport behavior under a strong electric field and a weak temperature gradient, we choose

$$\{P_m(x, t)\} = \{H_e(x, t), H_{\text{ph}}(x, t), N(x, t)\},$$

and

$$\{F_m(x, t)\} = \{\beta_e(x, t), \beta_r(x, t), -\beta_e(x, t)\mu(x, t)\},$$

where $\beta_e$ and $\mu$ are the reciprocal effective temperature and the chemical potential of the electrons and $\beta$ is the reciprocal temperature of the lattice. It should be noted here that $S(t,0)$ only contains the relative part of the electron Hamiltonian because a quasiequilibrium statistical operator is always independent of the center-of-mass motion. The time derivatives of the operators $F_m(x, t)$ appearing in $\tilde{S}(t,0)$ can be shown to satisfy the following equations:

$$\dot{P}_m(x, t) + \text{div} J_m(x, t) + i [P_m(x, t), H] = 0,$$

where $J_1(x, t)$ and $J_2(x, t)$ are the energy flux densities of electrons and phonons, respectively, and $J_3(x, t)$ is the electron-number flux density. Substituting Eq. (5) into Eq. (2) and integrating by parts, we obtain

$$\dot{S}(t, t') = \int dx \{\beta_e(x, t) H_e(x, t) + \beta_r(x, t) H_{\text{ph}}(x, t) \}
+ \sum_m \left[ J_m(x, t) - \langle J_m(x, t) \rangle_t \right] \cdot \nabla F_m(x, t),$$

where $H_e(x, t) = -i[H_e, H_{\text{el}}]$ and $H_{\text{ph}}(x, t) = -i[H_{\text{ph}}, H_{\text{el}}]$. It can be seen from Eq. (6) that the entropy production comes from two types of dissipative processes: the energy or particle flux, and the energy transfer from hot electrons to the lattice. One can see from Eq. (6) that all the terms in $\dot{S}(t, t')$ contain small parameters. In the first two terms both $H_e(x, t)$ and $H_{\text{ph}}(x, t)$ are proportional to the electron-impurity and the electron-phonon interactions, which have been regarded as being small in the hot-electron transport theory. Those terms that appear in $\nabla F_m(x, t)$ are also small for weak spatial inhomogeneity. We can thus expand $\rho(t)$ to the first order in $\dot{S}(t, t')$ (Ref. 7)

$$\rho(t) = \rho_t \left( 1 + \int_{-\infty}^{0} dt' e^{t't} \int_{0}^{1} dw e^{-\omega S(t,0)} \times \dot{S}(t + t', t') e^{\omega S(t,0)} \right).$$

Since the present approach is confined to the linear approximation in $\nabla F_m(x, t)$, we can replace all the parameters in Eq. (7) by their corresponding spatial average values. Under this approximation the statistical density matrix at a steady state can be written as

$$\rho = \rho_t \left( 1 + \int_{-\infty}^{0} dt' e^{t't} \int_{0}^{1} dw \left[ \beta_e H_e(t', t) + \beta_r H_{\text{ph}}(t', t) \right] \right. \left. + \sum_m [J_m(t', t) - \langle J_m(t', t) \rangle_t] \cdot \nabla F_m(t') \right) \right) \right) \right).$$

Here $\rho_t = \text{exp}[-\beta_e(E_e + \alpha H_{\text{ph}} - \mu N)]$ with $\alpha = T_e/T$, and $A(t, t', \omega) = \text{exp}[-\omega \beta_e(E_e + \alpha H_{\text{ph}})/A(t') + \alpha H_{\text{ph}}]$ with $A(t')$ having the Heisenberg time dependence. The four momentum $k$ and $\Omega_{\lambda}$ are the energy of the phonon in branch $\lambda$. Since $\nabla F_m(t')$ should be switched on adiabatically, $\lim_{t' \to -\infty} \nabla F_m(t') = 0$, it is reasonable to take $\nabla F_m(t') = F_m e^{t'/\tau}$ with $\tau$ the average relaxation time due to the electron-impurity and electron-phonon scattering as well as the interactions between electrons. For the terms involving $\nabla F_m(t')$ in Eq. (8), the integrations over $t'$ and $\omega$ are easily done, and so the NSO density matrix is given by

$$\rho = \rho_t \left( 1 + \int_{-\infty}^{0} dt' e^{t't} \int_{0}^{1} dw \left[ \beta_e H_e(t', t) + \beta_r H_{\text{ph}}(t', t) \right] \right. \left. + \sum_m \tau (J_m(x, t) - \langle J_m(x, t) \rangle_t) \cdot \nabla F_m \right) \right).$$
III. HOT-ELECTRON THERMOELECTRIC POWER

In the steady state, hot-electron transport is determined by the force and energy balance equations. The time derivatives of the center-of-mass momentum operator $\mathbf{P}$ and the Hamiltonian operator $H_e$ of the relative electron system are given by $\dot{\mathbf{P}} = NeE - i[\mathbf{P}, H_e]$ and $\dot{H}_e = -i[H_e, H_\text{el}]$ with $N$ the charge carrier density. Their statistical averages with respect to the density matrix given by Eq. (9) should vanish for the steady state, i.e., $\langle \mathbf{P} \rangle = 0$ and $\langle H_e \rangle = 0$. Taking $\mathbf{E}$ to be along the $x$ axis and $\nabla F_m$ along the $y$ axis, and using the standard Green’s function technique, we obtain the force and energy balance equations

$$N_eE + F(v_d, T_e) = 0,$$

$$N_eE v_d - W(v_d, T_e) = 0,$$

where $v_d$ is the drift velocity due to the applied electric field, and

$$F(v_d, T_e) = 2 \sum_{q, \lambda} |M(q, \lambda)|^2 q_e \Pi_2(q, q_e v_d + \Omega_{q\lambda})$$

$$\times \left[ n(\beta_e \Omega_{q\lambda}) - n(\beta_e (q_e v_d + \Omega_{q\lambda})) \right]$$

$$+ n_i \sum_{q} |u(q)|^2 \Pi_2(q, q_e v_d),$$

$$W(v_d, T_e) = 2 \sum_{q, \lambda} |M(q, \lambda)|^2 \Omega_{q\lambda} \Pi_2(q, q_e v_d + \Omega_{q\lambda})$$

$$\times \left[ n(\beta_e \Omega_{q\lambda}) - n(\beta_e (q_e v_d + \Omega_{q\lambda})) \right].$$

Here $M(q, \lambda)$ and $u(q)$ are, respectively, the electron-phonon and impurity-electron interaction matrix elements, and $n_i$ is the impurity concentration. $n(\omega)$ is the Bose factor. $\Pi_2(q, \omega) = \sum_{k, q} \Pi_2(k, q, \omega) = 2\pi \sum_{k} |f(k)|^2 \delta(\omega - \epsilon_k) \delta(\omega + \epsilon_k)$ is the imaginary part of the retarded electron density-density response function with $f(k)$ as the Fermi function at the temperature $T_e$. Its renormalization due to the Coulomb interaction between electrons can be easily obtained in the random-phase approximation. Equations (10)–(13) can be used to determine the solutions of $v_d$ and $T_e$ for given $E$ and $T$. In the low-field limit, taking $T_e = T$ and expanding $F(v_d, T)$ to the linear order in $v_d$: $F(v_d) = -N m \tau_{\text{tr}}^{-1} v_d$, from Eq. (10) we get the Ohmic resistivity $\rho = (m/Ne^2) \tau_{\text{tr}}^{-1}$, where the inverse transport lifetime $\tau_{\text{tr}}^{-1} = \tau_{\text{el}}^{-1} + \tau_{\text{ph}}^{-1}$ with

$$\tau_{\text{el}}^{-1} = \frac{n_i}{Nm} \sum_{k, q} |u(q)|^2 q_e d\Pi_2(k, q, \omega)$$

$$\left. \frac{d\Pi_2(k, q, \omega)}{d\omega} \right|_{\omega=0},$$

$$\tau_{\text{ph}}^{-1} = \frac{2}{NmT} \sum_{k, q} |M(q, \lambda)|^2 q_e^2 \Pi_2(k, q, \Omega_{q\lambda}) \frac{e^{\beta_e \Omega_{q\lambda}}}{e^{\beta_e \Omega_{q\lambda}} - 1}.$$
\[
\left[ e E v_x - v_y \left( \frac{\epsilon_k - \mu}{T} \nabla T + \nabla \mu \right) \right] \frac{\partial f}{\partial \epsilon_k} = \frac{\partial f}{\partial t} \bigg|_{\text{collision}}^{\text{one-body}} + \frac{\partial f}{\partial t} \bigg|_{\text{collision}}^{\text{two-body}}. \tag{20}
\]

All the drift terms due to external forces are placed on the left-hand side of the equation. The one-body and two-body collision terms describe the scattering of electrons by phonons and impurities and the scattering due to the electron-electron interaction, respectively. Under the relaxation-time approximation they are given by\(^{12,14}\)

\[
\frac{\partial f}{\partial t} \bigg|_{\text{collision}}^{\text{one-body}} = -\tau_k^{-1} [f(k) - f_0(k)], \tag{21}
\]

\[
\frac{\partial f}{\partial t} \bigg|_{\text{collision}}^{\text{two-body}} = -\tau_{ee}^{-1} [f(k) - f_0(k - m v_d)], \tag{22}
\]

where \(f_0(k)\) is the equilibrium distribution function. The effect of the two-body collision term is to drive the distribution at a rate \(1/\tau_{ee}\) towards a displaced Boltzmann distribution with a center-of-mass momentum \(m v_d\), while the one-body scattering is to drive the distribution at a rate \(1/\tau_k\) towards the equilibrium one. Substituting Eqs. (21) and (22) into Eq. (20) and expanding \(f_0(k - m v_d)\) to the first order in \(v_d\),\(^{12}\) we obtain the correction to the distribution function, \(g = f - f_0\), as

\[
g(k) = -\tau_{\text{eff}} \left[ e E v_x - v_y \left( \frac{\epsilon_k - \mu}{T} \nabla T + \nabla \mu \right) \right] \frac{\partial f_0}{\partial \epsilon_k} - m v_d \tau_{ee} \tau_{\text{eff}} \frac{\partial f_0}{\partial \epsilon_k}, \tag{23}
\]

where \(\tau_{\text{eff}} = \tau_k \tau_{ee}/(\tau_k + \tau_{ee})\). The electric current density can be calculated by \(J = (e/m) \sum_k k g(k)\). It then follows from Eq. (23) that

\[
J_x = \frac{\tau_{\text{eff}}}{\tau_k} \frac{\epsilon_k}{\epsilon_k} = \frac{N e^2 E}{m} \langle \tau_{\text{eff}} \rangle_0, \tag{24}
\]

\[
J_y = \frac{\tau_{\text{eff}}}{\tau_k} \frac{\epsilon_k}{\epsilon_k} = -\frac{N e}{m} \left[ \langle \tau_{\text{eff}} \epsilon_k \rangle_0 - \mu \langle \epsilon_k \rangle_0 \nabla T/T \right. \\
+ \left. \langle \epsilon_k \rangle_0 \nabla \mu \right], \tag{25}
\]

where

\[
\langle A(k) \rangle_0 = (2/3N) \sum_k A(k) \epsilon_k (-\partial f_0/\partial \epsilon_k). \tag{26}
\]

The results for \(\sigma\) and \(S\) are easily obtained from Eqs. (24) and (25). If \(\tau_{ee}\) is much greater than \(\tau_k\), one finds from \(\tau_{\text{eff}} \approx \tau_k\) that

\[
\sigma = \frac{N e^2}{m} \langle \tau_k \rangle_0, \tag{27}
\]

which are just the well-known Boltzmann results. On the other hand, the hot-electron transport problem belongs to the opposite limit, i.e., \(\tau_{ee} \ll \tau_k\). In this case, \(\tau_{\text{eff}} \approx \tau_{ee}\), taking it as a constant independent of \(k\), we have

\[
\sigma = \frac{N e^2}{m} \frac{1}{(1/\tau_k)_0}, \tag{29}
\]

\[
S = \frac{1}{eT} \left[ \left\langle \frac{\epsilon_k \epsilon_k}{2} \right\rangle_0 - \mu \right]. \tag{30}
\]

In obtaining Eq. (30) we have used the relations \(\langle \epsilon_k \rangle_0 = 5k_B T/2\) and \(\langle 1 \rangle_0 = 1\). Equation (29) is the formula for conductivity in the balance equation approach\(^{7-9,12}\) and Eq. (30) is found to be the same as Eq. (19) in the limit of \(T_e = T\).

V. NUMERICAL CALCULATION AND DISCUSSION

As an example, we study the hot-electron effect on \(S\) for \(p\)-type Ge. It is assumed that the transport is due to the heavy holes with a parabolic band and the scattering mechanisms include both acoustic and nonpolar optical phonons. All parameters we employ are exactly the same as those listed in Ref. 8. The procedure for calculating \(S\) is to first determine \(v_d\) and \(T_e\) from the force and energy balance equations [Eqs. (10)–(13)], and then to substitute them into Eq. (18) or (19) for obtaining \(S\). The calculated results for \(v_d\) and \(T_e\) as functions of electric field strength \(E\) at several lattice temperatures are shown in Fig. 1, where a comparison between the

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**FIG. 1.** Drift velocity \(v_d\) (solid line) and temperature ratio \(T_e/T\) of holes as functions of the electric field strength at different lattice temperatures [(a) 20 K, (b) 77 K, and (c) 220 K]. The black dots refer to the experimental data on \(p\)-type Ge from Ref. 13 for \(E\) along the (100) direction. All the parameters we employ are listed in Ref. 8.
Theoretical values for \( v_d \) (solid lines) and experimental data (black dots) of Reggiani et al.\(^{16} \) have been carried out. As shown by dashed lines in Fig. 1, the temperature ratio \( T_\text{e}/T \) is larger as the electric field becomes stronger and the lattice temperature becomes lower. Using the solutions for \( v_d \) and \( T_\text{e} \) and taking the density of carriers as \( 1.3 \times 10^{14} \text{ cm}^{-3} \), we have calculated \( S \) from Eq. (19). In Fig. 2, we show the \( T \) dependence of \( S \) at different \( E \), together with its zero-field result (the uppermost curve). With an increase in \( E \), the hot-electron \( S \) varies gradually towards the decreasing direction and becomes negative at higher electric fields. The chemical potential is always negative over the parameter range under consideration. It then follows from Eq. (18) that the sign reversal in \( S \) comes from the sign change of \( (\partial T_\text{e}/\partial T)_E \). The calculated results shown in Fig. 3 indicate that at high fields (e.g., \( E = 5000 \text{ V/cm} \)), the slope \( (\partial T_\text{e}/\partial T)_E \) becomes negative over a large temperature range, where negative \( S \) appears. In order to explain more explicitly why \( (\partial T_\text{e}/\partial T)_E \) becomes negative at high fields, we take a nondegenerate semiconductor with only the electron–phonon interaction as an example. Let us introduce a temperature-dimension parameter \( \theta_\text{e} = \hbar v_\text{e}^2 / m \) with \( v_\text{e} \) the sound speed and \( m \) the effective mass \( (\theta_\text{e} \simeq \theta_\text{e} \approx 1 \text{ K} \) for \( v_\text{e} = 5 \times 10^5 \text{ cm/s} \) and \( m = 0.6m_0 \)), then it is straightforward to show that under the limit \( T_\text{e}/\theta_\text{e} \ll 1 \) and to the lowest order in \( (T_\text{e}/\theta_\text{e})^{-1} \), the following equation can be derived from Eqs. (10)–(13):

\[
N_\text{e}E[v_d/v_\text{e} + 3(v_d/v_\text{e})^{-1}] = 2A(T_\text{e}/\theta_\text{e})^{3/2}, \tag{31}
\]

where \( A \) is a constant term. In the low-field limit: \( v_d/v_\text{e} \ll 1 \) and \( T_\text{e} \) close to \( T \), Eq. (31) reduces to \( N_\text{e}E = (2A/3)(v_d/v_\text{e})(T_\text{e}/\theta_\text{e})^{3/2} \), which is the well-known Ohm’s law with \( v_\text{e} \) varying linearly with \( E \). In this case, \( (\partial T_\text{e}/\partial v_d)_E < 0 \), and further taking into account that \( (\partial v_d/\partial T)_E < 0 \), one finds \( (\partial T_\text{e}/\partial T)_E = (\partial T_\text{e}/\partial v_d)_E(\partial v_d/\partial T)_E > 0 \). The \( (\partial v_d/\partial T)_E < 0 \) stems from the fact that for a constant \( E \), a decrease in \( T \) always leads to the decrease of phonon scattering, and so the drift velocity \( v_d \) increases. In the high-field case: \( v_d/v_\text{e} \gg 1 \), however, the situation is quite different. Equation (31) becomes \( N_\text{e}Ev_d = 2Av_\text{e}(T_\text{e}/\theta_\text{e})^{3/2} \) with \( (\partial T_\text{e}/\partial v_d)_E > 0 \) so that \( (\partial T_\text{e}/\partial T)_E < 0 \).

In summary, we have derived an analytic expression for the thermoelectric power of electrons under a strong electric field by using the NSO method and the balance equation approach. At zero field it reduces to the same form as that derived from the Boltzmann equation approach in the relaxation-time approximation provided that the condition \( \tau_\text{e} \gg \tau_\text{me} \) is well satisfied. The calculated results indicate that the hot-electron effect on \( S \) not only changes its magnitude but also may alter its sign.

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10 In the simplest case, if the hot-electron effect can be ignored ($T_e = T$) and the temperature distribution is uniform ($\nabla T = 0$), we have $S(t, 0) = \Psi + (H_e + H_{ph} - \mu N)/T$ and so $\rho_t$ reduces to the well-known equilibrium density operator of an electron-phonon system at temperature $T$.