# Tailoring interface roughness and superlattice period length in electron-filtering thermoelectric materials

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We quantify the effects of interface roughness and superlattice period on thermoelectric electron filtering in superlattices, using the nonequilibrium Green's function method with a realistic description of the interface. In contrast with previous suggestions, we find that rough interfaces do not enhance the power factor more than smooth ones. Quantitative results are provided in the case of InGaAs/InAlGaAs superlattices.

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## I. INTRODUCTION

The performance of a thermoelectric device is directly related to the dimensionless figures of merit of its constituent materials $ZT=\sigma S^2 T/\kappa$, where $T$ is the temperature, $\sigma$ is the conductivity, $S$ is the Seebeck coefficient, and $\kappa$ is the total thermal conductivity, respectively.[1] Thus, good thermoelectric materials are those with high $\sigma S^2$ (also known as power factor) and low thermal conductivity. Despite decades of efforts, the best commercial thermoelectric materials, such as $Bi_2Te_3$, have $ZT\sim 1$ at room temperature.

With the development of nanostructured materials, different strategies have been theoretically and experimentally investigated in order to develop materials with higher $ZT$s. A rough division can be made between strategies where the main effect is to decrease the thermal conductivity[2–7] and those aiming at increasing the power factor.[8,9] Electron filtering belongs to the second category. A related strategy, thermionic emission, had been proposed earlier.[10–13] However, later on it was realized that the power factor cannot be higher than the bulk value[14–16] because of the decrease in the $B$ factors.[1,15] Shakouri *et al.* then proposed to increase the power factor by using the electron-filtering effect. Emitters and collectors are made of the working materials and barriers are made wide enough to decrease the tunneling probabilities. Therefore, carriers with low energies are blocked and the sharp increase in the transmission function near the barrier height may increase the power factor.[17]

Vashaee and Shakouri[18] showed that enhanced power factors can be achieved by using electron filtering in superlattices (SLs). They also pointed out that the nonconservation of transverse momentum due to rough interfaces may cause a significant increase in power factors by increasing the number of particles contributing to the conduction process.[19] An experiment has shown an enhanced Seebeck coefficient in InGaAs/InAlGaAs superlattices, which has been attributed to electron filtering.[20] However, the effect of nonconservation of transverse momentum on the thermoelectric power factor still lacks a conclusive experimental proof.

The theoretical approach developed in Refs. 18 and 19 is illuminating, but it relied on an earlier argument by Meshkov[21] regarding the electron-tunneling probability across rough interface barriers. The applicability of Meshkov's argument to the case considered (where many of the electron energies are high above the barrier, and thus are not in the tunneling regime) was not rigorously considered. Furthermore, such a model only distinguishes between purely smooth and purely rough (which do not have strict meanings) interfaces. Thus, it cannot quantify the effect of different degrees of roughness on thermoelectric transport. In addition, the dependence of the power factor on the length of the wells, which is essential for optimizing the material, has never been addressed before. A detailed investigation of the electron-filtering effect by a more sophisticated model free of those shortcomings is therefore needed, and this is the aim of the present work.

If electron transport is elastic, the electrical conductivity and Seebeck coefficient can be expressed in the most general form as[22,23]

$$\sigma = \frac{e^2\ell}{h\mathcal{A}}\int dE\mathcal{T}(E)\left.\frac{\partial f}{\partial E}\right|_{\mu_0}, \tag{1}$$

and

$$S = \frac{1}{\sigma}\frac{e^2\ell}{h\mathcal{A}}\int dE\mathcal{T}(E)(E-\mu_0)\left.\frac{\partial f}{\partial E}\right|_{\mu_0}, \tag{2}$$

where $\ell$ and $\mathcal{A}$ are the length and the cross section of the system, respectively, $f(E)$ is the Fermi function, and $\mu_0$ is the chemical potential. $\mathcal{T}(E)$ is the transmission function (not to be confused with the transmission probability presented later), which contains all the information specific to the nanostructured material. The exact transmission function can be computed directly from the Green's function of the system as[24]

$$\mathcal{T}(E) = \mathrm{Tr}\{G^r\Gamma^L G^a \Gamma^R\}, \tag{3}$$

where $G^{r/a}$ is the retarded (advanced) Green's function and $\Gamma^{L/R}=\Sigma^{r,L/R}-\Sigma^{a,L/R}$ with $\Sigma^{r/a,L/R}$ the retarded (advanced) self-energy due to the left (right) lead. It is a simple exercise to verify that Eqs. (1) and (2) reduce to those used in Refs. 18 and 19 if the following approximation to the transmission function is used:

 

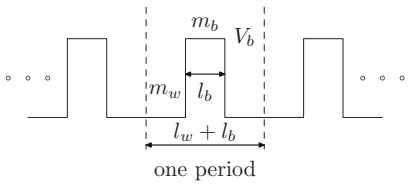


FIG. 1. Schematic diagram of the conduction-band edge in a superlattice. The well and barrier have the effective mass $m_w$ and $m_b$, respectively. The barrier height is $V_b$.

$$\mathcal{T}(E) \simeq \mathcal{A} \int \frac{d\mathbf{k}_\parallel}{(2\pi)^2} \frac{\lambda}{2\ell} T(\mathbf{k}_\parallel, E), \tag{4}$$

where $T(\mathbf{k}_\parallel, E)$ is the transmission probability across the barrier for an electron with total energy $E$ and wave vector parallel to the barrier $\mathbf{k}_\parallel$. Reference 18 used a WKB estimation for $T(\mathbf{k}_\parallel, E)$ in the $\mathbf{k}_\parallel$ conserving case and the Meshkov tunneling argument $T(\mathbf{k}_\parallel, E) \equiv T(E - V_b)$ in the $\mathbf{k}_\parallel$ nonconserving case. The intrinsic mean-free path (mfp) of the well material in terms of the electron mobility $\tilde{\mu}$ is $\lambda = \hbar k_z \tilde{\mu}/e$. This intuitive approximated expression was implicitly used in Refs. 18 and 19 without rigorous justification. By analytical arguments, one can actually show that this is a reasonable approximation if the partial transmission probabilities are close to 1. Equation (4) is, however, not valid in other cases. Rather than trying approximations of this sort, in the present paper we employ accurate transmission functions computed fully from the Green's function. Thus, our results do not rely on the validity or not of the Meshkov approximation. Furthermore, by using a realistic model of interface roughness, we are able to quantify its effect in terms of physical parameters. Finally, we also discuss inelastic electron-scattering effects, using the Keldysh formalism.[24–29]

## II. SYSTEM AND METHOD

The system that we consider is a SL connected to two perfect leads made of the well materials. The SL is described by a tight-binding model in the real space in the direction perpendicular to the interfaces and in the momentum space for the other two directions. The Hamiltonian of one period (cf. Fig. 1) is

$$H = H_w + H_b + H_I, \tag{5}$$

where the Hamiltonians $H_w$ and $H_b$ are those of the well and barrier, respectively,

$$H_{w/b} = \sum_{i,\mathbf{k}_\parallel} (\epsilon_{w/b} + E_{\mathbf{k}_\parallel, m_{w/b}}) c^\dagger_{i,\mathbf{k}_\parallel} c_{i,\mathbf{k}_\parallel} + \sum_{\langle i,j\rangle,\mathbf{k}_\parallel} t_{w/b} c^\dagger_{i,\mathbf{k}_\parallel} c_{j,\mathbf{k}_\parallel} + \text{H.c.}, \tag{6}$$

where the site index $i = -n_w/2, \ldots, 0$ and $i = n_b + 1, \ldots, n_b + n_w/2$ for well and $i = 1, \ldots, n_b$ for barrier with the site number in a well (barrier) $n_{w/b}$. Operators $c^\dagger_{i,\mathbf{k}_\parallel}/c_{i,\mathbf{k}_\parallel}$ are the creation (annihilation) operators. The on-site energy is $\epsilon_{w/b}$. The transverse energy is $E_{\mathbf{k}_\parallel, m_{w/b}} = \hbar^2 k_\parallel^2/2m_{w/b}$ with the effective mass $m_{w/b}$ and the transverse momentum $k_\parallel$. The hopping energy $t_{w/b}$ is related to the effective mass as $t_{w/b} = -\hbar/2a_0^2 m_{w/b}$ with the site distance $a_0$. The well and barrier lengths are $l_w = n_w a_0$ and $l_b = n_b a_0$, respectively. The barrier height is $V_b = \epsilon_b - \epsilon_w - 2(|t_b| - |t_w|)$. Symbol $\langle i, j\rangle$ means to sum over the nearest-neighbor sites. The Hamiltonian of the interface is

$$H_I = \sum_{i=0,n_b} t_I c^\dagger_{i,\mathbf{k}_\parallel} c_{i+1,\mathbf{k}_\parallel} + \sum_{i,j=0,n_b} \sum_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel} t^r_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel,i,j} c^\dagger_{i,\mathbf{k}_\parallel} c_{j,\mathbf{k}'_\parallel} + \text{H.c.}, \tag{7}$$

where $t_I$ is the hopping energy due to the smooth interface and is assumed to be $(t_w + t_b)/2$. The rough interface causes extra hopping energies $t^r_{\mathbf{k}_\parallel,\mathbf{k}'_\parallel,i,j}$, which mix the states with different transverse momenta.[30] We assume that the extra hopping energy due to the rough interface has a Gaussian form,[30]

$$t^r_{\mathbf{k},\mathbf{k}',i,j} = t_r e^{-|\mathbf{k} - \mathbf{k}'|^2 \xi^2/8},$$

where $\xi$ is the correlation length of the fluctuation in the rough interface and $t_r$ is related to both the variance of the fluctuation and $\xi$.

Scattering processes other than the interface scattering are efficiently included by using the Büttiker probe method.[24–29] In our calculations, a common Büttiker probe is attached to all sites with the same site indices but with different transverse momenta. This induces transitions between states with the same energy and different total momenta, which take place when scattering is due to impurities, disorder, or acoustic phonons. To mimic the inelastic-scattering process, we use a probe with the same structure as the perfect lead and use the parameter $U_b$, the coupling between the probe and the sites, to characterize the scattering strength.[26,28,29] The electric and heat currents and in turn the conductivity and the Seebeck coefficient are obtained by the nonequilibrium Green's function method.[24,31] For the elastic-scattering process, we assume that the self-energy due to a probe is a diagonal matrix with only imaginary elements $\Sigma^r_{ij} = -i\gamma_0 \delta_{ij}$, where $\delta_{ij}$ is the Kronecker delta function[24,32] and $\gamma_0$ is a parameter characterizing the elastic-scattering strength and related to the relaxation time by $\tau \sim \hbar/2\gamma_0$.

Computational requirements would in principle impose stringent limits on the maximum system size. However, it is possible to compute accurate transmission functions by employing a piecewise method based on the assumption of incoherence between scatterers.[24,33,34] We numerically verified that the approach is accurate by explicitly computing the exact transmission through three periods and comparing with the incoherent one. In all cases of interest, differences between the two transmissions were smaller than 3%. This permits us to investigate considerably long well SLs made of real materials, where the electron mean-free paths can reach hundreds of nanometers. In the case of inelastic scattering, we are unaware of any such piecewise approach, however. Thus for the inelastic scattering, one is limited to small wells with rather short mean-free paths, and the results are only qualitative.

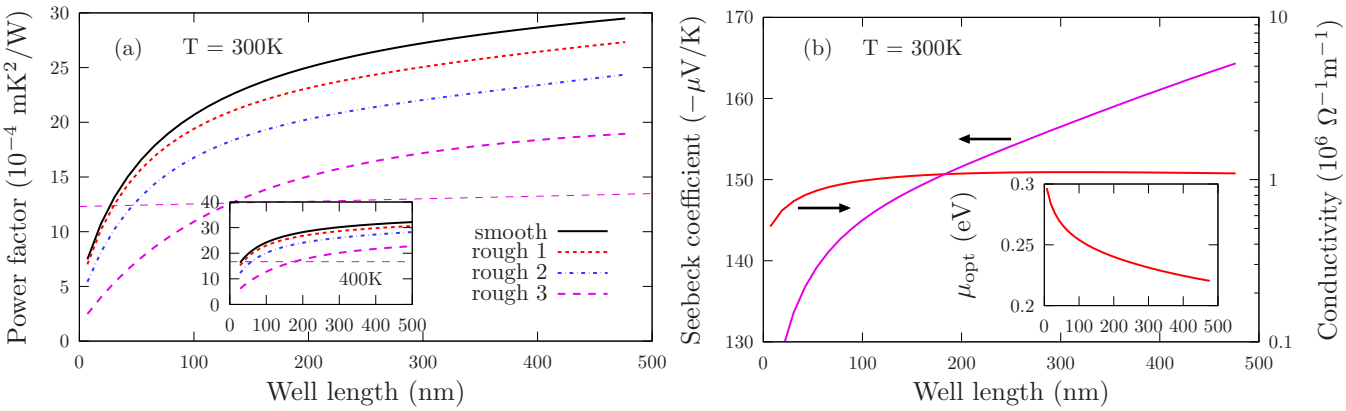


FIG. 2. (Color online) (a) Optimal power factors of superlattices of different well lengths with smooth and rough interfaces at 300 and 400 K (inset). The dashed line indicates the power factor of bulk InGaAs. (b) Optimal Seebeck coefficients and conductivities of different well lengths with smooth interface at 300 K. Inset: optimal chemical potentials with respect to the InGaAs band edge. Only the elastic-scattering process is included. The parameters of roughness are $t_r/t_I$=0.1(1), 0.2(2), and 0.5(3). The correlation length is $\xi$=0.5 nm. Other parameters are given in the text.

### III. RESULTS AND DISCUSSION

We calculate the optimal power factors due to the electron filtering in InGaAs/InAlGaAs SLs. The effective masses are $m_w$=0.043$m_e$ and $m_b$=0.058$m_e$ with electron mass $m_e$.[19] The grid length in the $z$ direction is $a_0$=1.17 nm and the barrier height is $V_b$=0.2 eV. The on-site energies are $\epsilon_w$ =−3.846 eV and $\epsilon_b$=−3.997 eV. The hopping energies are $t_w$=−0.64 eV and $t_b$=−0.48 eV. We use 400 states in the $\mathbf{k}_\parallel$ direction with the minimum difference of the wave vectors $6\times10^7$ m$^{-1}$.

We have to know the scattering strengths in order to determine the parameter of the elastic Büttiker probe. There are different types of elastic scatterings in semiconductors such as acoustic-phonon deformation-potential scattering, piezoelectric scattering, impurity and alloy scattering, and so on. The relaxation times due to different scatterings, in general, have different energy dependences.[35] To simplify the calculations, we do not include explicitly the different types of scatterings but use an energy-independent mean-free path to characterize the elastic scatterings.[36] Because of the constant mean-free-path assumption, the parameter of the elastic Büttiker probe $\gamma_0(E)=\gamma_0'\sqrt{E}$. The factor $\gamma_0'$ is determined by fitting the calculated power factors of the well and barrier bulks to the values obtained in Refs. 19 and 20. The fitted parameter of the Büttiker probe is $\gamma_0(E)=0.004\sqrt{E/e}$ eV in both the wells and the barriers with $E$ measured from the well and barrier conduction-band edges, respectively.

When the elastic-scattering process is included, the power factor with fixed well length monotonically increases as the period number $N$ increases and is saturated at large $N$. We take this saturated value as the power factor of a SL. The calculated power factors of SLs of different well lengths with barrier length $l_b$=9.4 nm at 300 K and 400 K are shown in Fig. 2(a) and the inset, respectively, for the case when only the elastic scattering is present. The straight dashed line indicates the calculated power factor of bulk InGaAs, which corresponds to $ZT\approx0.067$ with thermal conductivity $\kappa$ =5.5 W/mK.[19] Electron filtering increases the power factors in SLs with both smooth and rough interfaces. However, rough interfaces are always worse than the perfect ones. Optimal Seebeck coefficient and conductivity as functions of the well length are shown in Fig. 2(b) while the optimal chemical potentials of different well lengths are shown in the inset.

These results are in striking contrast with the conclusions of Refs. 18 and 19. The current flow is limited by scattering in the well. The barrier only dominates the flow rate when its transmission is smaller than the one across the well, i.e., when it is basically zero. Electrons in the well always have some probability of scattering into the lower $\mathbf{k}_\parallel$ states, which can traverse the barrier. The net effect is that electrons are filtered off below the barrier and allowed to pass above it, even in the smooth barrier case. Although roughness enhances transmission probabilities for the larger $\mathbf{k}_\parallel$ states, it actually reduces them for the low $\mathbf{k}_\parallel$ states. The resulting balance favors the smooth surface over the rough one, since the momentum mixing is already taking place before reaching the barrier. Despite these qualitative arguments, it would have been quite difficult to guess the actual behavior of the system without a calculation such as the one presented here.

Figure 3 shows the calculated power factors when only the inelastic scattering is present. As mentioned before, in this case the calculation is limited to smaller systems. We calculate the power factor of a single well/barrier/well system. As we can only give qualitative results for SLs with inelastic scatterings due to computing power limitations, we do not choose a Büttiker probe describing the inelastic polar-phonon scattering in semiconductors. Instead, we use the "auxiliary probe" approach[24,28,29] for the inelastic Büttiker probe as stated in Sec. II. The parameters used for the inelastic Büttiker probes are $U_b$=0.69$t_w$ in wells and $U_b$=0.069$t_w$ in barriers. In this inelastic case, electron filtering does not increase the power factor. However, the inelastic mfps employed are unrealistically short in this case (~5 nm), and it is difficult to draw quantitative conclusions. A proper account of inelastic scattering should be done in conjunction with the elastic mechanisms rigorously studied in the previous paragraphs. In the specific case of InGaAs, the inelastic mfp has been roughly estimated in the order of 1 $\mu$m by

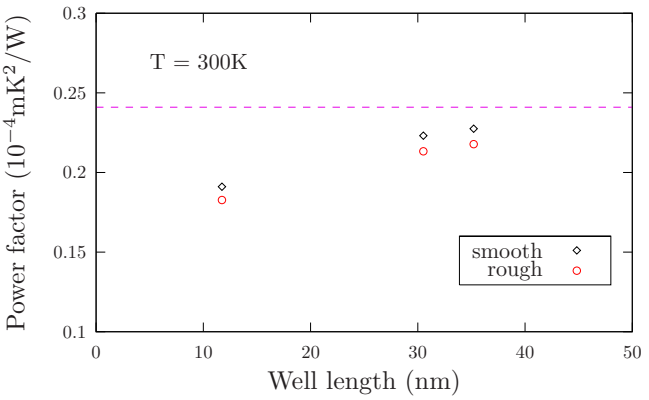


FIG. 3. (Color online) Optimal power factors of one period SLs of different well lengths with smooth and rough interfaces. The dashed line indicates the bulk power factor. Only the inelastic scattering is present. The parameter of roughness is $t_r/t_I=0.1$ and the correlation length is $\xi=0.5$ nm. Other parameters are given in the text.

comparing the different mobility contributions for different types of processes.[37,38] This means that the electron filtering in InGaAs/InAlGaAs should be effective for well lengths up to hundreds of nanometers with possible power factor enhancements of about 2.2.

## IV. CONCLUSIONS

In conclusion, by using the nonequilibrium Green's function method with a realistic description of the interfaces, we have calculated the power factor of electron-filtering superlattices, as functions of the degree of interface roughness and the well length. If the scattering is elastic, the electron filtering increases the power factors above the bulk value saturating toward an asymptotic value for long wells. The obtained power factors are larger in SLs with smooth interfaces than in SLs with rough ones, contrary to earlier suggestions. In the strongly inelastic-scattering limit, the electron filtering does not increase the power factor when the well length is longer than the inelastic mean-free path.

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