Phonon transport in atomic chains coupled by thermal contacts: The role of buffer layer

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In this work, ballistic phonon transport in atomic chain nanostructures is investigated by atomic nonequilibrium Green’s functions and embedded atom method. Bond length and strength modification in atomic chain (low-dimensional structure) was taken into consideration by using bond-order-length-strength correlation premise. We especially focus on the contact interface effects on phonon transmission and thermal conductance. It is found that the contact interfaces between an atomic chain and contact reservoir, i.e., neck region or buffer layers, play an important role in phonon transport. The more buffer layers the less thermal conductance. © 2010 American Institute of Physics. [doi:10.1063/1.3359708]

I. INTRODUCTION

Much attention has been focused on thermal properties of nanosystems in recent years1–3 due to their great importance in fundamental low-dimensional physics as well as in application of nanoscale devices. Thermal management in nanoscaled device and development of thermoelectric device urge a complete understanding of thermal properties in low-dimensional structure. The development of nonequilibrium atomic Green’s functions4–8 provides a systematic way to study phonon transport in the case that the mean free path of phonon is comparable to or greater than the characteristic dimensions of the systems under consideration. The advantage of the nonequilibrium atomic Green’s functions approach is that the regime of short phonon wavelength can be accurately treated, while the continuum model fails. Besides that, the nonequilibrium atomic Green’s functions could effectively handle important features of phonon transport, such as phonon density of state (DOS), transmission probability, thermal conductance, etc.

In a system, which is a nanostructure coupled by two thermal contacts at both sides, the surface bond reconstruction (SBR) of thermal contact affects phonon transport in nanoscale system significantly. The reason is that around the surface of thermal contact (normally the first few layers), bond strengths and interatomic distances are substantially different from the bulk case that leads to significant change of interatomic force at the surface of thermal contact. Also due to this change, surface phonon DOS is significantly modified causing dramatic change in thermal conductance. The effect of SBR is analyzed in details in our previous paper.9,10

One more important factor that affects phonon transport is the nonlinear interaction between atoms in atomic wire. Although there are lots of studies on phonon transport, most of them are considered ballistic phonon transport that leads to serious inaccuracy in phonon transport properties characterization and thermal conductance calculation. Nonlinear effect causes phonon scattering during the ballistic transport of phonons and hence affects thermal conductance in atomic wire systems.11

Recently a considerable effort has been devoted to the structural and transport properties of metallic nanowires and atomic chains.12–19 In practice, the atomic chains are connected to thermal contacts through a buffer layer instead of direction connection. This problem has not been addressed in any previous paper. So it is interesting to know about the fundamental thermal properties induced by phonons in such system. From theoretical point of view it is important to establish a realistic model to calculate thermal conductance of an atomic chain, and to provide accurate information on phonon transport. In this paper, we adopt nonequilibrium atomic Green’s functions by considering ballistic phonon transport to study the thermal transport properties in a system with the presence of buffer layers. The theoretical model is elaborated in Sec. II; interesting results from investigating the effect of contact interface (or neck region) are discussed in Sec. III, in the end, a short and important conclusion is drawn.

II. THEORY

Our modeled system formed by gold atoms is shown in Fig. 1, where the atomic chain is connecting semi-infinite thermal contacts through neck region (also known as buffer layers) at both sides.20 Gold atoms are in fcc structure inside the semi-infinite contacts, and the contact surface is set to (001) plane. The neck region is assumed in pyramidal form, thus a neck region with N buffer layers is constructed by (N+1)2 atoms. The notations used in our modeled system are shown in Fig. 2. LB (RB) represents thermal contact on left
mined by equations,

\[ K_{ij} = -Q_{ij} - F_i^p \hat{g}_{ij} \rho_j^p - F_j^p \hat{g}_{ij} \rho_i^p + \sum_{k \neq i,j} F_k^p \rho_j^p \rho_i^p \hat{r}_{ik}. \]

The quantities in the above expression are given below,

\[ Q_{ij} = \psi_{ij}^a \hat{r}_{ij} + \psi_{ij}^b (\hat{1} - \hat{r}_{ij} \hat{r}_{ij}) / r_{ij}, \quad \hat{g}_i = \sum_{j \neq i} \rho_j^p (r_{ij}) \hat{r}_{ij}, \]

where \( \hat{1} \) is unit tensor, \( \hat{r}_{ij} \) and \( r_{ij} \) are the unit vector and distance from atom \( i \) to atom \( j \), and \( \psi_{ij}^a \) and \( \psi_{ij}^b \) are determined by equations,

\[ \psi_{ij}^a = F_i^p(\rho_i) \rho_j^p + F_j^p(\rho_j) \rho_i^p + \phi_{ij}^a, \]

\[ \psi_{ij}^b = F_i^p(\rho_i) \rho_j^p + F_j^p(\rho_j) \rho_i^p + \phi_{ij}^b. \]

The embedded atomic function \( F_i(\rho) \) \( \text{Eq. 23} \)

\[ F_i(\rho) = -F_0 \left[ 1 - n \ln \left( \frac{\rho_i}{\rho_c} \right) \right] \left( \frac{\rho_i}{\rho_c} \right)^a + F_1 \left( \frac{\rho_i}{\rho_c} \right), \]

where \( \phi_{ij} = -\phi_{ij}^a \left[ 1 + \beta (r_{ij}/r_{ij} - 1) \right] e^{-\beta (r_{ij}/r_{ij} - 1)} \) (Rose-like pairwise interaction potential) \( \text{Eq. 23-26} \).

Cases of different number of atomic layers \( N \) in the buffer layers, view from [010] direction.

<table>
<thead>
<tr>
<th>TABLE I. Parameters of gold (Ref. 23).</th>
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<tr>
<td>( a_0 ) (Å)</td>
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<td>( r_s ) (Å)</td>
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<tr>
<td>( r_a ) (Å)</td>
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<tr>
<td>( F_0 ) (eV)</td>
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<td>( F_1 ) (eV)</td>
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<td>( \phi_0 ) (eV)</td>
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<td>( n )</td>
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<tr>
<td>( m \times 10^{-25} ) (kg)</td>
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<td>( \omega_0 ) (THz)</td>
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EAM is a very good potential to describe bulk metal, but it is not accurate for low-dimensional systems such as atomic chain. This problem has been pointed by Peláez and Serena.\textsuperscript{12} To overcome this problem, in this work, we adopt bond-order-length-strength (BOLS) correlation premise\textsuperscript{28,29} to obtain an averaged estimation of the equilibrium bond length $r_0$, bond energy $E_i$, and cohesive energy $E_{Bi}$ per atom in the atomic chain. According to BOLS, the bond contraction coefficient $c_i(z_i)$ is given by

$$c_i(z_i) = \frac{2}{1 + e^{(12-z_i)/\delta z_i}},$$

where $z_i$ is the effective atomic coordination number, and $z_i=2$ for atomic chain. The bond energy is determined by $E_i = c_i^2 E_0$, and cohesive energy is given by $E_{Bi} = z_i E_i$. The obtained parameters are then substituted into the Rose’s universal binding energy.\textsuperscript{26} So the interatomic potential of atomic chain is expressed as

$$E(r_{ij}) = -E_{Bi} \left[ 1 + \alpha \left( \frac{r_{ij}}{r_0} - 1 \right) \right] e^{-\alpha(r_{ij}/r_0)^{-1}},$$

where $E_{Bi} = 1.4628$ (eV) and $\alpha = 2.5537$ (Values are calculated based on parameters in Ref. 16). Hence, the dynamic matrix of atomic chain is obtained.

The retarded Green’s function of the atomic chain is

$$D_{ij} = \left[ \omega^2 - H_{DD} - \Sigma_L^I - \Sigma_R^I \right]^{-1},$$

where $H_{DD}$ is the dynamic matrix of the atomic chain and $\Sigma_L^I$ is the self-energy due to the coupling between the atomic chain and the LD (RD) region which has only one atom. The effect of buffer layers is involved in the self-energy $\Sigma'_L$, and they are obtained by

$$\Sigma'_L = H_{D,LD}(\omega^2 - H_{DD,LD} - \Sigma'_L)^{-1} H_{LD,DD},$$

where $\Sigma'_L$ is the self-energy due to the coupling between $LD$ (RD) region and the first buffer layer in the neck region,

$$\Sigma'_R = H_{D,RD}(\omega^2 - H_{RD,RD} - \Sigma'_R)^{-1} H_{RD,DD}.$$}

In Eq. (7), $H_{LD,C_1}$ is the coupling between buffer layer (denoted $C_1$) and the atom in LD; $W_{C_1}^{-1}$ is the Green’s function of the buffer layer $C_1$, in which the coupling between the buffer layer and the left contact is taken into account, as shown,

$$\begin{bmatrix}
\omega^2 - H_{C_1,C_1} - \Sigma_{0L_1} & -(H_{C_1,C_1}\tau_1 + \tau_{12}) & -(H_{C_1,C_1}\tau_1 + \tau_{13}) & -(H_{C_1,C_1}\tau_1 + \tau_{14}) \\
 -(H_{C_1,C_1}\tau_1 + \tau_{12}) & \omega^2 - H_{C_1,C_2} - \Sigma_{0L_2} & -(H_{C_1,C_2}\tau_2 + \tau_{23}) & -(H_{C_1,C_2}\tau_2 + \tau_{24}) \\
 -(H_{C_1,C_1}\tau_1 + \tau_{13}) & -(H_{C_1,C_2}\tau_2 + \tau_{23}) & \omega^2 - H_{C_1,C_3} - \Sigma_{0L_3} & -(H_{C_1,C_3}\tau_3 + \tau_{34}) \\
 -(H_{C_1,C_1}\tau_1 + \tau_{14}) & -(H_{C_1,C_2}\tau_2 + \tau_{24}) & -(H_{C_1,C_3}\tau_3 + \tau_{34}) & \omega^2 - H_{C_1,C_4} - \Sigma_{0L_4}
\end{bmatrix}.$$

With nonequilibrium Green’s function, the heat current due to phonon transport was determined,\textsuperscript{30}

$$J_{LR} = \int \frac{d\omega}{2\pi} \frac{\hbar}{\omega} \left[ \xi(\omega) \right] f_L(T_L) - f_R(T_R) d\omega/2,$$

where $f_L(T_L)$ and $f_R(T_R)$ are the Bose–Einstein distribution functions in the left and right contacts with the temperature $T_L$ and $T_R$. The transmission function $\xi(\omega)$ is given by $\xi(\omega) = \text{Tr}(\Gamma L D_0^{\omega} \Gamma R D_0^{\omega})$, where $D_0^{\omega}$ is retarded (advanced) Green’s function of the atomic chain, and $\Gamma_{LR}$ is the broad functions due to couplings between the atomic chain and the left (right) contact. The thermal conductance $\kappa$ is calculated according to the definition $\kappa = J/\Delta T$, where $\Delta T$ represents a small difference in temperature across the atomic chain, so

$$\kappa = \int_0^\infty \frac{d\omega}{2\pi} \frac{\hbar}{\omega} \left[ \frac{\partial f(\omega,T)}{\partial T} \right] \xi(\omega).$$

In our calculation, all physical quantities were treated dimensionless, so we paid more attentions to the relative changes of physical quantities rather than their absolute values.

### III. RESULTS AND DISCUSSIONS

Based on the theory mentioned above, phonon transport in atomic chains with different numbers of buffer layers $N$ in the neck region is evaluated. In this section, the results such as DOS, the transmission, and thermal conductance are discussed.

DOS, affecting thermal conductance through self-energy and the transmission function, is a very important physical quantity in phonon transport in the atomic chain. Figure 4(a)

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**FIG. 3.** (a) The direct lattice unit cell in the contact surface and (b) the first Brillouin zone. $a_0$ is the lattice constant.
is showing the bulk DOS of the gold contacts, which is in good agreement with the well-known experimental result and theoretical predictions. In Fig. 4(b), the solid line is the partial surface DOS \( g_{zz}(\omega) \) for polarization in \( z \) direction, it is obtained by evaluating the Green’s function, \( g_{zz}(\omega) = -\text{Im} \frac{1}{\kappa} \) \( D_{zz}(\omega) \). The dashed line is the surface DOS \( g_{yy}(\omega) \) for polarization in \( y \) direction, which is parallel to the contact surface. It is obvious that the shape of \( g_{zz}(\omega) \) is different from the bulk DOS, while \( g_{xx}(\omega) = g_{yy}(\omega) \) appears in a similar shape with bulk DOS. The DOS of atomic chain [Figs. 5(a)–5(d)] is modified dramatically in the presence of the contacts coupled to the atomic chain, but cutoff frequency appeared in all cases, beyond which phonons do not contribute to thermal conductance.  

Figure 6 shows thermal conductances of the system with different numbers of buffer layers \( N = 0, 1, 2, 3 \) as shown in Fig. 2. The temperature dependence of thermal conductance in nanowire is clearly shown by these curves. At low temperature, thermal conductance shows the linear behavior (proportional to temperature, see the inset of Fig. 6), which is consistent with the theoretical prediction and experimental confirmation of the quantized thermal conductance \( \pi k_B T / 3h \). The continuous variation of \( \kappa \) with \( T \) rather than steplike behavior in charge transport reflects the entropy transport in thermal current. When temperature is sufficiently high, the thermal conductance increases rapidly showing the Debye behavior. This is because more degrees of freedom are excited in the system. At even higher temperature, the thermal conductance approaches a saturation value, as all the excited states are occupied.  

From Fig. 6, we notice that with the increasing number of buffer layer, the thermal conductance is decreasing. This can be explained from the physical mechanism of phonon transport. As the contact reservoir injects phonons into atomic chain, according to the transmission function \( \zeta(\omega) \), a phonon with the frequency of \( \omega \) can transmit through the atomic chain whenever a channel mode of that frequency is available. For \( N = 0 \) (no buffer layer), DOS at the tip of the neck region (i.e., the atom in LD region) consists of a broad range of frequencies, as shown in Fig. 5(a). This is due to weak reflections for the incident phonons by the neck region. In this case the surface DOS covers all the peak frequencies in the DOS of atomic chain. Hence phonons are injected into all modes in the atomic chain, leading to high transmission probability [Fig. 7(a)] and large thermal conductance (Fig. 6). When \( N \) increases from zero, DOS at the tip of the neck region is reduced. As shown in Figs. 5(b)–5(d), the DOS has narrower frequency range and more sharp peaks, it is because incident phonons experienced stronger reflection in the pyramidal neck region. In these cases, DOS does not cover all the peak frequencies in the DOS of the atomic chain. Hence the number of phonons injected into atomic chain is much reduced leading to low transmission probability [Figs. 7(b)–7(d)] and small thermal conductance (Fig. 6).  

It is easy to understand why the incident phonons suffer stronger reflection in the neck region when \( N \) increases. When the number of buffer layers \( N \) increases (the physical size of the pyramidal neck region becomes larger), the incident phonons need to propagate through a longer distance before arriving at the atomic chain. Hence these phonons suffer stronger scattering in the neck region and reflection by the boundary of the neck region. Thus the transmission func-

FIG. 4. (Color online) (a) Bulk DOS of contact, (b) surface DOSs. Dashed line is DOS of \( g_{zz}(\omega) \) or \( g_{yy}(\omega) \) parallel to the contact surface. Solid line is DOS \( g_{zz}(\omega) \) perpendicular to the contact surface.

FIG. 5. (Color online) The solid lines are DOS of finite atomic chains (seven atoms) for structures (a)–(d) in Fig. 2. The dashed line in plot A is DOS of the infinite atomic chain.

FIG. 6. (Color online) Thermal conductances of different junction configurations. The numbers indicate the structures corresponding to Fig. 2. Inset is the thermal conductance as a function of buffer layer number \( N \) at Debye temperature and low temperature thermal conductance, respectively.
tion is reduced to finite number of narrow and sharp resonant peaks that lead to a weak thermal conductance. While $N$ continues to increase, at certain sufficiently large, thermal conductance, it is expected to approach a small value. Also the largest thermal conductance is when $N=0$. In this case the atomic chain directly connects to four surface atoms of the contact. In our calculation, we found that the thermal conductance is sensitive to the bonding in the buffer layers as well.

In the last, we investigated phonon transport for all points in the first Brillouin zone, which is equivalent to all individual phonon incident directions. In Fig. 3, the $\Gamma$ point ($\vec{k}_i=0$) corresponds to the phonon incident direction which is perpendicular to the contact surface. These phonons experienced less reflection (by the boundary of the neck region) and contribute more to thermal conductance, while phonons in other incident directions (represented by points of $\vec{k}_i \neq 0$ in the Brillouin zone) undergo strong reflection and contribute less to thermal conductance.

IV. CONCLUSION

In summary, we use the atomic nonequilibrium Green’s functions together with EAM and BOLS to investigate phonon transport properties in atomic chain with different numbers of buffer layers in neck region. The results show that longer neck region in pyramidal form has lower thermal conductance. The reason is phonon reflection and scattering are enhanced by the increasing size of buffer layer.

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