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Image: Ornamental multiplication of space-time figures of temperature transformation rules (adapted from T. S. Bíró and P. Ván 2010 EPL 89 30001; artistic impression by Frédérique Swist).
Nonlinearity enhanced interfacial thermal conductance and rectification

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Abstract – We study the nonlinear interfacial thermal transport across atomic junctions by the quantum self-consistent mean-field (QSCMF) theory based on the nonequilibrium Green’s function approach; the QSCMF theory we propose is very precise and matches well with the exact results from quantum master equation. The nonlinearity at the interface is studied by effective temperature-dependent interfacial coupling calculated from the QSCMF theory. We find that nonlinearity can provide an extra channel for phonon transport in addition to the phonon scattering which usually blocks heat transfer. For weak linearly coupled interface, the nonlinearity can enhance the interfacial thermal transport; with increasing nonlinearity or temperature, the thermal conductance shows nonmonotonical behavior. The interfacial nonlinearity also induces thermal rectification, which depends on the mismatch of the two leads and also the interfacial linear coupling.

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Introduction. – In modern electronics, due to the rapidly increasing power density, accumulation of heat becomes an obstacle for further progress of microelectronic devices; thus the heat dissipation and manipulation have been recognized to be a crucial issue in information and energy technologies [1]. Especially, as the dimensions of materials shrink into the nanoscale, interfaces dramatically affect thermal transport [2–5] making it a lucrative field to explore. At a rough interface the atomic mixing can enhance the thermal transport [6]; however, the role of nonlinearity at the interface is not clear, and it is also unknown whether nonlinearity can enhance the phonon transport. Recent progress in functional thermal devices [7–10] makes the emerging new field—phononics—very attractive [11]. One of the most fundamental property of phononic devices is thermal rectification, which is known to be realized by combining inherent anharmonicity of the system with structural asymmetry [12,13]. Whether the interface itself can induce thermal rectification is still an open question; if yes, the property of the interfacial rectification is quite interesting and helpful for both theorists and experimentalists.

To investigate thermal transport across an interface, the most widely applied models are the acoustic mismatch model [14] and the diffuse mismatch model [15]. Both models offer limited accuracy in nanoscale interfacial resistance predictions [16,17] because they make simple assumptions and neglect atomic details of actual interfaces. Classical molecular-dynamics simulation is another widely used method in phonon transport and has been applied to interfacial thermal transport [18–22]; however, due to its classical nature, it is not accurate below the Debye temperature and cannot capture the quantum effects. To study nonlinear (anharmonic) thermal transport, the effective phonon theory has been recently introduced in some dynamical models [23–25]; and the quantum correction one [26] can be used to study low-temperature thermal transport. Despite their successes, such theories
cannot be well applied to nonlinear interfacial transport due to the inherent weak system-bath coupling assumption required for the validity of the Feynman-Jensen inequality [27]. Another approach, the nonequilibrium Green’s function (NEGF) method which originates from the study of electronic transport [28], has been applied to study the quantum phonon transport [29–31], the phonon Hall effect [32] and the topological magnon insulator [33].

In this paper, based on the nonequilibrium Green’s function method, to avoid the perturbation approximation, we develop the QSCMF theory for nonlinear thermal transport, which can be applied to an interface with arbitrary nonlinear strength. Then we study the interfacial thermal transport for a model as shown in fig. 1(a): thermal conductance and rectification across the interface are studied with an effective temperature-dependent–harmonic interfacial coupling calculated from the QSCMF theory.

**Model and Hamiltonian.** We study interfacial thermal transport with nonlinear coupling at the solid-solid interface as shown in fig. 1(a). To manifest the effect of the interface we exclude the nonlinear phonon transport in the two materials, and there is neither disorder nor defect in the whole system, thus the only thermal resistance comes from the interface. Such a model really uncovers the thermal transport properties of the interface. To study the longitudinal transport, that is, the cross-plane interfacial transport, we simplify the problem further to a one-dimensional interface as shown in fig. 1(b), where two linear semi-infinite atomic chains (solid-line regimes) are connected by the interfacial linear coupling $k_{12}$ and nonlinear interaction $\lambda$. Our model is similar to the one-dimensional interfacial model in refs. [3,5] where there is only a linear coupling at the interface. The Hamiltonian of the total system is

$$H = H_1 + H_2 + \frac{1}{2} k_{12}(x_{1,1} - x_{2,1})^2 + \frac{1}{4} \lambda (x_{1,1} - x_{2,1})^4,$$  

(1)

with

$$H_\alpha = \sum_{i=1}^{N_\alpha} \frac{1}{2} m_\alpha \dot{x}_{\alpha,i}^2 + \sum_{i=1}^{N_\alpha-1} \frac{1}{2} k_\alpha (x_{\alpha,i} - x_{\alpha,i+1})^2,$$  

(2)

where $N_\alpha \to \infty$, $\alpha = 1, 2$. In our model, the scattering for phonons only comes from the interface while the phonon transport in the two semi-infinite leads is ballistic. Thus, we can partition the system into three parts $(L, C, R)$, where the atoms at the interface are regarded as center (dashed-line part) and the leads $L$ and $R$ (dash-dotted-line regimes) are harmonic as shown in fig. 1(b). Using a mass-normalized displacement $u_\alpha = \sqrt{m_\alpha} x_{\alpha,1}$, the center Hamiltonian can be written as

$$H_C = \frac{1}{2} U_c^T U_c + \frac{1}{2} U_c^T K^C U_c + \frac{1}{4} \sum_{i,j,k,l=1}^2 T_{ijkl} u_i u_j u_k u_l,$$  

(3)

where $U_c = (u_1, u_2)^T$, $T_{ijkl} = (-1)^{i+j+k+l} \lambda$ and

$$K^C = \left( \begin{array}{cc} \frac{k_1 + k_2}{m_1} & -\frac{1}{\sqrt{m_1 m_2}} \\ -\frac{1}{\sqrt{m_1 m_2}} & \frac{k_1 + k_2}{m_2} \end{array} \right).$$  

(4)

For the leads, the Hamiltonian is written as $H_\alpha = \frac{1}{2} U_\alpha^T U_\alpha + \frac{1}{2} U_\alpha^T K^\alpha U_\alpha$ with its coupling to the center $H_{\alpha C} = U_\alpha^T V_{\alpha C} U_C$, $\alpha = L, R$. Here the center-lead coupling is the same as the inter-atomic spring constant in the corresponding bath.

**Quantum self-consistent mean-field theory.** We discuss the QSCMF based on the NEGF method for a general system where the center Hamiltonian has a fourth-order nonlinear interaction as given in eq. (3). The equation of motion of Green’s function [31], without the nonlinearity, is $(\frac{\partial^2}{\partial \tau^2} + K^C)G_0(\tau, \tau') = -i \delta(\tau - \tau') - \int d\tau'' \Sigma(\tau, \tau'')G_0(\tau'', \tau')$, where $\Sigma(\tau, \tau'')$ is the self-energy due to the center-lead coupling. With the nonlinearity, the full Green’s function has the equation of motion as

$$\frac{\partial^2}{\partial \tau^2} G_{im}(\tau, \tau') + \sum_j K_{ij}^C G_{jm}(\tau, \tau')$$

$$+ \sum_{jkl} T_{ijkl} G_{jklm}(\tau, \tau, \tau', \tau''),$$  

(5)

where $G_{jklm}(\tau_j, \tau_k, \tau_l, \tau_m)$ is the four-point Green’s function. Under the self-consistent mean-field approximation, the four-point Green’s function can be represented by the two-point Green’s function, and
we have \(-\frac{i}{\hbar}G(\tau_j, \tau_k, \tau_l, \tau_m) = G(\tau_j, \tau_k)G(\tau_l, \tau_m) + G(\tau_j, \tau_l)G(\tau_k, \tau_m)\). Thus, the full Green's function satisfies

\[
\begin{align*}
\frac{\partial^2}{\partial \tau^2}G_{im}(\tau, \tau') + \sum_j K_{ij}^C G_{jm}(\tau, \tau') \\
+ 3i\hbar \sum_{jkl} T_{ijkl} G_{kl}(0) G_{jm}(\tau, \tau') \\
+ \sum_j \int d\tau'' \Sigma_{ij}(\tau, \tau'') G_{jm}(\tau'', \tau') = \\
- \delta(\tau - \tau') \delta_{im},
\end{align*} 
\]

(6)

using the symmetry of \(T_{ijkl}\) with respect to the permutation of the indices. Thus, we introduce an effective dynamic matrix

\[
K_{ij}^C = K_{ij}^\alpha + 3i\hbar \sum_{kl} T_{ijkl} G_{kl}(0) = K_{ij}^\alpha + 3 \sum_{kl} T_{ijkl} \langle u_k u_l \rangle.
\]

(7)

The nonlinearity only has the effect to modulate the dynamic matrix. It is important to note that the results are independent of the partition size of the center due to \(T_{ijkl} = 0\) everywhere except for the two atoms at the interface.

Equation (7) together with \(G^r = [(\omega + i\eta)^2 - K_j^C - \Sigma_j^{-1}, G^s = G^r \Sigma^r G^s, \langle u_k u_l \rangle = i\hbar G_{kl}(t = 0) = 2i\hbar \int_0^\infty G_{kl}(\omega) d\omega/(2\pi)\) can be self-consistently calculated. Since the problem is now effectively harmonic, the heat current still satisfies the Landauer formula \(J = \int_0^\infty \frac{\hbar}{\pi} T[\omega](f_L - f_R), f_0 = 1/(e^{\hbar\omega/(k_B T_0)} - 1)\), and the thermal conductance is defined as \(\sigma = |J/(T_L - T_R)|\), while the transmission \(T[\omega] = \text{Tr}(G^r \Gamma_L G^s \Gamma_R)\) is temperature dependent. In later calculations, we will set \(\hbar = 1, k_B = 1\) for simplicity. For the conversion from the dimensionless unit to physical units, we take the energy unit \([E] = 1\text{ meV}\), the length unit \([L] = 1\text{ Å}\), the temperature unit \([T] = 11.6\text{ K}\), the thermal conductance \([\sigma] = 20.9\text{ mW/mK}\), the spring constant unit \([k] = 1\text{ meV/Å}^2\) and the nonlinearity unit \([\lambda] = 1\text{ meV/Å}^4\).

Applying our QSCMF theory to the nonlinear interface problem of eq. (1), we find that the nonlinearity plays a role to modulate the interfacial linear coupling \(k_{12}\), and the effective one is

\[
k_{12}\text{eff} = k_{12} + 3\lambda \left( \frac{\langle u_1^2 \rangle}{m_1} - 2 \frac{\langle u_1 u_2 \rangle}{\sqrt{m_1 m_2}} + \frac{\langle u_2^2 \rangle}{m_2} \right).
\]

(8)

Thus, it is clear that all the scattering occurs only at the interface since all other parts are harmonic.

Comparison with the quantum master equation. – While the effective phonon theory and its quantum correction one are valid in the linear response region at weak system-bath coupling, our QSCMF theory can be used to study the nonequilibrium thermal transport under larger temperature bias at any system-bath coupling. For anharmonic systems with arbitrary strength of anharmonicity under the weak system-bath coupling approximation, Redfield quantum master equation is suitable to study thermal transport [34]. In our interface problem, we partition the atoms at the interface as a center, and the center-lead coupling is the same as the inter-atomic spring constant in the corresponding bath. In such a system the system-bath coupling is always strong, thus one cannot apply the quantum master equation method which is limited to the weak system-bath coupling regime. However, since the quantum master equation treats the nonlinearity exactly, we numerically compare our theory with the quantum master equation at a weak system-bath coupling, as shown in fig. 2. In fig. 2, the system-bath coupling is 0.1 while the inter-atomic spring constant of the leads is 1.0 such that the weak system-bath coupling condition is well satisfied. From fig. 2 we find that the results from QSCMF perfectly match those from the quantum master equation method for even strong nonlinearities, thus verifying our approach. However, it should be noted that the QSCMF is not limited to the weak-coupling regime and can be applied to systems with arbitrary system-bath coupling strengths.

Numerical results on interfacial thermal transport. –

Nonlinearity suppressed thermal transport in homogeneous systems. Using the QSCMF theory we proposed above, the interface nonlinearity can be studied for the interfacial thermal transport. For the homogeneous lattice with \(k_1 = k_2 = k_{12}\), we calculate the interfacial thermal conductance for different values of nonlinearity \(\lambda\) as shown in fig. 3(a). With zero nonlinearity, the thermal conductance increases with increasing temperature, and tends to a constant due to the saturation of phonon modes contributing to thermal transport. However, with nonzero interfacial nonlinearity the thermal conductance decreases at high temperatures due to the dominant scattering coming from the nonlinear interface coupling. In the low-temperature regime, the thermal conductance almost
coincides with the ballistic transport and the nonlinearity has no noticeable effect on thermal transport, as previously shown in ref. [34]. With increasing temperature, more phonon modes are excited suppressing the effect of nonlinear scattering and thus increasing the conductance. At a certain temperature, the conductance reaches its maximum, after which it decreases since the nonlinear scattering effect dominates the enhancement effect from the excited phonon modes. With increasing nonlinearity the thermal conductance decreases due to the larger phonon scattering at the interface. As shown in fig. 3(b), the nonlinearity always decreases the thermal transport for homogeneous systems. A large interfacial nonlinearity makes the system more nonhomogeneous such as to induce more scattering to the phonon transport.

**Nonlinearity-enhanced interfacial thermal conductance.**

In the weak-interfacial-coupling regime, that is, \( k_{12} < k_1 = k_2 \), we find that the nonlinear interaction at the interface can enhance thermal transport as shown in fig. 4. With increasing nonlinearity, the interfacial conductance initially increases and then decreases after a certain maximum, as shown in fig. 4(a). The maximum of the conductance coincides with the point where the effective coupling equals 1 and the whole system is homogeneous. If \( \tilde{k}_{12} \) increases further, the conductance decreases due to the larger scattering at the interface. At a fixed linear interfacial coupling \( k_{12} < 1 \), the interfacial nonlinearity makes the effective coupling \( \tilde{k}_{12\text{eff}} \) larger than \( k_{12} \). A larger \( \tilde{k}_{12} \) reduces the difference between the interface and the leads, thus decreasing the phonon scattering and allowing more phonons to transmit through the interface. Therefore, the nonlinearity introduces an extra channel to transport phonons, which enhances the thermal transport. In fig. 4(b), the maximum of the thermal conductance does not coincide with the position of \( \tilde{k}_{12\text{eff}} = 1 \); this is mainly because the increase in temperature causes more phonons to transport so as to delay the maximum of conductance. For a larger nonlinearity, the maximum of the thermal conductance is delayed further.

For a hetero-junction, that is, the two mismatched leads, the thermal conductance can also be enhanced by interfacial nonlinearity. For a completely harmonic system we know that the thermal conductance reaches a maximum when the interfacial coupling equals the harmonic average of the spring constants of the two semi-infinite chains [5]. As shown in fig. 5(a), the conductance of two general hetero-junctions reaches a maximum when the effective coupling \( k_{12\text{eff}} \) equals \( k_{12m} = 2k_1k_2/(k_1 + k_2) \); after which the thermal conductance decreases. The interfacial conductance can be enhanced in a wide nonlinearity range, as shown in fig. 5(a). The enhancement of the conductance due to the nonlinearity depends on the interfacial linear coupling. In order to show this, we plot the thermal conductance as a function of the linear coupling \( k_{12} \) in fig. 5(b). For a fixed nonlinearity, the largest thermal conductance enhancement is at zero \( k_{12} \), where the only channel to transport phonons comes from the nonlinearity. With increasing \( k_{12} \) the conductance increases, but the effective coupling due to the nonlinearity \( k_{12\text{eff}} - k_{12} \) decreases. This effect causes the overall enhancement to decrease. The inset of fig. 5(b) shows that the nonlinear interfacial conductance equals the linear thermal case just before \( k_{12} = k_{12m} = 1.5 \), where the scattering from nonlinearity cancels the contribution from the extra channel introduced by the nonlinearity. At \( k_{12} = k_{12m} = 1.5 \), the linear conductance maximizes and the nonlinearity only adds to the phonon scattering. Thus the conductance of the nonlinear interface is small. If \( k_{12} \gg \lambda \), the effective coupling from the nonlinearity \( k_{12\text{eff}} - k_{12} \) nearly vanishes making the interfacial nonlinearity not important anymore, thus the conductance is almost the same as the ballistic one.

**Nonlinearity-induced interfacial thermal rectification.**

From our QSCMF approach, the effective interfacial coupling is temperature dependent; if we reversed the two
temperatures of the leads, it would be different so that we could observe thermal rectification. If the interfacial coupling is linear, then the Landauer formula applies and the reverse temperature only changes the sign of the heat current, thus there is no rectification. The rectification is defined as [35]

\[ R = (J_+ - J_-)/\max\{J_+, J_-\} \]

where \( J_+ \) is the forward direction heat flux when \( T_L = T_h \), \( T_R = T_c \), and \( J_- \) is that of the backward direction when \( T_L = T_c \), \( T_R = T_h \). Here \( T_h \) and \( T_c \) correspond to the temperatures of the hot and cold baths, respectively.

With the asymmetric structure the nonlinear interface shows rectification, which depends on the interfacial nonlinearity \( \lambda \) and the linear coupling \( k_{12} \) as shown in Fig. 6. If the nonlinearity is zero, there is no rectification. With the increase of the nonlinearity, the effective coupling will be different in the forward and backward transport which causes the rectification to increase. When the nonlinearity increases further, the effective coupling will monotonically increase. Therefore, the scattering from the interface will play an important role to decrease the difference between the forward and backward heat flux, which causes the rectification to decrease as shown in Fig. 6(a). With larger linear interfacial coupling, the contribution to the phonon transport from the channel of \( k_{12} \) is larger as compared to the extra channel provided by the nonlinearity \( \lambda \). Therefore, the relative nonlinear effect is weakened and the rectification is reduced as shown in Fig. 6(b). In the opposite limit, if \( k_{12} \ll \lambda \) the rectification almost keeps fixed, here the nonlinearity dominates the thermal transport and the difference of the effective coupling between the forward and backward flow has almost no changes. When \( k_{12} \sim \lambda \), the rectification decreases rapidly due to the decrease in the value of \( k_{12,\text{eff}} - k_{12} \). If \( k_{12} \gg \lambda \), the rectification decreases to almost zero since the effective coupling from the nonlinearity \( k_{12,\text{eff}} - k_{12} \) nearly vanishes making the nonlinearity not important anymore.

**Discussion.** We use a simplified one-dimensional atomic model to investigate the underlying physics in interfacial thermal transport. We find that interfacial nonlinearity can enhance thermal transport and can induce thermal rectification. Our model could be applied to a realistic interface where a general chemical bonding strength across an interface could be represented by a linear and a nonlinear coupling part. However, to compare with experimental results on real materials, the simplified model needs to be generalized to two- or three-dimensional systems with the input of the real parameters.

Our QSCMF theory is a mean-field–like approximation based on the nonequilibrium Green’s function approach and is similar to the other mean-field approaches in the literature [28,36]. Corroboration with the quantum master equation approach in the weak system-bath coupling regime strongly suggests that the QSCMF approach is a good candidate to solve nonlinear problems with arbitrary nonlinearity. Importantly, we do not make any assumptions on the system-bath coupling strength thus making our approach equally valid for the strong-coupling regime. In the case of the interface problem with two atoms, the QSCMF has a very high accuracy in a wide range of temperature and nonlinearity. However, if more atoms are included, the accuracy decreases [37]. Thus the QSCMF approach is best suited for the two-atom interface, which is a reasonable approximation for short-range inter-atomic coupling. Even though our approach has its shortcomings,
one of the most challenging problems for nonlinear systems is to rigorously develop an approach valid for an arbitrary number of atoms at the interface in two and three dimensions. We hope our novel approach sheds some light on advancing quantum phononics to nonlinear systems.

**Conclusion.** – Based on the NEGF approach, an efficient QSCMF theory is developed to study the nonlinear interfacial thermal transport. We find that the nonlinearity can enhance the interfacial thermal transport at weak linear interfacial coupling, while the enhancement vanishes in the strong-linear-coupling regime. The enhancement can exist at large nonlinearity where the effective coupling is less than the harmonic average of the spring constants of the two semi-infinite chains. Although the leads are linear, the interfacial nonlinearity can induce rectification provided that the two leads are asymmetric. With increasing nonlinear coupling, the rectification first increases and then decreases. The interfacial rectification also depends on the interfacial linear coupling; it vanishes if the linear interfacial coupling increases far beyond the nonlinear coupling.

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