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Thermal conductivity in two-dimensional monatomic non-linear lattices

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Abstract. We investigate the validity of Fourier's law in a two-dimensional monatomic Toda lattice using the molecular dynamics method. The temperature profiles in the lattice exhibit an exponential behaviour with lattice position. The temperature dependence of thermal conductivity is necessarily derived from the spatial variation of the local temperature, and found to be inversely proportional to the local temperature. The validity of Fourier's law is confirmed by excluding the non-diffusive heat flow from the total heat currents.

1. Introduction

Heat transport in an electrically insulating crystal or amorphous solid is attributed to the lattice vibrations and is described in terms of the phenomenological theory referred to as Fourier's law. The first attempt to explain Fourier's law was performed by Peierls in 1929 [1]. He attributed the energy sharing among the normal vibrational modes to lattice anharmonicity and derived the law, applying the concepts for gas dynamics to phonons. In 1955, Fermi *et al* [2] examined the energy-sharing mechanism in an anharmonic lattice by means of numerical simulations and exhibited a lack of equipartition of energy among the degrees of freedom, which cast some doubt on the hypothesis of Peierls for irreversibility of phonon systems. After this work a number of computer simulations on heat transport have been performed to verify the phenomenological theory from first principles. However, most attempts have failed to present a normal thermal conductivity [3–11] except for some special cases, such as the ding-a-ling model [12], since the resultant magnitude of the thermal conductivity was dependent on system size.

Mokross and Büttner [13] suggested that one-dimensional diatomic Toda lattices would present a normal thermal conductivity. Jackson and Mistriotis [14] examined the size dependence of the thermal conductivity of similar lattices, of larger size than those of Mokross and Büttner, and reported a size-independent thermal conductivity in one-dimensional diatomic Toda lattices. Although their work seemed to present a successful explanation for normal thermal conductivity, some problems had been found in their work which will be considered in the following.

In the numerical simulations on heat transport, the resultant temperature profiles have often appeared not to be linear but to be concave [10, 13, 14]. However, the

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temperature profiles have been blindly believed to be linear and approximated with straight lines to estimate the thermal conductivity. Another troublesome phenomenon, overlooked in the numerical simulations so far, is that the energy is carried not only through the diffusive process but also through the non-diffusive one [10, 14], which would lead to the undesirable non-local property of heat transport. The present authors [15] re-examined the heat transport in the one- and two-dimensional diatomic Toda lattices, paying special attention to the non-linear variation of local temperature and to the non-diffusive heat flow, which should be excluded for an estimation of normal thermal conductivity. We found, for both cases, that the temperature profiles show an exponential behaviour with respect to lattice position. The curved temperature profiles require a temperature-dependent thermal conductivity, so that the heat flow becomes independent of lattice position in the steady non-equilibrium states. Thus we made it clear that the deviation of the temperature profiles from a straight line is closely related to the temperature dependence of the thermal conductivity. We estimated the magnitude of the thermal conductivity by excluding the contribution of the non-diffusive heat flow to the heat transport and obtained a size-independent thermal conductivity which is inversely proportional to local temperature. This is just the evidence for Fourier's law. We have also investigated the heat transport in one-dimensional quasi-periodic Toda lattices [16, 17], using the same molecular dynamics method, and confirmed the evidence for normal thermal conductivity. We found a similar temperature dependence of thermal conductivity to that of the one-dimensional diatomic Toda lattices.

We have verified the validity of Fourier's law for some non-linear lattices whose mass distribution is diatomic or quasi-periodic. However, Fourier's law is known to be actually satisfied for any real material, even in monatomic crystals (for a review see [18]). We are interested in whether Fourier's law is satisfied or not with only lattice anharmonicity. This is precisely the motivation for the present work. If we could exhibit the validity of Fourier's law in monatomic systems, we could study fairly the heat transport in more complex systems such as amorphous solids by introducing some kind of inhomogeneity to the system. In this paper, we apply the molecular dynamics method used for the previous systems [15–17] to investigate the validity of Fourier's law in simple monatomic non-linear square (two-dimensional) lattices with nearest-neighbour interactions.

We mention here another method to investigate the thermal conductivity. In some numerical simulations on heat transport [10, 19], the Green–Kubo integral [20, 21] is used to estimate the thermal conductivity. The Green–Kubo integral relates the thermal conductivity to the heat current autocorrelation function in equilibrium. As the validity of Fourier's law is postulated in order to derive the Green–Kubo integral, we cannot use the Green–Kubo integral to investigate the existence of Fourier's law. Generally speaking, the applicability of the Green–Kubo integral to the numerical simulations of lattice heat transport is not justified unquestioningly since there are few systems for which Fourier's law has been verified from first principles.

The layout of this paper is as follows. In section 2 we give a brief description of the two-dimensional monatomic non-linear lattices to be studied here. Details of numerical simulations are also described in this section. We search for the temperature region where the system shows irreversible behaviour. The resultant temperature profiles are analysed and fitted with an empirical formula of exponential type in section 3. In section 4 we derive the temperature dependence of thermal conductivity based on empirical temperature profiles. Considering that the heat flow

consists of diffusive and non-diffusive parts, we examine the validity of Fourier's law. The last section will be devoted to a summary and discussions.

2. Two-dimensional Toda lattices and numerical simulations

The lattice to be studied here is a two-dimensional square lattice. All masses are equal to m_0 and the nearest-neighbour atoms are linked with Toda potentials [22]. In this paper, we hereafter refer to this two-dimensional non-linear square lattice as a two-dimensional Toda lattice. As is well known, a one-dimensional Toda lattice is integrable and has solitons or cnoidal waves as the non-linear normal modes [22]. On the other hand, such non-linear normal modes are not found in the two-dimensional Toda lattice. Therefore we may expect a finite temperature gradient or a finite thermal conductivity of the lattices in the non-equilibrium states due to the non-integrability of the lattice.

A direct connection from the specimen lattice to the heat baths is known to cause a large deformation of temperature profile near the interfaces between the lattice and heat reservoirs. This is because of the thermal boundary resistance due to large mismatch of acoustic impedance between them [23], and it is hard to specify temperature profiles correctly which reflect the temperature dependence of the thermal conductivity. In order to avoid this difficulty, we put Mikhailov's lattices [24]† as buffer areas between the relevant lattice and the heat baths. Mikhailov's lattice is another kind of two-dimensional non-linear lattice which can be constructed by replacing Toda potentials along the y-direction by harmonic ones in the two-dimensional Toda lattice. One of the important features of Mikhailov's lattice is that the lattice is integrable and has exact solitary-wave solutions as the non-linear normal modes [24]. This means that no finite temperature gradients occur in Mikhailov's lattice, since the energy can propagate ballistically even in the non-equilibrium states. Thus, the two-dimensional Toda lattice is sandwiched by regions of definite temperature. In other words, we can say that Mikhailov's lattices as a whole act as heat baths. However, since there is no longer a mismatch in acoustic impedance between the specimen lattice and Mikhailov's one, the temperature profiles of the two-dimensional Toda lattices might be easily affected near the interfaces between the different lattices. In order to prevent temperature profiles near the interfaces from being deformed, we insert heavy atoms $\{m_1\}$ at the interfaces and recover the thermal boundary resistance a little.

Figure 1 shows the experimental situation for the numerical simulations. As is seen in this figure, N_x and N_y denote the sizes of the specimen lattice in the x- and y-directions, and each buffer area is of atomic length N_B, including the interface atoms. Thus our system has a total atomic length N $(= N_x + 2N_B)$. We set $N_B = 10$ and $N_y = 10$ throughout the present experiments due to the restriction of computer capacity. The length of specimen lattice N_x varies in the range from 200 to 400.

For simplicity, we assume the atomic displacement to be scalar. The total energy for this system including the buffer areas whose dimensions are $N \times N_y$ is written as

† Mikhailov's lattice is often called the two-dimensional Toda lattice owing to its integrability in some of the literature. However, in this paper we refer to the two-dimensional non-linear lattice linked by Toda potentials between nearest-neighbour atoms as the two-dimensional Toda lattice.

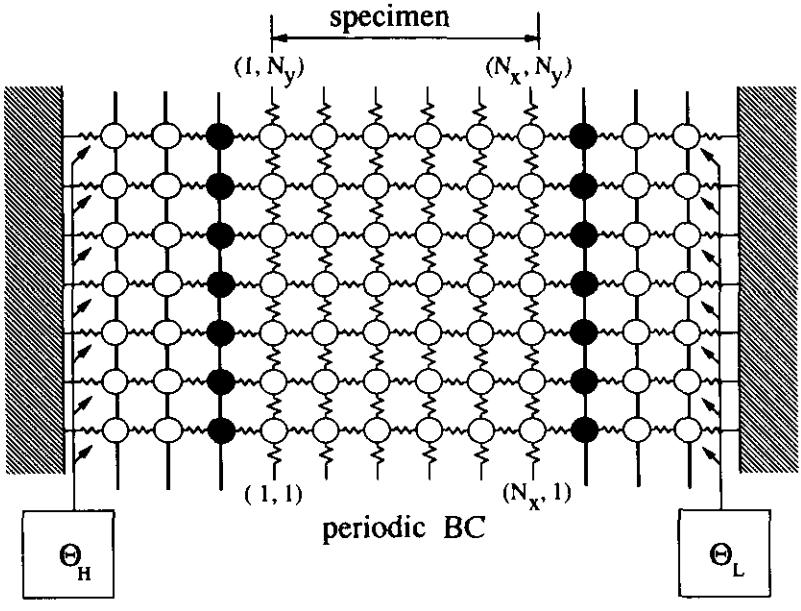

Figure 1. The model and experimental situation for the two-dimensional lattice. Open circles denote the atoms of mass m_0. The full circles denote the atoms of mass m_1 at the interfaces. All particles are connected to the Toda potentials along the x-direction. The atomic connection along the y-direction is also a Toda potential in the specimen lattice and an harmonic one in the buffer areas. The periodic boundary condition is used for displacement along the y-direction. The lattice is heated by the elastic collisions of atoms between the lattice ends and the heat reservoirs, whose temperatures are Θ_H and Θ_L, respectively.

$$H = \sum_{i=1}^{N}\sum_{j=1}^{N_y}\left(\frac{p_{ij}^2}{2m_i} + V(u_{ij} - u_{i-1j}) + W(u_{ij} - u_{ij-1})\right) + \text{interaction with heat reservoirs} \tag{1}$$

where p_{ij} and u_{ij} are the momentum and displacement of the atom at the (i, j) site, respectively. Masses $\{m_i\}$ for the two-dimensional Toda lattice and Mikhailov's one are set to be m_0 and those of the interface atoms to be m_1. In the present simulations, we set $m_0 = 1$ and $m_1 = 3$. The potential function $V(r)$ which connects all atoms along the x-direction is a Toda potential [22], such that

$$V(r) = (b/a)\exp(-ar) + br - b/a. \tag{2}$$

The other potential function $W(r)$, which connects the atoms along the perpendicular direction, is also a Toda potential $V(r)$ for the two-dimensional Toda lattice and is an harmonic potential $U(r)$ for Mikhailov's lattice, given by

$$U(r) = (c/2)r^2. \tag{3}$$

Here a, b and c are potential parameters which are all set to unity in this paper. The periodic boundary condition is used for the displacement of the end column

atoms, i.e. $u_{i1} = u_{iN_y+1}$. The equation of motion is integrated numerically using the Runge–Kutta–Gill method.

The energy exchange between the end atoms and the heat baths is supposed to occur through the elastic collisions of the end atoms with gas particles in the heat baths. The gas particles are also assumed to have a Maxwell distribution in velocity at the prescribed heat-bath temperatures, Θ_H and Θ_L, respectively.

At very low temperatures, the anharmonicity of the two-dimensional Toda lattices is extremely reduced and the dynamical behaviour of the lattices becomes reversible. With an increase in temperature the anharmonicity is enhanced, and above some critical temperature the dynamical behaviour changes drastically, to show stochastic or irreversible behaviour. Normal thermal conductivity is anticipated above such a critical temperature.

The divergence of trajectories starting from two points close in phase space has been investigated to check the establishment of thermal equilibrium states. The method has been improved for non-equilibrium states [14–17] since the dynamical behaviour must become stochastic within a short time interval, which is characteristic of the relevant system, as noted by Jackson and Mistriotis [14]. We will take a sound transit time interval T_s for a short time interval.

The type of divergence of trajectories is judged by the value of the quantity $\mu(T_s)$ defined by

$$\mu(T_s) = \int_0^{T_s} \mathrm{d}t\,[|d(t) - A(q_\Gamma, p_\Gamma, T_s)\exp(k(q_\Gamma, p_\Gamma, T_s)t)|^2 - |d(t) - B(q_\Gamma, p_\Gamma, T_s)t - C(q_\Gamma, p_\Gamma, T_s)|^2] \tag{4}$$

Here $d(t)$ is the distance between two trajectories α and β starting in the neighbourhood of a point (q_Γ, p_Γ) in the phase space, given by

$$d(t) = \left(\sum_{i=1}^{N}[(q_i^\alpha(t) - q_i^\beta(t))^2 + (p_i^\alpha(t) - p_i^\beta(t))^2]\right)^{1/2}. \tag{5}$$

The distance $d(t)$ is approximated for $0 < t < T_s$ by a function of the form $A(q_\Gamma, p_\Gamma, T_s)\exp[k(q_\Gamma, p_\Gamma, T_s)t]$ and also by a linear function of the form $B(q_\Gamma, p_\Gamma, T_s)t + C(q_\Gamma, p_\Gamma, T_s)$ using the least-squares method. q_i^α and p_i^α denote the position and momentum of the ith atom on the trajectory α, where the index i is abbreviated. Therefore, negative μ denotes that the divergence of trajectories is close to exponential behaviour and, namely, that irreversibility can be expected. We calculated the local rate of divergence 100 times at each value of energy per atom. The test was performed numerically and the ratio of the number of the exponential development M_1 to the total trials M was obtained for the energy per atom E/N.

Figure 2 exhibits the resultant probability that the system exhibits a stochastic behaviour versus E/N for the two-dimensional Toda lattice with $N_x = 125$ and $N_y = 10$ atoms, without the buffer areas. The sound transit time is taken as $T_s = 96$ from numerical experiments on the pulse propagation. The dynamical behaviour becomes stochastic at least at $E/N = 3.5$. As the critical energy per atom is reduced with system size and the energy per atom is almost equivalent to the temperature of the system, normal thermal conductivity is expected in the region $T > 3.5$ for $N_x \geqslant 200$.

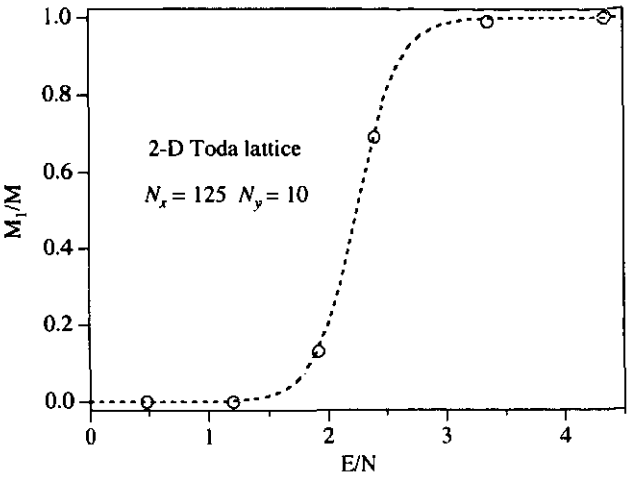

Figure 2. The probability that the system exhibits stochastic behaviour within a short time interval as a function of the energy-per-atom. The system examined is a two-dimensional Toda lattice with $N_x = 125$ and $N_y = 10$. The characteristic time interval is $T_s = 96$.

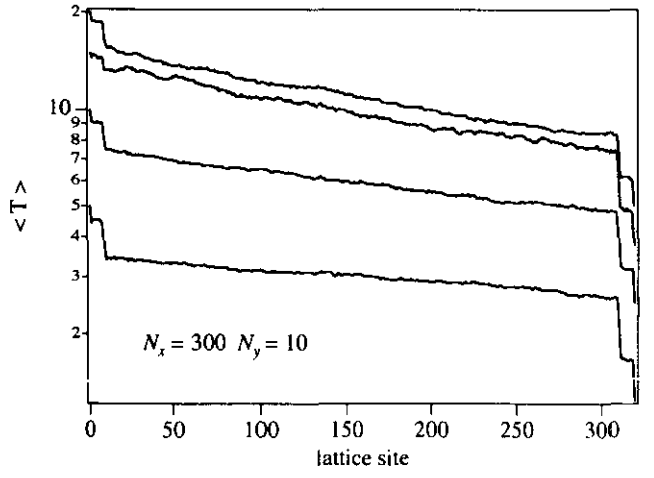

Figure 3. Temperature profiles for the two-dimensional monatomic Toda lattice with $N_x = 300$ and $N_y = 10$ atoms. The temperature profiles for $N_x = 200$ and $N_y = 10$, and $N_x = 400$ and $N_y = 10$ are similar to this figure.

3. Temperature profiles

We define the local temperature as twice the time average of the local kinetic energy. The temperature profiles along the x-direction of the lattice are obtained by averaging the local temperature over the lattice width

$$T_i = 2\left\langle \frac{1}{N_y}\sum_{j=1}^{N_y}\frac{p_{ij}^2}{2m_i}\right\rangle. \tag{6}$$

Here the angular brackets denote the time average. Figure 3 exhibits the temperature profiles of the two-dimensional Toda lattice with $N_x = 300$. The local temperature decreases exponentially with lattice positions. We also have similar temperature profiles for $N_x = 200$ and 400. Thus we can approximate the temperature profiles with an exponential function of the form

$$T(x) = T_H \left(\frac{T_L}{T_H}\right)^{x/N_x} \tag{7}$$

where T_H and T_L are not the temperatures of the heat reservoirs but those at the ends of the specimen lattice, and x measures the distance from the interface between the buffer area and the specimen lattice. Here we use the notation x instead of the subscript i, for convenience. The magnitudes of T_H and T_L are estimated by the least-squares method. In terms of the empirical formula for the temperature profile (7), the temperature gradient yields

$$dT/dx = -(1/N_x)\log(T_H/T_L)T. \tag{8}$$

4. Normal heat currents and thermal conductivity

The heat current J_x along the x-direction per width in the steady non-equilibrium state can be given by the following equation [15]:

$$J_x = \frac{1}{N_y}\sum_{j=1}^{N_y}\langle Q_{Hj}\rangle \tag{9}$$

where Q_{Hj} is the energy exchanged per unit time with heat reservoirs of high temperature. We consider that the heat current J_x consists of the normal heat current J_N, due to the local temperature gradients, and the ballistic or non-diffusive part J_B such that

$$J_x = J_N + J_B. \tag{10}$$

The normal heat current J_N is expected to obey the following Fourier's law

$$J_N = -\kappa\, dT/dx. \tag{11}$$

We assume here that the ballistic energy flow depends on the temperature difference of the buffer areas, and on the system size, since the buffer areas act as effective heat baths. The temperatures of the buffer areas $T_{B,H}$ and $T_{B,L}$ are slightly larger and smaller than T_H and T_L, respectively, because of the thermal boundary resistance. It should be noted here that the ballistic heat flow may not appear in real measurements of thermal conductivity since the temperature difference between the system ends is set to be quite small, in contrast to numerical experiments.

Substituting equations (8) and (11) into (10) yields

$$J_x = \kappa(1/N_x)\log(T_H/T_L)T + J_B(T_{B,H} - T_{B,L}, N_x). \tag{12}$$

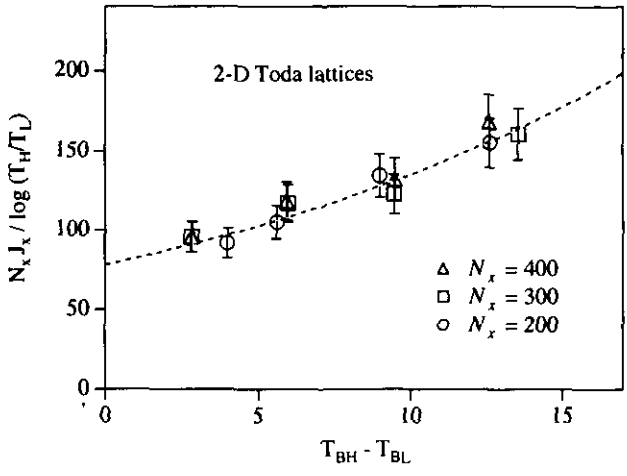

Figure 4. *The heat current per unit length* $J_x N_x / \log(T_H/T_L)$ *versus the temperature* difference of the buffer areas $T_{B,H} - T_{B,L}$. Open circles, squares and triangles denote the data for the lattices, with $N_x = 200$, 300 and 400, respectively. The broken curve is drawn using the least-squares method, to exhibit the extrapolation of the data to zero temperature difference. The extrapolated magnitude of ξ becomes 78.0.

Equation (12) requires that the thermal conductivity is proportional to the inverse of the local temperature, since the heat currents J_x and J_B do not depend on the local temperature. Thus we can write the thermal conductivity as follows:

$$\kappa = \xi / T \tag{13}$$

where ξ *is a constant.* The coefficient ξ must be independent of system size for the validity of Fourier's law since the thermal conductivity is an intensive quantity. In order to estimate ξ, and to examine the size dependence from the measurements, we have to exclude the contribution of the ballistic heat current from equation (12). If we could take the limit of the temperature difference to zero while keeping $\log(T_H/T_L)$ finite, we would obtain the normal heat current in the limit. However, experiments below the critical temperature are not available for an estimation of ξ because of the lack of irreversibility. Instead, utilizing the difference in temperature dependence between the heat currents J_x and J_B, we extrapolate ξ from the experimental data in the high-temperature regions. Substituting equation (13) into (12) yields

$$J_x N_x / \log(T_H/T_L) = \xi + J_B(T_{B,H} - T_{B,L}, N_x) N_x / \log(T_H/T_L). \tag{14}$$

We plot $J_x N_x / \log(T_H/T_L)$ versus temperature difference, $T_{B,H} - T_{B,L}$, for the two-dimensional Toda lattices with $N_x = 200$, 300 and 400 in figure 4. The magnitude of the left-hand side of equation (14) decreases drastically with the reduction of temperature difference and seems to become a positive and finite value at zero temperature difference. There is no difference in both the magnitude and its dependence on the temperature difference $T_{B,H} - T_{B,L}$ for all system sizes within numerical error. Therefore, we can obtain a unique value for the coefficient ξ ($= 78.0$) for two-dimensional Toda lattices, independent of the system size. This shows clear evidence for Fourier's law in the two-dimensional monatomic Toda lattice.

5. Summary and discussions

In this paper we examined heat transport in two-dimensional monatomic Toda lattices, to study the validity of Fourier's law by means of molecular dynamics. It has been shown that the temperature profiles along the x-direction exhibit not a linear but an exponential distribution independently of the system length. The deviation of the temperature profiles from a straight line requires the temperature-dependent thermal conductivity to maintain steady non-equilibrium states. We have derived the temperature dependence of the thermal conductivity, inversely proportional to the local temperature, necessarily from the exponential temperature profiles. In order to estimate the coefficient ξ of thermal conductivity, we have carefully eliminated the contribution of the non-diffusive heat flow to the total heat flow and determined the magnitude of ξ by extrapolating the experimental data for some system sizes in several temperature regions. The resultant thermal conductivity is inversely proportional to the local temperature and independent of the system length. The results give evidence in favour of the existence of Fourier's law in two-dimensional monatomic Toda lattices.

A remarkable result of this paper is the confirmation of the validity of Fourier's law only due to lattice anharmonicity, which offers a basic system for the study of thermal properties in more complex systems. As has been clarified so far, amorphous solids exhibit anomalous thermal conductivity behaviour (for a review see [25]). There have been some theoretical ideas, e.g. quasi-localized states [26], fractons [27], etc, to explain the plateau of thermal conductivity and the behaviour above the plateau region. It is not hard to introduce some structural inhomogeneities corresponding to additional mechanisms to the present system. We suppose that the present molecular dynamics method enables us to observe directly the temperature dependence of thermal conductivity from curved temperature profiles.

We would like to emphasize again here that the deviation of the temperature profiles from a straight line gives important information on the temperature dependence of thermal conductivity and should be always taken into account for the study of heat transport.

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