Non-diffusive heat transport and chaos in non-linear dielectric lattices

1998 J. Phys.: Condens. Matter 10 943

(http://iopscience.iop.org/0953-8984/10/5/004)

J. Phys.: Condens. Matter **10** (1998) 943–949. Printed in the UK PII: S0953-8984(98)86135-3

Non-diffusive heat transport and chaos in non-linear dielectric lattices

R H H Poetzsch and H Böttger
Institut für Theoretische Physik, Otto-von-Guericke-Universität, 39106 Magdeburg, Germany

Received 15 July 1997, in final form 31 October 1997

Abstract. Deviations from Fourier's law emerge from numerical simulations of various lattices modelling solids. Non-integrability and moreover chaotic motion are considered to be conditions of normal heat conduction. However, these are not sufficient conditions. Using a simple model as an example, we show that non-diffusive transport could occur even in the presence of disorder in the lattice and completely chaotic dynamics. We conclude that diffusive and non-diffusive transport can coexist, while the system is moving along a chaotic trajectory in the phase space.

1. Introduction

Normal thermal conduction in dielectric solids, which is formulated within Fourier's law, originates from diffusive heat transport. It has proved an enduring problem, still containing unresolved questions concerning the constructive properties of model systems yielding normal conduction [1–5]. It is now understood that non-linearity and non-integrability of the equations of motion do not necessarily result in ergodicity or even normal heat conduction [1, 2, 6–8]. There is a consensus that chaotic motion originating from non-linearity or, more precisely, non-integrability is essential for Fourier's law to be obeyed. But it is not known which features of a model guarantee chaotic motion and normal heat conduction, and whether or not chaotic motion is a sufficient condition for normal heat conduction. To confirm Fourier's law for a particular lattice model, one has to check the asymptotic behaviour of the heat flux as a function of the system size.

The purpose of this paper is to study the connection between normal heat conduction and chaotic motion. To this end we use a simple two-dimensional anharmonic model with mass disorder and solve the equations of motion numerically (section 2). We find an asymptotic behaviour of the heat flux which differs from the case of normal heat conduction. This indicates the existence of a non-diffusive contribution to the heat current (section 3). On the other hand, inspection of the dynamics of the model reveals chaotic motion with the probability of unity (section 4). This suggests that chaotic motion is not a sufficient condition for diffusive transport and normal heat conduction (section 5).

2. The model

The structure of the model system consists of a two-dimensional [9] regular triangular lattice with nearest-neighbour bonds and mass disorder. The masses m_i are equally distributed in the range $1-w, \ldots, 1+w$ and w is set to 0.8. The bonds are represented by pair potentials:

$$v(r_{ij}) = (r_{ij}-1)^2 - \varphi_3(r_{ij}-1)^3 + \varphi_4(r_{ij}-1)^4. \tag{1}$$

0953-8984/98/050943+07$19.50

The quantity r_{ij} is the instantaneous bond length of the nearest neighbours i and j. Their distance apart at rest is unity. Several values of the anharmonic coefficients $\varphi_3 \geqslant 0$ and $\varphi_4 \geqslant 0$ are chosen in such a way that they correspond to the Lennard-Jones potential as regards order of magnitude and that there is only one minimum in $v(r_{ij})$. With the help of this model, non-diffusive transport can be shown not to be a feature specific to ordered systems alone. Another advantage of the model is its completely chaotic motion even at temperatures corresponding to realistic vibrational amplitudes, i.e. $\Delta r_{ij} \propto \frac{1}{20}$ at $T = \frac{1}{200}$. Below we refer to this temperature T taken in energy units, i.e. Boltzmann's constant is set to unity. The units of all of the mechanical quantities are defined by setting the lattice constant, the mean mass and the factor of the right-hand side of equation (1) to unity.

The boundaries of the system are not fixed; instead they are allowed to vibrate freely. In order to calculate the current in the steady state, the lattice is attached at two opposite boundaries to heat baths. The two heat baths are simulated by the reversible Nosé–Hoover equations [10, 11]. At each of the boundaries all degrees of freedom within two layers are in contact with a heat bath. The parameter which controls the response time of the heat baths is suitably chosen to be $\tau = 3$ which is of the order of the mean period of vibration.

The equations of motion for thermal equilibrium and non-equilibrium are solved using the efficient velocity Verlet algorithm [12]. The details of the simulation technique that we used are also described in an earlier paper [13].

3. Heat conduction

The introduction pointed out the problem of normal heat conduction. For the calculation of heat conductivities it is necessary to check whether or not the lattice normally conducts heat. Usually, the coefficients κ_L obtained for systems of finite lengths L are treated as being intensive. Because the numerical simulations are restricted to fairly small systems, this criterion is often difficult to meet.

Consider the non-equilibrium case of a system which is coupled to two heat baths separated by the distance L. Assuming normal heat conduction, the calculated conductivity κ_L is related to the intrinsic one κ_∞ by

$$\frac{L}{\kappa_L} = \frac{L}{\kappa_\infty} + \varrho_1 + \varrho_2. \tag{2}$$

The quantities $\varrho_{1,2}$ are the resistances at the interfaces between the heat baths and the bulk of the system. The assumption of normal heat conduction implies that the intrinsic resistance L/κ_∞ can be defined independently of the properties of the surfaces of the sample. It is not valid, for example, for the harmonic limiting case, because the intrinsic resistance is not defined then and the quantity κ_L is mainly a function of the coupling at the interfaces [14].

Furthermore, it is presumed that κ_L is determined by

$$|j_L| = \kappa_L \frac{|T_2 - T_1|}{L} \tag{3}$$

formally corresponding to Fourier's law. Herein, j_L is the time-averaged heat flux in the steady state and $T_{1,2}$ are the temperatures of the heat baths. This equation involves an average of the reciprocal intrinsic conductivity over the distinct temperatures in the system, but this is not appropriate for our considerations. Obviously, even in the case of normal heat conduction the coefficient κ_L essentially depends on L unless $L/\kappa_\infty \gg \varrho_1 + \varrho_2$. A statement similar to equation (2) is valid if κ_L is calculated from the equilibrium fluctuations using e.g. the Kubo formula. Instead of interface resistances $\varrho_{1,2}$, a term taking scattering off the boundaries into account occurs [13].

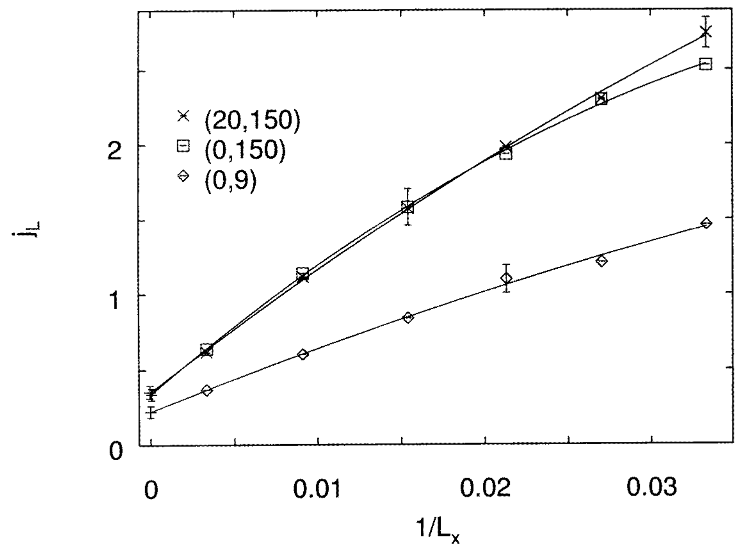

Figure 1. The heat current j_L in the non-equilibrium steady state as a function of the reciprocal of the sample length L_x for several pairs (f_3, f_4). The values for j_L are calculated for several numbers of layers L_x in the range 30–300. The symbols represent the values from the computer simulation. The lines are quadratic fitting functions. The temperatures are $T_1 = 1/400$ and $T_2 = 3/400$. The width of the systems is fixed at 20 layers. The values are given in reduced units.

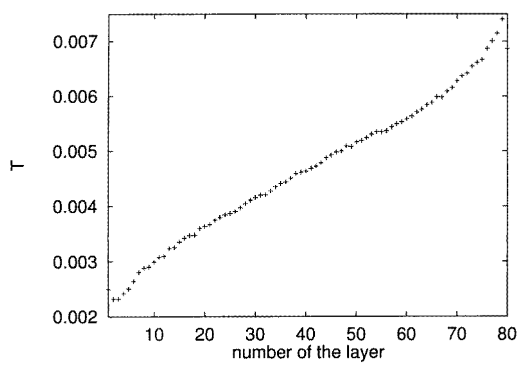

Figure 2. The profile of the temperature along the x-axis. The size of the system $N = 80 \times 20$; $(f_3, f_4) = (0, 9)$.

In the following we check our model system for the type of heat conduction. Thereby equation (2) is taken as an *ansatz*. With the help of this *ansatz* the quantity κ_L in equation (3) can be replaced. Then the expansion of the current j_L in terms of $1/L$ does not contain a constant term. Figure 1 displays the dependence of the calculated currents on $1/L$ at fixed bath temperatures $T_2/3 = T_1 = 1/400$. The symbols mark the values obtained by computer simulation with three pairs of anharmonic parameters (φ_3, φ_4) for the system defined above. Because the symbols may be connected by curves which are almost linear, the restriction of the fitting functions to second-degree polynomials is justified. This also means that the influence of the interface resistances is taken into account in the lowest order. It turns out that the constant term of each fitting function significantly differs from zero (cf. the error bars at $1/L = 0$), i.e. the current j_L cannot be represented by an analytic expansion in $1/L$ whose lowest-order term is the linear one. This contradicts Fourier's law. It indicates the existence of a non-diffusive contribution to the heat current.

It should be mentioned that the profiles of the temperature through the system are quite smooth for all of the pairs of the anharmonic parameters and for all of the sizes of the lattice that we used. There are no sharp jumps of the temperature near the boundaries. A typical profile is shown in figure 2.

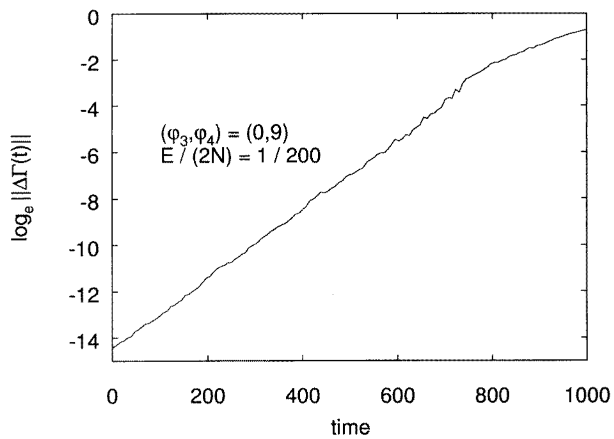

Figure 3. The distance (on a logarithmic scale) of two initially neighbouring trajectories on the energy surface $E/(2N) = 1/200$ in the phase space.

4. Characterization of the chaotic motion

Chaotic motion is considered to be a prerequisite for normal heat transport. Therefore the nature of the inner dynamics in the bulk of our lattice model without heat baths should be checked. Figure 3 shows how the distance of two initially neighbouring trajectories in the phase space typically evolves. The starting points in the phase space are arbitrarily chosen under the condition of vanishing total (angular) momentum. Almost all of the allowed starting points yield an evolution of the distance very similar to the one plotted in the figure. The distance diverges in an exponential manner over a few orders of magnitude. At long times the distance tends towards a constant mean value. This is due to the loss of correlation between the two trajectories. The slope of the exponential part of the graph corresponds to the maximum Lyapunov exponent. It is the maximum one because the numerical simulation acts like an iteration process. At the end of the initial phase (not plotted in figure 3) the distance vector in the phase space almost admits just one projection, which corresponds to the largest exponent. The value of this exponent is calculated from the slope of a linear function obtained by means of least-squares fitting.

The measure of the chaotic areas in the phase space at a given energy is determined in the following way [3].

A set of M initial states on the energy surface is arbitrarily chosen. For each initial state the distance $d(t)$ is computed and approximated by an exponential function $c\exp\{\lambda t\}$ as well as by a linear function $b_0 + b_1 t$. If the mean squared errors of the approximations obey the relation

$$\int_0^{\tilde{t}} \mathrm{d}t \ (c\mathrm{e}^{\lambda t} - d(t))^2 < \int_0^{\tilde{t}} \mathrm{d}t \ (b_0 + b_1 t - d(t))^2 \tag{4}$$

then the initial state is assumed to lie in a chaotic region. Let M_1 be the number of chaotic states. Then the frequency M_1/M may be taken as a measure of the chaotic regions in the phase space. The result is insensitive to the choice of the upper limit, $\tilde{t}$, of the integrals in relation (4) unless $\lambda\tilde{t} \ll 1$. With the value $\tilde{t} = 4/\lambda$, the distinction between linear and exponential divergence should be sufficiently reliable [15].

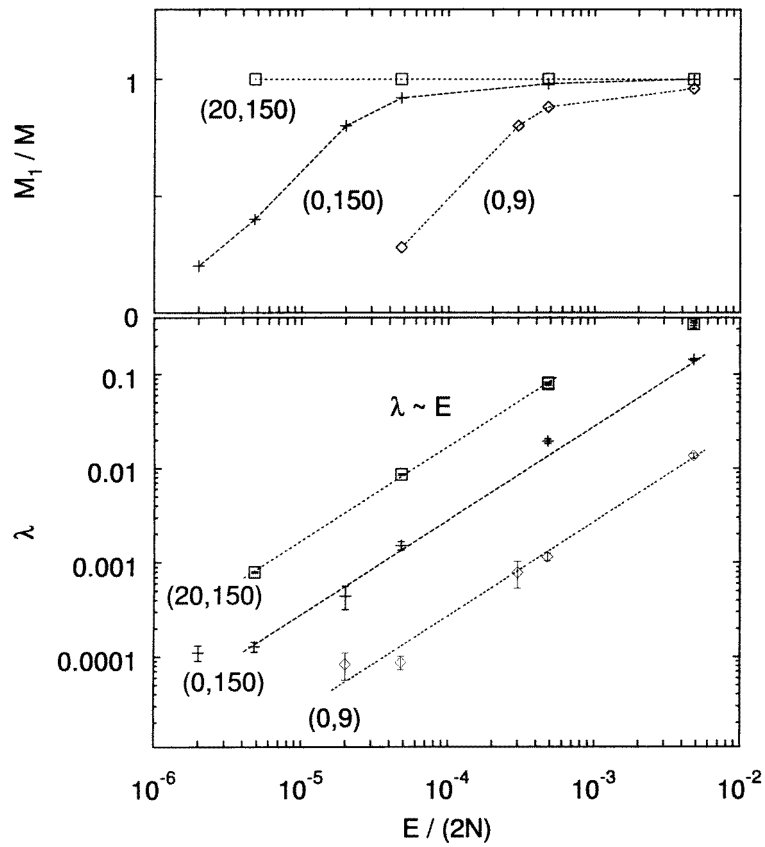

Figure 4. The measure M_1/M of the chaotic regions and the corresponding Lyapunov exponents λ. The values λ are averages merely over the chaotic regions. The error bars are the corresponding standard deviations. The size of the system $N = 10 \times 10$; the parameters (φ_3, φ_4) are given within the plot; $M = 50$.

Figure 4 shows the measure M_1/M and the corresponding maximum Lyapunov exponent as function of the mean energy per degree of freedom $E/(2N)$. The quantity N is the number of particles. At sufficiently high energies, let us say $E/(2N) \geqslant 1/200$, the measure M_1/M is unity, i.e. almost the whole of the energetically allowed phase space is chaotic. At small energies, M_1/M approaches zero, i.e. non-chaotic motion, because the harmonic limiting case is integrable. Within the scope of the model under consideration, the Lyapunov exponents are increasing functions of the parameters φ_3 and φ_4 as well as of the energy. Over a wide range of energy, $\lambda \sim E \sim T$ holds.

5. Conclusions

Now it becomes clear that the answer to the question [2] of whether chaotic motion is a sufficient condition for diffusive transport and normal heat conduction is in the negative. This can be deduced in an exemplary fashion from the model under consideration.

Despite the fact that $M_1/M = 1$ at high energies, the existence of *non*-chaotic trajectories cannot be ruled out. But the measure, i.e. the probability, of these trajectories is zero. Therefore, non-chaotic trajectories, even if they actually exist, do not contribute to thermal averages. In the case of chaotic motion with $M_1/M \neq 1$ one would not be surprised if there was a non-diffusive current. But in the case under consideration with $M_1/M = 1$, there is also a non-vanishing non-diffusive contribution to the heat current. As a result, trajectories with exponential divergence, i.e. chaotic ones, are responsible for non-diffusive transport. The means that the motion along a chaotic trajectory can result in diffusive and non-diffusive transport at the same time. This is consistent with the KAM theorem, which states that non-integrable motion may occur to some extent on persistent hyper-tori of the phase space. Due to the fact that the spectrum of Lyapunov exponents of a Hamiltonian system also contains non-positive exponents [16], there are always some projections of a state vector which do not contribute to the exponential divergence. Microscopic excitations causing non-diffusive transport should correspond to such projections. These projections may be viewed as degrees of freedom.

In other words, there may be a set of degrees of freedom which is not taking part in the chaotic motion of the system. Instead, these degrees of freedom are moving in a regular way. Therefore these degrees of freedom do not transport energy in a diffusive manner. Of course, this set can only have a weak coupling to all of the chaotically moving degrees of freedom. Given such a set of regularly moving degrees of freedom, we can observe non-diffusive transport while the system itself evolves chaotically.

The question of the kind and the nature of the non-diffusive excitations arises. First of all, we argue that the deviations from Fourier's law found are not simply due to short sample lengths allowing energy packets to propagate ballistically from one end of the sample to the other. It is guessed that soliton-like pulses are the most effectively transporting kind of non-linear wave packet [17–20]. This is also true for disorder of the masses [17]. In our two-dimensional models with $(\varphi_3, \varphi_4) = (0, 150)$ or $(20, 150)$ soliton-like pulses could exist, if linearly spreading fronts are forming. In the (20, 150) case, e.g., the decay time for merely dynamical scattering is about 17. The velocity of the pulses is approximately 2. Consequently, the mean scattering length is about 34. This is a short length compared to our largest systems with 300 layers.

The data given above do not allow any final decision to be reached regarding whether an expansion of the current j_L at $1/L = 0$ exists or not. In principle, j_L could also contain a power function with respect to $1/L$ in such a way that it yields $\lim_{1/L\to 0} j_L = 0$. However, a simple power law like that in the case of pure super-diffusion in harmonic lattices does not match the present data. Nonetheless, the current may consist of different contributions. One contribution could result from super-diffusion, i.e. from slowly decaying excitations of weakly localized eigenstates. This contribution should be drastically changed if the free boundaries are changed to the fixed boundaries of the lattice model. The proof of this is left to future investigations.

Finally, let us state once more the main result. The answer to the question posed by

Ford [2] regarding whether or not chaos is a sufficient condition for diffusive energy transport is in the negative [21]. Even in the case of measure-unity chaos, or, more specifically, in the case of evolution along a chaotic trajectory, non-diffusive and diffusive transport can coexist.

References

[1] Jackson E A 1978 *Rocky Mountain J. Math.* **8** 127
[2] Ford J 1992 *Phys. Rep.* **213** 272
[3] Jackson E A and Mistriotis A D 1989 *J. Phys.: Condens. Matter* **1** 1223
[4] Casati G, Ford J, Vivaldi F and Visscher W M 1984 *Phys. Rev. Lett.* **52** 1861
[5] Corezzi S, Bianucci M and Grigolini P 1995 *Phys. Rev.* E **52** 6881
[6] Fermi E, Pasta J and Ulam S 1955 *Los Alamos Report* LA-1940
[7] Nishiguchi N, Kawada Y and Sakuma T 1992 *J. Phys.: Condens. Matter* **4** 10 227
[8] MacDonald R A and Tsai D H 1978 *Phys. Rep.* **46** 1
[9] In the past the question was raised of whether transport coefficients exist at all in low-dimensional systems ($d \leqslant 2$). Meanwhile, there have been many indications that transport coefficients can exist even in one dimension (e.g. [3, 4, 22, 13]).
[10] Hoover W G 1985 *Phys. Rev.* A **31** 1695
[11] Holian B L, Hoover W G and Posch H A 1987 *Phys. Rev. Lett.* **59** 10
[12] Beemann D 1976 *J. Comput. Phys.* **20** 130
[13] Poetzsch R H H and Böttger H 1994 *Phys. Rev.* B **50** 15 757
[14] Ishii K 1973 *Suppl. Prog. Theor. Phys. Japan* **53** 77
[15] (i) The duration $\tilde{t}$ must not include late times without any correlation between the two trajectories. (ii) In reference [3] it is suggested that one relate $\tilde{t}$ to the 'sound transit time', i.e. the velocity multiplied by the size of the system. We cannot confirm this on the basis of our investigation. We found the Lyapunov exponents and the measure to be constant despite varying the size of the system by two orders of magnitude.
[16] See, e.g.,
Ebeling W, Engel H and Herzel H 1990 *Selbstorganisation in der Zeit* (Berlin: Akademie)
[17] Li Q, Pnevmatikos S, Economou E N and Soukoulis C M 1988 *Phys. Rev.* B **37** 3534
[18] Zabusky N J 1973 *Comput. Phys. Commun.* **5** 1
[19] Ilzuka T, Nakao T and Wadati M 1991 *J. Phys. Soc. Japan* **60** 4167
[20] Zavt G S, Wagner M and Lütze A 1993 *Phys. Rev.* E **47** 4108
[21] A recently published paper:
Lepri S, Livi R and Politi A 1997 *Phys. Rev. Lett.* **78** 1896
gives a negative answer to Ford's question, too. There, the authors make an assumption concerning the chaotic nature, but the decisive question concerning its measure remains open.
[22] von Beijeren H 1982 *Rev. Mod. Phys.* **54** 195