Normal transport at positive temperatures in classical Hamiltonian open systems

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Abstract

We study the transport properties of classical Hamiltonian models describing the motion of an unconfined particle coupled to vibrational degrees of freedom in thermal equilibrium at zero or positive temperature. We identify and discuss conditions under which, in such systems, the particle has a well-defined diffusion constant and mobility. We will in particular point out some marked differences with the situation where the particle is confined and described with a Caldeira-Leggett model. We will more specifically report on results obtained in a classical version of the Holstein molecular crystal model, speculate on their relevance in the corresponding quantum system and describe a number of open problems.

1   Introduction

A contribution in a volume dedicated to J.M. Combes can naturally be expected to address research issues concerning quantum mechanics, and to deal with them rigorously. In spite of that, the problems addressed here are set in the context of classical Hamiltonian dynamical systems and dealt with mostly numerically. Still, they are motivated by a set of interesting open questions in quantum transport theory explained in Section 4; since in addition they are directly related to some of Jean-Michel's main research interests over the last two decades, namely

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quantum dynamics and random Hamiltonians, we feel they certainly have their place in this collection of adventures in mathematical physics dedicated to him.

The behaviour of a system with a finite number of degrees of freedom, such as a single particle, in contact with an infinite dimensional system has been the subject of much research for several decades. There exists in particular an extensive literature (see [FKM] [CL1] [CL2] [DDLL] [JP3] [JP4] [CEFM] and references therein) in which the particular case of the behaviour of a classical or a quantum particle coupled to a heat bath modeled by harmonic degrees of freedom is studied. The goal in many of these studies is to shed light on various questions of non-equilibrium statistical mechanics : return to equilibrium, understanding possible sources of irreversible behaviour, derivation of an effective dynamics for the particle (such as Langevin or Fokker-Planck equations), computation of transport coefficients from microscopic models, *etc.* Those works have dealt almost exclusively with confined particles and used a dipole approximation to describe the interaction between the particle and the reservoir, leading to an interaction term in the Hamiltonian that is linear both in the bath variables and in the position coordinate of the particle. Those models – often referred to as Caldeira-Leggett models – are, as a result, not suitable for studying transport theory for unconfined particles in periodic or fully translationally invariant media. A class of models adapted to this physically important situation are easily constructed (Section 2) but their *dynamical* properties have been much less studied. In particular, very little is known rigorously about their dynamical behaviour, in particular at positive temperature or in the quantum mechanical context. The zero temperature situation is described for a model of radiation damping in [KKS1] [KKS2] [KS] [S] and for frictional damping in [BDB1].

In [DPS] [SPD] and [LPD] we are concerned with such a model in which the heat bath consists of a periodic array of *monochromatic* Einstein oscillators through which the particle moves. As such, the model can be seen as a classical version of the Holstein molecular crystal model (Section 3), a well-known model for electron-phonon interactions in molecular crystals [H] (see Section 4). Alternatively, it can be perceived as an inelastic Lorentz gas in which the particle scatters inelastically off periodically placed scatterers. It was found in these works that when no external field is applied to the particle ($F = 0$), it executes a normal diffusive motion, with a well-defined temperature dependent diffusion constant that was analysed in [SPD]. When $F \neq 0$, on the other hand, it was established that, on a suitable time scale, the particle reaches a limiting mean velocity linear in the applied field, with a well-defined low-field mobility compatible with the Einstein relation [LPD]. The model therefore possesses normal transport properties, a situation in sharp contrast to what happens in Caldeira-Leggett models, where it is well known such behaviour cannot occur when the bath is monochromatic. Indeed, two general ideas have emerged from the literature cited above concerning particles bilinearly coupled to an oscillator bath. First, for the harmonic heat bath to efficiently dissipate the particle's energy, and to induce a (generalized) Langevin or Fokker-Planck dynamics for the particle, it has to have a continuous frequency spectrum extending all the way down to zero. Second, if the particle couples too strongly to the low fre-

quency modes in its environment, the particle's damping will be very strong, and subdiffusive motion will occur, whereas if too few such modes are available, the particle will be insufficiently damped and its motion will be superdiffusive or even ballistic [SG] [MOBH]. I neither case does the particle have a diffusion constant or a mobility. In the classical Holstein molecular crystal model discussed below, which has a monochromatic heat bath and hence in particular no low-frequency modes, the situation is therefore radically different.

We end this note with some speculations on the pertinence of our findings to the dynamics of the Holstein molecular crystal and hence to the quantum transport problems alluded to above (Section 4).

2   A family of classical models

A family of classical Hamiltonian models in which a particle is coupled in a translationally invariant (hence non-linear) manner to the vibrational degrees of freedom of its environment was introduced in the unpublished Section 6 of [BDB2] (otherwise published as [BDB1]). These models include as special cases virtually all models typically studied in the literature in which a heat bath is described with vibrational degrees of freedom and they can serve to describe a variety of very different physical situations, as we will see below. They furthermore include a number of new models suitable for the study of transport properties, one of which will be detailed and studied in Section 3.

We consider a particle moving in $\mathbb{R}^n$. To model the vibrational degrees of freedom of the particle's environment, one considers a harmonic system characterized as usual by a positive operator Ω_{R}^2 on a real Hilbert space $\mathcal{K}_{\mathrm{R}} = L^2(\mathbb{R}^n, \mathrm{d}\nu, \mathcal{L})$ for some Borel measure ν, and a real Hilbert space $\mathcal{L}$. Very often, $\mathcal{L} = \mathbb{R}$, but for certain models, $\mathcal{L}$ is actually itself an infinite dimensional real Hilbert space, as we will see below. To guide one's intuition, one should keep two particular cases in mind: either ν is simply Lebesgue measure or it is the pure point measure concentrated on the lattice $\mathbb{Z}^n$. They are the only ones we will be interested in here. The operator Ω_{R}^2 is then typically a Laplacian or some finite difference operator. The Hamiltonian of the reservoir is

$$H_{\mathrm{R}}(q_{\mathrm{R}}, p_{\mathrm{R}}) = \frac{1}{2} p_{\mathrm{R}} \cdot p_{\mathrm{R}} + \frac{1}{2} q_{\mathrm{R}} \cdot \Omega_{\mathrm{R}}^2 q_{\mathrm{R}}.$$

Here $\cdot$ refers to the inner product on $\mathcal{K}_{\mathrm{R}}$ and q_{R} and p_{R} are the oscillator displacement (or field) and momentum respectively. It is often helpful to think of $q_{\mathrm{R}}(x) \in \mathcal{L}$, for each $x \in \mathbb{R}^n$ as being itself an oscillator field attached to the point x. The Hamiltonian of the combination particle+reservoir is then, with

$$X = (q_S, q_R, p_S, p_R),$$

$$H(X) = \frac{1}{2} p_S \cdot p_S + V(q_S) + \frac{1}{2} p_R \cdot p_R + \frac{1}{2} q_R \cdot \Omega_R^2 q_R + \int_{\mathbb{R}^n} d\nu(x) \rho(x - q_S) \cdot q_R(x). \quad (1)$$

Here the last term describes the interaction of the two systems and $\rho : \mathbb{R}^n \to \mathcal{L}$, called the "form factor", is typically a smooth rapidly decreasing function of its argument. The Hamiltonian equations of motion for such a model are

$$\dot{q}_S = p_S, \ \dot{p}_S = -\nabla V(q_S) + f(q_S, q_R),$$

and

$$\dot{q}_R = p_R, \ \dot{p}_R = -\Omega_R^2 q_R - \rho(\cdot - q_S),$$

where

$$f(q_S, q_R) = -\nabla_{q_S} \int_{\mathbb{R}^n} d\nu \rho(x - q_S) \cdot q_R(x)$$

is the force exerted by the reservoir on the particle. We make two remarks. First, even if the potential V is a second order polynomial, the model is non-linear because of the presence of the form factor. Second, if you substitute formally $\rho(x) = \delta(x)$ and $d\nu = dx$, the force f equals $-\nabla q_R(q_S)$; so the oscillator displacement $q_R(x)$ plays the role of a potential for the system. It is however in general impossible to prove existence and uniqueness of the solutions of the above equations without putting some regularity condition on ρ which amounts to making an ultraviolet cutoff.

Various models of this type have been studied in detail and they allow for the description of a wide variety of physical phenomena. Let me sketch two examples.

If, for example, you set $\mathcal{L} = \mathbb{R}$, $d\nu = dx$ and $\Omega_R^2 = -\Delta_x$, you obtain the standard model in which a scalar field is coupled to a moving particle. Its classical mechanics is studied in detail and with full rigour in [KS] [KKS1] [KKS2] [S], where it is proven this is a model for *radiation damping*. In particular, when $V = 0$, and for finite energy initial conditions, the particle reaches as $t \to \infty$ an asymptotic speed the value of which depends on the initial conditions. This reflects the fact that in such a model non-accelerating particles do not radiate, meaning they do not exchange energy with the field. To put it differently, the field exercises no force on the particle if the latter moves at constant speed. When in this model the potential V is confining, the particle will asymptotically come to rest at one of the critical points of the potential, as proven in [KS] [KKS1] [KKS2] for finite energy initial conditions. At positive temperatures, one would expect the full system to have the property of return to equilibrium, but this has not been proven rigorously, to the best of our knowledge. The property of return to equilibrium for quantum versions of this model has been proven in [BaFS] [JP1] [JP2], but it should be noted that the particle is then replaced by a system with a finite number of levels.

Now, in the case of a confining potential, it is reasonable to make a dipole approximation in this model, which then leads to a coupling term which is linear both in the particle position q_S and in the oscillator displacement q_R: in

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this form, possibly with different choices of the reservoir, the model is studied both classically and quantum mechanically and both at zero and positive temperatures in [FKM] [CL1] [CL2] [SG] [DDLL] [JP3] [JP4] [CEFM].

In [BDB1], a model for *frictional damping* by a medium with continuous translational invariance is introduced. This time we still have $d\nu = dx$, but now $\mathcal{L} = L_r^2(\mathbb{R}^d, dy)$ and $\Omega_R^2 = -c^2 \Delta_y$. The field q_R can then be seen as a function of $(x, y) \in \mathbb{R}^n \times \mathbb{R}^d$, and hence $q_R(x, y)$ thought of as representing, for each $x \in \mathbb{R}^n$, the displacement of a vibrating membrane. Since Ω_R does not contain any derivatives in the x-variables, the membranes at different points in the ambient space of the particle (which is $\mathbb{R}^n$) are not coupled, unlike what happens in the previous model for *radiation damping*. The behaviour of the system is very different from the one observed in the case of radiation damping. It is indeed proven in [BDB1] that if $d = 3$, $V = 0$, if c is large enough and if the initial conditions have finite total energy then the particle comes asymptotically to rest at some point in its ambient space $\mathbb{R}^n$. This is due to the fact that now the particle will lose energy to the field as long as it keeps moving, which in the end forces it to stop. Also, in the presence of an external driving potential of the type $V(q_S) = F \cdot q_S$, for some $F \in \mathbb{R}^n$, the particle reaches an asymptotic velocity v_F which depends on F but not on the initial conditions. In addition, as shown in [BDB1], this asymptotic velocity is proportional to the applied force F for small F. So this system exhibits a finite and non-zero low-field mobility $\mu := \lim_{F \to 0} v_F / F$ which depends on the coupling through the form factor ρ and on the reservoir via Ω_R. Finally, when the potential V is confining, the particle will come to rest at a critical point of the potential, with an exponential rate given by the mobility μ. In conclusion, the results of [BDB1] show that the particle behaves as if it obeyed the effective equation

$$\ddot{q}_S = -\nabla V(q_S) - \mu^{-1} \dot{q}_S. \tag{2}$$

So in this situation, the particle undergoes a friction force linear in its velocity, due to the presence of the environment. We remark that the conditions $d = 3$ and c large ensure in this case that the model is essentially Markovian: disturbances sent into the membranes by the particle are quickly evacuated to infinity. Note also that the frequency spectrum of the bath is continuous and stretches down all the way to zero.

The analysis of [BDB1] leaves several questions unanswered. First of all, it is restricted to the zero temperature case. This explains the absence of a random force term in the right hand side of (2) which would be present at positive temperatures and would then yield a Langevin equation. If one believes the Einstein relation $\mu = \beta D$ ought to be valid for this model, then the fact that the model has a non-zero mobility at zero temperature suggests that the particle behaviour must be diffusive for positive temperatures and that its diffusion constant must depend linearly on the temperature for small temperatures. This question is not addressed in [BDB1] since only finite energy initial conditions are considered there. Second, the analysis in [BDB1] strongly suggests that the existence of a finite mobility is directly related to both the complete translational

invariance of the model and to the low frequency behaviour of the oscillator bath. It is in particular necessary for the bath to contain sufficiently many low frequency components for it to be able to slow down slow particles sufficiently (See [BDB1] and Section 6 of [BDB2]). This of course is reminiscent of the findings in models with linear coupling described in the introduction.

In order to explore what happens at positive temperatures, to analyze the interesting situation when the medium is periodic, but has no continuous translational invariance, and to understand the role of possible cut-offs in the frequency distribution of the bath on the behaviour of the particle, we have studied in [DPS] [SPD] and [LPD] the transport properties of a model of the type (1), that can be understood as a classical version of the quantum mechanical Holstein molecular crystal model described in Section 4.

3 A classical version of the Holstein molecular crystal model

Consider a one-dimensional periodic array (with period a) of identical oscillators of frequency ω and a particle which interacts with the oscillator at ma if it is within a distance $\sigma < \frac{a}{2}$. The Hamiltonian of this system is

$$H = \frac{1}{2}p_{\mathrm{S}}^2 + \sum_{m \in \mathbb{Z}} \frac{1}{2}\left(p_m^2 + \omega^2 q_m^2\right) + \alpha \sum_m q_m n_m\left(q_{\mathrm{S}}\right) - F q_{\mathrm{S}}, \qquad (3)$$

where $n_m\left(q_{\mathrm{S}}\right)$ vanishes outside the interaction region associated with the oscillator at ma and is equal to unity inside it. This model clearly belongs to the family (1): the measure ν is now the sum of Dirac delta measures on the lattice $a\mathbb{Z}$ and the form factor ρ is the characteristic function of the interval $[-\sigma, \sigma]$. Comparing to the model of [BDB1], one sees that the continuous family of membranes is now replaced by a discrete family of single oscillators, all of the same frequency. Finally, the analogy of the above classical model with the Holstein molecular crystal model that will be discussed in Section 4 is equally obvious.

The dynamics generated by this Hamiltonian is easily described: the motion of the particle is decoupled from that of the oscillators, except when the particle crosses in and out of an interaction region $|q_{\mathrm{S}} - ma| < \sigma$. At those times, the particle sees a potential energy barrier $\pm \alpha q_m$ and its change in momentum is determined by conservation of energy. The oscillators all oscillate about their equilibrium position $q_m = 0$ when the particle is not within their interaction region, and about a displaced equilibrium position $q_{m*} = -\alpha/\omega^2$ when it is. These features make an efficient numerical simulation of the dynamics for very long times readily accessible, even at positive temperatures. Note that the Hamiltonian is bounded below by the energy $-\alpha^2/2\omega^2$, which corresponds to the situation where all oscillators are at rest, and the particle finds itself also at rest in one of the interaction regions. This corresponds to a self-trapped state of the particle in the sense that applying a small electric field to the particle will, in this situation, not allow it to escape from the cell in which it finds itself.

It follows that the zero temperature mobility of this model vanishes, unlike in the model described in [BDB1]. This is due to the fact that there are now free regions between the oscillators.

In order to study the transport properties of this model, a thermal distribution of particles (at inverse temperature β) was injected into the array of oscillators at $m = 0$, with the oscillators also drawn from their equilibrium distribution at the same temperature. The mean square displacement $\langle q_S^2(t) \rangle$ was then computed numerically and it was observed [SPD] that

$$\langle q_S^2(t) \rangle \sim 2Dt,$$

where the diffusion constant D depends on the temperature and on the two dimensionless parameters of the model, which are E_B/E_0 and $2\sigma/L$. Here $E_B = \alpha^2/2\omega^2$ is the binding energy of the particle, $E_0 = \sigma^2\omega^2$ and $L = a - 2\sigma$ is the size of the non-interacting region in a cell. At high temperatures ($\beta E_B >> 1$) the diffusion constant D behaves as $D \sim D_H^0 (\beta E_B)^{-5/2}$ where $D_H^0 = \frac{105}{16^2} a \sqrt{9\pi E_B/2} \left(\frac{E_B}{E_0} \right)$. At low temperatures the diffusion is thermally activated:

$$D \sim \frac{a^2}{2} \sqrt{\frac{E_B}{2\pi E_0}} (\beta E_B)^{-1/2} \exp{-\beta E_B}.$$

Although a rigorous proof of these observations is missing, the numerical results described give firm footing to the conjecture that the motion of the particle in this classical version of the Holstein molecular crystal model is indeed diffusive when the particle is in equilibrium with the oscillators at some strictly positive temperature. The appearance of the different power laws in temperature at low and high temperatures and of the thermal activation was theoretically explained in [SPD] by analyzing the Hamiltonian dynamics of the system. The high temperature behaviour is the easiest to understand. We give a simple version of the argument here, since we will use a variant of it in Section 4. The reasoning starts from the observation that at high temperatures the typical kinetic energy of the particle (which is kT) is much higher than the typical barrier height it encounters (which is of the order of $\sqrt{kT}$). As a result, typically a particle will traverse many cells before slowing down enough to receive a randomizing "kick" from one of the oscillators. This implies that it is reasonable to apply a relaxation time approximation in the following form. One computes easily (see [DPS]) that the average energy loss of a fast particle of velocity v when traversing one oscillator cell is

$$\frac{\Delta E}{\Delta n} = -\frac{4E_B E_0}{v^2}. \tag{4}$$

From this

$$\frac{\Delta v}{\Delta t} = \frac{\Delta v}{\Delta n} \frac{\Delta n}{\Delta t} = \frac{1}{v} \frac{\Delta E}{\Delta n} \frac{v}{a}.$$

Integrating this relation, the characteristic time a particle of high initial speed v needs to slow down is estimated to be

$$\tau(v) = \frac{v^3 a}{12 E_B E_0},$$

and the distance it travels while slowing down then turns out to be

$$\ell(v) = \frac{1}{4}v\tau(v).$$

Averaging this over the thermal distribution yields the following estimates for the mean free time and the mean free path squared of the particle:

$$\tau = \frac{a}{3E_0}\sqrt{\frac{E_B}{2\pi}}(\beta E_B)^{-3/2}, \qquad \ell^2 = 105\left(\frac{aE_0}{16E_B}\right)^2(\beta E_B)^{-4}$$

From this, one obtains for the diffusion constant the estimate

$$D = \frac{\ell^2}{2\tau} = \frac{105}{16^2}a\sqrt{9\pi E_B/2}\left(\frac{E_B}{E_0}\right)(\beta E_B)^{-5/2}.$$

This simple argument successfully explains the power law dependence on βE_B, and the numerical data presented in [SPD] show that the prefactor has the right order of magnitude. The simple derivation given above therefore contains the essential physics of the problem.

In [LPD] we study the motion of the particle when an external driving field F is applied, with the intent of checking if in this model linear response theory holds for the particle's average velocity and if, as a result, the Einstein relation between the mobility and the diffusion constant holds as well.

A first observation that we made in this context is that for this model, as for all models of the type (1), no stationary current can establish itself in the presence of the field, due to the following phenomenon. Indeed, we know from (4) that a particle of speed v will lose on average an energy $\Delta E \sim v^{-2}$ when traversing one cell. On the other hand, the particle picks up a potential energy Fa from the field when traversing the cell, so that whenever $Fa > \Delta E$, which will always be the case for fast enough particles, the result will be a net acceleration of the particle. Now any thermal distribution of particles contains particles with arbitrary high momenta, and those will therefore be accelerated indefinitely by the field F, however small it is. So one may expect that, for times beyond some critical time t_F, the thermally averaged particle position $\langle q_S(t, F)\rangle$ is of order t^2. Here $q_S(t, F)$ is the particle's position at time t, under the Hamiltonian evolution of the system in the presence of the driving field F.

In this sense, one may fear that models of this type can never have good transport properties. The situation is however more subtle than that, as we now explain. Indeed, as shown in [LPD], t_F behaves like $\exp(c/F)$ for some $c > 0$, and is therefore very large at small fields. It is then further shown numerically in [LPD] that on time scales shorter than the critical time t_F, the thermally averaged mean velocity $v_F(t)$ of the particle, defined via

$$v_F(t) = \frac{\langle q_S(t, F)\rangle}{t}$$

is constant in time, and proportional to the applied field. Here $\langle \cdot \rangle$ refers to the thermal averaging. In fact, linear response theory applied to this system yields

Kubo's formula in the form

$$\frac{v_F(t)}{F} = \beta \frac{\langle q_S^2(t, F = 0)\rangle}{2t} + O_t(F).$$

Our numerical findings indicate that the error term can be expected to be uniform for $t << t_F$. So, taking $t \to +\infty$ and $F \to 0$ in such a way that $t/t_F \to 0$, one may expect to find that the right hand side has a limit since in absence of the field the motion is diffusive, as shown above. This then yields the Einstein relation for the mobility μ of the system in the form:

$$\mu := \lim_{t \to \infty, t/t_F \to 0} \frac{\langle q_S(t, F)\rangle}{Ft} = \beta D.$$

Again, these statements lack rigorous proof, but are supported by the numerical evidence presented in [LPD].

4 Quantum transport in the Holstein molecular crystal model

The Holstein molecular crystal model is a well known, much studied and simple model for electron transport in the presence of electron-phonon interactions [H]. Its Hamiltonian is given by

$$\begin{aligned}
H_{\alpha,0} &= H_{\text{el}} + H_{\text{ph}} + \alpha H_{\text{int}} \\
&= V \sum_m (|m\rangle\langle m+1| + |m+1\rangle\langle m|) + \omega \sum_m a_m^\dagger a_m + \alpha \sum_m |m\rangle\langle m|Q_m,
\end{aligned}$$
(5)

where $Q_m = \frac{1}{\sqrt{2}}(a_m^\dagger + a_m)$ is the displacement of the oscillator at site m. This Hamiltonian describes the motion of a particle in a tight-binding band, linearly coupled at each site of a one-dimensional crystal to a single vibrational mode of frequency ω.

The Hilbert space $\mathcal{H}$ on which this Hamiltonian acts is

$$\mathcal{H} = \mathcal{H}_{\text{el}} \otimes \mathcal{H}_{\text{ph}} = \ell^2(\mathbb{Z}) \otimes \mathcal{F}(\ell^2(\mathbb{Z})).$$

The first factor $\ell^2(\mathbb{Z})$ corresponds to the Hilbert space for the electronic degree of freedom, whereas $\mathcal{F}(\ell^2(\mathbb{Z}))$ is the symmetric Fock space over $\ell^2(\mathbb{Z})$, describing the harmonic degrees of freedom representing the phonons. In the usual language of solid state physics, $a_m^\dagger$ and a_m correspond to the creation and annihilation operators for a phonon at site $m \in \mathbb{Z}$ of the lattice. We have used the bra-ket notation with $|m\rangle$ ($m \in \mathbb{Z}$) designating the canonical basis vectors of $\ell^2(\mathbb{Z})$, so that H_{el} designates the standard tight-binding Hamiltonian with nearest neighbour coupling only (*i.e.* the discrete Laplacian). We will systematically omit the tensor product notation when no confusion can arise. In particular,

when A is an operator on $\mathcal{H}_{el}$, we will write A as well for the operator $A \otimes \mathbb{1}$ on $\mathcal{H} = \mathcal{H}_{el} \otimes \mathcal{H}_{ph}$. Similarly, when B is an operator on $\mathcal{H}_{ph}$, we write B for $\mathbb{1} \otimes B$.

It is clear from the above that the Hamiltonian (5) has the typical structure of an open system Hamiltonian, in which a "small system", here the electron, is coupled to a heat bath, here modeled by the oscillators. It is also clear that the Hamiltonian (5) is a quantum mechanical version of (1), except for the fact that the kinetic energy term $p_S^2/2$ there is replaced here by a tight-binding dispersion relation of the form $2V(1 - \cos k)$ where $\hbar a k = p_S$.

A first question that comes to mind is the following one. If initially, the oscillators are in thermal equilibrium at inverse temperature β and the particle is injected into the lattice in an initially localized state close to $m = 0$, what is the asymptotic behaviour in time of the particle's mean square displacement $\langle X^2(t) \rangle$? Here X is the particle's position operator, defined on $\ell^2(\mathbb{Z})$ by

$$X = \sum_m m |m\rangle\langle m|.$$

Note that in the absence of the electron-phonon interaction (meaning that the coupling constant α in (5) vanishes), the answer to this question is trivial since then the motion is ballistic so that $X^2(t)$ behaves like t^2. But the coupling to the phonons introduces a scattering mechanism into the dynamics, and one may expect that now the motion of the particle becomes diffusive, meaning that

$$\langle X^2(t) \rangle \sim 2Dt,$$

for some diffusion constant $D > 0$, depending on β and on the parameters of the model, a situation that can be refered to loosely as "quantum Brownian motion". As far as we can tell, and in spite of the simplicity of the model, this expectation is not as yet corroborated by rigorous proof, nor even by numerical results. Indeed, if one works numerically on an M-site lattice and truncates the harmonic oscillator Hamiltonians at each site to N levels, one is still dealing with a linear space of size M^N which is too large to be tractable on today's computers, especially since one needs to study the dynamics for large M and long times.

The numerical results obtained for the classical model described in the previous section are an indication that the particle does indeed diffuse. One should nevertheless expect that the different band structure of the free particle Hamiltonian leads to a different temperature dependence of the diffusion constant, as we now argue for the high temperature regime. Consider for that purpose a semi-classical version of the Holstein molecular crystal model in which

$$H = 2V(1 - \cos \frac{a p_S}{\hbar}) + \sum_{m \in \mathbb{Z}} \frac{1}{2} \left(p_m^2 + \omega^2 q_m^2 \right) + \alpha \sum_m q_m n_m (q_S) - F q_S, \quad (6)$$

where $H_S = 2V(1 - \cos \frac{a p_S}{\hbar})$ is the standard one-band dispersion relation. Note that, beyond E_B and E_0, the model has an extra parameter with the dimension

of an energy, namely the bandwidth $4V$ and that the electron has a maximal possible speed, given by $\frac{2Va}{\hbar}$. This model reduces to (3) in the regime $\beta E_B << 1 << \beta V, \beta E_0 << 1$, in which the particle's thermal energy is much smaller than the bandwidth $4V$, but much larger than the other characteristic energies E_B, E_0 of the model. We will now consider a different parameter regime, as follows. We will first of all suppose the electron band is large, so that $V >> \hbar\omega$, which means that the fastest particles cross the interaction region in a time much shorter than the oscillator period. We consider in addition the situation where the temperature is high, in the sense that $\beta V << 1$. In this asymptotic regime the Boltzmann factor for the particle, given by $\frac{1}{Z}\exp-\beta H_S$, converges to a constant, so that the momentum is distributed uniformly in the first Brillouin zone. We finally suppose that the electron-phonon coupling is weak, meaning that $\frac{E_B}{V} << \beta V$. This last condition guarantees that the typical barrier height seen by the particle, which is of the order $(E_B kT)^{1/2}$ is much less than V, which is the typical particle energy in the regime considered. The analysis of the previous section concerning the energy loss of fast particles now applies and in particular (4) holds, but with $v = \frac{2Va}{\hbar}\sin\left(\frac{ap_S}{\hbar}\right)$. Since this time the Boltzmann distribution becomes temperature independent in the regime considered, this argument predicts that the mean free time, the mean free path and hence the diffusion constant also saturate to a temperature independent value. This leads for D to the order of magnitude estimate

$$D \sim \frac{a^4 V^4}{\hbar^3 E_B E_0}.$$

Let us now return to the full quantum mechanical model (5). Beyond the above question about diffusion, the second problem that presents itself naturally is what will happen to the particle dynamics if one adds an external electric field F to the Hamiltonian in (5) to obtain

$$H_{\alpha,F} = H_\alpha - FX. \tag{7}$$

It should be noted that this time again, when $\alpha = 0$, the dynamics can be computed explicitly and one has that

$$\sup_t \langle |X(t)| \rangle < +\infty,$$

a phenomenon due to the Bloch oscillations inherent to a tight binding model. So the dc electric field F actually localizes the particle, which had a ballistic behaviour before the field was turned on. This is of course very different from what happens in the classical model with Hamiltonian (3) but it also occurs for the semi-classical model (6). The common wisdom concerning the particle's asymptotic behaviour is now that the phonon scattering is responsible for a diffusion of the particle's quasi-momentum in the Brillouin zone (which here is simply $[0, 2\pi/a]$). This combines with a slight bias on the quasi-momentum induced by F, leading finally to a drift of the particle's average position, so that

$$\langle X(t) \rangle \sim v_F t.$$

11

A formal application of Kubo's linear response theory suggests that v_F should be linear in F at small F so that the mobility $\mu = \lim_{F \to 0} \frac{v_F}{F}$ is well defined. A further question is what the precise relation of this mobility to the diffusion constant is, in the present quantum context. Let us mention that in the semi-classical model above, we do not expect runaway to occur for small enough fields, since the condition $Fa < \Delta E$ can now be satisfied because ΔE has a strictly positive lower bound which is reached when the particle attains its maximal speed $2V$.

There are in fact, to the best of our knowledge, no quantum mechanical models at all in which such behaviour has been rigorously proven to occur. Indeed, in the quantum mechanical context, virtually all rigorous work in which an electron is coupled to a heat bath of oscillators is done under the hypothesis the electron is confined, or more restrictively still, only has a finite number of energy levels. A dipole approximation is also often made. As already pointed out, for the transport problems we have addressed here, this is inadequate: the particle is then not confined and it is natural to suppose that the interaction between the particle and the heat bath is translationally invariant, so that the dipole approximation is inappropriate. A rigorous approach to transport theory via quantum kinetic theory is reviewed in [B]. In this work the scattering mechanism with the phonons is however taken into account phenomenologically, through the relaxation-time approximation for example, rather than through a fully Hamiltonian treatment of the coupled system consisting of the electron and the phonons. In particular, transport coefficients such as the mean free time or the diffusion constants are not computed from first principles but added as free parameters to the models.

Several variations of the above questions are of obvious interest, in particular with respect to Anderson localization and random Hamiltonians. For that purpose one may consider the same Hamiltonian, to which an external random potential is added:

$$
\begin{aligned}
H_{\alpha,F}^{\lambda} &= H_{\alpha,F} + \lambda W \\
&= V \sum_m \left(|m\rangle\langle m+1| + |m+1\rangle\langle m| \right) + \lambda \sum_m w_m |m\rangle\langle m| - FX \\
&\qquad + \omega \sum_m a_m^\dagger a_m + \alpha \sum_m |m\rangle\langle m| Q_m. \quad (8)
\end{aligned}
$$

Here the w_m are a family of independent identically distributed random variables. The question of interest is now to understand how the behaviour of the particle changes when $\lambda \neq 0$ from the one described above when $\lambda = 0$. For that purpose, recall first that when $\alpha = 0 = F$, we recover the usual Anderson Hamiltonian for which dynamical localization is a well-understood phenomenon:

$$
\sup_t \langle |X(t)^2| \rangle < \infty. \quad (9)
$$

What happens now when the phonon scattering is turned on, meaning that $\alpha \neq 0$? Presumably, the electrons remain dynamically localized in the above

sense, or perhaps they will undergo a slow subdiffusive motion. And now, even if F is turned on, no drift should install itself, so that, at least

$$\limsup_{t\to+\infty} \frac{\langle X(t)\rangle_\beta}{t} = 0. \tag{10}$$

True to the common wisdom according to which one fool (and a fortiori three fools) can ask more questions than ten wise men can answer, it is easily seen that one can change the model by replacing the single oscillator at each site by a collection of such oscillators, or more generally by a free Bose field. For mathematically rigorous work the use of a continuous frequency spectrum can be expected to be important to ensure good ergodic properties of the heat bath. One can further consider the same set of questions when a time-dependent electric field is used or write down the analogous continuum models where the electron's configuration space is $\mathbb{R}$ rather than $\mathbb{Z}$. The answers in that latter case could be quite different since now the free particle undergoes a uniform acceleration when the field is turned on, in contrast to the confined motion induced by the Bloch oscillations.

At any rate, unable to answer any of the above queries, we have turned our attention to a similar set of questions, but in analogous classical Hamiltonian systems, and obtained a few answers, as described in the previous section.

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