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An impurity atom in a lattice of harmonically coupled atoms and the stationary generalized Langevin equation II

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Abstract. A generalized Langevin equation is set up for: (i) the velocity of an impurity atom; and (ii) the set of the displacement of an impurity atom and its velocity. The friction function $\Gamma^{\mathrm{I}}(t)$ for the former equation tends to a nonzero value at $t \to \infty$, if the impurity atom is localized in space and if the limiting value of $\Gamma^{\mathrm{I}}(t)$ at $t \to \infty$ exists. The friction function $\Gamma(t)$ for the latter equation always decays to zero, if its limit at large t exists. When the impurity atom is not localized in space, the latter equation is reduced to the former. The decay, and its rapidity, of the friction function $\Gamma(t)$ for the latter equation are investigated for an impurity atom in a lattice of harmonically coupled atoms, and the applicability of the stationary generalized Langevin equation is discussed for the impurity atom.

1. Introduction

In a preceding paper (Fukui and Morita 1971), the present authors derived the *stationary* generalized Langevin equation. In their succeeding paper (Fukui and Morita 1972, to be referred to as I), they discussed the applicability of that equation to the motion of an impurity atom in a lattice of harmonically coupled atoms. The system is a slight extension of the system investigated by Rubin (1960, 1961) and others as a model of the brownian motion. In Mori's formalism (Mori 1965), the generalized Langevin equation for the velocity $v(t)$ of the impurity atom is given by

$$\frac{\mathrm{d}}{\mathrm{d}t}v(t) = -\int_0^t \Gamma^{\mathrm{I}}(t-t')v(t')\,\mathrm{d}t' + R^{\mathrm{I}}(t). \tag{1.1}$$

The friction function $\Gamma^{\mathrm{I}}(t)$ is connected with the random force $R^{\mathrm{I}}(t)$ by the relation

$$\Gamma^{\mathrm{I}}(t) = \frac{\langle R^{\mathrm{I}}(t), R^{\mathrm{I}}\rangle}{\langle v, v\rangle}. \tag{1.2}$$

The present authors (Fukui and Morita 1971) argued that, if the friction functions occurring in the generalized Langevin equation decay to zero *fast*, the corresponding *stationary* generalized Langevin equation is valid. For the present case, if $\Gamma^{\mathrm{I}}(t)$ decays to zero fast, one obtains

$$\frac{\mathrm{d}}{\mathrm{d}t}v(t) = -\int_{-\infty}^t \Gamma^{\mathrm{I}}(t-t')v(t')\,\mathrm{d}t' + R_{\mathrm{s}}^{\mathrm{I}}(t) \tag{1.3}$$

and

$$\Gamma^{\mathrm{I}}(t) = \frac{\langle R_{\mathrm{s}}^{\mathrm{I}}(t), R_{\mathrm{s}}^{\mathrm{I}}\rangle}{\langle v, v\rangle}. \tag{1.4}$$

The present authors showed in I that the friction function $\Gamma^{\mathrm{I}}(t)$ decays to zero fast enough for the impurity atom in a one-dimensional lattice of infinite length if there exists no coupling of atoms to fixed points, and they concluded that the stationary generalized Langevin equation is obtained for this system. For the simple cubic lattice, the friction function $\Gamma^{\mathrm{I}}(t)$ is found not to decay to zero. For any lattice $\Gamma^{\mathrm{I}}(t)$ does not decay to zero if there exists a coupling of atoms to their respective equilibrium points. For the square lattice of infinite edge length, if there exists no coupling of atoms to fixed points, $\Gamma^{\mathrm{I}}(t)$ decays to zero but the decay is not fast enough.

In the present paper, we investigate an impurity atom in a system in thermal equilibrium. The generalized Langevin equation (1.1) is obtained under the assumption that

$$\langle \dot{v}, v \rangle = 0 \tag{1.5}$$

where $\dot{v} = \dot{v}(0)$ and $\dot{v}(t) = \mathrm{d}v(t)/\mathrm{d}t$. We further assume that the correlation function of the displacement $x(t)$ of the impurity atom decays to zero:

$$\lim_{t\to\infty} \langle x(t), x(0) \rangle = 0. \tag{1.6}$$

Discussion is given of this assumption at the end of § 2. We then prove that, if the limiting value of $\Gamma^{\mathrm{I}}(t)$ at large time t exists, it is given by

$$\lim_{t\to\infty} \Gamma^{\mathrm{I}}(t) = \frac{\langle v, v \rangle}{\langle x, x \rangle}. \tag{1.7}$$

(1.7) means that $\Gamma^{\mathrm{I}}(t)$ does not decay to zero as far as $\langle x, x \rangle$ is finite. Note that $\langle x, x \rangle$ can become infinity only for an infinite system. Thus we conclude that $\Gamma^{\mathrm{I}}(t)$ decays to zero only if the impurity atom is not localized in space in an infinite system.

Next, we set up the generalized Langevin equation for the set of the displacement $x(t)$ and its velocity $v(t)$. By assuming (1.5) and

$$\langle x, v \rangle = 0 \tag{1.8}$$

we obtain the following set of equations:

$$\frac{\mathrm{d}}{\mathrm{d}t} x(t) = v(t) \tag{1.9}$$

$$\frac{\mathrm{d}}{\mathrm{d}t} v(t) = -\frac{\langle v, v \rangle}{\langle x, x \rangle} x(t) - \int_0^t \Gamma(t-t') v(t') \, \mathrm{d}t' + R(t). \tag{1.10}$$

By assuming (1.6), we show that $\Gamma(t)$ always decays to zero:

$$\lim_{t\to\infty} \Gamma(t) = 0 \tag{1.11}$$

if the limit exists. The relation between $\Gamma(t)$ and $\Gamma^{\mathrm{I}}(t)$ is given by

$$\Gamma(t) = \Gamma^{\mathrm{I}}(t) - \frac{\langle v, v \rangle}{\langle x, x \rangle}. \tag{1.12}$$

Substitution of this relation into (1.10) and comparison of the result with (1.1) shows that

$$R^{\mathrm{I}}(t) = R(t) - \frac{\langle v, v \rangle}{\langle x, x \rangle} x(0). \tag{1.13}$$

By the relations (1.12) and (1.13), we see that (1.10) reduces to (1.1) if $\langle x, x \rangle = \infty$.

The above discussion, the details of which are given in §§ 2 and 3, shows that in the case when the impurity atom is localized in space and if the displacement is not included in the dynamical variables $X_j(t)$ for which the generalized Langevin equation is written, the memory of the *initial* displacement is included in the random part of the force and the correlation of that force, or equivalently $\Gamma^{\mathrm{I}}(t)$, cannot decay to zero. If we require that the correlation of random force should decay to zero, we have had to include the displacement in the set of dynamical variables $X_j(t)$.

In order to give the stationary generalized Langevin equation, a fast decay of $\Gamma(t)$ is required. We discuss the asymptotic behaviour of $\Gamma(t)$ for an impurity atom in harmonic lattices in §§ 4 and 5.

2. The generalized Langevin equation for the velocity of an impurity atom

We investigate the generalized Langevin equation for an impurity atom in a system in thermal equilibrium. We shall first set up the generalized Langevin equation for the velocity $v(t)$ of the impurity atom. Assuming (1.5), we obtain the equation (1.1) with the relations (1.2) and

$$\langle R^{\mathrm{I}}(t), v(0)\rangle = 0. \tag{2.1}$$

Taking a correlation of (1.1) with $v(0)$ and then a Laplace transform, we have the Laplace transform of the two-time correlation function $\langle v(t), v(0)\rangle$ in the following form:

$$\langle v, v\rangle_z = \frac{\langle v, v\rangle}{z+\Gamma^{\mathrm{I}}_z} \tag{2.2}$$

where $\langle v, v\rangle_z$ and Γ^{I}_z are the Laplace transforms of $\langle v(t), v\rangle$ and $\Gamma^{\mathrm{I}}(t)$, respectively.

Solving (2.2) for Γ^{I}_z, we have

$$\Gamma^{\mathrm{I}}_z = -z+\frac{\langle v, v\rangle}{\langle v, v\rangle_z}. \tag{2.3}$$

If the limiting value $\lim_{t\to\infty}\Gamma^{\mathrm{I}}(t)$ exists, it is related with the Laplace transform Γ^{I}_z by

$$\lim_{t\to\infty}\Gamma^{\mathrm{I}}(t) = \lim_{z\to 0^+} z\Gamma^{\mathrm{I}}_z \tag{2.4}$$

(Widder 1946)†. Using (2.3) in (2.4) we have

$$\lim_{t\to\infty}\Gamma^{\mathrm{I}}(t) = \left(\lim_{z\to 0^+}\frac{z}{\langle v, v\rangle_z}\right)\langle v, v\rangle. \tag{2.5}$$

Here we shall present an identity:

$$\langle x, x\rangle = \lim_{z\to 0^+}\frac{\langle v, v\rangle_z}{z}. \tag{2.6}$$

† Theorem 1 on p 181 or corollary 1*a* on p 182 of Widder's book (1946) shows that

$$\lim_{s\to 0^+} f(s) = \lim_{t\to\infty}\alpha(t).$$

whenever the limit on the right-hand side exists, where

$$f(s) = s\int_0^\infty \alpha(t)\,\mathrm{e}^{-st}\,\mathrm{d}t.$$

A proof of this identity is as follows:

$$\begin{aligned}\lim_{z\to 0^+}\frac{\langle v, v\rangle_z}{z} &= \lim_{t\to\infty}\int_0^t \mathrm{d}t_1 \int_0^{t_1} \mathrm{d}t_2\langle v(t_2), v(0)\rangle \\ &= \lim_{t\to\infty}\int_0^t \mathrm{d}t_1\langle x(t_1)-x(0), v(0)\rangle \\ &= \lim_{t\to\infty}\int_0^t \mathrm{d}t_1\langle x(0), v(-t_1)\rangle \\ &= \lim_{t\to\infty}\int_{-t}^0 \mathrm{d}t_1\langle x(0), v(t_1)\rangle \\ &= \langle x(0), x(0)\rangle - \lim_{t\to\infty}\langle x(0), x(-t)\rangle \\ &= \langle x, x\rangle.\end{aligned}$$

This proof shows that (2.6) follows if $\langle x, v\rangle = 0$ and

$$\lim_{t\to\infty}\langle x(t), x(0)\rangle = 0 \tag{2.7}$$

are assumed.

Substituting (2.6) into (2.5), we have

$$\lim_{t\to\infty}\Gamma^{\mathrm{I}}(t) = \frac{\langle v, v\rangle}{\langle x, x\rangle}. \tag{2.8}$$

This shows that $\Gamma^{\mathrm{I}}(t)$ decays to zero only if $\langle x, x\rangle = \infty$. As noticed in § 1, this occurs only for an infinite system when the impurity atom is not localized in space.

We consider that the assumption (2.7) is related to the choice of the origin of the coordinate x. If the system is ergodic and the left-hand side of (2.7) tends to $\langle x\rangle^2$, the origin of the coordinate must be chosen such that $\langle x\rangle = 0$. If an infinite system is investigated, the thermodynamic limit must be taken in a fashion that guarantees the assumption (2.7).

3. Equations for the velocity and the displacement of an impurity atom

In this section, we set up the generalized Langevin equation in Mori's formalism (Mori 1965) for the set of the displacement $x(t)$ of an impurity atom and its velocity $v(t)$. The notation used follows Fukui and Morita (1971). In setting up the generalized Langevin equation, the projection operator P is introduced, which satisfies

$$(1-P)v\rangle = 0$$

and

$$\langle v(1-P) = 0. \tag{3.1}$$

When the equation of motion is written for the set of $x(t)$ and its time derivative $v(t)$, the equation for $x(t)$ must naturally be given by

$$\frac{\mathrm{d}}{\mathrm{d}t}x(t) = v(t). \tag{3.2}$$

In fact the random force $R_x(t)$ corresponding to the variable $x(t)$ is identically zero:

$$R_x(t) \equiv 0 \tag{3.3}$$

as seen by a comparison of its definition:

$$\langle R_x(t) = \langle xL(1-P)\exp\{L(1-P)t\}$$
$$R_x\rangle = (1-P)Lx\rangle$$

with (3.1). Here L is the Liouville operator and $\langle xL = \langle v \text{ and } Lx\rangle = v\rangle$.

For the static correlation functions, we assume (1.5) and (1.8). By using (3.3), we find that the generalized Langevin equations for the set of $x(t)$ and $v(t)$ are given by (3.2) and the following equation:

$$\frac{\mathrm{d}}{\mathrm{d}t}v(t) = \Omega x(t) - \int_0^t \Gamma(t-t')v(t')\,\mathrm{d}t' + R(t) \tag{3.4}$$

where

$$\Omega\langle x, x\rangle = \langle \dot{v}, x\rangle = -\langle v, v\rangle \tag{3.5}$$

$$\Gamma(t) = \frac{\langle R(t), R\rangle}{\langle v, v\rangle}. \tag{3.6}$$

By taking a correlation of (3.2) and (3.4) with $v(0)$ and then a Laplace transform, we obtain

$$\langle v, v\rangle_z = \langle v, v\rangle\left(z + \frac{1}{z}\frac{\langle v, v\rangle}{\langle x, x\rangle} + \Gamma_z\right)^{-1} \tag{3.7}$$

or

$$\Gamma_z = -z + \frac{\langle v, v\rangle}{\langle v, v\rangle_z} - \frac{1}{z}\frac{\langle v, v\rangle}{\langle x, x\rangle}. \tag{3.8}$$

Comparison of (3.8) and (2.3) shows that

$$\Gamma_z = \Gamma_z^{\mathrm{I}} - \frac{1}{z}\frac{\langle v, v\rangle}{\langle x, x\rangle}. \tag{3.9}$$

By (2.6) and (2.3), this reads

$$\Gamma_z = \Gamma_z^{\mathrm{I}} - \frac{1}{z}\lim_{z\to 0^+}(z\Gamma_z^{\mathrm{I}}). \tag{3.10}$$

The inverse Laplace transform of (3.9) gives

$$\Gamma(t) = \Gamma^{\mathrm{I}}(t) - \frac{\langle v, v\rangle}{\langle x, x\rangle}. \tag{3.11}$$

If $\lim_{t\to\infty}\Gamma(t)$ exists, it is given by $\lim_{z\to 0^+} z\Gamma_z$. (3.10) then shows that

$$\lim_{t\to\infty}\Gamma(t) = \lim_{z\to 0^+} z\Gamma_z = 0. \tag{3.12}$$

$\Gamma(t)$ occurring in (3.4) always decays to zero, if its limit at $t \to \infty$ exists.

Substitution of (3.11) into (3.4) and comparison of the result with (1.1) show that

$$R^{\mathrm{I}}(t) = R(t) - \frac{\langle v, v\rangle}{\langle x, x\rangle} x(0). \tag{3.13}$$

Discussion of the above calculations is given at the end of § 1.

4. Application to an impurity atom in a *finite* harmonic lattice

In the preceding paper I, we investigated the time dependence of $\Gamma^{\mathrm{I}}(t)$ for an impurity atom in a harmonic lattice. In this section, we investigate $\Gamma(t)$ for an impurity atom in a *finite* harmonic lattice. The hamiltonian of the system is given by

$$H = \sum_R \left(\frac{p_R^2}{2m_R} + \frac{1}{2} k'_R x_R^2 \right) + \sum_R \sum_a \frac{k}{4} (x_{R+a} - x_R)^2 \tag{4.1}$$

where x_R and p_R are the displacement and the canonical conjugate momentum of the atom at the lattice site R. The atom at the origin is the impurity. $m_R = M(R = 0)$ is the impurity mass and $m_R = m(R \neq 0)$ is the mass of host atoms. The coupling constants $k'_R = K'(R = 0)$ and $k'_R = k'(R \neq 0)$ are zero or positive. The coupling constant k is positive. a is the vector from a lattice site to its nearest neighbours. The summation over a is taken over all nearest neighbours.

The equation of motion for the atom at the lattice site R is written as

$$m_R \ddot{x}_R(t) = k \sum_a x_{R+a}(t) - (z_c k + k'_R) x_R(t) \tag{4.2}$$

where z_c is the coordination number of the lattice. Taking a correlation of this equation with $\dot{x}_0(0)$ and a Laplace transform with respect to time, and then multiplying by z/m, one obtains

$$\left(z^2 + \frac{z_c k + k'}{m} \right) \langle \dot{x}_R, \dot{x}_0 \rangle_z - \frac{k}{m} \sum_a \langle \dot{x}_{R+a}, \dot{x}_0 \rangle_z$$
$$= \left[- \left\{ \left(\frac{M}{m} - 1 \right) z^2 + \frac{K' - k'}{m} \right\} \langle \dot{x}_0, \dot{x}_0 \rangle_z + \frac{zM \langle \dot{x}_0, \dot{x}_0 \rangle}{m} \right] \delta_{R0}. \tag{4.3}$$

The resolvent $R(z; R)$ is introduced by

$$\left(z^2 + \frac{z_c k + k'}{m} \right) R(z; R) - \frac{k}{m} \sum_a R(z; R + a) = \delta_{R0}. \tag{4.4}$$

We shall adopt the periodic boundary condition. Then (4.4) is solved for $R(z; R)$ as follows:

$$R(z; R) = \frac{1}{N+1} \sum_K \exp(\mathrm{i}K \cdot R) \left(z^2 + \frac{z_c k + k'}{m} - \frac{k}{m} \sum_a \exp(\mathrm{i}K \cdot a) \right)^{-1} \tag{4.5}$$

where $N + 1$ is the total number of the atoms in the system. Equation (4.3) is solved in terms of $R(z; R)$ as

$$\langle \dot{x}_0, \dot{x}_0 \rangle_z = \frac{zM \langle \dot{x}_0, \dot{x}_0 \rangle}{m} \left\{ \left(\frac{M}{m} - 1 \right) z^2 + \frac{K' - k'}{m} + \frac{1}{R(z; 0)} \right\}^{-1}. \tag{4.6}$$

Comparing this expression with (3.7) and identifying $\dot{x}_0$ with v, we obtain

$$\Gamma_z = \frac{m}{Mz}\left(\frac{1}{R(z;0)} - \frac{1}{R(0;0)} - z^2\right) \tag{4.7}$$

with the aid of the relation (3.12). This shows that Γ_z has simple poles at the zeros of $R(z;0)$. The total number of the zeros is $N+1$ or N for the linear chain according to whether $N+1$ is even or odd. One easily sees that the number is less than $N+1$ also for the two- and three-dimensional lattices. Those zeros are on the imaginary axis within the ranges from $\mathrm{i}(k'/m)^{1/2}$ to $\mathrm{i}\{(2z_c k+k')/m\}^{1/2}$ and from $-\mathrm{i}(k'/m)^{1/2}$ to $-\mathrm{i}\{(2z_c k+k')/m\}^{1/2}$. As a result, $\Gamma(t)$ is expressed as a linear combination of a finite number of functions of the form

$$\exp(\mathrm{i}y_c t) \tag{4.8}$$

where y_c is a real number. $\Gamma(t)$ is not expected to decay to zero and will always behave irregularly.

5. Application to an impurity atom in an *infinite* harmonic lattice

Behaviours of an impurity atom in an *infinite* harmonic lattice are obtained from the corresponding behaviours given in the preceding section for the finite lattice by taking the limit as $N \to \infty$. In particular, the Laplace transform Γ_z of the friction function $\Gamma(t)$, which occurs in the generalized Langevin equation of the velocity of the impurity atom, is obtained from (4.7) as follows:

$$\Gamma_z = \frac{m}{zM}\left(G_d\left(z^2 + \frac{z_c k+k'}{m};0\right)^{-1} - G_d\left(\frac{z_c k+k'}{m};0\right)^{-1} - z^2\right). \tag{5.1}$$

Here $G_d(\omega;R)$ is the lattice Green function which is the limit of (4.5):

$$G_d(\omega;R) = \frac{1}{v_d}\int_\Omega \mathrm{d}K \exp(\mathrm{i}K\,.\,R)\left(\omega - \frac{k}{m}\sum_a \exp(\mathrm{i}K\,.\,a)\right)^{-1}$$

where d is the dimension of the lattice. The integral is taken over the first one or several Brillouin zones and v_d is its volume. For the linear chain, Γ_z reads

$$\Gamma_z = \frac{m}{zM}\left\{\left(z^2+\frac{k'}{m}\right)^{1/2}\left(z^2+\frac{k'}{m}+\frac{4k}{m}\right)^{1/2} - \left(\frac{k'}{m}\right)^{1/2}\left(\frac{k'}{m}+\frac{4k}{m}\right)^{1/2} - z^2\right\}. \tag{5.2}$$

In fact, one obtains this result by substituting the Γ_z^{I} given in I into (3.10).

It has been proved that the lattice Green function $G_d(\omega;0)$ is analytic in the whole complex ω plane except for real ω within the range between $-z_c k/m$ and $z_c k/m$ (Morita and Horiguchi 1972). We show in appendix 1 that the same is true for the inverse $G_d(\omega;0)^{-1}$. The singularities of the lattice Green function, due to non-degenerate critical points, are given in the same paper. The singularities for the linear, square and simple cubic lattices are all due to non-degenerate critical points. From these singularities, we obtain the singularities of Γ_z.

For the linear and simple cubic lattices, the singularities of Γ_z occur on nonzero imaginary values of z. The singular behaviours are branch points of the form:

$$\Gamma_z \sim A(z-\mathrm{i}y_c)^{1/2} \qquad z \sim \mathrm{i}y_c \tag{5.3}$$

where y_c is real and nonzero and A is a constant. Here the branch of the square root is so chosen that $(z-iy_c)^{1/2}$ is real and positive when $z-iy_c$ is real and positive. We adopt a similar choice of the branch for $\ln(z-iy_c)$ in the following. The inverse Laplace transform gives

$$-\frac{A}{2\sqrt{\pi}}\frac{1}{t^{3/2}}\exp(iy_c t) \tag{5.4}$$

as a contribution to the asymptotic behaviour of $\Gamma(t)$ from (5.3). In particular, for the linear chain with $k' = 0$, we have

$$\Gamma(t) = \frac{2k}{M}\frac{J_1(2t\sqrt{(k/m)})}{t\sqrt{(k/m)}}. \tag{5.5}$$

The above result (5.4) shows that $\Gamma(t)$ for the linear and simple cubic lattices decay fast enough that

$$\int_0^\infty |\Gamma(t)|\,dt < \infty. \tag{5.6}$$

For the square lattice with $k' \neq 0$, the singularities of Γ_z occur only on nonzero imaginary values of z and they are branch points of the form:

$$\Gamma_z \sim \frac{A}{\ln(z-iy_c)} \qquad z \sim iy_c. \tag{5.7}$$

The asymptotic behaviour of the inverse Laplace transform due to this singularity is found to be given by

$$\frac{A}{t(\ln t)^2}\exp(iy_c t) \tag{5.8}$$

in appendix 2. The asymptotic behaviour of $\Gamma(t)$ for this case is a linear combination of terms of this form, and (5.6) is satisfied.

For the square lattice with $k' = 0$, $z = 0$ becomes a branch point of Γ_z and the singularity for it is

$$\Gamma_z \sim \frac{2\pi k}{Mz(-\ln z)} \qquad z \sim 0. \tag{5.9}$$

The asymptotic behaviour of the inverse Laplace transform of (5.9) is

$$\frac{2\pi k}{M \ln t}; \tag{5.10}$$

(cf appendix 2). The asymptotic behaviour of $\Gamma(t)$ is given by a linear combination of (5.10) and terms of the form (5.8). (5.10) takes a decisive role†, and (5.6) is not satisfied for this case.

The lattice Green functions for the FCC and BCC lattices involve singularities of the form $[\ln\{\omega+(4k/m)\}]^2$ and $(\ln\omega)^2$, respectively, in addition to the ones due to non-degenerate critical points (eg Morita and Horiguchi 1971). Those singularities give rise

† The coefficient of (5.9) in I is not correct. $ke\sqrt{(2\pi)}/M \ln t$ there should read $2\pi k/M \ln t$

Other errors in I are as follows. The minus sign in front of the second term in the braces of (4.8) should be replaced by a plus sign. (4.8) applies when $m/M < \frac{1}{2}$. When $\frac{1}{2} \leqslant m/M < 1$, the first term in the braces, the exponentially decaying term, should be omitted. For $1 < m/M$, a localized mode appears and $\langle \dot{x}_0(t), \dot{x}_0(0)\rangle$ keeps oscillating.

to branch points of the form

$$\Gamma_z \sim \frac{A}{\{\ln(z-iy_c)\}^2} \qquad z \sim iy_c \tag{5.11}$$

for Γ_z of those lattices, where y_c is real and nonzero. The asymptotic behaviour of the inverse Laplace transform of (5.11) is given by

$$-\frac{2A}{t(\ln t)^3}\exp(iy_c t) \tag{5.12}$$

(cf appendix 2). The asymptotic behaviour of $\Gamma(t)$ is now given by a sum of terms (5.12) and (5.4). (5.12) takes a decisive role. (5.6) is satisfied for the FCC and BCC lattices.

The above calculations show that the square lattice with $k' = 0$ is an exceptional case when (5.6) is not satisfied.

6. Conclusions

It is shown that if the generalized Langevin equation is set up for the variable $v(t)$, the friction function $\Gamma^{\mathrm{I}}(t)$ in that equation decays to $\langle v, v\rangle/\langle x, x\rangle$ if $\lim_{t\to\infty}\Gamma^{\mathrm{I}}(t)$ exists. Hence it can decay to zero only if $\langle x, x\rangle = \infty$, that is, for an infinite system in which the particle is not localized in space. If the equation is set up for the set of variables $v(t)$ and $x(t)$, the friction function $\Gamma(t)$ always decays to zero if $\lim_{t\to\infty}\Gamma(t)$ exists. When $\langle x,x\rangle = \infty$, this set of equations reduces to the previous one. The random force $R^{\mathrm{I}}(t)$ for the former equation is found to be a sum of the random force $R(t)$ for the latter equation and a constant $(-\langle v, v\rangle/\langle x, x\rangle)x(0)$. When the particle is localized in space but the spatial coordinate $x(t)$ is not included in the set of the dynamical variables for which the generalized Langevin equation is set up, the memory of the initial position is put in the random force $R^{\mathrm{I}}(t)$ and the correlation of $R^{\mathrm{I}}(t)$ and $R^{\mathrm{I}}(0)$ does not decay.

The asymptotic behaviour of $\Gamma(t)$ is investigated for an impurity atom in a harmonic lattice. The results for an infinite lattice are given as follows. For the linear and simple cubic lattices, the asymptotic behaviour of $\Gamma(t)$ is expressed as a linear combination of terms of the form $At^{-3/2}\exp(iy_c t)$. For the FCC and BCC lattices, it is given by a sum of the terms of the form $A\{t(\ln t)^3\}^{-1}\exp(iy_c t)$, and for the square lattice with $k' \neq 0$, by a sum of terms of the form $A\{t(\ln t)^2\}^{-1}\exp(iy_c t)$. For these cases, $\int_0^\infty |\Gamma(t)|\,dt$ converges and we consider that the *stationary* generalized Langevin equation of the following form is applicable to these systems:

$$\frac{d}{dt}x(t) = v(t) \tag{6.1}$$

$$\frac{d}{dt}v(t) = -\frac{\langle v, v\rangle}{\langle x, x\rangle}x(t) - \int_{-\infty}^{t}\Gamma(t-t')v(t')\,dt' + R_s(t). \tag{6.2}$$

The square lattice with $k' = 0$ is an exceptional case when $\Gamma(t) \sim A/\ln t$ and $\int_0^\infty |\Gamma(t)|\,dt$ does not converge. We cannot conclude the applicability of (6.2) for this case.

For an impurity atom in a finite lattice of $N+1$ atoms, $\Gamma(t)$ consists of at most $N+1$ terms of the form $\exp(iy_c t)$. It will stay finite forever, and (6.2) will not apply for this case.

Appendix 1. Analyticity of $G_d(\omega;0)^{-1}$

The lattice Green function at the origin takes the form

$$G_d(\omega;0)=\frac{1}{v_d}\int_\Omega \frac{1}{\omega-\epsilon(\boldsymbol{k})}\,\mathrm{d}\boldsymbol{k},$$

where $\epsilon(\boldsymbol{k})$ takes real values. The integral is taken over the first one or several Brillouin zones and v_d is its volume. The imaginary part of this expression is

$$\operatorname{Im} G_d(\omega;0) = -\operatorname{Im}\omega\frac{1}{v_d}\int_\Omega \frac{1}{(\operatorname{Re}\omega-\epsilon(\boldsymbol{k}))^2+(\operatorname{Im}\omega)^2}\,\mathrm{d}\boldsymbol{k}.$$

This quantity cannot be zero if Im ω is not zero. Thus we conclude that

$$G_d(\omega;0)\neq 0 \qquad \text{if} \qquad \operatorname{Im}\omega\neq 0.$$

Next, we consider real ω which is larger than the maximum ($\epsilon_{\max}$) of $\epsilon(\boldsymbol{k})$ and then

$$G_d(\omega;0)>0 \qquad \text{if} \qquad \omega>\epsilon_{\max}.$$

In a similar way, for real ω less than the minimum ($\epsilon_{\min}$) of $\epsilon(\boldsymbol{k})$, we have

$$G_d(\omega;0)<0 \qquad \text{if} \qquad \omega<\epsilon_{\min}.$$

As a result we conlude that $G_d(\omega;0)$ cannot have a zero in the whole complex ω plane excluding the real axis between $\epsilon_{\min}$ and $\epsilon_{\max}$. In the same region, $G_d(\omega;0)$ is known to be analytic (Morita and Horiguchi 1972). Hence we conclude that $G_d(\omega;0)^{-1}$ is also analytic in the same region.

The present proof can be extended to systems with many bands. In that case, there might occur one zero of $G_d(\omega;0)$ and hence one pole of $G_d(\omega;0)^{-1}$ between two successive bands. Except for those points, $G_d(\omega;0)^{-1}$ is analytic outside the bands.

Appendix 2. Long-time asymptotic behaviour of $\Gamma(t)$

We shall discuss the derivation of the long-time asymptotic behaviour of $\Gamma(t)$ from the analytic properties of its Laplace transform Γ_z. In § 5, we find that Γ_z is analytic except on the imaginary axis. Hence $\Gamma(t)$ is calculated from Γ_z by

$$\Gamma(t)=\frac{1}{2\pi\mathrm{i}}\int_C \Gamma_z\,\mathrm{e}^{zt}\,\mathrm{d}t. \tag{A.1}$$

Here C is the path from $\sigma-\mathrm{i}\infty$ to $\sigma+\mathrm{i}\infty$ parallel to the imaginary axis where $\sigma>0$. Let us assume for the purpose of illustration that Γ_z has four branch-points z_1, z_2, z_3 and z_4, and no poles, on the imaginary axis as shown in figure 1. We can easily confirm that Γ_z given by (5.1) tends to zero as $|z|\to\infty$. Then we deform the path C to the sum of the paths C_1, C_2, C_3, C_4 and C_5 and C_6 by virtue of Jordan's lemma. Here it is assumed that there occur two singularities z_5 and z_6 by the analytic continuation of the function Γ_z defined in the half-plane with positive real z to the left half-plane, when the cuts are introduced parallel to the negative real axis from the singular points as shown in figure 1. The real parts of z_5 and z_6 are always nonzero and negative, and the contribution of the integrals along C_5 and C_6 to $\Gamma(t)$ has an exponential damping factor and decays faster

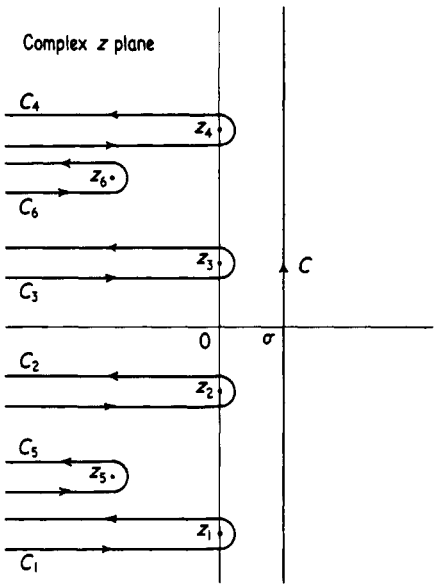

Figure 1. Deformation of the path of integral C to $C_1 \sim C_6$ in deriving the long-time asymptotic behaviour of $\Gamma(t)$ from the analytic behaviour of the Laplace transform Γ_z.

than the contributions from the paths C_1, C_2, C_3 and C_4. As a consequence, the long-time asymptotic behaviour of $\Gamma(t)$ is given by the sum of the contributions from the paths C_1, C_2, C_3 and C_4.

We now calculate the long-time asymptotic behaviour of the contribution from each branch-point on the imaginary axis. First we consider the case when Γ_z has a branch-point at the origin and behaves like

$$\Gamma_z = \frac{f(z)}{z^\alpha(-\ln z)^\beta} \tag{A.2}$$

where $f(z)$ is analytic in a neighbourhood of $z = 0$ and $f(0) \neq 0$. It is assumed that

$$\lim_{|z|\to\infty} \mathrm{e}^{zt} f(z) = 0 \qquad (\mathrm{Re}\, z < 0, t > 0). \tag{A.3}$$

α and β are arbitrary complex numbers. Then the contribution to $\Gamma(t)$ from this singularity is given by

$$I(\alpha, \beta; t) = \frac{1}{2\pi\mathrm{i}} \int_{C_0} \frac{f(z)}{z^\alpha(-\ln z)^\beta} \mathrm{e}^{zt}\, \mathrm{d}z \tag{A.4}$$

where C_0 denotes the path shown in figure 2. Changing the variable z to $z' = zt$, one has

$$I(\alpha, \beta; t) = \frac{t^{\alpha-1}}{(\ln t)^\beta} \frac{1}{2\pi\mathrm{i}} \int_{C_0} \frac{f(z/t)\, \mathrm{e}^z}{z^\alpha\{1-(\ln z/\ln t)\}^\beta} \mathrm{d}z.$$

If α is not equal to zero or a negative integer, we use Hankel's formula for the gamma function and obtain the following formula:

$$I(\alpha, \beta; t) \sim \frac{f(0)}{\Gamma(\alpha)} \frac{t^{\alpha-1}}{(\ln t)^\beta} \qquad t \to \infty. \tag{A.5}$$

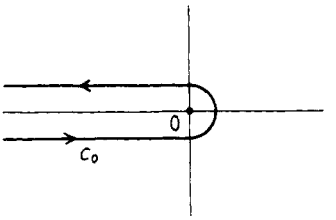

Figure 2. Path of integral C_0.

Here $\Gamma(\alpha)$ denotes the gamma function which must be discriminated from the friction function. It is obvious from the above derivation that the formula (A.5) is valid as far as $f(0) \equiv \lim_{z\to 0} f(z)$ does exist, even if $z = 0$ is a branch-point of the function $f(z)$.

When $\alpha = 0$, (A.4) reads

$$I(0, \beta; t) = \frac{1}{2\pi i} \int_{C_0} \frac{f(z)}{(-\ln z)^{\beta}} e^{zt}\, dz. \tag{A.6}$$

We integrate this expression by parts and obtain

$$I(0, \beta; t) = -\frac{1}{2\pi i} \int_{C_0} \frac{e^{zt}}{t} \frac{(\beta f(z) - z \ln z f'(z))}{z(-\ln z)^{\beta+1}}\, dz.$$

If $\beta \neq 0$, we use the formula (A.5) for $I(1, \beta+1; t)$ to this integral and obtain the following asymptotic behaviour:

$$I(0, \beta; t) \sim -\frac{\beta f(0)}{t(\ln t)^{\beta+1}} \qquad t \to \infty. \tag{A.7}$$

When α is zero or a negative integer, (A.4) reads

$$I(-n, \beta; t) = \frac{1}{2\pi i} \int_{C_0} \frac{z^n f(z)}{(-\ln z)^{\beta}} e^{zt}\, dz \tag{A.8}$$

where n is zero or a positive integer. If $\beta \neq 0$, one proves by a mathematical induction that

$$I(-n, \beta; t) \sim \left(-\frac{1}{t}\right)^{n+1} \frac{n!\beta f(0)}{(\ln t)^{\beta+1}} \qquad t \to \infty. \tag{A.9}$$

We first notice that this is true for $n = 0$ by comparing this with (A.7). If we integrate (A.8) by parts, we have

$$I(-n, \beta; t) = -\frac{1}{2\pi i} \int_{C_0} \frac{e^{zt}}{t} \left(\frac{z^{n-1}(nf(z) + zf'(z))}{(-\ln z)^{\beta}} + \frac{\beta z^{n-1} f(z)}{(-\ln z)^{\beta+1}} \right) dz \tag{A.10}$$

for positive n. We now suppose that (A.9) is valid for $I(-(n-1), \beta; t)$ and use it on the right-hand side of (A.10) and then we conclude that (A.9) is valid for $I(-n, \beta; t)$. As a consequence, we confirm (A.9) for zero and positive integers n when $\beta \neq 0$.

We now consider the case where the position of the branch-point is at iy_c and the behaviour of Γ_z is given by

$$\Gamma_z = \frac{f(z)}{\{-\ln(z - iy_c)\}^{\beta}} \tag{A.11}$$

where $f(z)$ is analytic in a neighbourhood of $z = iy_c$ and $f(iy_c) \neq 0$. By assuming (A.3), we obtain

$$-\frac{\beta f(iy_c)}{t(\ln t)^{\beta+1}} \exp(iy_c t) \qquad t \to \infty. \tag{A.12}$$

in place of (A.7), as the contribution due to the branch-point at $z = iy_c$ to the asymptotic behaviour of $\Gamma(t)$.

In the above expressions (A.5), (A.7), (A.9) and (A.12), only the leading terms are given. The terms which we have ignored are smaller only by a factor of order $(1/\ln t)$.

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