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Dynamic Structure Factor in a Random Diffusion Model

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Let $\{X_t: t \geqslant 0\}$ denote random walk in the random waiting time model, i.e., simple random walk with jump rate $w^{-1}(X_t)$, where $\{w(x): x \in \mathbb{Z}^d\}$ is an i.i.d. random field. We show that (under some mild conditions) the *intermediate scattering function* $F(q, t) = \mathbb{E}_0 e^{iqX_t}$ $(q \in \mathbb{R}^d)$ is completely monotonic in t ($\mathbb{E}_0$ denotes double expectation w.r.t. walk and field). We also show that the *dynamic structure factor* $S(q, \omega) = 2\int_0^\infty \cos(\omega t)\, F(q, t)\, dt$ exists for $\omega \neq 0$ and is strictly positive. In $d = 1, 2$ it diverges as $1/|\omega|^{1/2}$, resp. $-\ln(|\omega|)$, in the limit $\omega \to 0$; in $d \geqslant 3$ its limit value is strictly larger than expected from hydrodynamics. This and further results support the conclusion that the hydrodynamic region is limited to small q and small ω such that $|\omega| \gg D\,|q|^2$, where D is the diffusion constant.

KEY WORDS: Random walk in random environment; dynamic structure factor; hydrodynamic limit; long-time tail.

1. INTRODUCTION AND STATEMENT OF THE RESULTS

An important quantity in the study of liquids is the (self-part of the) *dynamic structure factor* $S(q, \omega)$.[1] This quantity monitors the motion of a tracer particle in the liquid. In good approximation the particle performs Brownian motion with diffusion constant D,[2] which implies

$$S(q, \omega) \approx S_0(q, \omega) \tag{1.1}$$

with

$$S_0(q, \omega) = \frac{2DF_0(q)}{D^2 F_0(q)^2 + \omega^2} \tag{1.2}$$

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where $F_0(q)$ equals $|q|^2/2d$ for Brownian motion. Measurements of $S(q, \omega)$ by means of neutron scattering on liquids[3] show that (1.1) is verified within 10% accuracy. The small discrepancies can be understood because one expects that, as $q \to 0$ and $\omega \to 0$,

$$S(q, \omega) \sim z(\omega)\, S_0(q, \omega) \tag{1.3}$$

where $z(\omega)$ is proportional to the Fourier transform of the velocity autocorrelation function (VACF) (see ref. 4, Chapter 11). The *long-time tail* (LTT) in the VACF, if present, produces a nonanalyticity in $z(\omega)$ at $\omega = 0$.[5] The region of validity of (1.3) is called the *hydrodynamic region*. Montfrooij and de Schepper[6] have argued that (1.3) is in fact a good approximation if and only if $|\omega|$ is small but not too small, namely

$$2D\, |q|^2 \leqslant |\omega| \ll 1 \tag{1.4a}$$

The region where

$$|\omega| \ll 2D\, |q|^2 \ll 1 \tag{1.4b}$$

then corresponds to the *static* limit. The difference between results in the static limit and in the hydrodynamic limit is investigated in the present paper. The distinction between them is of experimental relevance and leads to the paradoxical statement that measurements on a too slow time scale deviate from hydrodynamics.

A LTT can also occur in models of *random walk in random environment* (RWRE), though in a less pronounced way (a $t^{-5/2}$-decay in time t instead of the $t^{-3/2}$-decay observed in liquids). In the model studied here there is no LTT correction to diffusion. As a consequence the function $z(\omega)$ in (1.3) is identically one. Still, the correction to (1.3) for finite q and ω is such that the results in the static and in the hydrodynamic limits disagree (see Theorems 2 and 3 below). In fact, in $d = 1, 2$ the correction term diverges in the static limit, while in $d \geqslant 3$ it converges to a nonvanishing constant.

1.1. Model

In the present paper we continue our study of the random waiting time model.[7,8] Let

$$w = \{w(x) \colon x \in \mathbb{Z}^d\} \tag{1.5}$$

be a collection of random variables taking values in $(0, \infty)$ according to an i.i.d. distribution μ satisfying

$$\int w^{-1}(0)\, \mu(dw) < \infty \tag{1.6a}$$

$$\int w^{2}(0)\, \mu(dw) < \infty \tag{1.6b}$$

Given w, let $X = \{X_t\colon t \geqslant 0\}$ be simple random walk with jump rate $w^{-1}(X_t)$. Then the random waiting time model is defined as the combined process (X, w). This is an example of RWRE.

Let M and V^2 denote mean and variance of $w(0)$ under μ:

$$M = \int w(0)\, \mu(dw) \tag{1.7a}$$

$$V^2 = \int [w(0) - M]^2\, \mu(dw) \tag{1.7b}$$

Let μ_0 denote the probability measure defined by

$$\mu_0(dw) = \frac{w(0)}{M}\, \mu(dw) \tag{1.8}$$

Then μ_0 is stationary for the environment process associated with $\{X_t\colon t \geqslant 0\}$ (starting at $X_0 = 0$).[8] Let $\mathbb{E}$ and $\mathbb{E}_0$ denote double expectation w.r.t. $\{X_t\colon t \geqslant 0\}$ and w.r.t. w distributed according to μ, resp. μ_0.

Given the configuration w, let $p_t^w(x, y)$ denote the probability that the walk moves from x to y in time t. It is easy to show that the *detailed balance condition* holds:

$$w(0)\, p_t^w(0, x) = w(x)\, p_t^w(x, 0), \qquad t \geqslant 0, \quad x \in \mathbb{Z}^d \tag{1.9}$$

1.2. Definitions

For more background on the following definitions the reader is referred to refs. 9–11. The *intermediate scattering function* $F(q, t)$ is given by

$$F(q, t) = \mathbb{E}_0 e^{iqX_t}, \qquad q \in \mathbb{R}^d \tag{1.10}$$

The *dynamic scattering function*, also called the *dynamic structure factor*, is given by

$$S(q, \omega) = 2 \int_0^\infty \cos(\omega t)\, F(q, t)\, dt, \qquad \omega \in \mathbb{R}, \quad q \in \mathbb{R}^d \tag{1.11}$$

whenever the integral converges. The *form factor* F_0 is given by

$$\begin{aligned} F_0(q) &= \sum_{x \in \mathbb{Z}^d} p(0, x)(1 - e^{iqx}), \qquad q \in \mathbb{R}^d \\ &= \frac{2}{d} \sum_{i=1}^{d} \sin^2 \frac{q_i}{2} \end{aligned} \tag{1.12}$$

Here, $p(x, y)$ denotes the transition kernel of simple random walk, i.e., $p(x, y) = 1/2d$ if $|x - y| = 1$ and zero otherwise.

1.3. Results

Throughout the paper we assume that there exists $a > 0$ such that $\mu(w(0) \geqslant a) = 1$. This is a technical condition which will be needed in the proofs. We have three theorems.

Our first result reads:

Theorem 1. *$F(q, t)$ is completely monotonic in t, for every q.*

Theorem 1 implies that there exists a representation of $F(q, t)$ as the Laplace transform of a positive measure. More precisely, by Bernstein's theorem (see ref. 12, Theorem 12a, Section IV.12) there exists a non-decreasing right-continuous function α_q such that for all $t \geqslant 0$

$$F(q, t) = \int_0^\infty e^{-\lambda t}\, d\alpha_q(\lambda) \tag{1.13}$$

Incidentally, in the physics literature the existence of this representation is always assumed when discussing real-time correlation functions, e.g., in dynamic light scattering experiments. Often the approximation $F(q, t) \simeq \exp(-D\, |q|^2\, t/2d)$ is used, with D the diffusion constant. Now, for the continuous-time simple random walk with mean exponential waiting time D^{-1} one has

$$F(q, t) = e^{-DF_0(q)t} \tag{1.14}$$

which is obviously completely monotonic in t. However, for the discrete-time simple random walk one has $\mathbb{E}e^{iqX_n} = [1 - F_0(q)]^n$, which is *not* a positive function. Therefore Theorem 1 is not a trivial property. In fact, it is essential in the proof of theorems 2 and 3 below.

From (1.11) and (1.13) it follows that $S(q, \omega)$ exists for $\omega \neq 0$ and has the often-used representation

$$S(q, \omega) = \int_0^\infty \frac{2\lambda}{\lambda^2 + \omega^2} \, d\alpha_q(\lambda) \tag{1.15}$$

In particular, there follows

$$|\omega'| \geqslant |\omega| \Rightarrow S(q, \omega) \geqslant S(q, \omega') \geqslant (\omega^2/\omega'^2) \, S(q, \omega) \tag{1.16}$$

which implies continuity of $\omega \to S(q, \omega)$ for $\omega \neq 0$.

As a consequence of Theorem 1, the behavior of

$$\int_0^\infty e^{izt} F(q, t) \, dt = \int_0^\infty \frac{1}{\lambda - iz} \, d\alpha_q(\lambda) \tag{1.17}$$

as $z \to 0$ in $\{z \in \mathbb{C}\colon \operatorname{Im} z \geqslant 0\}$ depends on the asymptotics of $\alpha_q(\lambda)$ as $\lambda \to 0$, which can be derived from the behavior of (1.17) as $z \to 0$ along the imaginary axis. This important fact is used to prove our second result, which concerns $S(q, \omega)$ in the static region.

Theorem 2. Let $G(z)$ denote the Green's function of simple random walk on $\mathbb{Z}^d$, i.e., $G(z) = \sum_{n \geqslant 0} z^n p_n(0, 0)$ with $p_n(x, y)$ the probability that the walk moves from x to y in n steps. Then for every q such that $F_0(q) \neq 0$

$$\lim_{t \to \infty} F(q, t) = 0 \tag{1.18a}$$

and

$$S(q, \omega) = 2 \operatorname{Re} \left\{ \frac{1}{M^{-1} F_0(q) + i\omega} + \frac{V^2}{M} [1 + o(\omega^0)] \, G\left(\frac{1}{1 + iM\omega} \right) \right\} \tag{1.18b}$$

as $\omega \to 0$ in $\mathbb{R}$.

Incidentally, one cannot expect an easy proof of (1.18a), because $F(q, t)$ decays algebraically in t due to the presence of the LTT. An easy inequality which follows from Theorem 1 (and which will be proved at the end of Section 2) is

$$F(q, t) \geqslant e^{-DF_0(q) t} \tag{1.19}$$

Since for $z \to 0$ in $\{z \in \mathbb{C}\colon \operatorname{Re} z \geqslant 0\}$

$$d = 1\colon \qquad G(1 - z) \sim (1/2z)^{1/2}$$

$$d = 2\colon \qquad G(1 - z) \sim -(1/\pi) \ln z$$

$$d \geqslant 3\colon \qquad G(1 - z) \sim G(1)$$

there follows from (1.18b) as $\omega \to 0$

$$d=1: \qquad S(q,\omega)=\frac{V^2}{M}\frac{1}{(M\,|\omega|)^{1/2}}+o(|\omega|^{1/2}) \tag{1.20a}$$

$$d=2: \qquad S(q,\omega)=-2\frac{V^2}{M}\frac{1}{\pi}\ln(M\,|\omega|)+o(\ln(|\omega|)) \tag{1.20b}$$

$$d\geqslant 3: \qquad S(q,\omega)=2\left[\frac{1}{M^{-1}F_0(q)}+\frac{V^2}{M}G(1)\right]+o(\omega^0) \tag{1.20c}$$

Our third and final result concerns $S(q,\omega)$ in the hydrodynamic region. Using the central limit theorem of refs. 13 and 14, one can show that the rescaled random walk

$$(\varepsilon X_{\varepsilon^{-2}t})_{t\geqslant 0} \tag{1.21}$$

as $\varepsilon\to 0$ converges weakly in path space to Brownian motion with diffusion constant $D=M^{-1}$. This implies that

$$\varepsilon^2 S(\varepsilon q,\varepsilon^2\omega)=S_0(q,\omega)+o(\varepsilon^0) \tag{1.22}$$

with $S_0(q,\omega)$ given by (1.2). The latter result can be sharpened to:

Theorem 3. As $q\to 0$

$$S(q,\omega)\sim S_0(q,\omega) \tag{1.23}$$

uniformly inside the region

$$\alpha D F_0(q)^\beta \leqslant |\omega| \tag{1.24}$$

for any $\alpha>0$ and $0<\beta<1$.

The condition $\alpha>0$ is necessary as is evident from Theorem 2. Equation (1.22) is implied by Theorem 3 because of (1.16).

2. PROOF OF THEOREM 1 and (1.19)

We start with two preparatory sections, Sections 2.1 and 2.2, in which we use some Hilbert space techniques. In Sections 2.3 and 2.4 we prove Theorem 1 and (1.19).

2.1. Hilbert Space Decomposition

Let us denote by p_t the function

$$p_t\colon (x,w)\to p_t^w(0,x) \tag{2.1}$$

and by $\mathscr{D}$ the linear space spanned by the set $\{p_t: t \geq 0\}$. It is natural to consider the inner product $(\cdot, \cdot)$ on $\mathscr{D}$ defined by

$$(f, g) = \int \mu_0(dw) \sum_x f(x, w)\, \overline{g(x, w)} \tag{2.2}$$

and to construct a Hilbert space $\mathscr{H}$ by completion of $\mathscr{D}$ w.r.t. $(\cdot, \cdot)$. However, in what follows we need an integral decomposition of $\mathscr{H}$ into wavevector-dependent Hilbert spaces $\mathscr{H}_q$.

Introduce the (degenerate) sesquilinear form $(\cdot, \cdot)_q$ defined by

$$(f, g)_q = \int \mu_0(dw) \sum_{x, y} e^{iq(x-y)} f(x, w)\, \overline{g(y, w)} \tag{2.3}$$

Let $\mathscr{H}_q$ denote the Hilbert space obtained by dividing out the null space $\mathscr{N}_q$ of $(\cdot, \cdot)_q$ and completing $\mathscr{D}/\mathscr{N}_q$ w.r.t. $(\cdot, \cdot)_q$.

Lemma 2.1. For all s, $t \geq 0$

$$(p_s, p_t)_q = \mathbb{E}_0 e^{iqX_{s+t}} \tag{2.4}$$

Proof. One has (recall (1.8))

$$\mathbb{E}_0 e^{iqX_{s+t}} = \int \mu_0(dw) \sum_x e^{iqx} p^w_{s+t}(0, x)$$

$$= \frac{1}{M} \int \mu(dw)\, w(0) \sum_x e^{iqx} \sum_y p^w_s(0, y)\, p^w_t(y, x) \tag{2.5}$$

Using detailed balance (1.9), one obtains

$$\mathbb{E}_0 e^{iqX_{s+t}} = \frac{1}{M} \int \mu(dw) \sum_x e^{iqx} \sum_y w(y)\, p^w_s(y, 0)\, p^w_t(y, x) \tag{2.6}$$

Using translation invariance of μ, together with the observation that $p^{\tau_{-y} w}_s(y, 0) = p^w_s(0, -y)$ and $p^{\tau_{-y} w}_t(y, x) = p^w_t(0, x-y)$, one gets

$$\mathbb{E}_0 e^{iqX_{s+t}} = \frac{1}{M} \int \mu(dw)\, w(0) \sum_{x, y} e^{iqx} p^w_s(0, -y)\, p^w_t(0, x-y)$$

$$= \int \mu_0(dw) \sum_{x, y} e^{-iq(x-y)} p^w_s(0, x)\, p^w_t(0, y)$$

$$= (p_t, p_s)_q = (p_s, p_t)_q \quad \blacksquare \tag{2.7}$$

An immediate consequence of (2.4) is that $\mathbb{E}_0 e^{iqX_t} = (p_{t/2}, p_{t/2})_q$ is nonnegative. The following lemma allows us to relate results for $\mathbb{E}_0 e^{iqX_t}$ and $\mathbb{E} e^{iqX_t}$.

Lemma 2.2. For all $t \geq 0$

$$\frac{\partial}{\partial t} \mathbb{E}_0 e^{iqX_t} = -M^{-1} F_0(q)\, \mathbb{E} e^{-iqX_t} \tag{2.8}$$

Proof. The function p_t satisfies the equation

$$\frac{\partial}{\partial t} p_t^w(0, x) = \sum_y \frac{1}{w(y)} p_t^w(0, y)[p(y, x) - \delta_{y,x}] \tag{2.9}$$

Let $\mathbb{E}_w$ denote expectation w.r.t. the random walk in the fixed environment w. Using (2.9), one calculates (recall (1.12))

$$\begin{aligned}
\frac{\partial}{\partial t} \mathbb{E}_w e^{iqX_t} &= \frac{\partial}{\partial t} \sum_x e^{iqx} p_t^w(0, x) \\
&= \sum_x e^{iqx} \sum_y \frac{1}{w(y)} p_t^w(0, y)[p(y, x) - \delta_{y,x}] \\
&= -F_0(q) \sum_y e^{iqy} \frac{1}{w(y)} p_t^w(0, y) \\
&= -F_0(q)\, \mathbb{E}_w \frac{1}{w(X_t)} e^{iqX_t}
\end{aligned} \tag{2.10}$$

Integrating (2.10) over μ_0, using detailed balance (1.9) and using the translation invariance of μ, one gets (2.8). ■

Lemma 2.3. For $n \geq 1$

$$\left| \frac{\partial^n}{\partial t^n} \bigg|_{t=0} (p_t, p_0)_q \right| \leq F_0(q)\, a^{-n} 2^{n-1} \tag{2.11}$$

where a is the constant such that $\mu(w(0) \geq a) = 1$.

Proof. Using (2.4) and (2.9), one calculates

$$\begin{aligned}
\frac{\partial^n}{\partial t^n} \bigg|_{t=0} (p_t, p_0)_q &= \frac{\partial^n}{\partial t^n} \bigg|_{t=0} \int \mu_0(dw) \sum_x e^{iqx} p_t^w(0, x) \\
&= (-1)^n F_0(q) \int \mu_0(dw) \\
&\qquad \sum_{x_1, \dots, x_{n-1}} \frac{e^{iqx_1}}{w(x_1)} \Delta_{x_1, x_2} \frac{1}{w(x_2)} \times \cdots \times \Delta_{x_{n-1}, 0} \frac{1}{w(0)}
\end{aligned} \tag{2.12}$$

where $\Delta_{x,y} = \delta_{x,y} - p(x, y)$. This implies (2.11) because of $\mu(w(0) \geqslant a) = 1$ and $\sum_x |\Delta_{x,y}| \leqslant 2$ for all y. ∎

2.2. A Contracting Semigroup

We show now that a contracting semigroup $\{S(t): t \geqslant 0\}$ is defined by $S(t)p_{t'} = p_{t+t'}$, $t, t' \geqslant 0$. We do not know how to show this in a direct manner and so we proceed as follows.

Proposition 2.4. There exists a self-adjoint operator L on $\mathscr{H}_q$ satisfying:

(i) $\mathscr{D} \subset \mathrm{Dom}(L)$.

(ii) $(\partial/\partial s)(p_s, p_t)_q = (Lp_s, p_t)_q$ for all $s, t \geqslant 0$.

(iii) $p_t = e^{Lt}p_0$ for all $t \geqslant 0$.

Proof. (i), (ii) Let $\mathbb{E}_w$ be as before. From (2.3) it follows that

$$\frac{\partial}{\partial s}(p_s, p_t)_q = \frac{\partial}{\partial s}\int \mu_0(dw)\, \mathbb{E}_w(e^{iqX_s})\, \mathbb{E}_w(e^{-iqX_t}) \tag{2.13}$$

Via (2.10), this becomes

$$\frac{\partial}{\partial s}(p_s, p_t)_q = -F_0(q)\int \mu_0(dw)\, \mathbb{E}_w\left(\frac{1}{w(X_s)}\, e^{iqX_s}\right) \mathbb{E}_w(e^{-iqX_t}) \tag{2.14}$$

By Schwarz's inequality therefore

$$\begin{aligned}
&\left|\frac{\partial}{\partial s}\left(p_s, \sum_i \lambda_i p_{t_i}\right)_q\right|^2 \\
&\quad \leqslant F_0(q)^2\left[\int \mu_0(dw)\left|\mathbb{E}_w\left(\frac{1}{w(X_s)}\, e^{iqX_s}\right)\right|^2\right] \\
&\qquad \times\left[\int \mu_0(dw)\left|\sum_i \lambda_i \mathbb{E}_w(e^{-iqX_{t_i}})\right|^2\right] \\
&\quad \leqslant F_0(q)^2\, a^{-2}\left(\sum_i \lambda_i p_{t_i}, \sum_i \lambda_i p_{t_i}\right)_q
\end{aligned} \tag{2.15}$$

where again a is the constant such that $\mu(w(0) \geqslant a) = 1$. Hence the linear map $\sum_i \lambda_i p_{t_i} \to (\partial/\partial s)(p_s, \sum_i \lambda_i p_{t_i})$ is continuous. Therefore, by Riesz's theorem, there exists a function η_s in $\mathscr{H}_q$ such that

$$\frac{\partial}{\partial s}(p_s, p_t)_q = (\eta_s, p_t)_q \qquad \text{for all} \quad s, t \geqslant 0 \tag{2.16}$$

Which clearly satisfies $\|\eta_s\|_q \leqslant F_0(q)\, a^{-1}$.

From Lemma 2.1 it follows that $(\eta_s, p_t)_q = (p_s, \eta_t)_q$ for all $s, t \geqslant 0$. Hence $\sum_i \lambda_i p_{t_i} = 0$ implies $\sum_i \lambda_i \eta_{t_i} = 0$ for any finite linear combination. Therefore the map L defined by $Lp_t = \eta_t$ extends to a symmetric linear operator with domain $\mathscr{D}$, and extends further to a self-adjoint operator on $\mathscr{H}_q$.

(iii) From Lemma 2.3 it follows that p_0 belongs to the analytic domain of L. Hence

$$(L^n p_0, p_t)_q = \left.\frac{\partial^n}{\partial s^n}\right|_{s=0} (p_s, p_t)_q \qquad \text{for all} \quad n \geqslant 0 \tag{2.17}$$

This implies $e^{Ls} p_0 = p_s$. ■

2.3. Proof of Theorem 1

From Lemma 2.1, Proposition 2.4 and spectral theory it follows that there exists a nondecreasing right-continuous function α_q on $\mathbb{R}$ such that

$$F(q, t) = \mathbb{E}_0(e^{iqX_t}) = (e^{Lt} p_0, p_0)_q = \int_{\mathbb{R}} e^{-\lambda t}\, d\alpha_q(\lambda) \tag{2.18}$$

Because the l.h.s. of the above expression is obviously bounded as $t \to \infty$, the support of α_q is a subset of $\mathbb{R}^+$. Hence (2.18) is completely monotonic in t. ■

2.4. Proof of (1.19)

The proof runs via two more lemmas.

Lemma 2.5. $t \to \mathbb{E}e^{iqX_t}/F(q, t)$ is nonincreasing. Hence

$$\mathbb{E}e^{iqX_t} \leqslant F(q, t) \tag{2.19}$$

Proof. $t \to F(q, t)$ is completely monotonic on $[0, \infty)$ and hence log-convex. This implies

$$0 \leqslant \frac{\partial^2}{\partial t^2} \ln(F(q, t)) = -DF_0(q) \frac{\partial}{\partial t} \frac{\mathbb{E} e^{iqX_t}}{F(q, t)} \tag{2.20}$$

where we use (2.8) with $M^{-1} = D$ and note that $\mathbb{E} e^{-iqX_t} = \mathbb{E} e^{iqX_t}$. ∎

Lemma 2.6. $t \to e^{DF_0(q)t} F(q, t)$ is nondecreasing.

Proof. Let $t \geqslant t_0 \geqslant 0$. From (1.13) there follows by Jensen's inequality

$$\begin{aligned} F(q, t)/F(q, t_0) &\geqslant \exp\left[-(t - t_0) \int_0^\infty \lambda e^{-\lambda t_0}\, d\alpha_q(\lambda)/F(q, t_0) \right] \\ &= \exp\left[(t - t_0) \frac{\partial}{\partial t_0} F(q, t_0)/F(q, t_0) \right] \end{aligned} \tag{2.21}$$

By (2.8) and (2.19) this becomes

$$\begin{aligned} F(q, t)/F(q, t_0) &\geqslant \exp[-(t - t_0)\, DF_0(q)\, \mathbb{E} e^{iqX_{t_0}}/F(q, t_0)] \\ &\geqslant \exp[-(t - t_0)\, DF_0(q)] \quad ∎ \end{aligned} \tag{2.22}$$

Lemma 2.6 implies (1.19).

3. PROOF OF THEOREM 2

In this section we make an expansion of the Laplace transform of $\mathbb{E} e^{iqX_t}$ for small λ and fixed q. The main result is given in Proposition 3.3 below and leads immediately to the proof of Theorem 2.

3.1. Two Preparatory Lemmas

First, we rewrite the Laplace transform in terms of quantities which depend only on the underlying simple random walk. Let $\mathbb{E}_Y$ denote expectation w.r.t. to the discrete-time simple random walk $Y = (Y_n)_{n \geqslant 0}$ starting at $Y_0 = 0$. Denote the local time of Y at site x after k steps by

$$l_Y(x, k) = \sum_{n=0}^{k} \mathbb{1}_{\{Y_n = x\}} \tag{3.1}$$

Lemma 3.1. For $\operatorname{Re} \lambda > 0$

$$\int_0^\infty e^{-\lambda t}\, d\mathbb{E}\, e^{iqX_t} = \sum_{k \geqslant 0} \mathbb{E}_Y \left[(e^{iqY_{k+1}} - e^{iqY_k}) \prod_x I(\lambda, l_Y(x, k)) \right] \tag{3.2}$$

with

$$I(\lambda, l) = \int \mu(dw)[1 + \lambda w(0)]^{-l} \tag{3.3}$$

Proof. Condition on the environment w and on the embedded walk Y (which is the discrete-time skeleton of our continuous-time RWRE X). Let $T_0 = 0$ and let T_k denote the time of the kth step, $k = 1, 2,...$. The probability that $T_k \leqslant t$ equals the probability that the sum of k independent exponential waiting times with averages $w(Y_0)$, $w(Y_1),..., w(Y_{k-1})$ is smaller than or equal to t. Hence we obtain

$$\int_0^\infty e^{-\lambda t}\, d\mathbb{P}_w(T_k \leqslant t \mid Y) = \prod_{m=0}^{k-1} \frac{1}{1 + \lambda w(Y_m)} \qquad (k \geqslant 1) \tag{3.4}$$

Combining this with the identity

$$\mathbb{P}_w(X_t = x) = \sum_{k \geqslant 0} \mathbb{E}_Y\big(\mathbb{1}_{\{Y_k = x\}} \mathbb{P}_w(T_k \leqslant t < T_{k+1} \mid Y)\big) \tag{3.5}$$

we obtain, after a straightforward calculation,

$$\begin{aligned} \int_0^\infty e^{-\lambda t}\, d\mathbb{E}_w e^{iqX_t} &= \sum_x e^{iqx} \int_0^\infty e^{-\lambda t}\, d\mathbb{P}_w(X_t = x) \\ &= \sum_{k \geqslant 0} \mathbb{E}_Y\left[(e^{iqY_{k+1}} - e^{iqY_k}) \prod_{m=0}^{k} \frac{1}{1 + \lambda w(Y_m)}\right] \end{aligned} \tag{3.6}$$

Now note that

$$\prod_{m=0}^{k} \frac{1}{1 + \lambda w(Y_m)} = \prod_x [1 + \lambda w(x)]^{-l_Y(x,k)} \tag{3.7}$$

Hence, by averaging (3.6) over w and using the i.i.d. property of $\{w(x): x \in \mathbb{Z}^d\}$ we obtain (3.2). ■

Second, we formulate a large-deviation estimate.

Lemma 3.2. For any $\delta \in (0, 1)$ there exist positive constants L and K such that for all $k \geqslant 0$

$$\mathbb{P}(\sup_x l_Y(x, k) \geqslant k^{(1+\delta)/2}) \leqslant L e^{-Kk^{\delta/4}} \tag{3.8}$$

Proof. See ref. 8, Lemma 1. ■

3.2. Asymptotic Expansion

The basic result of the present section is the following.

Proposition 3.3. For $\lambda \downarrow 0$ along the reals

$$\int_0^\infty e^{-\lambda t}\, d\mathbb{E}\, e^{iqX_t}$$
$$= -\frac{F_0(q)}{F_0(q)+\lambda M}\left[1+\frac{V^2\lambda^2[1+o(\lambda^0)]}{F_0(q)+\lambda M}\, G\left(\frac{1}{1+\lambda M}\right)\right] \tag{3.9}$$

Proof. The proof consists of three steps.

Step 1. Pick $\alpha > 1$ and split the sum appearing in (3.2) into two parts:

$$\sum_{k \geqslant 0} = \sum_{k=0}^{\lfloor \lambda^{-\alpha} \rfloor} + \sum_{k=\lfloor \lambda^{-\alpha}\rfloor + 1}^{\infty} \tag{3.10}$$

The second sum in (3.10) goes to zero as $\lambda \downarrow 0$ faster than any polynomial. To see why, substitute the estimate [recall that $\mu(w(0) \geqslant a) = 1$]

$$I(\lambda, l) \leqslant (1+\lambda a)^{-l} \tag{3.11}$$

and the identity

$$\sum_x l_Y(x, k) = k+1 \tag{3.12}$$

into (3.2) to get the upper bound

$$2 \sum_{k=\lfloor \lambda^{-\alpha}\rfloor+1}^{\infty} \frac{1}{(1+\lambda a)^{k+1}} = O(e^{-\lambda^{1-\alpha}}) \tag{3.13}$$

To prepare for the computation of the first sum in (3.10), pick $\beta \in (1/2, 1)$ and let

$$V_k^\lambda = \{\sup_x l_Y(x, k) \leqslant \lambda^{-\alpha\beta}\} \tag{3.14}$$

Lemma 3.2 with $(1+\delta)/2 = \beta$ implies that

$$\mathbb{P}\left(\left(\bigcap_{0 \leqslant k \leqslant \lfloor \lambda^{-\alpha}\rfloor} V_k^\lambda\right)^c\right) = \mathbb{P}(V^\lambda_{\lfloor \lambda^{-\alpha}\rfloor})^c \leqslant L \exp(-K \lfloor \lambda^{-\alpha} \rfloor^{(2\beta-1)/4}) \tag{3.15}$$

Hence, neglecting a term which as $\lambda \downarrow 0$ tends to zero faster than any polynomial, we may insert the indicator $\mathbb{1}_{V_k^\lambda}$ under the expectation $\mathbb{E}_Y$ in the first sum in (3.10).

Step 2. The standard estimate for Taylor expansion up to second order gives [recall (1.7)]

$$I(\lambda, l) = (1+\lambda M)^{-1}\{1+\tfrac{1}{2}V^2\lambda^2 l(l+1) + o((\lambda l)^2)\} \tag{3.16}$$

Assume that $\alpha\beta < 1$. Then we obtain on the event V_k^λ

$$\begin{aligned} I(\lambda, l_Y(x,k)) &= (1+\lambda M)^{-l_Y(x,k)}\{1+\tfrac{1}{2}V^2\lambda^2 l_Y(x,k)[l_Y(x,k)+1] + o(\lambda^0)\} \\ &= (1+\lambda M)^{-l_Y(x,k)} \\ &\quad \times \exp\left\{\tfrac{1}{2}V^2\lambda^2 l_Y(x,k)[l_Y(x,k)+1][1+o(\lambda^0)]\right\} \end{aligned} \tag{3.17}$$

where the $o(\lambda^0)$ term tends to zero uniformly in $k \in [0, \lfloor \lambda^{-\alpha} \rfloor]$ and x as $\lambda \to 0$. Next assume that $2-\alpha-\alpha\beta > 0$. Then, via (3.12) and conditioned on the event V_k^λ (recall that $k \leqslant \lfloor \lambda^{-\alpha} \rfloor$),

$$\begin{aligned} &\prod_x I(\lambda, l_Y(x,k)) \\ &\quad = (1+\lambda M)^{-k-1}\exp\left\{\tfrac{1}{2}V^2\lambda^2[1+o(\lambda^0)]\sum_x l_Y(x,k)[l_Y(x,k)+1]\right\} \\ &\quad = (1+\lambda M)^{-k-1}\left\{1+\tfrac{1}{2}V^2\lambda^2[1+o(\lambda^0)]\sum_x l_Y(x,k)[l_Y(x,k)+1]\right\} \end{aligned} \tag{3.18}$$

Note that the factor $[1+o(\lambda^0)]$ can be brought in front of the sum over x because it is uniform in x.

Substituting (3.18) into (3.2), we arrive at the following intermediate result:

$$\int_0^\infty e^{-\lambda t}\, d\mathbb{E}\, e^{iqX_t} = A_1(\lambda) + \tfrac{1}{2}V^2\lambda^2[1+o(\lambda^0)]\, A_2(\lambda) \tag{3.19}$$

with

$$\begin{aligned} A_1(\lambda) &= \sum_{k\geqslant 0}(1+\lambda M)^{-k-1}\,\mathbb{E}_Y(e^{iqY_{k+1}} - e^{iqY_k}) \\ A_2(\lambda) &= \sum_{k\geqslant 0}(1+\lambda M)^{-k-1}\, B_k(\lambda) \\ B_k(\lambda) &= \mathbb{E}_Y\left[(e^{iqY_{k+1}} - e^{iqY_k})\sum_x l_Y(x,k)[l_Y(x,k)+1]\right] \end{aligned} \tag{3.20}$$

Step 3. It remains to work out (3.20). Using that (recall (1.12))

$$\mathbb{E}_Y e^{iqY_k} = [1 - F_0(q)]^k \tag{3.21}$$

the A_1-term can be evaluated explicitly. The result is

$$A_1(\lambda) = -\frac{F_0(q)}{F_0(q) + \lambda M} \tag{3.22}$$

The calculation of the A_2-term is less obvious. By means of the identity

$$\sum_x l_Y(x, k)[l_Y(x, k) + 1] = 2 \sum_x \sum_{0 \leqslant i \leqslant j \leqslant k} \mathbb{1}_{\{Y_i = x\}} \mathbb{1}_{\{Y_j = x\}} \tag{3.23}$$

the B_k-terms can be written as

$$\begin{aligned} B_k(\lambda) &= 2 \sum_x \sum_{0 \leqslant i \leqslant j \leqslant k} \mathbb{E}_Y[(e^{iqY_{k+1}} - e^{iqY_k}) \mathbb{1}_{\{Y_i = x\}} \mathbb{1}_{\{Y_j = x\}}] \\ &= 2 \sum_x \sum_{0 \leqslant i \leqslant j \leqslant k} \mathbb{E}^x_Y(e^{iqY_{k+1-j}} - e^{iqY_{k-j}}) \, p_i(0, x) \, p_{j-i}(x, x) \end{aligned} \tag{3.24}$$

where $\mathbb{E}^x_Y$ denotes expectation w.r.t. Y starting at $Y_0 = x$ and $p_k(x, y)$ denotes the probability that Y moves from x to y in k steps. Because

$$\mathbb{E}^x_Y e^{iqY_k} = \mathbb{E}_Y e^{iq(Y_k + x)} = e^{iqx} \mathbb{E}_Y e^{iqY_k} \tag{3.25}$$

it follows via (3.21) that

$$\begin{aligned} B_k(\lambda) &= 2 \sum_{0 \leqslant i \leqslant j \leqslant k} \mathbb{E}_Y(e^{iqY_{k+1-j}} - e^{iqY_{k-j}}) \, E_Y e^{iqY_i} p_{j-i}(0, 0) \\ &= 2 \sum_{0 \leqslant i \leqslant j \leqslant k} \{[1 - F_0(q)]^{k+1-j} - [1 - F_0(q)]^{k-j}\} \\ &\quad \times [1 - F_0(q)]^i \, p_{j-i}(0, 0) \\ &= -2F_0(q) \sum_{i=0}^{k} \sum_{j=0}^{k-i} [1 - F_0(q)]^{k-j} \, p_j(0, 0) \end{aligned} \tag{3.26}$$

Next, define the truncated Green's function

$$G_i(z) = \sum_{j=0}^{i} z^j p_j(0, 0) \tag{3.27}$$

Then

$$z^k \sum_{i=0}^{k} G_i(z^{-1}) = \sum_{i=0}^{k} \sum_{j=0}^{k-i} z^{k-j} p_j(0, 0) \tag{3.28}$$

and hence, combining (3.20) and (3.26), we arrive at

$$\begin{aligned} A_2(\lambda) &= -2\frac{F_0(q)}{1-F_0(q)} \sum_{k\geq 0} \left(\frac{1-F_0(q)}{1+\lambda M}\right)^{k+1} \sum_{i=0}^{k} G_i([1-F_0(q)]^{-1}) \\ &= -2\frac{F_0(q)}{1-F_0(q)} \frac{1+\lambda M}{F_0(q)+\lambda M} \\ &\quad \times \sum_{i\geq 0} \left(\frac{1-F_0(q)}{1+\lambda M}\right)^{i+1} G_i([1-F_0(q)]^{-1}) \\ &= -2\frac{F_0(q)(1+\lambda M)}{[F_0(q)+\lambda M]^2} G\left(\frac{1}{1+\lambda M}\right) \end{aligned} \tag{3.29}$$

In the last line appears $G(z) = G_\infty(z)$, the Green's function of simple random walk. Putting (3.19), (3.22), and (3.29) together, we get (3.9). ■

3.3. Proof of Theorem 2

Assume $F_0(q) \neq 0$. From (1.10), (2.8) and (3.9) it follows that

$$\begin{aligned} &\int_0^\infty e^{-\lambda t}\, dF(q,t) \\ &\quad = -M^{-1}F_0(q) \int_0^\infty e^{-\lambda t} \mathbb{E} e^{iqX_t}\, dt \\ &\quad = -M^{-1}\lambda^{-1}F_0(q) \left[1 + \int_0^\infty e^{-\lambda t}\, d\mathbb{E}\, e^{iqX_t}\right] \\ &\quad = \frac{-F_0(q)}{F_0(q)+\lambda M} + \frac{V^2}{M}\lambda[1+o(\lambda^0)]\, G\left(\frac{1}{1+\lambda M}\right) \end{aligned} \tag{3.30}$$

This implies

$$\lim_{\lambda\downarrow 0} \int_0^\infty e^{-\lambda t}\, dF(q,t) = -1 \tag{3.31}$$

and hence $\lim_{t\to\infty} F(q,t) = 0$ (note that $F(q,0) = 1$ and recall Theorem 1), which proves the first part of the theorem.

Recalling (1.13), we see that (3.30) becomes

$$\int_0^\infty \frac{1}{\lambda+s}\, d\alpha_q(s) = \frac{1}{M^{-1}F_0(q)+\lambda} + \frac{V^2}{M}[1+o(\lambda^0)]\, G\left(\frac{1}{1+M\lambda}\right) \tag{3.32}$$

In $d \geq 3$ the r.h.s. of (3.32) converges as $\lambda \downarrow 0$. Hence, because the l.h.s. is an absolutely convergent Laplace transform, we have (see ref. 15, Satz 2, p. 157)

$$\lim_{\lambda\to 0}\int_0^\infty \frac{1}{\lambda+s}\,d\alpha_q(s)=\frac{1}{M^{-1}F_0(q)}+\frac{V^2}{M}G(1),\qquad \operatorname{Re}\lambda\geqslant 0 \tag{3.33}$$

In particular, using (1.15), we get

$$\begin{aligned}\lim_{\omega\downarrow 0} S(q,\omega)&=\lim_{\omega\downarrow 0} 2\operatorname{Re}\int_0^\infty \frac{1}{i\omega+s}\,d\alpha_q(s)\\&=\frac{2}{M^{-1}F_0(q)}+2\frac{V^2}{M}G(1)\end{aligned} \tag{3.34}$$

which is (1.20c).

In $d=1$ the r.h.s. of (3.32) diverges like $V^2/M(2M\lambda)^{1/2}$ as $\lambda\downarrow 0$ and the Tauberian theorem for Stieltjes transforms (ref. 12, Chapter V, Theorem 7) can be applied. One obtains

$$\alpha_q(s)\sim\frac{2}{\pi}\frac{V^2}{M}(2M^{-1}s)^{1/2}\qquad \text{as}\quad s\downarrow 0 \tag{3.35}$$

From (1.15) we have

$$S(q,\omega)=\int_0^\infty dx\, e^{-x\omega^2}\int_0^\infty 2se^{-xs^2}\,d\alpha_q(s) \tag{3.36}$$

Inserting (3.35) and applying twice the Abelian theorem for Laplace transforms, one obtains (1.20a).

In $d=2$ expression (3.32) reads

$$\int_0^\infty \frac{1}{\lambda+s}\,d\alpha_q(s)=-\frac{V^2}{M}[1+o(\lambda^0)]\frac{1}{\pi}\ln(\lambda) \tag{3.37}$$

Taking the derivative of (3.37) w.r.t. λ is allowed because

$$\lambda\to\int_0^\infty \frac{\lambda}{(\lambda+s)^2}\,d\alpha_q(s)$$

asymptotically is a nonincreasing function which converges to a constant as $\lambda\downarrow 0$. By an Abelian argument this constant then necessarily equals $V^2/\pi M$. By ref. 12, Chapter V, Theorem 7, it then follows that

$$\alpha_q(s)\sim\frac{V^2}{M}\frac{s}{\pi}\qquad \text{as}\quad s\downarrow 0 \tag{3.38}$$

Proceeding as in $d=1$, one obtains

$$\frac{d}{d\omega}S(q,\omega)\sim -\frac{2}{\pi\omega}\frac{V^2}{M}\qquad \text{as}\quad \omega\downarrow 0 \tag{3.39}$$

The latter implies (1.20b).

Eqs. (1.20a–c) prove the second part of the theorem. ■

4. PROOF OF THEOREM 3

Theorem 3 is in fact an easy consequence of Theorem 1. We need a preparatory lemma.

Lemma 4.1. $\mu(w(0)\geqslant a)=1$ implies

$$\left|\int_0^\infty \cos(\omega s)\, d\mathbb{E}\, e^{iqX_s}\right| \leqslant \frac{\pi}{2a}\frac{F_0(q)}{|\omega|} \tag{4.1}$$

Proof. By Lemma 2.2 and Theorem 1, $\mathbb{E}e^{iqX_t}$ is completely monotonic [recall (1.10)]. Hence, if $\omega\neq 0$, then

$$\begin{aligned}\left|\int_0^\infty \cos(\omega s)\, d\mathbb{E}\, e^{iqX_s}\right| &\leqslant \left|\int_0^{\pi/2|\omega|} \cos(\omega s)\, d\mathbb{E}\, e^{iqX_s}\right| \\ &\leqslant 1-\mathbb{E}e^{iqX_{\pi/2|\omega|}}\end{aligned} \tag{4.2}$$

On the other hand, from (2.10) it follows that

$$\begin{aligned}-\frac{\partial}{\partial t}\mathbb{E}e^{iqX_t} &= F_0(q)\,\mathbb{E}\frac{1}{w(X_t)}e^{iqX_t} \\ &\leqslant a^{-1}F_0(q)\end{aligned} \tag{4.3}$$

Moreover, by convexity of $t\to e^{iqX_t}$ it follows that

$$\mathbb{E}e^{iqX_t}\geqslant 1+t\left.\frac{\partial}{\partial t}\right|_{t=0}\mathbb{E}e^{iqX_t} \tag{4.4}$$

Equations (4.3) and (4.4) together give

$$1-\mathbb{E}e^{iqX_{\pi/2|\omega|}}\leqslant a^{-1}F_0(q)\frac{\pi}{2|\omega|} \tag{4.5}$$

Equations (4.2) and (4.5) combine to give (4.1). ■

Proof of Theorem 3. From (1.10) and (1.11), by doing two partial integrations and using Lemma 2.2, one obtains

$$\frac{1}{2}S(q,\omega)=\omega^{-2}DF_0(q)\left[1+\int_0^\infty \cos(\omega s)\, d\mathbb{E}\, e^{iqX_s}\right] \tag{4.6}$$

It follows that (recall (1.2))

$$\frac{S(q,\omega)}{S_0(q,\omega)}=[1+\omega^{-2}D^2F_0(q)^2]\left[1+\int_0^\infty \cos(\omega s)\, d\mathbb{E}\, e^{iqX_s}\right] \tag{4.7}$$

If ω is in the set defined by (1.24), then by Lemma 4.1 one obtains

$$\left|1-\frac{S(q,\omega)}{S_0(q,\omega)}\right| \leqslant \frac{F_0(q)^{2(1-\beta)}}{\alpha^2}+\frac{\pi F_0(q)^{1-\beta}}{2a\alpha D} \tag{4.8}$$

which tends to zero as $q \to 0$. ■

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