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On the Dynamics of Excitations in Disordered Systems

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A new, time-local (TL) reduced equation of motion for the probability distribution of excitations in a disordered system is developed. To $O(k^2)$ the TL equation results in a Gaussian spatial probability distribution, i.e, $\langle P(\mathbf{r}, t)\rangle = [(2\pi\xi)^{1/2}]^{-d}\exp(-r^2/2\xi^2)$, where $\xi = \xi(t)$ is a correlation length, and $r = |\mathbf{r}|$. The corresponding distribution derived from the Hahn–Zwanzig (HZ) equation is more complicated and assumes the asymptotic $(r \to \infty)$ form: $\langle P(\mathbf{r}, s)\rangle \sim (s\xi^d)^{-1}\exp(-r/\xi)\cdot(r/\xi)^{(1-d)/2}$ where $\xi = \xi(s)$, d is the space dimensionality, and s is the Laplace transform variable conjugate to t. The HZ distribution generalizes the scaling form suggested by Alexander *et al.* for $d = 1$. In the Markov limit $\xi(t) \sim \sqrt{t}$, $\xi(s) \sim 1/\sqrt{s}$, and the two distributions are identical (ordinary diffusion).

KEY WORDS: Disordered system; diffusion; master equations; non-Markovian dynamics.

The problem of the dynamics of particles or excitations in systems that exhibit various types of randomness but are translationally invariant on the mean is currently under active study.[1–7] Some examples are energy transfer and spectral diffusion among randomly scattered impurities in a solid or fluid, electrical conductivity in disordered lattices, the vibrations of a disordered chain, etc. A natural starting point for the theoretical treatment of many of these systems is a master equation for the probability distribution $P(\mathbf{r}, t)$ of finding the particle (or excitation) at point $\mathbf{r}$ at time t, i.e.,

$$\frac{dP(\mathbf{r}, t)}{dt} = \sum_{\mathbf{r}'} \mathsf{W}(\mathbf{r}, \mathbf{r}')P(\mathbf{r}', t) \tag{1}$$

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where conservation of probability requires that $\sum_{\mathbf{r}} \mathrm{W}(\mathbf{r}, \mathbf{r}') = 0$. (We are using here a discrete notation for $\mathbf{r}$ and $\mathbf{r}'$; in the continuous, long-wavelength limit the summations will be replaced by integrations.) $\mathrm{W}(\mathbf{r}, \mathbf{r}')$ are taken to be random variables which have a given probability distribution and statistical properties which depend on the problem at hand. We denote ensemble averaged quantities by $\langle \cdots \rangle$, e.g., $\langle P \rangle$, $\langle \mathrm{W} \rangle$, etc. We wish to evaluate $\langle P(\mathbf{r}, t) \rangle$, given the statistical properties of W and the initial condition: $\langle P(\mathbf{r}, 0) \rangle = \delta_{\mathbf{r},0}$. Since the ensemble-averaged system is translationally invariant it is convenient to switch to $\mathbf{k}$ space by defining

$$\langle P(\mathbf{k}, t) \rangle = \sum_{\mathbf{r}} \langle P(\mathbf{r}, t) \rangle \exp(i\mathbf{k} \cdot \mathbf{r}) \tag{2}$$

The equation of motion derived by Hahn and Zwanzig (HZ)[5] is

$$\frac{d\langle P(\mathbf{k}, t) \rangle}{dt} = -\int_0^t d\tau \langle R(\mathbf{k}, t-\tau) \rangle \langle P(\mathbf{k}, t) \rangle \tag{3}$$

where

$$\langle R(\mathbf{k}, \tau) \rangle = -\langle W(\mathbf{k}) \rangle \delta(\tau) - \sum_{\mathbf{r}} \langle 0 | \mathrm{W} \exp(\hat{Q} \mathrm{W} t) \hat{Q} \mathrm{W} | \mathbf{r} \rangle \exp(i\mathbf{k} \cdot \mathbf{r}) \tag{3a}$$

and

$$\langle W(\mathbf{k}) \rangle = -\sum_{\mathbf{r}} \langle \mathrm{W}(\mathbf{r}) \rangle \exp(i\mathbf{k} \cdot \mathbf{r}) \tag{3b}$$

$\hat{P}$ is a projection operator which performs the ensemble averaging $\hat{P}A \equiv \langle A \rangle$ and $\hat{Q}$ is the complementary projection, $\hat{Q} \equiv 1 - \hat{P}$. We wish to suggest here an alternative equation. Instead of (3) we write the time local (TL) equation:

$$\frac{d\langle P(\mathbf{k}, t) \rangle}{dt} = -\int_0^t d\tau \, \tilde{R}(k, \tau) \cdot P(\mathbf{k}, t) \tag{4}$$

where

$$\langle \tilde{R}(\mathbf{k}, \tau) \rangle = -\langle W(\mathbf{k}) \rangle \delta(\tau) - \frac{d^2 \ln \langle \phi(\mathbf{k}, \tau) \rangle}{d\tau^2} \tag{4a}$$

and where

$$\langle \phi(\mathbf{k}, t) \rangle = \sum_{\mathbf{r}} \langle \mathbf{0} | \exp(\mathrm{W} t) | \mathbf{r} \rangle \exp(i\mathbf{k} \cdot \mathbf{r}) \tag{4b}$$

Equations of this type were proven recently very useful for other problems[8] and the proof of Eq. (4) is formally identical with the formalism developed elsewhere[8] where the starting equation was the Liouville equation [instead of (1)] and the averaging $\langle \cdots \rangle$ had a different meaning.

The HZ equation is most easily solved in the frequency domain, by performing a Laplace transform:

$$\langle P(\mathbf{k},s)\rangle \equiv \int_0^\infty d\tau \, \langle P(\mathbf{k},\tau)\rangle \exp(-s\tau) \tag{5a}$$

$$\langle R(\mathbf{k},s)\rangle = \int_0^\infty d\tau \, \langle R(\mathbf{k},\tau)\rangle \exp(-s\tau) \tag{5b}$$

we then have

$$\langle P(\mathbf{k},\tau)\rangle = \frac{1}{2\pi}\int_{-\infty}^{\infty} d\omega \exp(-i\omega\tau)\langle P(\mathbf{k},s=-i\omega)\rangle \tag{6a}$$

where

$$\langle P(\mathbf{k},s)\rangle = \frac{1}{s+R(\mathbf{k},s)} \tag{6b}$$

and where

$$\langle R(\mathbf{k},s)\rangle = k^2 D_2(s) + k^4 D_4(s) + \cdots \tag{6c}$$

On the other hand the solution of the TL equation is

$$\langle P(\mathbf{k},t)\rangle = \exp\left[-\int_0^t d\tau \,(t-\tau)\langle \tilde{R}(\mathbf{k},\tau)\rangle\right] \tag{7a}$$

where

$$\tilde{R}(\mathbf{k},\tau) \equiv k^2\tilde{D}_2(\tau) + k^4\tilde{D}_4(\tau) + \cdots \tag{7b}$$

In Eqs. (6c) and (7b) we have assumed that our system is isotropic so that only even powers of $k (k = |\mathbf{k}|)$ appear.

Both solutions (6) and (7) are formally exact but as we shall see they may yield very different results for $\langle P(\mathbf{k},t)\rangle$ once the kernels $\langle R\rangle$ or $\langle \tilde{R}\rangle$ are evaluated approximately such as when the expansions (6c) or (7b) are truncated.

In order to compare Eqs. (6) and (7) let us define the nth moment of the distribution $\langle P(\mathbf{r},t)\rangle$, i.e.,

$$M_n(t) \equiv \int d\mathbf{r}\, r^n \langle P(\mathbf{r},t)\rangle \tag{8}$$

where by construction $M_0 = 1$. The various moments may be obtained directly from $\langle P(\mathbf{k},t)\rangle$ using the identity

$$M_n = -i^n \frac{d^n}{dk^n}\langle P(k,t)\rangle_{k=0} \tag{9}$$

where $k = |\mathbf{k}|$. For one dimension $\langle P(\mathbf{k},t)\rangle = \langle P(k,t)\rangle$ but for higher dimensionalities one should add an appropriate phase space factor.

Using the expansion (6c) and (7b) it is clear that if we truncate them at nth order (i.e., retaining terms up to k^n) then the first n moments $M_1 \ldots M_n$ will be exact for both expansions. However, the two expansions will have different predictions regarding the higher moments. The choice of the equation [either Eq. (3) or Eq. (4)] is therefore equivalent to an ansatz regarding the behavior of the higher moments. We shall now explore this hidden ansatz by considering the shape of the distribution $\langle P(\mathbf{r},t)\rangle$ for the common case where we truncate the expansions to $O(k^2)$ (i.e., the long-wavelength limit). Using Eq. (9) it is clear that all the odd moments M_1, M_3, etc. vanish identically in our case since no odd powers of k appear in $\langle P(k,t)\rangle$.

For the HZ equation (6) we have

$$\langle P(\mathbf{r},s)\rangle = \int \frac{k^{d-1} dk \exp(-i\mathbf{k}\cdot\mathbf{r})}{s + k^2 D_2(s)} \tag{10}$$

which may be represented as

$$\langle P(r,s)\rangle = (s\xi^d)^{-1} F(r/\xi) \tag{11a}$$

where

$$\xi(s) = \left[\frac{D_2(s)}{s}\right]^{1/2} \tag{11b}$$

and

$$F(x) \propto \int_0^\infty dy \exp(iyx) \frac{y^{d-1}}{1+y^2} \frac{J_{d/2-1}(y)}{y^{d/2-1}} \tag{11c}$$

Here $J_p(y)$ is a Bessel function of the first kind. An asymptotic evaluation of $F(x)$ for $x \to \infty$ results in[9]

$$F(x) \xrightarrow[x\to\infty]{} \frac{\mathrm{xxp}(-x)}{x^{(d-1)/2}} \tag{11d}$$

Turning now to the TL equation we have upon truncating $\langle \tilde{R}(k,t)\rangle$ to $O(k^2)$

$$\langle P(\mathbf{k},t)\rangle = \exp(-\tfrac{1}{2}k^2\xi^2) \tag{12}$$

which gives

$$\langle P(\mathbf{r},t)\rangle = \left[(2\pi)^{1/2}\xi\right]^{-d} \exp\left[-r^2/2\xi^2\right] \tag{13}$$

where

$$\xi^2 = M_2(t) = 2\int_0^t d\tau\,(t-\tau)\tilde{D}_2(\tau) \tag{13a}$$

In conclusion we note the following:

1. When the only available information is $M_2(t)$ [either from experiment or from a truncated diagramatic expansion of $\langle R \rangle$ or $\langle \tilde{R} \rangle$ to $O(k^2)$] then the TL equation predicts a Gaussian spatial probability distribution [Eq. (13)]. This prediction is consistent with the maximum entropy distribution (i.e., the least biased probability distribution with the given second moment). In contrast the distribution obtained from the HZ equation [Eq. (11)] is more complicated, and in general is very different.

2. The higher moments predicted by the second-order TL equation are very simple:

$$M_{2p}^{(\mathrm{TL})}(t) = \frac{(2p)!}{p!\,2^p}\left[M_2(t)\right]^p \qquad (\text{all } d) \tag{14a}$$

This result holds for all dimensionalities d. On the other hand the predictions of the HZ equation are more complicated. For one-dimensional problems we have

$$\begin{aligned} M_{2p}^{(\mathrm{HZ})}(t) = \frac{(2p)!}{2^p} \int_0^t d\tau_1 \int_0^{\tau} d\tau_2 \cdots \int_0^{\tau_{p-2}} d\tau_{p-1}\, M_2(t-\tau_1) \\ \times \dot{M}_2(\tau_1-\tau_2)\cdots \dot{M}_2(\tau_{p-2}-\tau_{p-1})\dot{M}_2(\tau_{p-1}) \qquad (d=1) \end{aligned} \tag{14b}$$

where $\dot{M} = dM/dt$. In particular, for M_4 we have

$$M_4^{(\mathrm{HZ})}(t) = 6\int_0^t d\tau\, M_2(t-\tau)\dot{M}_2(\tau) \qquad (d=1) \tag{15a}$$

$$M_4^{(\mathrm{TL})}(t) = 3M_2^2(t) \qquad (\text{all } d) \tag{15b}$$

3. In the Markovian limit we assume

$$D_2(s) \cong D_2 = \text{const} \tag{16a}$$

$$\tilde{D}_2(t) = D_2\delta(t) \tag{16b}$$

so that $\xi^2(t) = 2D_2t$ and $\xi^2(s) = D_2/s$. In this case both equations reduce to the ordinary diffusion equation:

$$\frac{d\langle P(\mathbf{k},t)\rangle}{dt} = -k^2D_2\langle P(k,t)\rangle \tag{17a}$$

whose solution is

$$P(r,t) = (4\pi D_2t)^{-d/2}\exp(-r^2/4D_2t) \tag{17b}$$

and

$$M_{2p}(t) = \frac{(2p)!}{p!}(D_2t)^p, \qquad p \geqslant 1 \tag{18}$$

4. The results (11) and (13) hold when we truncate our expansions (6c) or (7b) to $O(k^2)$. By going to higher orders (k^n) we may guarantee that both formulations will agree for the first n moments. At infinite order they are both exact.

5. The form (11a) together with (11d) and in particular the relation $\langle P(r=0,s)\rangle \sim (\xi s)^{-1}$ for $d=1$ was suggested recently,[4,10,11] as an Ansatz based on a scaling argument. In the present formulation this is a straightforward result of the second-order HZ equation and Eqs. (11) generalize this result to all d. Moreover, Eq. (13) provides an alternative prediction, i.e., $P(r=0,t)=[(2\pi)^{1/2}\xi(t)]^{-d}$. Experiments or numerical simulations should be used to decide whether (11) or (13) are to be preferred.

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