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Classical Transport in Modulated Structures

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Classical transport coefficients in a d-dimensional medium with a potential $V(\mathbf{x})$ and/or conductivity $a(\mathbf{x})$ are found to vary discontinuously as functions of the "wavelengths" of the inhomogeneities. For example, with a potential depending on one direction only, say $V(\mathbf{x}) = \cos 2\pi x_1 + \cos 2\pi k x_1$, the effective diffusion coefficient $D(k)$ has the same value D^* for all *irrational* k, but differs from D^* and depends on k for k *rational*. Thus $D(k)$ is discontinuous at rational k. Moreover, $D(k)$ is continuous at irrational k. This pathology is reflected in the time scales on which the diffusion approaches its limiting behavior.

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1. Introduction.—There is current interest in transport in random systems and in modulated structures.[1-4] Here we describe the very discontinuous dependence of the effective diffusion constant D, and other transport coefficients, on the wavelengths of the inhomogeneities in a periodic, quasiperiodic, or random medium. These discontinuities arise from the infinite-time and -volume limits involved in computing stationary transport coefficients. The pathologies also show up, however, in the variations of the length and time scales on which the system approaches its limiting behavior, and necessitate the use of caution in carrying out and interpreting computer simulations. They also affect the low-frequency transport properties of the medium.

We consider first the motion of a particle in a diffusive medium with a spatially varying drift proportional to a force (Smoluchowski regime). Different transport coefficients and/or inhomogeneities behave in a similar way. The results also hold for lattice systems.

Let $\mathbf{X}(t)$ be the position of a particle at time t diffusing in R^d in a medium with a bounded (sufficiently smooth) potential V according to

$$d\mathbf{X}(t) = -\sigma_0 \nabla V(\mathbf{X}(t))\,dt + (2D_0)^{1/2} d\mathbf{W}(t), \quad (1)$$

where $\mathbf{W}(t)$ is standard Brownian motion, $\langle W_i(t) W_j(t)\rangle = \delta_{ij} t$, $i,j = 1,\ldots,d$, and σ_0 and D_0 are the "bare" mobility and diffusion constants. The density $\rho(x,t)$ associated with (1) satisfies the diffusion equation

$$\partial\rho/\partial t = D_0 \Delta\rho + \nabla\cdot[\sigma_0(\nabla V)\rho], \quad (2)$$

which has the equilibrium density $\rho \sim \exp[-\beta V]$, $\beta = \sigma_0/D_0$, as per the Einstein relation. For $\mathbf{X}(0) = \mathbf{x}_0$, $\rho(\mathbf{x},0) = \delta(\mathbf{x}-\mathbf{x}_0)$.

It is known[5-7] that for V periodic, quasiperiodic, or stationary random ergodic,

$$\mathscr{D}_{ij}(t,V) = (2t)^{-1}\langle \delta X_i(t)\,\delta X_j(t)\rangle_V \underset{t\to\infty}{\longrightarrow} D_{ij}(V), \quad i,j = 1,\ldots,d. \quad (3)$$

Here $\delta X_i = X_i(t) - X_i(0)$, $\langle\ \rangle_V$ denotes average over Brownian motion paths in (1), and $\mathbf{D}(V)$ is a positive-definite effective-diffusion tensor. The actual trajectories are asymptotically Brownian with diffusion tensor $\mathbf{D}(V)$, i.e.,

$$\epsilon\mathbf{X}(t/\epsilon^2) \underset{\epsilon\to 0}{\longrightarrow} \mathbf{W}_{D(V)}(t). \quad (4)$$

The computation of $\mathbf{D}(V)$ involves spatial averages of functionals of V so that $\mathbf{D}(V)$ (but not $\mathscr{D}$) is independent of the starting position $\mathbf{X}(0)$ and (with probability one) of the realization of the random potential V. In particular, when the potential varies only in one direction, $V=V(x_1)$, i.e., the medium is layered, then $D_{ij}=D_{ii}\delta_{ij}$, $D_{ii}=D_0$, $i\neq 1$, while $D_{11}=D(V)$ is given (see, e.g., Ref. 7 and Ferrari, Goldstein, and Lebowitz[8]) by

$$D(V)/D_0=\langle e^{\beta V}\rangle^{-1}\langle e^{-\beta V}\rangle^{-1}. \tag{5}$$

Here,

$$\langle \phi(V)\rangle = \lim_{N\to\infty}\frac{1}{2N}\int_{-N}^{N}\phi(V(x))dx. \tag{6}$$

We note that when $V(x)$ is periodic with period L, $V(x)=\hat V(x/L)$, $\hat V(x)=\hat V(x+1)$, then $\langle\phi(V)\rangle =\int_0^1\phi(\hat V(x))dx$ and D is independent of L. The same is true if $V(x)$ is a random potential with a scale L; e.g., if $V(x)=\pm\epsilon$ in intervals of random lengths s with a density $p(s)=L^{-1}e^{-s/L}$, then (5) gives $D(V)/D_0=(\cosh\beta\epsilon)^{-2}$.

The scale invariance of $\mathbf{D}$ follows generally from (1) or (2). Letting $V_\lambda(\mathbf{x})=V(\lambda\mathbf{x})$, $\mathbf{D}(\lambda)=\mathbf{D}(V_\lambda)$, we find

$$\mathscr{D}(t,V_\lambda)=\mathscr{D}(\lambda^2 t,V)\underset{t\to\infty}{\longrightarrow}\mathbf{D}(\lambda)=\mathbf{D}(V), \tag{7}$$

independent of λ, for $\lambda\neq 0$. However, $\mathbf{D}(\lambda)$ is discontinuous at $\lambda=0$ since $D_{ij}(\lambda=0)=D_0\delta_{ij}$ for any V. This can be understood from the fact, seen explicitly in (7), that the time t_λ that it takes $\mathscr{D}(t,V_\lambda)$ to approach its limiting value $\mathbf{D}(V)$ goes as λ^{-2}, the square of the length scale of the inhomogeneity. Note that the discontinuity in $\mathbf{D}(\lambda)$ at $\lambda=0$ appears only in the infinite-time limit since $\mathscr{D}(t,V)$ for finite t is, in fact, continuous in V.

There are, however, more interesting and pathological discontinuities in $\mathbf{D}(V)$ which are less readily made explicit. We shall therefore describe them first for $d=1$ (or the layered structure) in the next section where everything can be obtained directly from Eqs. (5) and (6). The case of inhomogeneous conductivity is also described in Sec. 2. We consider more dimensions in Sec. 3, while the approach to the asymptotic behavior, is discussed in Sec. 4.

2. Discontinuities in d=1.—We illustrate the discontinuous dependence of $D(V)$ on V by considering the example $V(x)=\cos 2\pi x+b\cos 2\pi kx$. Equations (5) and (6) yield

$$D(k)=\begin{cases} D_0\{\int_0^1\exp[\beta(\cos 2\pi px+b\cos 2\pi qx)]dx\}^{-1}\{\int_0^1\exp[-\beta(\cos 2\pi px+b\cos 2\pi qx)]dx\}^{-1}, & k=q/p,\\ D_0(\int_0^1 e^{\beta\cos 2\pi x}dx)^{-2}(\int_0^1 e^{\beta b\cos 2\pi x}dx)^{-2}=D^*, & k \text{ irrational},\end{cases} \tag{8}$$

where p and q are integers. We see that $D(k)$ has the same value D^* for all irrational k, but differs from D^* and depends on k for (almost) all rational k. Furthermore, if $k_n\to k$ $(k_n\neq k)$, then $D(k_n)\to D^*$, regardless of the rationality of k_n or k. Thus $D(k)$ is discontinuous at (almost) all rational k but continuous at the irrationals.

The mathematical origin of the bizarre behavior exhibited in (8) is very simple. For functions of the form $V(x)=\hat V(k_1x,\dots,k_nx)=\hat V(\mathbf{k}x)$, where $\hat V(y_1,\dots,y_n)$ has period 1 in each y_i and $\mathbf{k}=(k_1,\dots,k_n)$, the spatial averages in (6) correspond by the ergodic theorem to averages over T^n, the n-dimensional torus. In fact, with $\hat\phi(\mathbf{y})=\phi(\hat V(\mathbf{y}))$,

$$\langle\phi(V)\rangle=\langle\hat\phi(\mathbf{y})\rangle=\int_{T^n}\hat\phi(y_1,\dots,y_n)\mu_{\mathbf{k}}(y_1,\dots,y_n)d^ny, \tag{9}$$

where

$$\mu_{\mathbf{k}}(\mathbf{y})=\lim_{T\to\infty}\frac{1}{T}\int_0^T\delta(\mathbf{y}-\mathbf{k}t)dt \tag{10}$$

is the (weak) limit of the δ function along a trajectory of the flow $\dot y_i=k_i$ on T^n starting at the origin. When $\mathbf{k}$ is "irrational," i.e., $\sum m_ik_i=0$ has no nontrivial solutions for integers m_i, then $\mu=1$ on T^n. However, when $\mathbf{k}$ is "rational," $\mu_{\mathbf{k}}d^ny$ is uniformly distributed on an l-dimensional torus, $l<n$, embedded in T^n (and is zero elsewhere on T^n). The discontinuous dependence of $\mu_{\mathbf{k}}$ on $\mathbf{k}$ renders $D(\mathbf{k})$ discontinuous, in the same way as $D(k)$ above.

To obtain a more physical understanding of the phenomenon, we observe that if in our example we introduce a phase $\boldsymbol\theta=(\theta_1,\theta_2)$ by setting

$$V(x,\boldsymbol\theta)=\cos 2\pi(x+\theta_1)+b\cos 2\pi(kx+\theta_2),$$

then $D(k,\boldsymbol\theta)$ will depend on $\boldsymbol\theta$ for k rational but not for k irrational. It should be noted, however, that even after we average $D(k,\boldsymbol\theta)$ over $\boldsymbol\theta$, with weight $\sim\exp[-\beta(\cos 2\pi\theta_1+b\cos 2\pi\theta_2)]$, the result $\bar D(k)$ for k rational is still unequal to D^*. In fact, $\bar D(k)>D^*$ for all rational k.

We can also treat the more general case of a random phase change (or noise in the wave vector), e.g., $\boldsymbol\theta=(0,\theta(x))$, $\theta(x)=\int_0^x\epsilon(y)dy$, where $\epsilon(x)$ is itself some stationary process with mean zero. Then, if and only if $\epsilon(x)$ is "good" noise (having good mixing properties) will $D(k,\boldsymbol\theta)=D^*$ regardless of k.

The above results all follow from (5). In fact it is easy to see from (5) that if $V = V_1 + V_2$, where V_1 and V_2 are "independent" potentials, then

$$\frac{D(V_1+V_2)}{D_0} = \frac{D(V_1)}{D_0}\frac{D(V_2)}{D_0}. \tag{11}$$

Equation (8) for irrational k may thus be regarded as a special case of (11), where $V_1 = \cos 2\pi x$ and $V_2 = b\cos 2\pi kx$. For k rational, on the other hand, V_1 and V_2 are not "sufficiently independent." [Equation (11) is true when the direct product of the spatial translation groups arising from V_1 and V_2 is ergodic, which holds, e.g., when V_2 is weakly mixing or when V_1 and V_2 are quasiperiodic with incommensurate wave vectors.]

We may also consider the effect of adding to the potential in (8) an "independent" potential, say the one described in Sec. 1. In view of (9), the resulting diffusion constant would then be

$$D = D(k)[\cosh\beta\epsilon]^{-2} \underset{\epsilon\to 0}{\longrightarrow} D(k).$$

Thus *additive* noise does not destroy the difference between k rational and irrational. It is also worth noting that D in $d=1$ is invariant under a random scaling, i.e., if $\tilde{V}(x) = V(\int_0^x \lambda(y)dy)$, then $D(\tilde{V}) = D(V)$ under sufficiently strong mixing conditions on V and λ.

As mentioned previously, our analysis applies equally well to other transport coefficients, e.g., heat conduction in a medium with a spatially varying conductivity $a(\mathbf{x})$. Then the analog of (2) takes the form

$$\partial u/\partial t = \nabla\cdot[a(\mathbf{x})\nabla u], \tag{12}$$

where u is the temperature. The effective conductivity matrix $\mathbf{A}$ is again equal to spatial averages of some functional of a. For $d=1$ we again have an explicit formula $A(a) = \langle a^{-1}\rangle^{-1}$, which leads to results analogous to those obtained for $D(V)$. The same formula holds for the effective diffusion constant of the discrete version of the diffusion process associated with (12), so that similar discontinuous behavior occurs for lattice systems.[7,9]

3. More dimensions.—There are no explicit formulas like (5) for effective transport coefficients when the inhomogeneities depend on more than one space dimension, and we cannot give general proofs of the existence of the discontinuities. Instead we will give, below, examples of two types of higher-dimensional systems which exhibit the discontinuous behavior. However, it would be surprising if this behavior is *not* generic, since the origins of it discussed for $d=1$ persist in more dimensions. In particular, the diffusion matrix has the representation[7]

$$D_{ij}(V) = D_0\delta_{ij} - \frac{\sigma_0^2}{2}\int_0^\infty \left\langle\left\langle \frac{\partial V}{\partial x_i}(\mathbf{X}(0))\frac{\partial V}{\partial x_j}(\mathbf{X}(s))\right\rangle\right\rangle_V ds, \tag{13}$$

where $\langle\langle\ \rangle\rangle_V$ denotes average over Brownian motion paths and initial position {with weight $\sim\exp[-\beta V(\mathbf{X}(0))]$}. For

$$V(\mathbf{x}) = V_\mathbf{k}(x_1,\ldots,x_d) = \hat{V}(k_1^1x_1,\ldots,k_{n_1}^1x_1,\ldots,k_1^dx_d,\ldots,k_{n_d}^dx_d),$$

the spatial average contained in (13) corresponds to an average over T^n with respect to a d-dimensional version of (10) (which involves d-dimensional t and extra weight $e^{-\beta\hat{V}}$), where $n = \sum_{i=1}^d n_i$ and

$$\mathbf{k} = (k_1^1,\ldots,k_{n_1}^1,\ldots,k_1^d,\ldots,k_{n_d}^d).$$

As before, $\mu_\mathbf{k}$ is nonzero only on an l-dimensional torus inside T^n, with $l=n$ when the k_i^j, for each fixed $1\le j\le d$, are rationally independent, and $l<n$ otherwise. Thus while V is continuous in $\mathbf{k}$, $\mu_\mathbf{k}$ is not, so that D_{ij} in (13) should generally be discontinuous in $\mathbf{k}$. Furthermore, for irrational $\mathbf{k}$, D_{ij} does not depend on the phase of V [see the sentence following Eq. (4)], while for $\mathbf{k}$ rational ($l<n$), D_{ij} should in general be phase dependent, again implying discontinuity. In fact, it is easy to see that given any rational $\mathbf{k}_0$, there exists a $\hat{V}_0$ on T^n such that $D_{ij}(V_{0,\mathbf{k}})$ is phase dependent, and hence discontinuous, at least at $\mathbf{k}=\mathbf{k}_0$.

Another example which exhibits discontinuous behavior in more dimensions (say, $d=2$) is the following. As mentioned before, Sec. 2 applies verbatim to layered structures varying in the x_1 direction. For spatially varying conductivity $a(\mathbf{x})$, it is easy to construct such an example by slight modulation of the layered environment. For example, consider

$$a(x_1,x_2) = [3+\cos 2\pi x_1 + \cos 2\pi kx_1][1+\epsilon f(x_1,x_2)],$$

where $0\le f(x_1,x_2)\le 1$, $\epsilon>0$. Now $a_1(\mathbf{x})\le a_2(\mathbf{x})$ implies that the corresponding bulk conductivities satisfy $A_1\le A_2$. Then for any fixed rational k_0, the bulk conductivity associated with our $a(\mathbf{x})$ will be discontinuous at k_0 when $\epsilon < |A^* - A(k_0)|/\min[A^*, A(k_0)]$, where A^* $[A(k_0)]$ is the bulk conductivity of the layered environment for irrational k (rational k_0).

4. Approach to the limiting behavior and low-frequency transport.—Let V be a general ergodic potential in R^d. Then $\mathscr{D}(t,V)$ defined in (3) is equal to $D_0\mathbf{I}$ for $t=0$, where $\mathbf{I}$ is the identity, and approaches $\mathbf{D}(V)$ as $t\to\infty$. The time τ_V necessary for $\mathscr{D}(t,V)$ to approach its asymptotic value will depend on the length scale L on which V varies. It is only for $t \gg \tau_V = L^2/D_0$ that the particle knows what V is really like, as can be seen explicitly via (7).

Consider now $\mathscr{D}(t,k)$ for the potential considered in (8). Let $k_n = k+\epsilon_n$, $0\neq\epsilon_n\to 0$ as $n\to\infty$, with k rational. The time τ_n after which $\mathscr{D}(t,k_n)\sim D(k_n)$ will grow with n. More precisely, for $\delta > 0$, let

$$t_\delta(k) = \inf\{t>0 \,|\, \sup_{s\geq t} |\mathscr{D}(s,k) - D^*| \leq \delta\}.$$

Then $t_\delta(k)\to\infty$ as $k\to k'$, k' rational, whenever $|D(k')-D^*|>\delta$, since $\mathscr{D}(t,k)$ is continuous in k for finite t. Furthermore, if k is "well approximated" by rationals k_n, then we expect that there is a sequence of times $t_n(k)$ such that $\mathscr{D}(t,k)\sim D(k_n)$ for $t\sim t_n(k)$.

Thus we see that the time dependence of the system is quite sensitive to the nature of the inhomogeneities. This sensitivity for $a(\mathbf{x})$ (now electrical conductivity) will affect the low-frequency ac conductivity $\sigma(\nu)$, which is proportional to the one-sided Fourier transform of the mean square displacement,[2,10]

$$\sigma(\nu)\sim \nu^2\int_0^\infty e^{-i\nu t}\langle\langle |\mathbf{X}(t)-\mathbf{X}(0)|^2\rangle\rangle\, dt. \qquad (14)$$

Here $\langle\langle\ \rangle\rangle$ denotes averaging over Brownian motion paths and over the initial position $\mathbf{X}(0)$. The *structure* of $\sigma(\nu)$ for small ν will depend markedly on the modulation of $a(\mathbf{x})$. This dependence can be studied through analysis of the spectral properties of the generator $L=\nabla\cdot a(\mathbf{x})\nabla$ of the process $\mathbf{X}(t)$. For periodic $a(\mathbf{x})$, L on the appropriate Hilbert space has (negative) discrete spectrum, which renders $\sigma(\nu)$ analytic in a neighborhood of $\nu=0$. For quasiperiodic $a(\mathbf{x})$, we believe that the origin is a limit point of the spectrum of L, so that $\sigma(\nu)$ near $\nu=0$ is presumably analytic only in a neighborhood which excludes $\nu=0$ and the negative imaginary axis near 0. Details will appear elsewhere.

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