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On the approximate solution of non-deterministic heat and mass transport problems

JOHN G. GEORGIADIS

Department of Mechanical Engineering and Materials Science, Duke University, Durham, NC 27706, U.S.A.

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Abstract—Uncertainties inherent to transport processes in realistic heterogeneous media can be described by non-deterministic equations with random coefficients. In this paper, we undertake an analytical study of three classes of heat and mass transfer phenomena described by convection–diffusion reaction continuum models and discrete models: (1) unsteady dispersion in a random filtration velocity field; (2) anomalous diffusion in media with random reaction sites; (3) size effect on thermal conductivity of isotropically disordered solid lattices. Using small perturbation analysis, we solve three non-trivial problems described by differential equations with random coefficients. Although the random part of the parameters is much smaller than the deterministic (weak disorder), the effect of randomness on the behavior of the averaged quantities is both important and counterintuitive.

1. INTRODUCTION

THE ANALYTICAL study of heat and mass transfer processes in media involving very short or very large length scales (e.g. microelectronics or geophysical media) can introduce non-deterministic systems. The urgent nature of underground waste management (as it pertains to the environmental restoration) has focused the attempts to predict solute movement in aquifers and field solids. Although such phenomena involve field scale distances and times, the present body of experimental data is neither extensive nor precise as a result of the large-scale nonuniformity of geological strata. In other cases, randomness can be introduced to facilitate the description of a complex system. There has always been an interest to model transport processes in physiological media of complicated geometry (e.g. cardiovascular or respiratory networks) but the inherent randomness of their structure calls for special methods. Occasionally, disorder is desirable. For example, the presence of impurities and other defects in solids, generate a wider range of properties than would be available with 'perfect' lattices.

In most man-made and natural systems the source of uncertainty is threefold: (a) model uncertainty (limited sample), (b) parameter uncertainty (sampling error), and (c) input error (error in measuring initial and boundary conditions or system inputs). In this work, we study systems with type (b) uncertainty. Uncertainties in structural or thermophysical parameters can be most economically described by statistics. For example, transport processes in heterogeneous media of disordered microstructure (e.g. fluid-saturated porous media) have been described by continuum models with stochastic parameters, cf. Sposito *et al.* [1]. In order for stochastic models to be useful, one needs to relate microscopic statistical information to macroscopic (average) properties. At first order, the following problem needs to be solved: given an a priori statistical description of the stochastic field, predict the average (or 'effective') value. Three classes of methods are available for the solution of such non-deterministic problems: (i) Monte Carlo simulation, (ii) perturbation analysis, cf. Keller [2], and (iii) direct numerical methods, cf. Padovan and Guo [3]. In this work, approximate methods of class (ii) will be employed in the solution of differential equations with stochastic coefficients that model certain heat and mass transport problems.

We present a very brief overview of available methodologies for the solution of stochastic heat and mass transfer problems. One group of researchers have implemented classical methods of the field of applied mechanics. Samuels [4] obtained analytical solutions for the one-dimensional heat conduction equation with random boundary conditions and random heat generation. Ahmadi [5] solved the unsteady heat conduction equation with spatially random heat conductivity following a perturbation scheme developed by Keller [2]. Tzou [6, 7] derived the statistics of one-dimensional temperature fields in solids with random conductivity. Employing n-point probability functions, another group of researchers (with statistical mechanics background) have developed a class of techniques to obtain rigorous bounds of the effective transport coefficients for linear transport processes in two-phase random media under steady-state conditions. Weissberg [8] was the first to derive lower-order variational bounds for the effective diffusion coefficient through a bed of spheres. Torquato and Lado [9] obtained high-order bounds for the effective conductivity of composite media. Recently, Rubinstein and Torquato [10] derived rigorous bounds of effective properties associated with the diffusion-controlled reaction equation. A critical

NOMENCLATURE

$A\{\cdot\}$, $\tilde{A}\{\cdot\}$ differential operators, equation (5)
c species concentration
D transport coefficient
k wave number
m_n random mass of nth mode
N number of nodes in the harmonic chain, Fig. 3
q mean velocity, equation (2)
Q, R, S correlation functions
$\mathbf{r}$ position vector
t time
w longitudinal component of velocity
$\mathbf{x}$ position vector in the transverse direction
z longitudinal coordinate.

Greek symbols
α species annihilation rate
β species production rate
γ spring constant of harmonic chain
$\delta(z)$ delta function
ε amplitude of random fluctuation, equation (2)
λ localization factor
μ random mass variation, equation (22) [dimensionless]
σ^2 variance, $\langle\mu^2\rangle$
τ correlation time
Ψ Schrödinger eigenfunction
ω radial frequency.

Special symbols
$*$ complex conjugate
$O(\ldots)$ order of ...
$o(\ldots)$ much less than ...
$\langle\cdot\rangle$ ensemble average, expected value.

review of the available literature suggests that the stochastic approaches adopted are determined by the problem to be solved. Extending the models to account for time-dependency or two- or three-dimensionality is not a trivial task.

In this work, elegant methods from the applied physics literature will be applied to three non-deterministic systems: (1) convective transport in columnar packed beds, (2) diffusive transport in the presence of a first-order autocatalytic reaction, and (3) a one-dimensional imperfect-lattice model of heat conduction in solids. We start from stochastic governing equations with random coefficients (of given statistics) and follow rigorous procedures of small perturbation analysis. The only empirical element of this approach is the statistical behavior of the random coefficients. Our methodology has been successfully used to derive the effective energy equation for convective transport in random packed beds, cf. ref. [11]. The appendix of the above article contains a short exposition of the necessary theoretical background for problems (1) and (2) which involve continuum models. Problem (3) involves a discrete model of heat conduction which can also have applications in solid mechanics.

Although we try to keep in mind the obvious technological applications of the present work, our current objective is to extend the applicability of stochastic methodologies borrowed from applied physics into the field of heat and mass transfer and to improve the predictive capacity of transport models. The physical problems suggested during the development of the solutions serve as a mere motivation to pursue this study. Furthermore, it is hoped that this work will contribute to further development of the mathematical tools borrowed from applied physics.

2. PROBLEM: APPROXIMATE SOLUTION OF CONVECTION–DISPERSION STOCHASTIC EQUATIONS

Assume that the concentration $c(z, t)$ of a conservative solute in an unsteady unidirectional velocity field inside a fully saturated porous medium is described by the following convection–dispersion equation, cf. Sposito *et al.* [1]

$$\frac{\partial c}{\partial t}+w(\mathbf{x},t)\frac{\partial c}{\partial z}=\frac{\partial}{\partial z}\left(D_{\mathrm{L}}\frac{\partial c}{\partial z}\right) \tag{1}$$

where z is the axial coordinate and $\mathbf{x}$ the two-component vector representing the coordinates in the transverse direction. The equation above describes the axial distribution of the concentration of a solute which has been introduced uniformly over the transverse direction at $t = 0$. Such a situation arises, for example, in stratified flow in a typical sedimentary rock. The longitudinal transport coefficient D_{L} depends on the local filtration velocity, cf. ref. [11]. The macroscopic variation of permeability in a z-stratified porous medium produces non-uniform filtration velocity (which is unsteady at large Reynolds numbers) and transport coefficient distributions which are modelled by random processes. We can separate the velocity and the transport coefficient into deterministic and random parts

$$w(\mathbf{x},t)=q+\varepsilon Q'(\mathbf{x},t),\quad D_{\mathrm{L}}(\mathbf{x},t)=D+\varepsilon D'(\mathbf{x},t) \tag{2}$$

where q and D are constants. Assuming stochastic uniformity in space and time, we can define the following correlation functions:

$$\langle Q'(\mathbf{x}_1,t_1)Q'(\mathbf{x}_2,t_2)\rangle = Q(|\mathbf{x}_1-\mathbf{x}_2|,|t_1-t_2|)$$
$$\langle Q'(\mathbf{x}_1,t_1)D'(\mathbf{x}_2,t_2)\rangle = R(|\mathbf{x}_1-\mathbf{x}_2|,|t_1-t_2|)$$
$$\langle D'(\mathbf{x}_1,t_1)D'(\mathbf{x}_2,t_2)\rangle = S(|\mathbf{x}_1-\mathbf{x}_2|,|t_1-t_2|). \quad (3)$$

Due to equations (2), the operators in equation (1) can be separated into deterministic and stochastic parts

$$\frac{\partial c}{\partial t} = A\{c\}+\varepsilon\tilde{A}(\mathbf{x},t)\{c\} \quad (4)$$

where the deterministic and stochastic operators are defined as follows:

$$A\{\ \} = -q\frac{\partial}{\partial z}\{\ \}+D\frac{\partial^2}{\partial z^2}\{\ \}$$
$$\tilde{A}(\mathbf{x},t)\{\ \} = -Q'(\mathbf{x},t)\frac{\partial}{\partial z}\{\ \}+D'(\mathbf{x},t)\frac{\partial^2}{\partial z^2}\{\ \}. \quad (5)$$

Equations (4) and (5) define a multiplicative stochastic process, cf. Fox [12]. Now let τ be the maximum of the correlation times of functions (3). Following van Kampen [13] and Fox [12], we can obtain the solution of equation (4) in terms of a time-ordered cumulant expansion and then take its ensemble average. The result is the solution of an evolution equation which can be truncated to give the following equation correct to order $O(\varepsilon^2\tau)$:

$$\frac{\partial\langle c\rangle}{\partial t} = A\{\langle c\rangle\}+\varepsilon^2K_2\{\langle c\rangle\}. \quad (6)$$

Formally, equation (6) is an integro-differential equation. For large times $t \gg \tau$ and because of the cluster property [14], the second operator on the right-hand side of equation (6) becomes

$$K_2\{\ \} = \int_0^\infty \langle\tilde{A}(\mathbf{x},t)\{\ \}\tilde{A}(\mathbf{x},t-t_1)\{\ \}\rangle\,\mathrm{d}t_1$$
$$= D_Q\frac{\partial^2}{\partial z^2}\{\ \}+D_S\frac{\partial^4}{\partial z^4}\{\ \}-2D_R\frac{\partial^3}{\partial z^3}\{\ \} \quad (7)$$

with the following renormalized transport coefficients:

$$D_Q = \int_0^\infty Q(0,t)\,\mathrm{d}t,\quad D_S = \int_0^\infty S(0,t)\,\mathrm{d}t,$$
$$D_R = \int_0^\infty R(0,t)\,\mathrm{d}t. \quad (8)$$

The evolution equation (6) becomes exact for all times in the case of delta-correlated temporal fluctuations ($\tau \to 0$).

Due to equations (5) and (7), the average transport equation (6) can be rearranged as follows:

$$\frac{\partial}{\partial t}\langle c\rangle+q\frac{\partial}{\partial z}\langle c\rangle = (D+\varepsilon^2D_Q)\frac{\partial^2}{\partial z^2}\langle c\rangle$$
$$+\varepsilon^2D_S\frac{\partial^4}{\partial z^4}\langle c\rangle-2\varepsilon^2D_R\frac{\partial^3}{\partial z^3}\langle c\rangle. \quad (9)$$

The randomness of the velocity field brings an enhancement of the transport coefficient by a hydrodynamic component ε^2D_Q (first term on the right-hand side of equation (9)). In addition, due to the statistical correlation between the velocity and axial transport coefficient as given by equations (3), higher order derivative terms appear in equation (9) and the transport equation for the average becomes different in form from the Fickian local transport equation (1). However, in the case of finite transport coefficients (8), the solution of equation (9) becomes Fickian for asymptotically large times. For example, the solution of the transport equation for the delta pulse initial condition $\langle c\rangle = \delta(z)$, reaches the Fickian solution as $t \to \infty$. The rate at which this asymptotic limit is approached depends on the magnitude of ε^2D_S and ε^2D_R (Burnett coefficients). The derivation of this asymptotic limit can be obtained by following Fox and Barakat [14] who solved an equation similar to equation (9) in terms of an asymptotic series. To the author's knowledge, this is the first time that an equation like equation (9) has been proposed for convective transport. Gelhar *et al.* [15], who considered steady velocity field and included transverse diffusion, arrived at a third-order equation which also exhibits Fickian behavior at large times.

It is important here to examine whether non-Fickian transport can occur early in the process described by equation (9). We will still assume that 'early' is not inconsistent with our postulate $t \gg \tau$ and that coefficients (8) are constant. Of course, the physics of the problem will dictate the correlation functions in equations (3). However, it is traditional in the theory of stochastic equations to introduce the statistics by postulation. We solve equation (9) subject to a step-like initial condition (Heaviside distribution): $\langle c\rangle = 1.0$ for $0 < z < 1$ and $\langle c\rangle = 0$ for $1 < z < 2$. We use a fourth-order accurate finite-difference scheme (grid size $\Delta z = 0.1$) and the parameters shown in Fig. 1. These parameters, as well as others in applications in the following sections, do not correspond

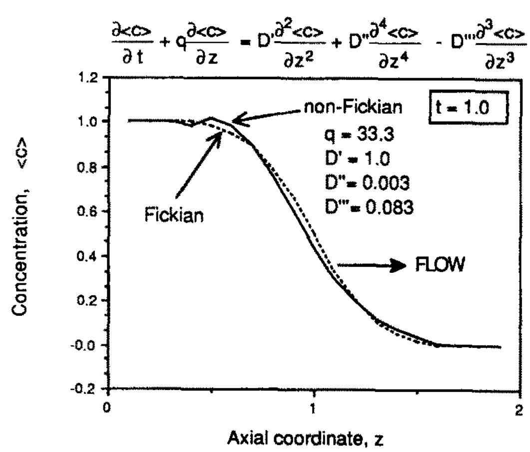

FIG. 1. Numerical solution of the fourth-order non-Fickian transport equation (9).

to actual physical data but serve to give a graphical representation of the solution. The solution is plotted in Fig. 1 for a certain time instant. The significant deviation (in the form of a damped wave upstream from the front) from the Fickian distribution ($D'' = D''' = 0$) can be attributed to the third-order dispersive term in equation (9).

This dispersive (in the classical sense) phenomenon in random fields is unexpected in view of the dissipative nature of the solute transport equation (1). In practice, one would expect that equation (9) holds when the velocity (and the transport coefficient which depends on it) fluctuates according to equation (4). Flow experiments through packed beds (made of spherical beads) reveal a disorganized temporal behavior of the interstitial velocity when the Reynolds number (based on the bead diameter) exceeds approximately 100. Sundaresan *et al.* [16] have previously proposed heuristic hyperbolic transport equations after examining dispersive transport of tracers in columnar porous beds for very high Reynolds numbers. Their observation of finite speed of signal propagation provides some corroborative evidence towards the justification of similar hyperbolic transport equations (such as equation (9)).

3. PROBLEM: EFFECT OF RANDOMLY DISTRIBUTED HOMOGENEOUS REACTION SOURCES

Consider the diffusion problem in a medium of random structure contained in a volume V in which a conserved species is produced by a homogeneous first-order reaction. Assume that the species concentration is governed by the following reaction–diffusion equation:

$$\frac{\partial}{\partial t}c(\mathbf{r},t) = D\nabla^2 c(\mathbf{r},t) + [\beta(\mathbf{r}) + \alpha(t)]c(\mathbf{r},t);$$

$$\mathbf{n}\cdot\nabla c = 0 \quad \text{on} \quad \partial V \tag{10}$$

where $\mathbf{r}$ is the position vector in V, ∂V the boundary of V, and $\mathbf{n}$ the normal unit vector. In addition to chemical processes, the above equation can be used to model several biophysical processes (bioheat transport or production and diffusion of metabolites in perfused tissues, population dynamics, etc.). The diffusion coefficient D characterizes the homogenized medium. The species is generated according to the spatially varying production rate $\beta(\mathbf{r})$. We have also assumed in equation (10) that the species is absorbed (annihilated) uniformly at a rate $\alpha(t)$ so that its volume content in V remains constant, cf. Zel'dovich [17]

$$\text{constant } c_0 = \int_V c(\mathbf{r},t)\,\mathrm{d}V. \tag{11}$$

The absorption term $\alpha(t)$ can be readily suppressed by applying a simple integral transformation which is given in this section (equation (19)).

The conservation constraint (11) and the zero-flux boundary condition for the concentration in equation (10) imply that the absorption rate is equal to the average production rate

$$\alpha(t) = -\frac{1}{c_0}\int_V \beta(\mathbf{r})c(\mathbf{r},t)\,\mathrm{d}V. \tag{12}$$

Using the simple exponential transformation

$$b(\mathbf{r},t) = c(\mathbf{r},t)\exp\left\{-\int_0^t [\beta(\mathbf{r})+\alpha(t')]\,\mathrm{d}t'\right\} \tag{13}$$

equation (10) is transformed to the following 'interaction picture' [12]:

$$\frac{\partial b(\mathbf{r},t)}{\partial t} = \tilde{B}(\mathbf{r},t)b(\mathbf{r},t) \tag{14}$$

with

$$\tilde{B}(\mathbf{r},t) = \exp\{-t\beta(\mathbf{r})\}\{D\nabla^2\}\exp\{t\beta(\mathbf{r})\}. \tag{15}$$

We can now approximate the operator (15) for small times and smooth $\beta(\mathbf{r})$ by expanding the exponentials in Taylor series:

$$\tilde{B}(\mathbf{r},t) \sim D\{[t\nabla^2\beta + t^2\nabla\beta\nabla\beta + O(t^3\beta\nabla\beta\nabla\beta)] + 2[t\nabla\beta + O(t^3\beta\nabla\beta)]\nabla + \nabla^2\}. \tag{16}$$

The solution to equations (13)–(15) can be given in terms of the time-ordered exponential [12]. The latter is identical to the ordinary exponential since $\tilde{B}(\mathbf{r}, t_1)$ commutes with $\tilde{B}(\mathbf{r}, t_2)$. Consequently, the exact solution of equations (13)–(15) can be formally written as follows:

$$c(\mathbf{r},t) = \exp\left\{\int_0^t [\beta(\mathbf{r})+\alpha(t')]\,\mathrm{d}t'\right\} \times \exp\left\{\int_0^t \tilde{B}(\mathbf{r},t)\,\mathrm{d}t\right\}c(\mathbf{r},0). \tag{17}$$

In order to examine the behavior of solution (17), we consider a delta-pulse initial condition $c(\mathbf{r},0) = c_0\delta(\mathbf{r}-\mathbf{r}')$, where $\mathbf{r}$ belongs to the three-dimensional space. Equations (16) and (17) give

$$c(\mathbf{r},t) \sim \exp\left\{\int_0^t [\beta(\mathbf{r})+\alpha(t')]\,\mathrm{d}t' + D\frac{t^2}{2}\nabla^2\beta + D\frac{t^3}{3}\nabla\beta\nabla\beta + O(Dt^4\beta\nabla\beta\nabla\beta)\right\} \times \frac{c_0}{(4\pi Dt)^{3/2}}\exp\left\{-\frac{1}{4Dt}[\mathbf{r}-\mathbf{r}' + t^2\nabla\beta + O(t^4\beta\nabla\beta)]^2\right\}. \tag{18}$$

The first factor on the right-hand side of expression (18) shows that the concentration increases faster in areas where the production rate $\beta(\mathbf{r})$ is larger than the average (12), while the second factor represents normal diffusion which is Fickian for small times.

Since the leading behavior of the first term is exponential in time, the species is accumulated in regions of high values of $\beta(\mathbf{r})$. An interesting result concerning the topology of such 'dense' regions has been obtained by Zel'dovich [17] for weak diffusion ($D \cong 0$). Assuming that the spatial variation of $\beta(\mathbf{r})$ follows the Gaussian distribution and using percolation theory, Zel'dovich [17] proved that the regions of high concentration become unconnected (islands). The resulting structure consists of dense inclusions imbedded in a 'dry' porous matrix.

Equation (18) is a novel asymptotic formula for the concentration distribution, correct to $O(Dt^4\beta\nabla\beta\nabla\beta)$. We perform a two-dimensional numerical simulation of equation (10) aimed at understanding the long time behavior of the solution. Starting with a pulse-like initial disturbance in a random field $\beta(\mathbf{r})$, we can observe that the disturbance follows an unpredictable pattern from one local maximum to another. After it diffuses according to expression (18), the peak of the disturbance 'tunnels' to the nearest maximum of $\beta(\mathbf{r})$ which is not necessarily the absolute maximum, as shown in Fig. 2. It is apparent that diffusion destroys the island structure predicted by Zel'dovich [17]. To examine this phenomenon, we solve the problem for an ensemble of numerically-generated random fields $\beta(\mathbf{r})$ following the Gaussian distribution. The results are surprising: after forming islands, the initial disturbance spreads in a random walk fashion for long times. Despite the fact that the above is a qualitative observation based on hundreds of numerical simulations (such as the one presented in Fig. 2), it indicates that the transport process modelled by the stochastic equation (10) does not exhibit a simple diffusive behavior.

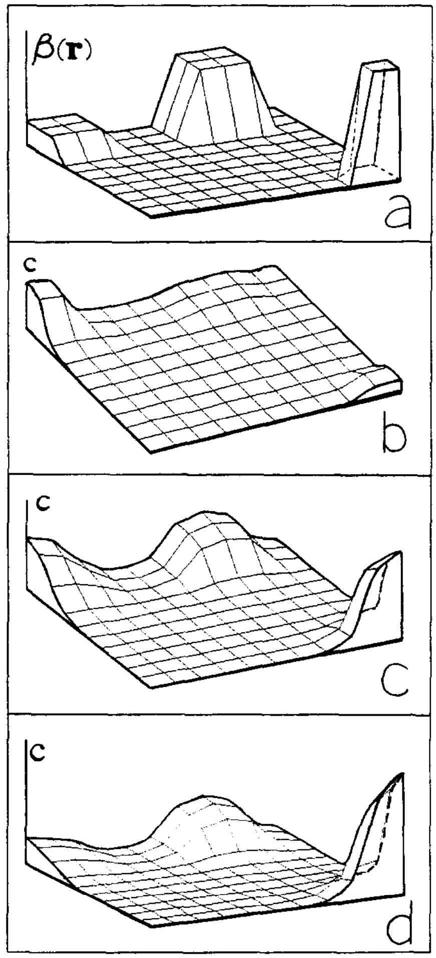

FIG. 2. Numerical simulation of the diffusion reaction equation with variable reaction rate distribution: (a) plot of the production rate distribution $\beta(\mathbf{r})$; (b)–(d) plots of the solution $c(\mathbf{r}, t)$ for three consecutive times.

To investigate whether this atypical behavior is caused by the random variation of the reaction rate $\beta(\mathbf{r})$, we apply the following transformation to the solution of equation (10):

$$C(\mathbf{r}, t) = c(\mathbf{r}, t) \exp\left\{-\int_0^t \alpha(t')\,\mathrm{d}t'\right\} \tag{19}$$

which yields

$$\frac{\partial}{\partial t}C(\mathbf{r}, t) = D\nabla^2 C(\mathbf{r}, t) + \beta(\mathbf{r})C(\mathbf{r}, t);$$

$$\mathbf{n}\cdot\nabla C = 0 \quad \text{on} \quad \partial V. \tag{20}$$

The solution of equation (20) can be written as a linear superposition of terms of the form $\exp\{\lambda_n t\}\Psi_n(\mathbf{r})$, where λ_n and Ψ_n are the eigenvalues and eigenfunctions, respectively, of the Schrödinger eigenvalue problem

$$[D\nabla^2 + \beta(\mathbf{r})]\Psi_n(\mathbf{r}) = \lambda_n \Psi_n(\mathbf{r}). \tag{21}$$

There is a growing body of knowledge concerning the type of the eigenfunctions Ψ_n, for various classes of random potentials $\beta(\mathbf{r})$, cf. Souillard [18]. One of the most interesting spectral properties of the random Schrödinger operator in equation (21) concerns the appearance of 'localized' states (eigenfunctions). This is related to the Anderson [19] localization phenomenon (since equation (21) also models the motion of electrons subjected to random potential fluctuations): certain electronic states can acquire a local character and thus an electric charge does not diffuse away.

We now return to the reaction–diffusion equation (10). We postulate that the existence and spatial distribution of these localized states is intimately connected with the degree of disorder of the potential $\beta(\mathbf{r})$. The quantification of the degree of disorder is beyond the scope of the present work. The non-diffusive behavior of our numerical results presented above can be qualitatively explained in terms of competition between localized states. This is a novel theoretical result. The prediction of localized 'hot spots' has important implications in connection with the anomalous dispersion of chemical or radioactive pollutants in the environment (pollutants generated by nonpoint sources) and the initiation of dry-out in fluid-saturated debris beds.

4. PROBLEM: LOCALIZATION OF LATTICE WAVES IN SOLIDS

In this section, we consider heat conduction by lattice waves in real solids with dimensions that are comparable to the characteristic physical length (e.g. the phonon mean free path). We focus our attention to the estimation of the thermal conductivity across solid firms of submicrometer thickness which is of growing importance to superconductor technology [20] and the evolving field of micromachinery. The classical microscopic model of heat conduction involves the concept of phonon scattering in the form of lattice waves which collide with electrons, with grain and external boundaries, and with each other. However, in the presence of crystal defects, the model of phonons is controversial because lattice vibrations become localized, cf. Toda [21].

Even in perfect crystals there is intrinsic thermal resistance due to common isotropic impurities. In order to model this intrinsic resistance, we assume that the crystalline structure is homogeneous, i.e. the crystal contains no grain boundaries. We will study a one-dimensional model of an isotropically disordered lattice consisting of a chain of N particles of varying mass

$$m_n = \langle m\rangle[1+\mu_n]; \quad n = 1,2,3,\ldots,N \to \infty \tag{22}$$

connected by harmonic springs of constant γ. The equation of motion at each node is $m_n\,\mathrm{d}^2u_n/\mathrm{d}t^2 = \gamma(u_{n+1}-2u_n+u_{n-1})$, where $u_n(t)$ is the displacement of the node. The governing equation for a monochromatic wave of frequency ω (which corresponds to a normal mode of vibration) can be written in the following transmission form:

$$\begin{bmatrix} u_{n+1} \\ u_n \end{bmatrix} = [T_n]\begin{bmatrix} u_n \\ u_{n-1} \end{bmatrix};$$

$$[T_n] = \begin{bmatrix} 2-\omega^2 m_n/\gamma & -1 \\ 1 & 0 \end{bmatrix}. \tag{23}$$

The corresponding ordered (periodic) system can transmit waves without attenuation only in the following frequency range:

$$0 \leqslant \omega\sqrt{(\langle m\rangle/\gamma)} \leqslant 2; \quad \cos k = 1-\omega^2\langle m\rangle/2\gamma \tag{24}$$

where k is the wave number. In the disordered system, these waves are not perfectly transmitted but decay exponentially with the distance in the lattice

$$|u_n| \approx \mathrm{e}^{-\lambda n}. \tag{25}$$

In the Appendix, we present the formulation of the system transfer matrix and the very useful theoretical result of Baluni and Willemsen [22] concerning localization. In order to apply that result, we first need to put the transfer matrix into the proper Hermitian form via the following similarity transformation:

$$[S]^{-1}[T_n][S] = \begin{bmatrix} (1+\mathrm{i}\Omega_n)\mathrm{e}^{\mathrm{i}k} & \mathrm{i}\Omega_n \\ -\mathrm{i}\Omega_n & (1-\mathrm{i}\Omega_n)\mathrm{e}^{-\mathrm{i}k} \end{bmatrix};$$

$$[S] = \begin{bmatrix} \mathrm{e}^{\mathrm{i}k} & 1 \\ 1 & \mathrm{e}^{\mathrm{i}k} \end{bmatrix} \tag{26}$$

where $\Omega_n = \omega^2\langle m\rangle\mu_n/(2\gamma\sin k)$. According to equations (A1)–(A3) in the Appendix, the localization factor is given by

$$\lambda = \tfrac{1}{2}\sigma^2\frac{\partial}{\partial\mu^2}\ln\left|\left(1+\frac{\omega^4\langle m\rangle^2\mu^2}{4\gamma^2\sin^2 k}\right)^{1/2}\right| + o(\sigma^2) \tag{27}$$

which reduces to

$$\lambda = \sigma^2\frac{\omega^2\langle m\rangle}{8\gamma}\left(1-\frac{\omega^2\langle m\rangle}{4\gamma}\right)^{-1} + o(\sigma^2) \tag{28}$$

where $\sigma^2 = \langle\mu^2\rangle$ is the (dimensionless) variance of the modal mass distribution according to equation (22). The following small wave number approximation of equation (28) can be easily obtained:

$$\lambda \to \sigma^2\frac{\omega^2\langle m\rangle}{8\gamma} \quad \text{as} \quad \omega \to 0. \tag{29}$$

The plot of the localization factor vs (dimensionless) frequency is given in Fig. 3. Expression (28) is valid for $\sigma^2 \ll 1$; this means that waves with frequencies in the range $1.4 < \omega(\langle m\rangle/\gamma)^{1/2} < 2.0$ are attenuated within a few hundred or thousand lattice nodes. For example, if the masses vary by 1% ($\sigma = 0.01$), a wave with frequency $\omega = 1.8(\gamma/\langle m\rangle)^{1/2}$ will decay by a factor of 10 after approximately 2300 nodes. Although no dissipation is included, the model exhibits dissipative behavior when large spatial scales are considered. Since lattice vibrations of all frequencies ultimately decay as $N \to \infty$, the thermal conductivity is finite in the case of disordered harmonic lattices, while it is infinite in the ordered system. The asymptotic relationship (29) is the well-known low frequency limit which has been obtained earlier with tedious analysis [23]. Based on this limit, Toda [21] derived an approximate expression for the thermal conductivity in terms of the number of normal modes which are extended through the lattice.

We use here the full expression of the localization factor (28) (valid for all frequencies in the passband (24)) and follow the same procedure [21, 23] to derive the following expression for the lattice conductivity:

$$K \approx ck_{\mathrm{B}}fN(N\sigma^2+2)^{-1/2} \tag{30}$$

where c denotes a constant which is $O(1)$, k_{B} the Boltzmann constant, and f the friction constant of the random (Langevin) force that characterizes the coupling between the lattice and heat reservoirs at

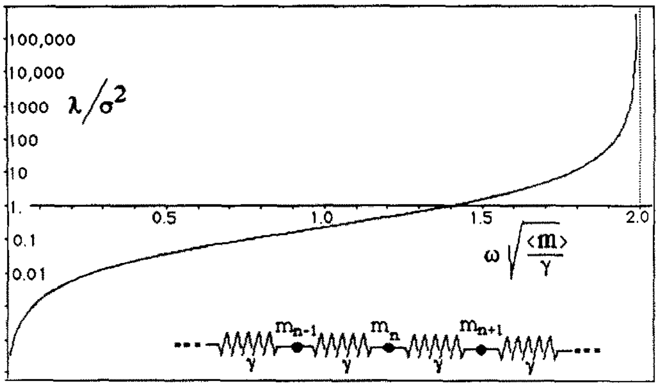

FIG. 3. Plot of the localization factor λ vs frequency of vibration ω for a system of springs-masses (randomly varying mass, $\sigma \ll 1$).

both ends of the one-dimensional lattice. In the limit of long lattice ($N \gg 1/\sigma^2$), equation (30) approximates the Toda [21] limit

$$K_\infty = ck_B f \frac{1}{\sigma}\sqrt{N}. \tag{31}$$

In Fig. 4, we plot expression (30) for the apparent conductivity renormalized by limit (31). For short lattices, K is a linear function of the size N, while the conductivity remains unbounded for all N. In real crystals, the conductivity eventually attains its constant (bulk) value since the lattice waves will also interact with grain interfaces, external boundaries, and more complicated crystal defects. Our simple model also does not account for two- or three-dimensional effects and anharmonic lattice forces.

In order to get a feeling of the length scales involved, let us assume a lattice constant of 1 nm. Then, in the presence of 10% lattice impurity ($\sigma = 0.1$), Fig. 4 shows that the conductivity reaches its asymptotic value after approximately 1000 nodes which is equivalent to a film of 1 μm thickness. Unfortunately, there are no measurements of apparent conductivity for heat conduction normal to thin films reported in the literature; the only available data are for conduction *along* the film. However, the behavior of the

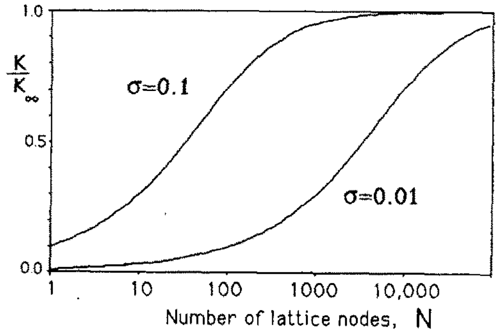

FIG. 4. Variation of predicted thermal conductivity K with lattice length N. K_∞ is the conductivity for $N \to \infty$.

scaled conductivity K/K_∞ as a function of the lattice size N is in qualitative agreement with the predictions by Flik and Tien [24] which were obtained with the classical phonon model.

5. CONCLUDING REMARKS

In the present work, we obtain the solution of three general non-deterministic problems in an elegant manner by simply exploiting rigorous analytical methods available in the literature of applied physics. These methods are applicable when the mathematical model of the system and the underlying statistics concerning its parameters are known a priori. The results we derive describe the behavior of an *ensemble* of systems. In other words, they describe the average behavior of a set of different realizations of the stochastic process. In practice, we are interested in the average solution in a single realization (one experiment). We can consider ensemble and spatial averages to be equivalent if the process is statistically homogeneous (stationary) and the relevant length scales are much larger than the covariance scales.

In the following, we summarize the original contributions of this manuscript. The general thrust of this work is to show the effect of microscopic disorder on macroscopic transport properties. The model of Section 2, which involves convective transport in a 'turbulent' field, shows Fickian behavior as $t \to \infty$. In contrast, the driven dissipative system of Section 3 shows the same as $t \to 0$. Furthermore, the behavior of the second system for long times is very anomalous. This suggests that diffusion can exhibit atypical properties in non-deterministic fields. In Section 4, we have extended the result of Toda [21] for the intrinsic conductivity in disordered harmonic lattices to the whole spectrum of frequencies in the passband. Our predictions for the size effect of the (renormalized) conductivity show qualitative agreement with previously reported [24] theoretical results obtained with phonon models.

The general mathematical problems treated in this work can be mapped onto a variety of other mathematical or physical problems. The methodology we applied to the convection–dispersion equation can also be implemented to solve a host of multiplicative stochastic problems (transport equations with statistically varying parameters) in a straightforward manner. The analysis of both the reaction–diffusion system with random reaction sites (parabolic equation) and the discrete model of lattice vibrations (hyperbolic system) demonstrates the effect of localization in thermal systems. Analogous phenomena can be encountered in computational domains where partial differential equations are approximated with finite differences or finite elements, cf. Vichnevetsky [25]. We finally observe that the method outlined in the Appendix can be applied to any one-dimensional monocoupled structure encountered in solid mechanics problems.

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APPENDIX: LOCALIZATION FACTOR FOR ALMOST PERIODIC STRUCTURES

The transfer matrix of an elastic structure consisting of N finite elements can be written as a product of N transfer matrices: $T_N T_{N-1} \cdots T_n \cdots T_3 T_2 T_1$. If each matrix is a function of a random variable μ, this formulation represents a Markovian chain acting on the input vector. In the case of mono-coupled systems [26], the transfer matrix is 2×2 and it can be transformed in the following Hermitian form with a simple similarity transformation:

$$[S]^{-1}[T_n][S] = \begin{bmatrix} g_{11} & g_{12} \\ g_{21}^* & g_{22}^* \end{bmatrix}. \tag{A1}$$

The elements of the transfer matrix are, in general, functions of the random variable μ. In contrast to the periodic system, vibrations in an almost periodic system without dissipation can be localized: wavetrains initiated at a site in the system are attenuated according to relation (25) which also expresses their envelope. The concept of localization, which is due to Anderson [19], has been first applied to elastic systems by Hodges [27] and was further investigated by Pierre and Dowell [28] for a finite chain of single-degree-of-freedom coupled oscillators. The following definition of the localization factor is almost universally accepted in the engineering community:

$$0 \leqslant \lambda = \lim_{N \to \infty} \frac{1}{N} \ln \| T_N T_{N-1} \cdots T_n \cdots T_3 T_2 T_1 \| \tag{A2}$$

where $\| \cdot \|$ is the matrix norm.

In general, the dependence of the localization factor on the wave number or the degree of disorder is complicated. If the random transfer matrices depend on the same random variable with common probability distribution, analytic expressions can be obtained with perturbation methods. Baluni and Willemsen [22] derived the following closed-form expression for the localization factor

$$\lambda = \tfrac{1}{2}\sigma^2 \frac{\partial^2}{\partial \mu^2} \ln |g_{11}(\mu)| + o(\sigma^2) \tag{A3}$$

which is valid for small variance σ^2 (weak-disorder regime).

SUR LA SOLUTION APPROCHEE DE PROBLEMES NON DETERMINISTES DE CHALEUR ET DE MASSE

Résumé—Les incertitudes inhérentes aux mécanismes de transport dans les milieux hétérogènes réalistes peuvent être décrites par des équations non déterministes avec des coefficients aléatoires. On étudie analytiquement trois classes de phénomènes de chaleur et de masse décrits par des modèles continus conduction–diffusion-réaction et des modèles discrets : (1) dispersion variable dans un champ aléatoire de vitesse de filtration ; (2) diffusion anormale dans des milieux avec des sites aléatoires de réaction ; (3) effet de taille sur la conductivité thermique de trames solides en désordre isotrope. En utilisant une analyse de petite perturbation, on résout trois problèmes intéressants décrits par des équations aux dérivées partielles avec des coefficients aléatoires. Bien que la part aléatoire des paramètres soit plus faible que celle déterministe (faible désordre) son effet sur le comportement des quantités moyennes est à la fois important et contraire à l'intuition.

ÜBER NÄHERUNGSLÖSUNGEN FÜR NICHT-DETERMINISTISCHE PROBLEME DER WÄRME- UND STOFFÜBERTRAGUNG

Zusammenfassung—Die bei den Transportvorgängen in realistischen heterogenen Medien auftretenden Unsicherheiten können durch nicht-deterministische Gleichungen mit Zufallskoeffizienten beschrieben werden. In der vorliegenden Arbeit werden drei Klassen von Phänomenen der Wärme- und Stoffübertragung analytisch untersucht, welche durch kontinuierliche und diskrete Modelle mit Konvektion, Diffusion und Reaktion beschrieben werden: (1) nichtstationäre Dispersion in einem ungerichteten Geschwindigkeitsfeld bei Filtration; (2) unregelmäßige Diffusion in Medien mit zufällig verteilten Reaktionen; (3) Einfluß der Größe auf die Wärmeleitfähigkeit von isotopisch ungeordneten festen Gittern. Unter Verwendung des Störungsverfahrens werden drei nicht-triviale Probleme gelöst, welche durch Differentialgleichungen mit Zufallskoeffizienten beschrieben werden. Obwohl der Anteil an zufälligen Parametern wesentlich kleiner ist als derjenige deterministischer (schwach ungeordneter), ist der Zufallseinfluß auf das Verhalten der gemittelten Größen wichtig, und er widerspricht der Anschauung.

ПРИБЛИЖЕННОЕ РЕШЕНИЕ НЕДЕТЕРМИНИРОВАННЫХ ЗАДАЧ ТЕПЛО- И МАССОПЕРЕНОСА

Аннотация—Неопределенности, присущие процессам переноса в реальных неоднородных средах, могут быть описаны недетерминированными уравнениями со случайными коэффициентами. В данной работе аналитически исследуются три класса явлений тепло- и массопереноса, описываемые моделями конвекции–диффузии—реакции в сплошных средах и дискретными моделями: (1) нестационарное диспергирование в случайном поле скорости фильрации; (2) аномальная диффузия в средах со случайным распределением реакционноспособных участков; (3) влияние размеров образца на коэффициент теплопроводности разупорядоченных изотопами решеток твердых тел. С использованием анализа малых возмущений решаются три нетривиальные задачи, описываемые дифференциальными уравнениями со случайными коэффициентами. Несмотря на то, что вклад случайных параметров намного меньше, чем определяемых (слабая неупорядоченность), влияние случайности на поведение усредненных величин является весьма существенным.