Nonvalidity of the telegrapher's diffusion equation in two and three dimensions for crystalline solids

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We use a classical analog of two-dimensional (2D) and 3D quantum S-matrix scattering theory to study classical mesoscopic diffusion in isotropic, crystalline, solids. The individual collisions include transmission, reflection, and lateral scattering probabilities. The resulting stochastic process is a second-order Markov process in phase space, which is known in the literature as 2D (3D) persistent random walk. In striking contrast with the 1D case, in the continuum limit, the 2D and 3D total densities *do not* satisfy the telegrapher's diffusion equation. We explain this fact deriving the anomalous Maxwell-Cattaneo equation in the case of discrete diffusion processes. We find that inertial memory, giving the forward scattering a preferential direction, breaks the x-y symmetry. [S1063-651X(97)06301-0]

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I. INTRODUCTION

The so-called telegrapher's equation has recently been a subject of many studies concerning transport properties in fluids and solids. Although well known since the last century in electrodynamics [1], the use of the telegrapher's equation in transport theory dates scarcely from 1951, after its derivation by Goldstein [2] using a stochastic process called the one-dimensional persistent random walk (1D-PRW). Its assessment as a transport equation was nicely discussed by Weymann [3] and its applications to a wide variety of problems have been recently reviewed by a number of authors [4–6]. In particular, Masoliver *et al.* [7,8] pointed out that in two dimensions a persistent random walk describes a motion which in the continuum limit does not obey two-dimensional the (2D) telegrapher's equation. Furthermore, they conjectured that it is not possible to derive the telegrapher's equation in 2D (or in 3D) by following Goldstein's procedure starting from a persistent random walk and passing to the continuum limit.

In this paper we explicitly wish to prove this last assertion and furthermore to clearly point out why the Fickian diffusion ceases to hold true in 2D and 3D. In fact, since the probabilities of making a forward and sideways transition are different with respect to a chosen direction, the probabilities of finding the walker at a certain point in the lattice are not symmetrical. The continuum limit of 2D (3D) persistent random walk leads to a diffusive type equation very much like the one derived by Masoliver *et al.* [7,8].

II. THE 1D-PRW MODEL

Since the pioneering work by Goldstein [2] we know that 1D mesoscopic diffusion equations can be obtained using a one-dimensional persistent random walk [4]. Among the different 1D-PRW models present in the literature there is one called quantum random walk (QRW) which is derived directly from the 1D quantum S-matrix scattering theory [9,10]. Scattering with energies below (tunneling) and above the potential barrier are described indistinctly with the transmission and reflection coefficients (T,R).

The QRW is a coherent (having interference) diffusive process, where elastic scatterings of wave packets, of constant (average) energy, against a crystalline lattice are considered. In a recent article [11] it was proved that if a time average is taken on the quantum probabilities described in QRW, the quantum interference contributions can be neglected, and a set of incoherent (classical) 1D-PRW equations are obtained. This quantum-derived incoherent process describes a succession of 1D classical scatterings in a lattice where all particles incident upon any potential barrier are scattered with forward (transmission) and backward (reflection) probabilities (T,R), respectively. Conservation of particles demands that $T+R=1$. Usually $T>R$, and this expresses the inertial memory of particles under scattering.

Assuming the particles to be described only at the mid-valleys between potential barriers, the quantum-derived classical 1D-PRW equations may be rewritten relating the classical incoming probabilities $P_1(x,t)$ and $P_2(x+1,t)$ with the corresponding outgoing ones $P_1(x+1,t+1)$ and $P_2(x,t+1)$. The subscripts in the probabilities denote the direction of motion (1=right, 2=left). The 1D-PRW equations become

$$\begin{pmatrix} P_1(x+1,t+1) \\ P_2(x,t+1) \end{pmatrix} = \begin{pmatrix} T & R \\ R & T \end{pmatrix} \begin{pmatrix} P_1(x,t) \\ P_2(x+1,t) \end{pmatrix}. \qquad (2.1)$$

Equation (2.1) describes a classical process where all particles have the same average speed $c\equiv\Delta x/\Delta t$. Having constant energy, in 1D the velocity has only two values: $\pm c$. The effect of elastic collisions is simply to change their directions of motion. $P_1(x,t)$ and $P_2(x,t)$ describe the joint probability of finding the particle at position x at time t with positive and negative velocities, respectively. Thus the 1D-PRW process (2.1) describes in phase space a Markovian random walk with *internal degrees of freedom*. Each individual probability P_1 and P_2 in Eq. (2.1) is hence a *second-order Markov process* [4].

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In Eq. (2.1), for arbitrary values of R and T, the continuum limit $(\Delta x,\Delta t)\to(0,0)$, such that $\Delta x/\Delta t\equiv c$ is kept constant, *does not exist* [4]. However, the continuum limit does exist in the particular case of the weak-scattering limit (WSL) where, besides $(\Delta x,\Delta t)\to(0,0)$ with $\Delta x/\Delta t\equiv c$, the coefficients (R,T) have to satisfy the supplementary conditions: $R\equiv\Delta t/2\theta\sim 0$, and $T=1-R\sim 1$, where θ is a constant relaxation time characteristic of the solid [4].

The corresponding equations in the 1D-WSL are

$$\frac{\partial P_1}{\partial t}+c\,\frac{\partial P_1}{\partial x}=\frac{1}{2\theta}\,(P_2-P_1), \tag{2.2a}$$

$$\frac{\partial P_2}{\partial t}-c\,\frac{\partial P_2}{\partial x}=\frac{1}{2\theta}\,(P_1-P_2). \tag{2.2b}$$

The two equations (2.2) may be rewritten in terms of two new functions, the density $\rho(x,t)\equiv P_1(x,t)+P_2(x,t)$ and its associated current $J(x,t)/c\equiv P_1(x,t)-P_2(x,t)$; they are found to be

$$\frac{\partial\rho}{\partial t}+\frac{\partial J}{\partial x}=0,\quad J=-D\,\frac{\partial\rho}{\partial x}-\theta\,\frac{\partial J}{\partial t}, \tag{2.3}$$

where $D\equiv c^2\theta$ is the diffusion coefficient. Equation (2.3a) describes the conservation of mass, and Eq. (2.3b) the Maxwell-Cattaneo equation [12]. After eliminating the current J in Eqs.(2.3) we get a closed equation for the 1D density, the telegrapher's equation

$$\frac{1}{c^2}\,\frac{\partial^2\rho}{\partial t^2}+\frac{1}{D}\,\frac{\partial\rho}{\partial t}=\frac{\partial^2\rho}{\partial x^2}. \tag{2.4}$$

Likewise, if one does not choose to assume that the random walker moves at constant speed, still further types of evolution equations are possible [13].

In the next sections we will prove, using the classical analog of the 2D (and 3D) quantum S-matrix scattering theory, that in 2D (and 3D) the resulting equations are (i) a 2D (3D) PRW stochastic process, and (ii) in the WSL, in striking contrast with the 1D case, the diffusion equation for the density $\rho(x,y,t)$ *is not* given by the 2D (3D) version of Eq. (2.4).

That is, at least for our classical S-matrix scattering model of diffusion, the telegrapher's equation analogous to Eq. (2.4) *is not* the correct equation for describing 2D and 3D classical mesoscopic diffusion.

Finally, notice that for arbitrary values of (T,R) and keeping $\Delta x^2/\Delta t=$const (the same limit which takes the usual random walk into a parabolic equation), we may also take in Eq. (2.1) the continuous limit. However in doing so we arrive, the same as in the parabolic equation, at a diffusion process with an infinite propagation speed for diffusion. This is an unphysical result.

III. DIFFUSION IN A 2D SQUARE LATTICE

To begin with, consider a 2D, isotropic, square lattice. Assuming the scattered particles follow the same lattice symmetry, we describe any single 2D classical collision with three parameters (T,R,L) which are the forward, backward, and lateral scattering probabilities, respectively. Conservation of particles demands $T+R+2L=1$.

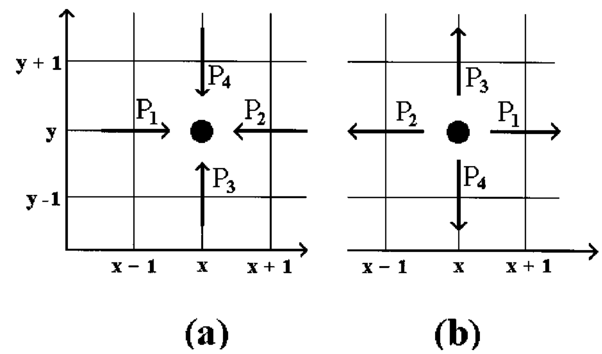

FIG. 1. (a) Input at time (t) and (b) output at time $(t+1)$ for a 2D square lattice.

In a square lattice we have four probability densities: $P_1(x,y,t)$, $P_2(x,y,t)$, $P_3(x,y,t)$, and $P_4(x,y,t)$, where the subscripts denote the directions of motion (1 =right, 2=left, 3=up, 4=down).

In a single 2D classical scattering process, we can find by inspection from Fig. 1 the most general relation between input and output probability densities, namely,

$$\begin{pmatrix} P_1(x+1,y,t+1)\\ P_2(x-1,y,t+1)\\ P_3(x,y+1,t+1)\\ P_4(x,y-1,t+1)\end{pmatrix}=\begin{pmatrix} T & R & L & L\\ R & T & L & L\\ L & L & T & R\\ L & L & R & T\end{pmatrix}\begin{pmatrix} P_1(x-1,y,t)\\ P_2(x+1,y,t)\\ P_3(x,y-1,t)\\ P_4(x,y+1,t)\end{pmatrix}. \tag{3.1}$$

Notice that in the model described by Eq. (3.1), since we have elastic scattering and every scattering process has the same mean free path, made of two displacements each one of the same size $(|\Delta x|=|\Delta y|)$ for input and output respectively, this implies that the mean collision time Δt is the same in every scattering process. From a statistical point of view, Eq. (3.1) describes a stochastic process called 2D-persistent-random-walk (2D-PRW). Notice that this is a *second-order* Markov process since at any time one needs to have two pieces of information, the position and direction of travel. The symmetry of the transition matrix reflects the isotropy of the solid. Eq. (3.1) represents a set of recursive equations for P_i $(i=1,2,3,4)$ whose solution depends strongly on the initial and boundary conditions.

IV. THE 2D WEAK-SCATTERING LIMIT

In order to get the continuum limit of Eq. (3.1) consider, as an example, the first equation of the set, namely

$$P_1(x,y,t+1)=TP_1(x-2,y,t)+RP_2(x,y,t)$$
$$+LP_3(x-1,y-1,t)+LP_4(x-1,y+1,t). \tag{4.1}$$

Next, let us perform in Eq. (4.1) a first-order Taylor series expansion around the point (x,y,t). After some simplifications we have that

$$\frac{\partial P_1}{\partial t}=-\frac{R}{\Delta t}(P_1-P_2)+\frac{L}{\Delta t}(P_3+P_4-2P_1)-T\,\frac{2\Delta x}{\Delta t}\frac{\partial P_1}{\partial x}+L\left(-\frac{\Delta x}{\Delta t}\frac{\partial P_3}{\partial x}-\frac{\Delta y}{\Delta t}\frac{\partial P_3}{\partial y}-\frac{\Delta x}{\Delta t}\frac{\partial P_4}{\partial x}+\frac{\Delta y}{\Delta t}\frac{\partial P_4}{\partial y}\right)+O(\Delta^2/\Delta t). \tag{4.2}$$

As we can clearly see from Eq. (4.2), analogously to the 1D case, for arbitrary values of (T,R,L), the continuum limit ($\Delta x=\Delta y=\Delta t\rightarrow 0$) keeping $\Delta x/\Delta t=\Delta y/\Delta t=$const does not exist. However, we do have a continuum limit in the case of the 2D weak-scattering limit (2D-WSL), where, besides $\Delta x=\Delta y=\Delta t\rightarrow 0$, we define a constant speed c and two constant times θ_R and θ_L such that $(R,L)\rightarrow(0,0)$, in such a way that

$$2\Delta x/\Delta t=2\Delta y/\Delta t\equiv c,\quad R/\Delta t\equiv 1/(2\theta_R),$$
$$L/\Delta t\equiv 1/(2\theta_L),\quad T\rightarrow 1. \tag{4.3}$$

Here, θ_R and θ_L are two characteristic relaxation times of the solid. The times are associated with the backward and lateral scatterings, respectively. In this 2D-WSL, Eq. (4.2), the first of the set describing classical mesoscopic diffusion, becomes

$$\frac{\partial P_1}{\partial t}+c\,\frac{\partial P_1}{\partial x}=-(P_1-P_2)\,\frac{1}{2\theta_R}+(P_3+P_4-2P_1)\,\frac{1}{2\theta_L}. \tag{4.4a}$$

Analogously, the other three continuum equations arising from Eq. (3.1) in the 2D-WSL are given by

$$\frac{\partial P_2}{\partial t}-c\,\frac{\partial P_2}{\partial x}=+(P_1-P_2)\,\frac{1}{2\theta_R}+(P_3+P_4-2P_2)\,\frac{1}{2\theta_L}, \tag{4.4b}$$

$$\frac{\partial P_3}{\partial t}+c\,\frac{\partial P_3}{\partial y}=-(P_3-P_4)\,\frac{1}{2\theta_R}+(P_1+P_2-2P_3)\,\frac{1}{2\theta_L}, \tag{4.4c}$$

$$\frac{\partial P_4}{\partial t}-c\,\frac{\partial P_4}{\partial y}=+(P_3-P_4)\,\frac{1}{2\theta_R}+(P_1+P_2-2P_4)\,\frac{1}{2\theta_L}. \tag{4.4d}$$

Analogously to the 1D case, we will rewrite Eqs. (4.4) in terms of densities $\rho_x\equiv P_1+P_2,\rho_y\equiv P_3+P_4$, and their associated currents $J_x\equiv c(P_1-P_2),J_y\equiv c(P_3-P_4)$. Adding and subtracting Eqs. (4.4a), (4.4b), (4.4c), and (4.4d) we find that

$$\frac{\partial\rho_x}{\partial t}+\frac{\partial J_x}{\partial x}=-(\rho_x-\rho_y)\,\frac{1}{\theta_L}, \tag{4.5a}$$

$$J_x=-c^2\theta_R\,\frac{\partial\rho_x}{\partial x}-\theta_R\,\frac{\partial J_x}{\partial t}, \tag{4.5b}$$

$$\frac{\partial\rho_y}{\partial t}+\frac{\partial J_y}{\partial y}=+(\rho_x-\rho_y)\,\frac{1}{\theta_L}, \tag{4.5c}$$

$$J_y=-c^2\theta_R\,\frac{\partial\rho_y}{\partial y}-\theta_R\,\frac{\partial J_y}{\partial t}. \tag{4.5d}$$

Individually, Eqs. (4.5a) and (4.5c) express the nonconservation of mass moving along each direction. Every direction of motion has lateral scatterings and therefore there is a lateral flux of particles. As we expected in an isotropic solid, the loss of mass in any direction becomes exactly the gain of mass for the perpendicular direction, so the total mass in the process is conserved. Indeed, adding Eqs. (4.5a) and (4.5c) we have the total conservation of mass,

$$\frac{\partial(\rho_x+\rho_y)}{\partial t}+\frac{\partial J_x}{\partial x}+\frac{\partial J_y}{\partial y}=0. \tag{4.6}$$

Finally, Eqs. (4.5b) and (4.5d) for the diffusive currents exhibit the clearest distinction of the 2D-WSL model compared with the 1D-WSL case. Notice that even if we still have the partial time derivative of the current itself (this is the hallmark of mesoscopic diffusion), Fick's law in 2D *is not* satisfied. In Eqs. (4.5b) and (4.5d) we do not have the gradient of the *total* density $(\rho_x+\rho_y)$. Therefore Eqs. (4.5b) and (4.5d) describe an anomalous diffusion process and consequently may be regarded as anomalous Maxwell-Cattaneo equations. This fact has deep consequences in the diffusion equation as we shall see next. Attempting to obtain the telegrapher's equation, we next eliminate in Eqs. (4.5) the two current components J_x and J_y and we arrive at the following equation:

$$\frac{1}{c^2}\,\frac{\partial^2(\rho_x+\rho_y)}{\partial t^2}+\frac{1}{D}\,\frac{\partial(\rho_x+\rho_y)}{\partial t}=\frac{\partial^2\rho_x}{\partial x^2}+\frac{\partial^2\rho_y}{\partial y^2}, \tag{4.7}$$

where the diffusion coefficient $D\equiv c^2\theta_R$ is the same as in the 1D case. This striking 2D-PRW result, trivially extended to 3D-PRW, shows very clearly that mesoscopic diffusion in 2D and 3D *is not* described by the telegrapher's equation. In fact, Eq. (4.7) is not even a closed equation for the total density $(\rho_x+\rho_y)$, and it is by itself useless. In this PRW model, in 2D and 3D, one cannot bypass the evaluation of the diffusive current $\mathbf{J}(\mathbf{r},t)$ in the process of getting the mesoscopic diffusion solution for $\rho(\mathbf{r},t)$. What is very important to realize is that the set of four simultaneous equations (4.5) for (ρ_x,ρ_y,J_x,J_y) become now the fundamental mesoscopic diffusion equations for 2D lattices.

V. THE ANOMALOUS MAXWELL-CATTANEO EQUATION

The above continuum limit has the undesirable property of being only valid in the 2D-WSL, where the scattering coefficients satisfy the properties $(T,R,L)\sim(1,0,0)$. Due to this weak scattering, the solution of Eq. (4.5) describes, for an initial cluster of particles moving in the same direction, a ballistic motion leaving behind a cloud of particles moving backwards and sideways. For short times, the WSL solution resembles more the sublimation of a comet than a diffusion process.

Next, we derive Fick's law without taking the WSL. If we want to know the diffusion coefficient for arbitrary values of (T,R,L), we restrict ourselves to the discrete case. Keeping constant the discrete values of $\Delta x=\Delta y=l/2$, where l is the square lattice constant, and keeping the speed definition $c\equiv 2\Delta x/\Delta t=2\Delta y/\Delta t$, consider in Eq. (3.1) the first equation for P_1; after a Taylor series expansion and keeping only the first-order term we may rewrite it as

$$\frac{\partial P_1}{\partial t} = -(P_1 - P_2)\,\frac{R}{\Delta t} + (P_3 + P_4 - 2P_1)\,\frac{L}{\Delta t} - Tc\,\frac{\partial P_1}{\partial x} - \frac{Lc}{2}\,\frac{\partial}{\partial x}\,(P_3 + P_4) - \frac{Lc}{2}\,\frac{\partial}{\partial y}\,(P_3 - P_4). \tag{5.1}$$

From Eq. (3.1), the second equation for P_2 can be similarly obtained, namely

$$\frac{\partial P_2}{\partial t} = +(P_1 - P_2)\,\frac{R}{\Delta t} + (P_3 + P_4 - 2P_2)\,\frac{L}{\Delta t} + Tc\,\frac{\partial P_2}{\partial x} + \frac{Lc}{2}\,\frac{\partial}{\partial x}\,(P_3 + P_4) - \frac{Lc}{2}\,\frac{\partial}{\partial y}\,(P_3 - P_4). \tag{5.2}$$

Subtracting Eq. (5.2) from Eq. (5.1) and substituting the probabilities (P_1, P_2, P_3, P_4) for the densities and currents $(\rho_x, \rho_y, J_x, J_y)$ we get after some simplifications that

$$J_x = -\frac{c^2 \Delta t}{2R + 2L}\,\frac{\partial}{\partial x}\,[T\rho_x + L\rho_y] - \frac{\Delta t}{2R + 2L}\,\frac{\partial}{\partial t}\,J_x. \tag{5.3a}$$

In a similar way, from the third and fourth equations in Eq. (3.1), we find that

$$J_y = -\frac{c^2 \Delta t}{2R + 2L}\,\frac{\partial}{\partial y}\,[T\rho_y + L\rho_x] - \frac{\Delta t}{2R + 2L}\,\frac{\partial}{\partial t}\,J_y. \tag{5.3b}$$

Equations (5.3), valid for arbitrary values of (T,R,L), show the exact problem with 2D (also 3D) classical mesoscopic diffusion. As long as we have some inertial memory $(T \neq L)$, Fick's equation is not valid. What we have is an anomalous diffusion process with a tensor diffusion coefficient D. We may define two diffusion coefficients: a parallel coefficient $D_\parallel$ and a perpendicular coefficient $D_\perp$, where

$$D_\parallel \equiv \frac{c^2 \Delta t T}{2R + 2L}, \quad D_\perp \equiv \frac{c^2 \Delta t L}{2R + 2L}. \tag{5.4}$$

With this notation we can write the 2D anomalous Maxwell-Cattaneo equation as

$$J_x = -\frac{\partial}{\partial x}\,(D_\parallel \rho_x + D_\perp \rho_y) - \theta_{2\mathrm{D}}\,\frac{\partial}{\partial t}\,J_x, \tag{5.5a}$$

$$J_y = -\frac{\partial}{\partial y}\,(D_\parallel \rho_y + D_\perp \rho_x) - \theta_{2\mathrm{D}}\,\frac{\partial}{\partial t}\,J_y, \tag{5.5b}$$

where

$$\theta_{2\mathrm{D}} \equiv \frac{\Delta t}{2R + 2L} = \frac{D_\parallel}{c^2 T} = \frac{D_\perp}{c^2 L} \tag{5.5c}$$

is the relaxation time associated to a 2D mesoscopic diffusion process. It is clear from Eq. (5.5) that since $T \neq L$ the probabilities of finding the walker at a certain point in the lattice are not symmetrical. This explains why the x-y symmetry of the telegrapher's equation is no longer valid in the 2D continuous case, where in the 2D-WSL we certainly have $(T \sim 1) \neq (L \sim 0)$. Clearly, this result is easily extrapolated to the 3D case.

VI. THE 3D-WSL RESULTS

The above 2D results can be trivially extended to the 3D case. For a simple cubic lattice, the three coefficients (T,R,L) satisfy the condition $T + R + 4L = 1$. In this 3D case we have six probability densities P_i ($i = 1, \ldots, 6$), and the continuous result in the 3D-WSL for the apparent telegrapher's equation is

$$\frac{1}{c^2}\,\frac{\partial^2(\rho_x + \rho_y + \rho_z)}{\partial t^2} + \frac{1}{D}\,\frac{\partial(\rho_x + \rho_y + \rho_z)}{\partial t} = \frac{\partial^2 \rho_x}{\partial x^2} + \frac{\partial^2 \rho_y}{\partial y^2} + \frac{\partial^2 \rho_z}{\partial z^2}, \tag{6.1}$$

where $D \equiv c^2 \theta_R$.

For arbitrary values of (T,R,L) the discrete case gives, for the 3D diffusive current $\mathbf{J}(\mathbf{r},t)$, an anomalous Maxwell-Cattaneo equation where one typical component J_x looks like

$$J_x = -\frac{\partial}{\partial x}\,[D_\parallel \rho_x + D_\perp(\rho_y + \rho_z)] - \theta_{3\mathrm{D}}\,\frac{\partial}{\partial t}\,J_x, \tag{6.2}$$

and the 3D diffusion coefficients D and relaxation time $\theta_{3\mathrm{D}}$ are found to be

$$D_\parallel \equiv \frac{c^2 \Delta t T}{2R + 4L}, \quad D_\perp \equiv \frac{c^2 \Delta t L}{2R + 4L}, \quad \theta_{3\mathrm{D}} \equiv \frac{\Delta t}{2R + 4L}. \tag{6.3}$$

From Eq. (6.2) we see that as long as we can distinguish the forward from the lateral scattering coefficients, the symmetry x-y-z for diffusion will be broken.

As a final conclusion, the inertial memory, which makes valid in 1D the telegrapher's equation, is the same property which prevents it in 2D and 3D.

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