Generalized Landauer equation: Absorption-controlled diffusion processes

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The exact expression of the one-dimensional Boltzmann multiple-scattering coefficients, for the passage of particles through a slab of a given material, is obtained in terms of the single-scattering cross section of the material, including absorption. The remarkable feature of the result is that for multiple scattering in a metal, free from absorption, one recovers the well-known Landauer result for conduction electrons. In the case of particles, such as neutrons, moving through a weak absorbing media, Landuer's formula is modified due to the absorption cross section. For photons, in a strong absorbing media, one recovers the Lambert-Beer equation. In this latter case one may therefore speak of absorption-controlled diffusive processes.

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I. INTRODUCTION

One of the problems that has most attracted the attention of solid state physicists in the past three decades is that of mesoscopic diffusion in nanostructures. This process is due to the multiple scattering of electrons with the lattice forming the material. The field was pioneered by Landauer [1], who in 1957 obtained his already famous formula for the diffusion coefficient for conduction electrons in a one-dimensional (1D) solid, namely,

$$D = cL\frac{T}{2R}, \tag{1}$$

where c is the Fermi velocity for the electrons and L is the length of the solid. The solid, considered as a single complex scattering center, has multiple-scattering transmission and reflection coefficients T and R, respectively.

Although many of the derivations of Eq. (1) are of a quantum-mechanical nature [2], however, the quantum derivation assumes that the potentials are measured some distance away from the scatterers and it assumes that this measurement is incoherent, which implies not taking into account the interference of the incident and the reflected wave. Once the interference is neglected, the result so obtained can, therefore, also be derived with incoherent stochastic processes as it has been shown several times [3,4]. Thus, the ratio T/R appearing in Eq. (1) is not a fully quantum-mechanical result, a fact that has long been recognized by many authors [5–8].

However, not all incoherent processes are, necessarily, in agreement with classical mechanics. What is surprising is the fact that Eq. (1) may also be derived from a strictly *classical* transport equation. Indeed, in the present paper we derive the Landauer result (1) from the 1D version of the Boltzmann transport equation. This derivation implies that the Landauer result is not only an incoherent, but also a *classical* result.

On the other hand, it is well known that the Boltzmann transport equation can also be used to study the diffusion of particles in materials having absorption. For example, in the case of photon migration where absorption is strong, astronomers have long been using the 1D Boltzmann case, under the name of two stream theory, to study radiation transfer in a stellar atmosphere [9,10]. Another example is the case of neutrons diffusing with weak absorption in the moderator of a neutron chain reactor. The Boltzmann equation has been a cornerstone in the design of nuclear reactors [11,12].

For neutrons and photons the presence of an absorbing cross section in the diffusion process is rather relevant. And in fact, for absorption dominated diffusion (photons), one would expect that the Lambert-Beer law, which describes the absorption C of light passing through a slab of material of size x, given by $C(x) = \exp[-\alpha x]$ and regarded as one of the basic laws in photochemistry [13,14], holds true.

In the language of transmission and reflection coefficients T and R, we know that for conduction electrons, where no absorption is present, $T+R=1$ due to particle conservation. However, in the passage of particles through an absorbing medium, $T+R<1$ and, in fact, we could define the absorption coefficient C at the end of the sample as $C(L) \equiv T$.

Examination of these facts leads immediately to one question. Since the processes (i) electrons scattering in a metal, according to the Landauer theory, and (ii) particle diffusion in an absorptive medium (photons and neutrons, for example), can both be regarded as classical diffusive scattering processes against fixed targets, namely a Lorentz gas [15], the question is, can they be unified within a single theoretical framework?

The purpose of the present paper is to offer an *affirmative* reply to this question. In fact, we show that both results, the Landauer and Lambert-Beer, are just the opposite limits of

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the same general 1D equation, which stems from the exact solution of the Boltzmann transport equation with absorption. This general result leads to a formula containing a relationship between the diffusion coefficient D, the multiple-scattering coefficients T and R, and the absorption cross section Σ_a; we call this general result the "generalized Landauer equation." We show that in the absorption-free diffusion limit it will reduce to the Landauer result Eq. (1), whereas in an absorption-dominated process, passage of photons through matter yields the Lambert-Beer equation. For neutrons with weak absorption, for example, the generalized Landauer equation gives an explicit—absorption-dependent—correction to Eq. (1).

The new feature of this calculation is that it gives a unified picture of what one might think are unrelated results, and we can visualize them now as particular cases of an absorption-controlled diffusion process.

II. BOLTZMANN'S TRANSPORT EQUATION

Let us consider Boltzmann's transport equation free of external forces [11],

$$\left[\frac{1}{c}\frac{\partial}{\partial t}+\mathbf{\Omega}\cdot\mathrm{grad}_{\mathbf{r}}+\Sigma_s+\Sigma_a\right]f(\mathbf{r},\mathbf{\Omega},t)=\int d\mathbf{\Omega}'\,f(\mathbf{r},\mathbf{\Omega}',t)\,\Sigma_s(\mathbf{\Omega}'\to\mathbf{\Omega}). \quad (2)$$

Here f is the distribution of independent, *monoenergetic* particles moving in a homogeneous and isotropic medium. Elastic collisions are only against fixed targets. The constant speed is denoted by c, and $\mathbf{\Omega}$ is a unit vector in the direction of motion of the particles. $\Sigma_s(\mathbf{\Omega}'\to\mathbf{\Omega})$ denotes the macroscopic scattering cross section defined as the microscopic, or atomic, differential cross section multiplied by the density of target atoms. For isotropic scattering, $\Sigma_s(\mathbf{\Omega}'\to\mathbf{\Omega})$ depends only on the deflection angle θ_0 between the $\mathbf{\Omega}'$ and $\mathbf{\Omega}$ directions, that is, $\cos\theta_0\equiv\mathbf{\Omega}\cdot\mathbf{\Omega}'$. The quantities Σ_s and Σ_a denote the total macroscopic scattering and absorption cross sections, respectively,

$$\Sigma_s\equiv\int d\mathbf{\Omega}\,\Sigma_s(\mathbf{\Omega}'\to\mathbf{\Omega}).$$

In order to derive a generalized Landauer equation, we need the one-dimensional (1D) version of the Boltzmann transport equation. A strict 1D transport theory has only two directions of motion: "right" and "left." This means that the 1D velocity direction $\mathbf{\Omega}$ has only two components $+1$ or -1, written $(+,-)$ for short. Then, the scattering cross section has only two possibilities: forward or backward scattering $\theta_0\equiv(0 \text{ or } \pi)$. We define

$$t\equiv\Sigma_s(+\to+)/\Sigma_s=\Sigma_s(-\to-)/\Sigma_s, \quad (3)$$

$$r\equiv\Sigma_s(+\to-)/\Sigma_s=\Sigma_s(-\to+)/\Sigma_s, \quad (4)$$

as the microscopic forward and backward ("transmission" and "reflection") single-scattering probabilities, respectively. These probabilities are properly normalized $t+r=1$. In this 1D theory, the right- and left-moving densities of particles are denoted by $P_1(x,t)\equiv f(x,+,t)$ and $P_2(x,t)\equiv f(x,-,t)$, respectively. Along the x axis, the 1D Boltzmann equation then becomes a system of two equations written as

$$\left[\frac{1}{c}\frac{\partial}{\partial t}+\frac{\partial}{\partial x}\right]P_1=\Sigma_s r(P_2-P_1)-\Sigma_a P_1, \quad (5)$$

$$\left[\frac{1}{c}\frac{\partial}{\partial t}-\frac{\partial}{\partial x}\right]P_2=\Sigma_s r(P_1-P_2)-\Sigma_a P_2. \quad (6)$$

Both Eqs. (5) and (6) are called the two stream theory, and are used by astrophysicists in connection with radiation transfer [9,10].

The set of equations (5) and (6) may be rewritten in terms of two new functions: the total density $n(x,t)\equiv P_1(x,t)+P_2(x,t)$ and its associated current $J(x,t)/c\equiv P_1(x,t)-P_2(x,t)$. The new set of equivalent equations is

$$\frac{\partial n}{\partial t}+\frac{\partial J}{\partial x}=-c\Sigma_a n, \quad (7)$$

$$J=-\frac{c}{\Sigma_a+2\Sigma_s r}\left[\frac{\partial n}{\partial x}+\frac{1}{c^2}\frac{\partial J}{\partial t}\right]. \quad (8)$$

Equation (7) is simply the 1D continuity equation; it expresses, due to absorption, the nonconservation of mass. Equation (8) is the 1D Maxwell-Cattaneo equation [16], which is clearly a departure from Fick's law and has important physical consequences. The $\partial J/\partial t$ term is the distinctive property of the mesoscopic diffusion regime; for short times it describes the correct ballistic diffusion regime with constant velocity, and for long times it leads to the hydrodynamic diffusion regime. Equations (7) and (8), which are *exact* in 1D, are only the "P_1 approximation" of the spherical harmonic expansion method in 3D [11,12].

Equation (8) shows that the 1D Boltzmann diffusion coefficient is given by the well known result

$$D=\frac{c}{\Sigma_a+2\Sigma_s r}. \quad (9)$$

We call this diffusion coefficient a *local* result, because it involves microscopic single-scattering cross sections $(\Sigma_a,\Sigma_s r)$. The Landauer equation (1), on the other hand, is a *nonlocal* result since it involves coefficients (T,R) for multiple scattering in the bulk of the solid. Therefore, to get the generalized Landauer equation we need to express the single-scattering cross sections $(\Sigma_a,\Sigma_s r)$ as a function of the multiple-scattering coefficients (T,R).

III. BOLTZMANN'S MULTIPLE-SCATTERING COEFFICIENTS WITH ABSORPTION

Let us consider a slab of scattering material of size L. To find the multiple-scattering transmission and reflection coefficients (T,R) of the whole slab, we proceed just as is done in quantum mechanics; we assume a steady-state regime, with a unitary incoming flux of particles incident *only* at the left end. Therefore, we must solve the steady-state case of the 1D Boltzmann equations (5) and (6),

$$\frac{\partial P_1}{\partial x} = -(\Sigma_s r + \Sigma_a) P_1 + \Sigma_s r P_2, \tag{10}$$

$$\frac{\partial P_2}{\partial x} = -\Sigma_s r P_1 + (\Sigma_s r + \Sigma_a) P_2, \tag{11}$$

subject to the boundary conditions: $P_1(0)=1$, $P_2(L)=0$. If we solve this homogeneous linear system, for $P_1(x)$ and $P_2(x)$, the outgoing fluxes at the right and left end are, by definition, the multiple-scattering transmission and reflection coefficients: $T \equiv P_1(L)$, $R \equiv P_2(0)$, respectively.

Trivially, the solutions for $P_1(x)$ and $P_2(x)$, satisfying the boundary conditions, are given in terms of the eigenvalue of the coefficient matrix $\lambda \equiv \sqrt{\Sigma_a^2 + 2\Sigma_s r \Sigma_a}$. The solutions for $P_1(x)$ and $P_2(x)$ are thus written as

$$P_1(x) = \frac{\lambda \cosh[\lambda(L-x)] + (\Sigma_s r + \Sigma_a)\sinh[\lambda(L-x)]}{\lambda \cosh(\lambda L) + (\Sigma_s r + \Sigma_a)\sinh(\lambda L)}, \tag{12}$$

$$P_2(x) = \frac{\Sigma_s r \sinh[\lambda(L-x)]}{\lambda \cosh(\lambda L) + (\Sigma_s r + \Sigma_a)\sinh(\lambda L)}. \tag{13}$$

With these solutions, Boltzmann's multiple-scattering coefficients defined as $T \equiv P_1(L)$, $R \equiv P_2(0)$ are given by

$$T(\Sigma_a) = \frac{\lambda}{\lambda \cosh(\lambda L) + (\Sigma_s r + \Sigma_a)\sinh(\lambda L)}, \tag{14}$$

$$R(\Sigma_a) = \frac{\Sigma_s r \sinh(\lambda L)}{\lambda \cosh(\lambda L) + (\Sigma_s r + \Sigma_a)\sinh(\lambda L)}. \tag{15}$$

For the particular case of $\Sigma_a = 0$, Eqs. (14) and (15) become

$$T(0) = \frac{1}{1 + \Sigma_s r L}, \quad R(0) = \frac{\Sigma_s r L}{1 + \Sigma_s r L}, \tag{16}$$

where, as expected, the mass conservation is well satisfied: $T(0) + R(0) = 1$. On the other hand, having any amount of absorption $(\Sigma_a > 0)$, obviously in Eqs. (14) and (15) the mass conservation is not satisfied and we have $T(\Sigma_a) + R(\Sigma_a) < 1$.

IV. GENERALIZED LANDAUER EQUATION

We now proceed to obtain, from the results of the preceding sections, what we may call the *generalized Landauer equation:* a relation between (D, Σ_a, T, R). For this purpose we now take the ratio of both coefficients in Eqs. (14) and (15) to get

$$\frac{R(\Sigma_a)}{T(\Sigma_a)} = \frac{\Sigma_s r \sinh[L\sqrt{\Sigma_a^2 + 2\Sigma_s r \Sigma_a}]}{\sqrt{\Sigma_a^2 + 2\Sigma_s r \Sigma_a}}. \tag{17}$$

Notice that from the individual expression for both scattering coefficients (14) and (15), Eq. (17) is the simplest expression we can get in terms of $(\Sigma_a, \Sigma_s r)$. Substituting now the (positive definite) factor $2\Sigma_s r = c/D - \Sigma_a \geqslant 0$ obtained from the Boltzmann local diffusion coefficient (9) into Eq. (17), we finally get

$$\frac{2R(\Sigma_a)}{T(\Sigma_a)} = \left(\frac{c}{D} - \Sigma_a\right) \frac{\sinh\left[L\sqrt{\frac{\Sigma_a c}{D}}\right]}{\sqrt{\frac{\Sigma_a c}{D}}}. \tag{18}$$

This is one form of the generalized Landauer result we have been looking for. Notice that in Eq. (18) we have a transcendental equation for D so we cannot get, explicitly, D as a function of $(T/R, \Sigma_a)$. However, the important point here is that the inverse problem, the value of R/T as an explicit function of (D, Σ_a) given by Eq. (18), is *exact* in 1D. We can now get from Eq. (18) some limiting cases.

(i) For conduction electrons in a metal, $\Sigma_a = 0$. In this case Eq. (18) reduces to the Landauer result:

$$D = Lc\frac{T(0)}{2R(0)}, \quad T(0) + R(0) = 1. \tag{19}$$

(ii) For neutrons, in the moderator of a nuclear reactor with weak absorption, $L^2\Sigma_a c/D \ll 1$. We get now, from Eq. (18), a small correction, by absorption, to the Landauer result:

$$D \approx Lc\frac{T(\Sigma_a)}{2R(\Sigma_a)} \frac{1}{1 + L\Sigma_a \frac{T(\Sigma_a)}{2R(\Sigma_a)}}, \quad T(\Sigma_a) + R(\Sigma_a) < 1. \tag{20}$$

(iii) For photons, in a turbid medium with strong absorption, $L^2\Sigma_a c/D \gg 1$. In this limit we get, again from Eq. (18), an implicit equation for D:

$$\frac{T(\Sigma_a)}{2R(\Sigma_a)} \approx 2\left[\sqrt{\frac{c}{D\Sigma_a}} - \sqrt{\frac{D\Sigma_a}{c}}\right]^{-1} \exp\left(-L\sqrt{\frac{\Sigma_a c}{D}}\right). \tag{21}$$

As we can see, in the general form for the ratio R/T given by Eq. (18), even though we have the simplest expression in terms of (D, Σ_a), we still have a transcendental equation for the diffusion coefficient D. Therefore, if at the end we still have to resort to numerical calculations to obtain D as a function of $(T/R, \Sigma_a)$, then it is clear that Eq. (18) is not a wise choice for the generalized Landauer result. This is because in Eq. (18) we need the experimental knowledge of both coefficients (T, R). Therefore, it is better to have a single-scattering coefficient, let us say T as a function of (D, Σ_a), and have another form of the generalized Landauer result. Indeed, the transmission coefficient T can be rewritten as

$$T(\Sigma_a, D) = \left\{ \cosh\left[L\sqrt{\frac{\Sigma_a c}{D}}\right] + \frac{1}{2}\left(\Sigma_a + \frac{c}{D}\right) \frac{\sinh\left[L\sqrt{\frac{\Sigma_a c}{D}}\right]}{\sqrt{\frac{\Sigma_a c}{D}}} \right\}^{-1}. \tag{22}$$

From this equation we get the following two limiting cases.

(i) Zero absorption, which yields again the Landauer result

$$D(T,\Sigma_a=0)=\frac{cL}{2}\frac{T(0)}{1-T(0)}. \tag{23}$$

(ii) Strong absorption, where we have now the Lambert-Beer equation for the absorption coefficient,

$$C(L)\equiv T(\Sigma_a,D)\approx \exp(-\Sigma_a L). \tag{24}$$

These equations give the surprising result that two apparently uncorrelated results, Landauer and Lambert-Beer, are in fact just opposite limits of the same equation: the generalized Landauer equation (22). Now we have a single theoretical frame for both results.

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