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Corrections to the Boltzmann mean free path in disordered systems with finite size scatterers

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Abstract

The mean free path is an essential characteristic length in disordered systems. In microscopic calculations, it is usually approximated by the classical value of the elastic mean free path. It corresponds to the Boltzmann mean free path when only isotropic scattering is considered, but it is different for anisotropic scattering. In this paper, we work out the corrections to the so-called Boltzmann mean free path due to multiple scattering effects on finite size scatterers, in the s -wave approximation, i.e. when the elastic mean free path is equivalent to the Boltzmann mean free path. The main result is the expression of the mean free path expanded in powers of the perturbative parameter given by the scatterer density.

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1. Introduction

The mean free path l is an important parameter which allows characterization of the different regimes in disordered systems [1, 2]. When the disorder is strong, there appears a localized phase for $Kl \ll 1$, where K is the wave number, whereas the diffusive phase is characterized by $Kl \gg 1$. The strength of disorder is thus measured by the parameter l/Kl .

In most theoretical approaches, the randomly distributed impurities are often approximated by point-like potentials [3–5]. Although this type of potential greatly simplifies the analytical calculations, it has been found that this approximation can lead to incorrect results, in particular as far as causality violation is concerned [6]. The scattering theory with finite size potentials does not encounter any problem with causality [7]. Therefore, it is important to use finite size potentials in order to describe the multiple scattering effects. Effects due to the finiteness of the potential size are generally neglected even when the description is in terms of scattering t -matrices, which describe finite size potentials [8, chapters 4, 7], [9]. As we show, contributions coming from the momentum dependence of the t -matrix elements and

from spatial correlations can be neglected. Nevertheless, one has to take care of the way the corrections due to multiple scattering are calculated. It is shown in this paper that the range of the potential plays an important role in the calculation of corrections to the Boltzmann mean free path in the diffusive regime, where the scatterer density is low. Finally, the mean free path is expanded in powers of the scatterer density up to the first subleading order in two and three space dimensions.

2. Formalism

We consider the propagation of an electron in a disordered system, which is described by the following Hamiltonian,

$$H = -\frac{\hbar^2}{2m}\Delta + V \quad (1)$$

where the potential $V = \sum_{i=1}^N v_i$ is the sum of the N randomly distributed individual potentials. The simplest finite range interaction, which one can imagine, is the hard sphere potential defined by

$$v_i(\mathbf{r}) = v(|\mathbf{r} - \mathbf{R}_i|) = \begin{cases} \infty & \text{for } |\mathbf{r} - \mathbf{R}_i| \leq a \\ 0 & \text{for } |\mathbf{r} - \mathbf{R}_i| > a. \end{cases} \quad (2)$$

Like the point-like scatterer given by a delta function, this type of scatterer has no internal structure (the wavefunction vanishes inside the potential), but it has a finite size given by the range of the potential a . The electron propagator can be cast in the form

$$G = G_0 + G_0 \left(\sum_{i=1}^N v_i \right) G = G_0 + G_0 \mathcal{T} G_0 \quad (3)$$

where G_0 is the free propagator and \mathcal{T} is the global scattering matrix of the system. This \mathcal{T} -matrix is given in terms of the individual scattering \mathbf{t} -matrices by the Watson series [10]

$$\mathcal{T} = \sum_{i=1}^N \mathbf{t}_i + \sum_{i,j \neq i} \mathbf{t}_i G_0 \mathbf{t}_j + \sum_{\substack{i,j \neq i, \\ k \neq j}} \mathbf{t}_i G_0 \mathbf{t}_j G_0 \mathbf{t}_k + \dots \quad (4)$$

Each term of this operator series represents a sequence of multiple scattering. The insertion of the closure relation $\int \frac{d\mathbf{k}}{(2\pi)^3} |\mathbf{k}\rangle \langle \mathbf{k}| = 1$ between the operators introduces off-shell \mathbf{t} -matrix elements. The hard sphere off-shell \mathbf{t} -matrix elements, $\langle \mathbf{k}' | \mathbf{t}_K | \mathbf{k} \rangle$ with $k \neq k' \neq \sqrt{2mE}$, can be derived from the off-shell barrier potential of finite height [11] by the extrapolation of its height to infinity. Their expression is given in appendix A. The analytic expression of each term of the electron propagator in Fourier space, $G(E, \mathbf{k})$, given by (3), is obtained by inserting the momentum closure relation in the scattering series (4). The momentum integrations are then performed with the use of expressions (A.1) and (A.2) for the \mathbf{t} -matrix elements in two and three dimensions and the integrals (B.1) and (B.2) of appendix B. Throughout the calculation, one has to take care that the scatterers do not overlap. This gives rise to correlations between scatterers which have to be accounted for when taking the ensemble average of the propagator.

In contrast to the point-like scatterers case where there is no spatial correlation, the ensemble average, noted with a bar, of a quantity Q reads as

$$\bar{Q} = \frac{1}{Z} \int \prod_{i=1}^N d\mathbf{R}_i \prod_{j>i} \Theta(R_{ij} - 2a) Q(\{\mathbf{R}_i\}_{i=1,\dots,n}) \quad (5)$$

where

$$R_{ij} \equiv |\mathbf{R}_{ij}| \equiv |\mathbf{R}_i - \mathbf{R}_j|$$

and

$$\mathcal{Z} = \int \prod_{i=1}^N d\mathbf{R}_i \prod_{j>i} \Theta(R_{ij} - 2a)$$

is a normalization constant, the same as the spatial term of the partition function of a 3D hard sphere or 2D hard disc gas. The Heaviside step function Θ accounts for the spatial correlations.

Using results from the virial expansion, especially the Kirkwood superposition approximation [12, chapter 17], the spatial correlation function can be written as a product of two-body correlation functions g

$$\frac{1}{\mathcal{Z}} \prod_{j>i} \Theta(R_{ij} - 2a) = \frac{1}{V^n} \prod_{j>i} g(R_{ij}). \quad (6)$$

Then, the average of Q can be approximated by

$$\bar{Q} = \frac{1}{V^n} \int \prod_{i=1}^n d\mathbf{R}_i \prod_{j>i} g(R_{ij}) Q(\{\mathbf{R}_i\}_{i=1..n}) \quad (7)$$

where the two-body correlation function g is expanded in powers of the scatterer density ρ

$$g(R_{ij}) = g_0(R_{ij}) + \rho g_1(R_{ij}) + \rho^2 g_2(R_{ij}) + \dots \quad (8)$$

$g_0, g_1, g_2 \dots$ being derived from the virial expansion [12]. In the case of hard spheres, $g_0(R_{ij}) = \Theta(R_{ij} - 2a)$ describes the fact that two hard spheres at positions \mathbf{R}_i and \mathbf{R}_j cannot overlap and the other terms $g_1, g_2 \dots$ take into account the average effect coming from the presence of other scatterers in the system.

In the case of point-like scatterers $g_0(R_{ij}) = 1$ and $g_{i \geq 1}(R_{ij}) = 0$. Note that using this approximation, i.e. neglecting the spatial correlations, for the calculation of $\langle \mathbf{k}' | \mathbf{t}_i \mathbf{G}_0^{(+)}(E) \mathbf{t}_j \mathbf{G}_0^{(+)}(E) \mathbf{t}_i | \mathbf{k} \rangle$ with the expressions (A.2) and (A.1) leads to a divergence instead of the correct result [13, chapter 2]. This divergence is due to the convergence conditions of integrals (B.1) and (B.2). It shows that the use of off-shell \mathbf{t} -matrix elements is restricted to non-overlapping scatterers. This can be understood by observing that the existence of off-shell \mathbf{t} -matrix elements is related to the finiteness of the size of the scatterer. Therefore it makes no sense to use these off-shell \mathbf{t} -matrix elements in a point-like scatterer approximation.

In order to calculate the average of the propagator analytically, one needs an additional approximation, which neglects the correlations between non-successive scatterers which appear in the scattering series (4). Then

$$\bar{Q} = \frac{1}{V^n} \int d\mathbf{R}_n \int \prod_{i=1}^{n-1} d\mathbf{R}_{i,i+1} g(R_{i,i+1}) Q(\{\mathbf{R}_i\}_{i=1..n}). \quad (9)$$

With the help of this approximation, each sequence of the Watson series (4) containing only distinct scatterers can be calculated. The summation of this class of terms is usually referred to as *independent scatterer approximation* [14]. It gives the first-order contribution in the density of the self-energy. The approximation (9) is justified as long as the self-energy Σ of the average propagator

$$\bar{G}(E, \mathbf{k}) = G_0(E, \mathbf{k}) + G_0(E, \mathbf{k}) \Sigma(E, \mathbf{k}) \bar{G}(E, \mathbf{k})$$

is expanded up to order ρ^2 ,

$$\Sigma(E, \mathbf{k}) = \underbrace{\Sigma^{(1)}(E, \mathbf{k})}_{\mathcal{O}(\rho)} + \underbrace{\Sigma^{(2)}(E, \mathbf{k})}_{\mathcal{O}(\rho^2)} + \dots$$

Indeed, corrections coming from the overlapping of two non-successive scatterers are at least of the order ρ^3 , because they correspond to scattering sequences containing at least three scatterers.

In the s -wave approximation, where $Ka \ll 1$, the first-order term of the self-energy is simply given by the off-shell \mathbf{t} -matrix element

$$\Sigma^{(1)}(E, \mathbf{k}) \approx \rho(\mathbf{k}|\mathbf{t}(E)|\mathbf{k}). \tag{10}$$

Note that this expression is the one obtained in the usual independent scatterer approximation. Its imaginary part is related to the individual scattering cross section via the optical theorem and leads to the Boltzmann mean free path l_0 [8],

$$l_0 = -\frac{K}{\text{Im}\Sigma^{(1)}} \approx \begin{cases} \frac{K \ln^2 Ka}{\rho\pi^2} & \text{in 2D} \\ \frac{1}{4\pi\rho a^2} & \text{in 3D} \end{cases} \tag{11}$$

where the self-energy is taken on the energy shell, $\Sigma^{(1)} \equiv \Sigma^{(1)}(E, \mathbf{k})$ with $|\mathbf{k}| = K = \frac{\sqrt{2mE}}{\hbar}$.

The second term of the self-energy contains the contribution coming from the re-summation of the infinite series of all multiple scattering terms involving two distinct scatterers and another term coming from the excluded volume. It appears that this latter term is negligible in comparison with the other terms of order ρ^2 , because of order a^d , where d is the spatial dimension. The two-body scattering term is calculated in appendix C and reads as

$$\Sigma^{(2)} \approx \frac{\hbar^2}{2m} \left(\frac{\pi\rho}{K}\right)^2 \left\{ \frac{2}{(\ln K'a)^3} + \frac{3\pi i}{(\ln K'a)^4} \right\} \tag{12}$$

where $K' = Ke^\gamma/2$ and $\gamma \simeq 0.5772$ is the Euler constant in 2D, and

$$\Sigma^{(2)} \approx \frac{\hbar^2}{2m} \frac{(4\pi)^2 a^3}{K} \rho^2 \left\{ \frac{i}{2} + Ka \left(\ln Ka + 3 \ln 2 + \ln 3 + \gamma - 3 - \frac{i\pi}{2} \right) \right\} \tag{13}$$

in 3D.

3. Corrections to the Boltzmann mean free path

The mean free path l is defined as the decreasing rate of the average propagator

$$\bar{G}(E, \mathbf{r} - \mathbf{r}') \propto e^{-\frac{|\mathbf{r}-\mathbf{r}'|}{2l}}.$$

In order to compare with the case of point-like scatterers [4], we consider the deviation of the mean free path with respect to the Boltzmann mean free path l_0

$$\frac{l}{l_0} = 1 - \left(\frac{\text{Re}\Sigma^{(1)}}{2K^2} - \frac{\text{Re}\Sigma^{(1)'}}{K} + \frac{\text{Im}\Sigma^{(2)}}{\text{Im}\Sigma^{(1)}} \right) + \mathcal{O}(\rho^2) \tag{14}$$

where $\Sigma^{(1)'} \equiv \frac{d}{dk} \Sigma^{(1)}(E, \mathbf{k})|_{k=K}$. This formula is easily derived from the definition of the mean free path,

$$l = \frac{1}{2\text{Im}\tilde{K}} \quad \text{where} \quad \tilde{K}^2 = K^2 - (\Sigma^{(1)} + \Sigma^{(2)}). \tag{15}$$

For point-like scatterers, the term $\Sigma^{(1)'}$ vanishes; there is no off-shell contribution.

In two dimensions, for low density ρ and low energy $Ka \ll 1$, one obtains

$$\frac{l}{l_0} \approx 1 + \frac{\pi\rho}{K^2} \left(\frac{1}{\ln K'a} + \frac{3}{(\ln K'a)^2} \right) \tag{16}$$

where $K' = Ke^{\nu/2}$. The excluded volume term appears to be of the same order in Ka as the off-shell term $\Sigma^{(1)'$, i.e. $\mathcal{O}((Ka)^2)$. These two terms are also of the same order in Ka as the first partial wave term, which is the term for $m = \pm 1$ in expression (A.1). They are therefore neglected here.

The leading correction term in (16) is given by the on-shell \mathbf{t} -matrix $\Sigma^{(1)}$ and the subleading correction term comes from the first term of the two-body series. These corrections lead to a decrease of the mean free path with respect to the Boltzmann value. Neglecting the subleading corrections in $1/\ln^2 Ka$, expression (16) can be written in terms of $1/Kl_0$ and reads

$$\frac{l}{l_0} \approx 1 + \frac{\ln Ka}{\pi} \frac{1}{Kl_0}. \tag{17}$$

This expression shows that the corrections are not only written in terms of $1/Kl_0$, but that there appears a logarithmic correction involving the size of the scatterer.

In three dimensions, the excluded volume and the off-shell corrections are also negligible for the same reasons. In contrast to the two-dimensional case, the leading term is not given by the on-shell \mathbf{t} -matrix $\Sigma^{(1)}$. This latter is compensated by the first term of the two-body series $\Sigma^{(2)}$, given by (C.1). It emerges that the leading term of the corrections to the Boltzmann mean free path is given by the second term (C.2) of the two-body series. As in two dimensions, the mean free path is smaller than the Boltzmann expression

$$\frac{l}{l_0} \approx 1 - \rho \left(\frac{\pi^2 a^2}{K} + \mathcal{O}(a^3) \right). \tag{18}$$

The expression (14) for the deviation of the mean free path already exists for point-like scatterers in three dimensions [4]. Instead of being of the form $\rho a^2/K$ as obtained in (18), the corrections to the Boltzmann mean free path for scalar scatterers are proportional to ρ/K^3 . This shows that the behaviour of the scalar point-like scatterers is very different from that of finite size scatterers. It is much more sensitive to small wave numbers. For finite size potentials, the effect of two-body scattering is much less important than it is in the case of point-like potentials. Written in terms of the disorder strength $1/Kl_0$, expression (18) reads as

$$\frac{l}{l_0} \approx 1 - \frac{\pi}{4} \frac{1}{Kl_0}. \tag{19}$$

For the scalar point scatterers at resonance, a similar expression is found except that the numerical factor is not $\pi/4$ but 0.375 (see equation (3.14a) from [4]). The effect of the disorder is more important for finite size scatterers than for point-like scatterers.

4. Conclusion

In this paper we have worked out the corrections to the Boltzmann mean free path of infinite disordered systems composed of hard disc scatterers in two dimensions and hard sphere scatterers in three dimensions. The calculations with finite size scatterers present several differences compared with the calculations with point-like scatterers. One is led to use off-shell \mathbf{t} -matrix elements. The excluded volume has to be taken into account, yielding spatial correlations when averaging over disorder. These two effects appear to be of the same order in Ka and are negligible with respect to a more important effect due to multiple scattering. They become important when one is interested in non-isotropic scattering, i.e. when more than one partial wave has to be taken into account.

To the knowledge of the author, the result obtained in two dimensions is new and has not been evaluated even for point-like potentials. The corrections to the mean free path cannot be given only in terms of $1/Kl_0$; indeed, they contain a logarithmic prefactor depending on the size of the scatterer. In three dimensions, the comparison of our result with the result obtained for point-like potentials shows a discrepancy in the numerical factor in front of the disorder strength $1/Kl_0$. The decrease of the mean free path is more important for finite size scatterers than for point-like potentials.

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Appendix A. Off-shell t-matrix

Following [11], the off-shell scattering t-matrix of a quantum particle with energy $E = \frac{\hbar^2 K^2}{2m}$ takes the form

$$\begin{aligned} \langle \mathbf{k}' | \mathbf{t}(E) | \mathbf{k} \rangle &= \frac{\hbar^2}{2mV} 2\pi a \sum_{m=-\infty}^{+\infty} e^{im(\theta_{k'} - \theta_k)} \\ &\times \left\{ \frac{k^2 - K^2}{k'^2 - k^2} (kJ_m(k'a)J_{m-1}(ka) - k'J_{m-1}(k'a)J_m(ka)) \right. \\ &\left. + kJ_m(k'a)J_{m-1}(ka) - KJ_m(ka)J_m(k'a) \frac{H_{m-1}^{(1)}(Ka)}{H_m^{(1)}(Ka)} \right\} \end{aligned} \quad (\text{A.1})$$

in two dimensions and

$$\begin{aligned} \langle \mathbf{k}' | \mathbf{t}(E) | \mathbf{k} \rangle &= \frac{2\pi\hbar^2}{mV} a^2 \sum_{l=0}^{+\infty} (2l+1) P_l(\cos(\widehat{\mathbf{k}'}, \widehat{\mathbf{k}})) \\ &\times \left\{ \frac{k^2 - K^2}{k'^2 - k^2} [kj_l(k'a)j_{l-1}(ka) - k'j_{l-1}(k'a)j_l(ka)] \right. \\ &\left. + kj_l(k'a)j_{l-1}(ka) - Kj_l(k'a)j_l(ka) \frac{h_{l-1}^{(+)}(Ka)}{h_l^{(+)}(Ka)} \right\} \end{aligned} \quad (\text{A.2})$$

in three dimensions. The j_l and $h_l^{(+)}$ are the spherical Bessel and Hankel functions and J_m and $H_m^{(1)}$ are the ordinary Bessel and Hankel functions [15]. In three dimensions, the angular part is taken into account by the Legendre polynomial P_l .

These expressions enter the calculation of each term of the scattering series (4) in the expression of the electron propagator (3).

Appendix B. Useful integrals

In order to derive the expressions of each term of the scattering series (4) in Fourier space, one has to compute the products of the propagators G_0 with t-matrices. The following integrals are the basic integrals appearing in these products.

The integral used in the calculation of each scattering sequence of the Watson series (4) in two dimensions reads as

$$\int_0^{+\infty} \frac{k^{d-1} dk}{K^2 - k^2 + i\epsilon} \prod_{i=1}^n J_{\mu_i}(ka_i) J_m(kR) = -i\frac{\pi}{2} K^{d-2} \prod_{i=1}^n J_{\mu_i}(Ka_i) H_m^{(1)}(KR). \quad (\text{B.1})$$

It is valid under the following conditions:

$$\begin{cases} d + m + \sum_{i=1}^n \mu_i = 2p & p \in \mathbb{Z} \\ R > \sum_{i=1}^n a_i \\ d + \sum_{i=1}^n |\mu_i| > |m|. \end{cases}$$

In three dimensions, one gets

$$\int_0^{+\infty} \frac{k^{d-1} dk}{K^2 - k^2 + i\epsilon} \prod_{i=1}^n j_{\mu_i}(ka_i) j_l(kR) = -\frac{\pi}{2} K^{d-2} \prod_{i=1}^n j_{\mu_i}(Ka_i) h_l^{(+)}(KR) \tag{B.2}$$

if the conditions

$$\begin{cases} d + 1 + l + \sum_{i=1}^n \mu_i = 2p & p \in \mathbb{Z}^+ \\ R \geq \sum_{i=1}^n a_i \\ d - 1 + \sum_{i=1}^n \mu_i > l \end{cases}$$

are verified.

Appendix C. Two-body scattering

The two-body scattering term consists of the summation of all the multiple scattering terms with two distinct scatterers

$$(iji) + (ijij) + (ijiji) + (ijijj) + \dots \tag{22}$$

where (ij) is a symbolic notation for $\sum_{i,j \neq i} \mathbf{t}_i G_0 \mathbf{t}_j$.

In two dimensions, the average of the first term of this series is the leading term and can be calculated exactly [15].

$$\begin{aligned} \Sigma^{(2)} &\approx -8i\pi\rho^2 \frac{\hbar^2}{2m} \left(\frac{J_0(z)}{H_0^{(1)}(z)} \right)^3 \int_{2a}^{\infty} r dr H_0^{(1)}(Kr) H_0^{(1)}(Kr) \\ &= 16i\pi\rho^2 a^2 \frac{\hbar^2}{2m} \left(\frac{J_0(z)}{H_0^{(1)}(z)} \right)^3 \left[(H_0^{(1)}(2z))^2 + (H_1^{(1)}(2z))^2 \right] \end{aligned}$$

where $z = Ka$. When $Ka \rightarrow 0$, expression (12) is recovered. The average of the other terms appearing in the series (22) can be evaluated by a saddle-point approximation of $\int r dr \left(H_0^{(1)}(Kr) \right)^n$. They lead to an infinite series of powers of $t = \frac{J_0(z)}{H_0^{(1)}(z)} \approx \frac{1}{1 + \frac{2}{\pi} \ln \frac{z}{2} e^\gamma}$ for the self-energy

$$\Sigma^{(2)} \approx -\frac{8i\pi}{K^2} \rho^2 \frac{\hbar^2}{2m} t \sum_{n \geq 2} (-)^n a_n t^n \tag{23}$$

with $a_n \approx -\frac{2}{e^{2\gamma}} \left(\frac{i}{\pi} \right)^n n!$.

In three dimensions, the geometrical series is first resummed before averaging. Then the order ρ^2 term of the self-energy is given by the average of this re-summation and reads as

$$\Sigma^{(2)} = \frac{\hbar^2 (4\pi)^2}{2m K} \rho^2 \left(\frac{j_0(z)}{h_0^{(+)}(z)} \right)^3 \int_{2a}^{\infty} \frac{r^2 dr h_0^{(+)}(Kr)^2 \left(1 - \frac{j_0(z)}{h_0^{(+)}(z)} j_0(Kr) h_0^{(+)}(Kr) \right)}{1 - \left[\frac{j_0(z)}{h_0^{(+)}(z)} h_0^{(+)}(Kr) \right]^2}.$$

The integral is decomposed into three parts, namely $I^{(1)}$, $I^{(2)}$ and $I^{(3)}$.

The first contribution represents the scattering sequence (*iji*)

$$I^{(1)} \equiv \frac{1}{z^2} \int_2^{\infty} dx e^{2izx} = \frac{ie^{4izx}}{2z^3} \approx \frac{i}{2z^3} - \frac{2}{z^2}. \quad (\text{C.1})$$

The second contribution is the scattering sequence (*ijij*)

$$I^{(2)} \equiv -\frac{\sin ze^{-iz}}{z^4} \int_2^{\infty} \frac{dx}{x^2} e^{3izx} \sin zx \approx \frac{1}{z^2} \left(\ln z + 4 \ln 2 + \gamma - 1 - \frac{i\pi}{2} \right). \quad (\text{C.2})$$

In the limit $z \rightarrow 0$, the remainder of the series can be evaluated by taking $z = 0$. This gives

$$I^{(3)} = \frac{1}{z^2} \int_2^{\infty} \frac{dx}{x(x+1)} = \frac{1}{z^2} \ln \frac{3}{2}.$$

Combining these three expressions, one obtains expression (13) for the self-energy.

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