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Edited by **J. M. Marshall, N. Kirov** *and* **A. Vavrek**

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TRANSPORT PHENOMENA IN SYSTEMS WITH ROUGH BOUNDARIES

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ABSTRACT

We discuss a very simple and versatile formalism for transport problems in systems with rough boundaries. We reduce a transport problem near rough walls to a completely equivalent bulk problem with ideal flat walls, but with certain bulk imperfections. The latter problem can be easily analyzed by standard perturbative techniques. All the calculations are very simple and straightforward. As examples, we consider diffusion and conductivity in rough films, single-particle diffusion (a "bouncing ball"), quantum interference (localization) corrections to conductivity, *etc.*

1. Introduction

Motion of particles near the walls is determined by the surface-induced changes in energy, such as changes in energy spectra or potential reliefs, and by the profile of the walls. The latter factor becomes non-trivial if the walls are rough with random inhomogeneities of different scales. Often the effects of roughness and energy changes can be separated. Below we will discuss the effects of boundary roughness on transport of particles near the walls. Later, we will see that some of the energy changes can be easily incorporated into our formalism.

Reflection of particles from random surface inhomogeneities leads to an additional randomization of motion near the walls. This means that the boundary roughness imposes additional restrictions on the mean free path along the wall. Then the mean free path should be expressed via the parameters of the correlation function of surface inhomogeneities. We will discuss a simple method of calculation for this mean free path, and will apply this method to a wide variety of problems.

A standard approach to transport problems in restricted geometry is to solve the transport equation with a corresponding boundary condition. In the case of rough boundaries with random profile, this integro-differential equation becomes practically unsolvable. Then the only remaining option is to use some over-simplified boundary condition.

We developed an alternative approach to transport problems with rough boundaries[1,2]. It is known that sometimes the analysis of systems with complicated boundaries becomes more transparent if one performs a coordinate transformation to simplify the shape of the boundaries. Some of the examples are the Migdal transformation in nuclear physics or wave scattering from a random surface[3-6]. We will use a similar approach for transport of particles in thin films with rough surfaces. By a proper canonic transformation we will make both walls flat. The price will be the appearance of some complicated random terms in the *bulk* Hamiltonian. These small random terms can be treated perturbatively within a standard transport equation as any other bulk imperfections or impurities. As a result, we will get simple expressions for transport coefficients via the correlation function of surface inhomogeneities.

2. Formalism

Let us consider a thin film of an average thickness L with rough boundaries, $x = L/2 - \xi_1(y,z)$ and $x = -L/2 + \xi_2(y,z)$. [In principle, most of the results are also applicable to narrow *2D* strips with random linear boundaries.] The inhomogeneities are small, $\xi_{1,2} << L$, and random, $<\xi_{1,2}> = 0$, with the correlation function $\zeta_{ik}(|s_1-s_2|) = <\xi_i(s_1)\xi_k(s_2)>$,

$$\zeta = \zeta_{11} + \zeta_{22} + 2\zeta_{12}, \quad \zeta_{ik}(\boldsymbol{q}) = \int d^2 s e^{i\boldsymbol{q}\cdot\boldsymbol{s}} \zeta_{ik}(\boldsymbol{s})$$

Typical examples are the Gaussian correlation of surface inhomogeneities of an average height λ,

$$\zeta(\boldsymbol{s}) = \lambda^2 \exp(-s^2/2R^2), \quad \zeta(\boldsymbol{q}) = 2\pi\lambda^2 R^2 \exp(-q^2R^2/2),$$

and its limit for small correlation radii R, *i.e.*, the δ-type correlation,

$$\zeta(\boldsymbol{s}) = \lambda^2 R^2 \delta(s)/s, \quad \zeta(\boldsymbol{q}) = 2\pi\lambda^2 R^2.$$

As we will see, the transition between these two correlation functions occurs when the de Broglie wavelength of particles becomes larger than the correlation radius of surface inhomogeneities.

The boundaries can be made flat, $X = -L/2$ and $X = L/2$, by the non-linear coordinate transformation

$$X = \frac{x - \xi_1(y,z)/2 + \xi_2(y,z)/2}{1 - \xi_1(y,z)/2L - \xi_2(y,z)/2L}, \quad Y = y, \quad Z = z$$

This canonical coordinate transformation should be supplemented by the conjugate momentum transformation, $\boldsymbol{p} \to \boldsymbol{P}$. As a result, the Hamiltonian $\epsilon(\boldsymbol{p})$ in new variables acquires a small random part,

$$\epsilon(\boldsymbol{p}) = \epsilon(\boldsymbol{P}) + V, \quad \hat{V} = V_x \hat{P}_x \xi/L + [\hat{P}_x \boldsymbol{V}_q \cdot \xi'_q X + \boldsymbol{V}_q \cdot \xi'_q X \hat{P}_x]/2L,$$
$$V_x = \partial\epsilon/\partial P_x, \quad \boldsymbol{V}_q = \partial\epsilon/\partial \boldsymbol{Q}, \quad \xi'_q = \partial\xi/\partial \boldsymbol{s}, \quad \xi = \xi_1 + \xi_2, \quad \boldsymbol{Q} = \{P_y, P_z\}$$

The problem should be approached differently depending on whether the film is thick and the motion across the film is classical (or *WKB*), or the film is very thin with a large separation between levels of quantized motion. For example, one of the most important cases is the classical motion of particles with quadratic energy spectrum, $\epsilon = p^2/2m$. The above coordinate transformation demonstrates that the motion of such particles between rough walls is equivalent to the motion between perfect flat walls of particles with anisotropic coordinate-dependent random effective mass

$$\left(\frac{1}{m^*}\right)_{xx} = \frac{1}{m}\left(1 + \frac{2\xi}{L}\right), \quad \left(\frac{1}{m^*}\right)_{xy} = \frac{X\xi'_y}{mL}, \quad \left(\frac{1}{m^*}\right)_{xz} = \frac{X\xi'_z}{mL}$$

(other components are not affected). In the opposite case of strong quantization with suppressed transitions between widely separated states $P_x^{(j)} = 2\pi j/L$, the motion of particles along the film is equivalent to *2D* motion in the weak random potential

$$V_j(y,z) = \left(\frac{2\pi\hbar}{L}\right)^2 \frac{\xi(y,z)}{mL}$$

with all standard localization implications.

In general, the above "perturbation" V leads to transitions between different bulk (momentum) states with the probability $W(\boldsymbol{P},\boldsymbol{P}')$ which is determined by the matrix elements of V:

$$W(\boldsymbol{P},\boldsymbol{P}') = (2\pi/\hbar)<|V_{qP_x,q'P'_x}|>\delta(\epsilon-\epsilon') =$$
$$\frac{\zeta(\boldsymbol{q}-\boldsymbol{q}')}{4\pi L^2}[2V_x^2 P_x^2 \delta(P'_x - P_x) + \Omega V_x P_x \delta'(P'_x - P_x) + \frac{\Omega^2}{8}\delta''(P_x' - P_x)]\delta(\epsilon-\epsilon'),$$
$$\Omega = (P_x \boldsymbol{V}_q - P'_x \boldsymbol{V}_{q'})\cdot(\boldsymbol{Q}-\boldsymbol{Q}')$$

For simplicity, let us assume that the scattering from the rough walls is the sole source of particle diffusion in thin films thus neglecting scattering by bulk impurities or collisions of particles with each other. Then the above transition probability immediately gives us the diffusion coefficient in momentum space,

$$D_{ik}^{(p)} = \frac{1}{2}\int K_i K_k W(\boldsymbol{P},\boldsymbol{P}') d^3K, \quad \boldsymbol{K} = \boldsymbol{P} - \boldsymbol{P}',$$

for the Focker-Plank equation,

$$\partial_t n = \partial_p \cdot [\boldsymbol{G} n + \partial_p (\hat{D}^{(p)} n)], \quad \boldsymbol{G} = \int \boldsymbol{K} W(\boldsymbol{P},\boldsymbol{P}') d^3K,$$

or the collision integral,

$$L_{coll}\{n(\boldsymbol{P})\} = \int W(\boldsymbol{P},\boldsymbol{P}')[n(\boldsymbol{P}) - n(\boldsymbol{P}')] V_0 d^3P/(2\pi\hbar)^3$$

for the Boltzmann transport equation,

$$\partial_t n + \boldsymbol{v}\cdot\partial_r n + \boldsymbol{F}\cdot\partial_p n = L_{coll}\{n\}$$

where $\boldsymbol{v} = \partial\epsilon/\partial\boldsymbol{P}$ is the velocity, $\boldsymbol{F}$ is the external force, $n(\boldsymbol{P})$ is the distribution function. The transport problem with this collision integral is similar to standard problems with impurity scattering, and the consequent calculation of transport coefficients becomes rather straightforward.

3. Transport coefficients

Below we will give only the values of transport coefficients for particles with quadratic, $\epsilon = p^2/2m$, and linear, $\epsilon = cp$, spectra; data for more complicated spectra can be found elsewhere.[2] The conductivity (or mobility) along the film for particles with quadratic spectrum is

$$\sigma = -\frac{4e^2L^2}{\pi}\int\frac{\partial n_0}{\partial\epsilon}\frac{(2m\epsilon)^{1/2}}{\zeta_0-\zeta_1}\frac{d\epsilon}{\cos^2\theta}\frac{d\cos\theta}{\alpha+4\tan^4\theta}$$

where $\alpha = (\eta_0-\eta_1)/(\zeta_0-\zeta_1)$, $\eta(P,\theta,\phi) = \zeta(P\cos\theta,\phi)[1-\cos\phi]^2$, $\zeta_{0,1}$, ζ_1 and $\eta_{0,1}$ are the angular harmonics of the correlation function. This equation can be easily analyzed for different correlation functions and in different temperature regimes. For example, in the case of Gaussian correlations the conductivity along the film is equal to

$$\sigma = \frac{2e^2L^2}{\pi^2p_F}\frac{f_F(p_FR/\hbar)}{\lambda^2R^2},\quad f_F(x) = \int\frac{\exp(x^2\cos^2\theta/2)}{\cos^2\theta}\frac{d\cos\theta}{3/2+4\tan^4\theta}$$

for degenerate systems, $T << T_F$. Comparison with the standard expression for conductivity, $\sigma = e^2\Lambda N/p_F$ demonstrates that scattering by surface inhomogeneities restricts the free path along the surface by

$$\Lambda = 2L^2f_F(p_FR/\hbar)/\pi\lambda^2R^2N$$

where N is the density of particles inside the film. For Boltzmann systems the expression for mobility is similar,

$$\sigma = \frac{4}{\sqrt{\pi}}\frac{e^2L^2N\hbar^3}{\lambda^2R^2m^2T^2}f_B(mTR^2/\hbar^2),\quad f_B(x) = \int\exp[-z(1-x)]\frac{z^{-1/2}dz}{\cos^2\theta}\frac{d\cos\theta}{3/2+4\tan^4\theta}$$

Since in this case $\sigma = e^2\Lambda N/(2mT)^{1/2}$, the effective mean free path along the film is

$$\Lambda = \frac{16}{\sqrt{\pi}}\frac{L^2\hbar^3}{\lambda^2R^2(2mT)^{3/2}}f_B(mTR^2/\hbar^2)$$

The variable for the functions f_F and f_B is the ratio of the correlation radius of the surface inhomogeneities, R, to the de Broglie wavelength of particles. The transition from Gaussian to δ-correlations occurs when this ratio becomes small, and the argument x in the above exponents is close to zero. In the opposite limit, when the correlation radius becomes large on the scale of the wavelength, the mean free path increases. This increase is a natural consequence of the fact that at large correlation radii of surface inhomogeneities, R, the surface becomes effectively flat,

and the reflection of particles from such a surface is nearly specular and does not cause chaotization of motion.

The diffusion coefficient can be easily calculated using the above expressions for conductivity:

$$D_{yy} = D_{zz} = \frac{\pi^2\hbar^3\sigma}{2me^2}/\int\frac{\partial n_0}{\partial\epsilon}(2m\epsilon)^{1/2}d\epsilon$$

The limiting values in degenerate and Boltzmann cases for Gaussian correlations are

$$D_{yy} = D_{zz} = \frac{L^2\hbar^3}{mp_F^2}\frac{f_F(p_FR/\hbar)}{\lambda^2R^2}$$

and

$$D_{yy} = =D_{zz} = \frac{1}{\sqrt{\pi}}\frac{L^2\hbar^3}{\lambda^2R^2m^2T}f_B(mTR^2/\hbar^2)$$

These equations are consistent with the above expressions for the surface-induced mean free path.

The expression for the single-particle diffusion (*i.e.* for the Brownian motion of a ball bouncing between two rough walls - the so-called bouncing ball problem) is somewhat similar. Here the diffusion coefficient along the film depends on the particle energy and the average value of momentum across the film:

$$D_{yy} = D_{zz} = \frac{\hbar^3L^2}{\lambda^2R^2}\exp[(\epsilon-\frac{P_x^2}{2m})\frac{R^2}{2\hbar^2}]\frac{\epsilon-P_x^2/2m}{3(2m\epsilon-P_x^2)^2/8+P_x^4}$$

However, this expression assumes that the motion of a ball is diffusive. There are some numerical indications that the motion of a bouncing ball could slightly deviate from a classical diffusive picture; this question is not settled yet. Here our approach can provide an alternative and more simple computational method.

Let us give also an expression for diffusion coefficient of a particle with linear energy spectrum, $\epsilon = cp$ (a classical phonon or photon):

$$D_{yy} = D_{zz} = \frac{\pi\hbar^3c^2L^2}{2P^2\lambda^2R^2}\frac{(\epsilon^2/c^2-P_x^2)\exp[(\epsilon^2/c^2-P_x^2)R^2/2\hbar^2]}{3(\epsilon^2/c^2-P_x^2)^2/8+P_x^4}$$

4. Quantum interference corrections to transport

Interference between different scattering channels for scattering by random surface inhomogeneities leads to quantum interference (or localization) effects in

transport exactly in the same way as usual scattering by random bulk impurities. This analogy becomes especially transparent after we reduce, by means of canonic coordinate transformation, the problem of transport between rough walls to an equivalent problem of transport with random bulk perturbation V.

As usual, quantum interference corrections are determined by the diffusion coefficient. In *3D* films

$$\frac{\Delta\sigma}{\sigma} = -\int_{\tau}^{\tau_\phi} \nu\lambda_B^2 (D_{yy} t)^{-3/2} dt \sim -\frac{8\pi^2\hbar^2}{m^2 D_{yy}^2}$$

where τ and τ_ϕ are momentum and energy relaxation times, λ_B is the de Broglie wavelength of particles, and the last expression is valid if $\tau_\phi \to \infty$.

The *2D* case (a *2D* strip restricted by random linear boundaries) is different. One of the reasons is the divergence of the above integral at $\tau_\phi \to \infty$. However, in this case we can get an exact explicit expression for the density of states $\nu(E)$ when the boundary inhomogeneities are δ-correlated,

$$\langle \xi(y_1)\xi(y_2)\rangle = \sqrt{2\pi}\lambda^2 R\delta(y_1 - y_2),$$

and the transition between the states with different quantum numbers j, $P_x^{(j)} = 2\pi j\hbar/L$, are effectively suppressed. Then[7]

$$\nu^{-1}(E) = \frac{2\pi^2 (j^4\lambda^2 R)^{1/3}}{L^2}\int_0^\infty \frac{dz}{\sqrt{z}} \exp\left(-\frac{z^3}{24} - z\tilde{E}\right), \quad \tilde{E} = \frac{EmL^2}{(2\pi)^3\hbar^2}\frac{L^2}{(j^4\lambda^2 R)^{2/3}}$$

This expression gives the conductivity threshold for narrow clean *2D* film strips with rough boundaries.

As it was mentioned in Sec. 2, the localization properties of motion of particles along the film in the ultra-quantum *3D* case with suppressed transitions between the discrete levels the film, are the same as for any *2D* motion in weak random potential $V_j(y,z) = (2\pi j\hbar/L)^2 \xi(y,z)/mL$.

5. Summary and discussion

The main advantages of our formalism of solving transport problems in systems with rough boundaries are its simplicity and versatility. The calculations are very straightforward, and can be easily performed for different types of transport problems.

Above we presented different transport results mainly for classical particles with quadratic dispersion law, $\epsilon = p^2/2m$. Similar results for particles with different spectra and for quantized motion across the film can be found in Ref.[2]

The main deficiency of the results obtained so far is that we neglected bulk relaxation and collisions. As a result, the mean free path along the film is restricted only by scattering on surface inhomogeneities. It is relatively simple to include the effect of scattering by bulk impurities. The presence of bulk impurities is equivalent

to the random term $U(r)$ in the Hamiltonian. After the coordinate transformation the effective random scattering field has the form

$$V_{eff} = U(R) + \delta U(R) + V,$$

where V is the perturbation from Sec. 2, and δU is the change in U as a result of transformation $r \to R$. The matrix element of the perturbation V_{eff} is linear in these three terms, while its square will contain, after averaging over bulk impurities and surface inhomogeneities, only the terms

$$|V_{eff\,P,P'}|^2 = |U_{P,P'}|^2 + |V_{P,P'}|^2 + |\delta U_{P,P'}|^2 + \delta U^*_{P',P} V_{P,P'} + \delta U_{P,P'} V^*_{P',P}$$

After the substitution into the equations for scattering probability and collision integral, the first term will reproduce a usual collision frequency with impurities, the second term will give the surface-induced collision integral from Sec.2, while the last three interference terms will be linear in *both* impurity concentration and the surface correlator ζ/L. If the impurity concentration is small and these three terms can be neglected, the transport parameters will obey a simple Matthiessen's rule for independent bulk and surface-induced collisions. Otherwise, the interference terms will somewhat complicate the picture. In any case, the calculations are very straightforward. The situation will be much more complicated for large concentration of impurities or for strong interaction potential when the wave functions for calculation of matrix elements should include effects of impurity scattering. The situation, when the bulk relaxation is determined by particle-particle collisions, is less clear.

Practically the same modification of the calculations will take place if we have to take into account possible energy distortions near the boundaries, $U\,U_0(x-L/2+\xi_1) + U_0(x+L/2-\xi_2)$. Then, after the coordinate transformation, the matrix element in the scattering probability will contain the perturbation $V_{eff} = \delta U + V$, while the potential $U_0(X-L/2) + U_0(X+L/2)$ should be used for calculation of wave functions and be included into the *l.h.s.* of the transport equation. The consequences of change $V \to V_{eff}$ in the expressions for scattering probability are obvious.

The appearance of the new length scale, namely, the mean free path along the boundaries restricted by scattering by surface inhomogeneities, affects the scale of size effects (see, *i.e.*, Ref.[8]) near the boundaries. Our preliminary results[9] show that the corresponding calculation requires the simultaneous analysis of the effects of surface roughness and bulk relaxation.

Our method can also be applied for derivation of effective boundary conditions in different systems[10,11]. Another possibility of continuation of this work is the application to layered systems. In this case the main modification assumes the change of the boundary condition $\Psi = 0$ on the walls.

In addition, the results of this paper provide a new method for numerical study of the bouncing ball problem. There are some indications that the asymptotic motion of such a particle differs from a standard diffusion law $l \sim Dt^2$. Our method can simplify the calculations and clarify the situation.

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