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On the Theory of Brownian Motion. VIII. A Brownian Particle in a Dilute Gas with Inelastic Collisions

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The Fokker–Planck equation for a heavy particle in a dilute gas of light particles is derived from a Boltzmann equation. Inelastic collisions between the heavy and light particles are considered, and explicit quadratures for the frictional coefficients are given in terms of the scattering kernels. It is shown that the general formulation reduces properly to known results in the cases of pure elastic and pure diffuse scattering, respectively.

KEY WORDS: Brownian motion; Fokker–Planck equation; Boltzmann equation; diffuse scattering; inelastic collisions.

1. INTRODUCTION

This paper considers the problem of determining the kinetic equation for a heavy particle immersed in a medium that is a dilute gas. The temperature of the particle may differ from that of the ambient gas. This question has obvious interest with respect to the behavior of particulate matter in the atmosphere.

The problem has already been treated by Slinn *et al.*[1,2] from the point

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of view of a stochastic analysis of the random impacts of the gas molecules on the heavy particle (henceforth B-particle or Brownian particle). The model was that of a spherical B-particle, a fraction α of whose collisions with gas particles were elastic, and a fraction $1 - \alpha$ perfectly diffuse. We intend to treat the problem from the point of view of the Boltzmann equation. Our purpose is not to rederive a known result (though, of course, that is a by-product), but to permit situations of more general particle shape, and more general collision laws, to be treated. An analysis of the case where the ambient medium is dense is presented elsewhere.[3]

The present problem is clearly a case of Brownian motion. The derivation of the Fokker–Planck equation from the Boltzmann equation for a B-particle with no internal structure (elastic collisions) was first given by Wang Chang and Uhlenbeck,[4] but existed only in the form of an unpublished report until recently. Many years ago the present author derived it independently, but never published the derivation (only mentioning it in passing[5]) because he had heard of, but not seen, the Wang Chang–Uhlenbeck report. More recently, Montgomery has published a derivation,[6] the final results of which are restricted to a hard sphere interaction. An exposition is also given by Harris.[7] The aim of this paper is to generalize the derivation of a Fokker–Planck equation from the Boltzmann equation to the case of a general inelastic scattering law.

In order to avoid possible confusion, let us emphasize the following points. If inelastic collisions take place, there must be internal degrees of freedom in the colliding particles. These will describe overall rotations of the particles as well as their internal vibrations. However, we shall consider here *only* the translational motion. The distribution functions introduced in Section 2 are one-body translational distribution functions. Of course, these are affected by the existence of the internal degrees of freedom, and this must be accounted for in the structure of the relevant Boltzmann equation. In the development presented here, the effects of the internal degrees of freedom are supposed incorporated into the collision kernels, or scattering laws, W, introduced in Section 2.

A Brownian particle is itself a rather complicated many-body system A basic premise of the theory is that the mass of the Brownian particle is much greater than the mass of the particles of the medium. Since common atoms have more or less similar masses, within a factor ten or so, this implies that the Brownian particle must contain very many atoms, all strongly interacting to hold the particle together. This being the case, it is not possible to treat the internal degrees of freedom dynamically in any practical way. Rather, we shall describe inelastic collisions by a scattering function giving the probability of a given momentum transfer on collision, without specifying which internal degrees of freedom actually participate in the transfer.

This scattering law may well be, and in most cases probably will be, phenomenological in nature, and be based on the models used in boundary value problems in the kinetic theory of gases. In this latter theory, the boundary condition on the distribution function of a gas at a surface is specified in terms of a linear operator transforming the distribution function of incoming gas particles into that for outgoing particles. Since our Brownian particle is a large (compared to gas molecules), solid object, we feel justified in applying this model in the present case. From the point of view of practicality, this subterfuge is actually an advantage, since current dynamical knowledge of gas–surface interactions is so meagre that phenomenological descriptions are all we really have to work with.

A referee of an earlier version of this paper has drawn attention to a series of papers by Williams.[8] These papers consider a variety of problems concerning the motion of small spheres in gases; they go a great deal further, in most respects, than does the present paper. However, Williams does not consider the Fokker–Planck equation. The work here reported, though done in ignorance of Ref. 8, could thus be considered as supplementary to that of Williams.

In the cases to be considered here, we shall assume that the B-particle is being pumped by some external energy source, e.g., light, and that the absorbed energy is rapidly distributed among the internal degrees of freedom These degrees of freedom thus constitute a heat bath at temperature T_B, only negligibly perturbed by collisions with ambient gas molecules.

2. FORMULATION OF THE PROBLEM

Our system is a collection of particles of mass m (medium, or m-particles) with number density n_m, and particles of mass M (B-particles) with number density n_B. We require that

$$M \gg m; \qquad n_m \gg n_\mathrm{B} \tag{1}$$

The one-body distribution function of the m-particles will be called ϕ, that of the B-particles, f. These functions are assumed to satisfy the coupled Boltzmann equations

$$\frac{D\phi}{Dt} \equiv \frac{\partial\phi}{\partial t} + \mathbf{v}\cdot\nabla\phi + \frac{\mathbf{F}_1}{m}\cdot\nabla_\mathbf{v}\phi = n_m J_{mm}(\phi, \phi) + n_\mathrm{B} J_{m\mathrm{B}}(\phi, f) \tag{2a}$$

$$\frac{Df}{Dt} \equiv \frac{\partial f}{\partial t} + \mathbf{c}\cdot\nabla f + \frac{\mathbf{F}_2}{M}\cdot\nabla_\mathbf{c} f = n_m J_{\mathrm{B}m}(f, \phi) + n_\mathrm{B} J_{\mathrm{BB}}(f, f) \tag{2b}$$

The velocity of the m-particles is denoted by $\mathbf{v}$, that of the B-particles by $\mathbf{c}$. Here $\mathbf{F}_1$ and $\mathbf{F}_2$ are the external forces on the m- and B-particles, respectively.

The J's are the collision operators, to be specified below. Equations (2a) and (2b) are the starting equations of our development; we do not attempt to justify them from more basic hypotheses.

We shall consider the situation where $n_B \ll n_m$, i.e., the B-particles are very dilute. Thus we will neglect the second terms on the right-hand sides of Eq. (2a) and (2b). Clearly, then, if we were to solve (2a), we would not get the correct m-particle distribution function for those m-particles leaving collisions with a B-particle. But our main interest is in (2b), and, as we shall see, all we need there is the m-particle distribution function for m-particles *entering* collisions with the B-particle. So we restrict our attention to J_{Bm}.

We write

$$J_{Bm}(f, \phi) = -\int W(\mathbf{c}', \mathbf{v}'|\mathbf{c}, \mathbf{v})f(\mathbf{c})\phi^-(\mathbf{v})\, d\mathbf{c}'\, d\mathbf{v} + \int W(\mathbf{c}, \mathbf{v}|\mathbf{c}', \mathbf{v}')f(\mathbf{c}')\phi^-(\mathbf{v}')\, d\mathbf{c}'\, d\mathbf{v}' \tag{3}$$

ϕ^- is the distribution function of m-particles entering collisions with a B-particle. The W notation, which is very convenient for our problem, is due to Waldmann.[9] We shall assume that, aside from a term expressing momentum conservation, the transition kernel W depends only on the relative velocities $\mathbf{g}$ and $\mathbf{g}'$ before and after collision, respectively,

$$\mathbf{g} = \mathbf{v} - \mathbf{c}, \qquad \mathbf{g}' = \mathbf{v}' - \mathbf{c}' \tag{4}$$

So, setting $\mathbf{V} = (M + m)^{-1}(M\mathbf{c} + m\mathbf{v})$, the center-of-mass velocity, we obtain

$$W(\mathbf{c}', \mathbf{v}'|\mathbf{c}, \mathbf{v}) = w(\mathbf{g}'|\mathbf{g})\, \delta(\mathbf{V} - \mathbf{V}') \tag{5}$$

The delta function in (5) arises from conservation of momentum in collisions, even inelastic ones. This is an expression of Galilean invariance.

Waldmann has shown that the ordinary Boltzmann collision term can be written in the form (3). The main difference between his case and ours is that we consider inelastic collisions, with a consequent loss of symmetry properties of W.

3. THE FOKKER–PLANCK EQUATION

The transition from the Boltzmann equation to the Fokker–Planck equation is effected by recognizing that, because $\gamma^2 = m/M \ll 1$ (by hypothesis), on the average the velocity of the B-particle is much less than the velocity of an m-particle, $c = O(\gamma v)$. Furthermore, the *velocity change* on collision $\mathbf{c}' - \mathbf{c}$ is very small. In fact, by conservation of momentum, $\mathbf{c}' - \mathbf{c} = \gamma^2(\mathbf{v}' - \mathbf{v})$.

Therefore we expand $f(\mathbf{c}')$ about $f(\mathbf{c})$:

$$f(\mathbf{c}') = f(\mathbf{c}) + (\mathbf{c}' - \mathbf{c})\cdot\nabla_c f(\mathbf{c}) + \tfrac{1}{2}(\mathbf{c}' - \mathbf{c})(\mathbf{c}' - \mathbf{c}):\nabla_c\nabla_c f + O[(\mathbf{c}' - \mathbf{c})^3] \tag{6}$$

Let us note that, because of the delta function in (5), we may write both integrals in (3) as integrals over $\mathbf{c}'$, $\mathbf{v}'$, and $\mathbf{v}$. So, inserting (6) in (3) and using (2b), we obtain

$$\begin{aligned}\frac{Df}{Dt} &= \left\{\int [w(\mathbf{g}|\mathbf{g}')\phi^-(\mathbf{v}') - w(\mathbf{g}'|\mathbf{g})\phi^-(\mathbf{v})]\,\delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{v}\,d\mathbf{v}'\,d\mathbf{c}'\right\} f(\mathbf{c})\\ &+ \left\{\int w(\mathbf{g}|\mathbf{g}')\phi^-(\mathbf{v}')(\mathbf{c}' - \mathbf{c})\,\delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{v}\,d\mathbf{v}'\,d\mathbf{c}'\right\}\cdot\nabla_c f(\mathbf{c})\\ &+ \tfrac{1}{2}\left\{\int w(\mathbf{g}|\mathbf{g}')\phi^-(\mathbf{v}')(\mathbf{c} - \mathbf{c}')(\mathbf{c} - \mathbf{c}')\,\delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{v}\,d\mathbf{v}'\,d\mathbf{c}'\right\}:\nabla_c\nabla_c f(\mathbf{c})\\ &+ \cdots\end{aligned} \tag{7}$$

The terms indicated by ellipses are of higher order than those exhibited, and will be neglected. Our object now is to simplify the three terms on the right-hand side of (7).

Let us first investigate the first term, a scalar, evaluating it to order γ^2. To do this, we change variables to $\mathbf{g}$, $\mathbf{g}'$, $\mathbf{V}'$, noting that

$$d\mathbf{v}\,d\mathbf{v}'\,d\mathbf{c}' = d\mathbf{g}\,d\mathbf{g}'\,d\mathbf{V}' \tag{8}$$

Furthermore, we expand

$$\phi^-(\mathbf{v}) = \phi^-(\mathbf{g}) + \mathbf{c}\cdot\nabla_g\phi^-(\mathbf{g}) + \cdots \tag{9}$$

and similarly for $\phi^-(\mathbf{v}')$. Thus the first term on the right-hand side of (7) becomes

$$\begin{aligned}&\int \delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{V}'\,[w(\mathbf{g}|\mathbf{g}')\phi^-(\mathbf{g}') - w(\mathbf{g}'|\mathbf{g})\phi^-(\mathbf{g})]\,d\mathbf{g}\,d\mathbf{g}'\\ &\quad + \int \delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{V}'\,[w(\mathbf{g}|\mathbf{g}')\mathbf{c}'\cdot\nabla_{g'}\phi^-(\mathbf{g}') - w(\mathbf{g}'|\mathbf{g})\mathbf{c}\cdot\nabla_g\phi^-(\mathbf{g})]\,d\mathbf{g}\,d\mathbf{g}'\end{aligned} \tag{10}$$

The first term of (10) vanishes by symmetry, $\mathbf{g}'$ and $\mathbf{g}$ being dummy variables of integration. The second term may be written, after some manipulation, as

$$\int \delta(\mathbf{V} - \mathbf{V}')\,d\mathbf{V}'\,w(\mathbf{g}|\mathbf{g}')(\mathbf{c}' - \mathbf{c})\cdot\nabla_{g'}\phi^-(\mathbf{g}')\,d\mathbf{g}\,d\mathbf{g}' \tag{11}$$

Now

$$\mathbf{c}' - \mathbf{c} = [\gamma^2/(1 + \gamma^2)](\mathbf{g} - \mathbf{g}') \approx \gamma^2(\mathbf{g} - \mathbf{g}') \tag{12}$$

So, to order γ^2, the coefficient of $f(\mathbf{c})$ in (7) is given by

$$\gamma^2 \int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')\cdot\nabla_{g'}\phi^-(\mathbf{g}')\, d\mathbf{g}\, d\mathbf{g}' \tag{13}$$

We now go on to the second term on the right of (7), the vector term. Using (9) and (12) this term becomes

$$\gamma^2 \int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')\phi^-(\mathbf{g}')\, d\mathbf{g}\, d\mathbf{g}'$$
$$+ \gamma^2\left[\int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')\cdot\nabla_{g'}\phi^-(\mathbf{g}')\, d\mathbf{g}\, d\mathbf{g}'\right]\cdot\mathbf{c} + O(\gamma^4) \tag{14}$$

The first term of (14), which is independent of $\mathbf{c}$, is just m^{-1} times the negative of the mean momentum transferred to a stationary B-particle by collisions with the m-particles. Thus we write this term as $-\gamma^2\langle\mathbf{F}\rangle/m = -\langle\mathbf{F}\rangle/M$. It vanishes if $\phi^-(\mathbf{g}') = \phi^-(|\mathbf{g}'|)$, the collisions are elastic, and $w(\mathbf{g}|\mathbf{g}') = w(\mathbf{g}'|\mathbf{g})$ (existence of inverse collisions), but not in general.

The last term in (7), the tensor term, is already of order γ^4, so we may approximate $\phi^-(\mathbf{v}')$ by $\phi^-(\mathbf{g}')$ in it. After these computations, we can write down the Fokker–Planck equation for the B-particle

$$\frac{\partial f}{\partial t} + \mathbf{c}\cdot\nabla f + \frac{(\mathbf{F}_2 + \langle\mathbf{F}\rangle)}{M}\cdot\nabla_c f = Bf + (\mathbf{B}\cdot\mathbf{c})\cdot\nabla_c f + \mathbf{D}\colon\nabla_c\nabla_c f \tag{15}$$

where

$$\langle\mathbf{F}\rangle/M = -\int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')\phi^-(g')\, d\mathbf{g}\, d\mathbf{g}' \tag{16a}$$

$$\mathbf{B} = \gamma^2 \int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')\nabla_{g'}\phi^-(\mathbf{g}')\, d\mathbf{g}\, d\mathbf{g}' \tag{16b}$$

$$\mathbf{D} = \tfrac{1}{2}\gamma^4 \int \delta(\mathbf{V} - \mathbf{V}')\, d\mathbf{V}'\, w(\mathbf{g}|\mathbf{g}')(\mathbf{g} - \mathbf{g}')(\mathbf{g} - \mathbf{g}')\phi^-(\mathbf{g}')\, d\mathbf{g}\, d\mathbf{g}' \tag{16c}$$

$$B = \mathrm{Tr}\mathbf{B}$$

Let us note that, in fact, the $\mathbf{V}'$ integration can be carried out at this stage, and just yields unity. Equation (15) is the Fokker–Planck equation we have been seeking.

4. SPECIAL CASES

Since the coefficients defined by Eq. (16a)–(16c) look rather abstract, it will be useful, both for didactic purposes and for purposes of checking, to show that (15) reduces to the proper form in the two special cases for which the result is already known. These are (1) elastic collisions with spherically symmetric potentials,[4,6] and (2) purely diffuse collisions.[1] We take these in turn, giving only the outlines of the computation. The details are quite tedious.

4.1. Elastic Collisions with Spherically Symmetric Potentials

This is the case of the ordinary Boltzmann equation. For this case, Waldmann[9] has shown that

$$w(\mathbf{g}|\mathbf{g}') = \sigma(\theta, g')\,\delta\left(\frac{g^2 - g'^2}{2}\right) \tag{17}$$

where $\theta = \cos^{-1}(\mathbf{g}\cdot\mathbf{g}'/g^2)$, the angle of scattering. σ is the differential cross section. For this case

$$w(\mathbf{g}|\mathbf{g}') = w(\mathbf{g}'|\mathbf{g}) \tag{18}$$

If we let ϕ^- be the Maxwellian distribution (which we must if we are to compare with the known cases)

$$\phi^-(\mathbf{g}) = (m/2\pi kT)^{3/2}\exp(-mg^2/2kT) \tag{19}$$

then one can easily show, using the symmetry of w, that

$$\mathbf{B} = M\mathbf{D}/kT \tag{20}$$

Thus it is sufficient to evaluate $\mathbf{D}$.

We have done this as follows. Write the $\mathbf{g}$ and $\mathbf{g}'$ integrals in polar coordinates, using the $\mathbf{g}$ direction as the polar axis for $\mathbf{g}'$. First integrate over the magnitude of $\mathbf{g}'$, using the delta function in w. Then do the angular integrations over the direction of $\mathbf{g}'$. The computation is rather long, though each step is elementary, and the final result is

$$\mathbf{D} = D\mathbf{1} \tag{21}$$

$$D = \gamma^4(8\pi^2/3)(m/2\pi kT)^{3/2}\int_0^\infty g'^5\exp(-mg^2/2kT)\,dg' \times \int_0^\pi \sigma(\theta, g')(1 - \cos\theta)\sin\theta\,d\theta \tag{22}$$

which is the known result.[4]

4.2. Purely Diffuse Scattering

Purely diffuse scattering refers to the case where the particles leaving a collision have a Maxwellian velocity distribution appropriate to the temperature of the B-particle T_B (which may be different from the ambient gas temperature T) independently of their initial relative velocity $\mathbf{g}$. For this case we take

$$w(\mathbf{g}|\mathbf{g}') = (1/2\pi)(m/kT_{\mathrm{B}})^2 \int \mathbf{g}\cdot\mathbf{n} \exp(-mg^2/2kT) g' b \, db \, d\epsilon \tag{23}$$

where $\mathbf{n}$ is the unit outward normal to a surface element of the B-particle, assumed spherical for our example. The integration region is restricted by $\mathbf{g}\cdot\mathbf{n} > 0$, $\mathbf{g}'\cdot\mathbf{n} < 0$. Here b is the impact parameter of the impinging particle, and ϵ is the associated polar angle. For the justification of this form for w, see Ferziger and Kapper[10] (especially p. 348ff).

Again, we take ϕ^- to be a Maxwellian at temperature T [Eq. (19)]. Let us outline the computation of $\mathbf{D}$. First we integrate over $\mathbf{g}$, using polar coordinates with $\mathbf{n}$ as the polar axis, remembering that $\mathbf{g}\cdot\mathbf{n} > 0$. Then we do the $b\,db\,d\epsilon$ integral using $\mathbf{g}'$ as the polar axis. If a is the radius of the spherical B-particle and $\chi = \cos^{-1}(\mathbf{n}\cdot\mathbf{g}'/g')$, then

$$b = a \sin\chi, \qquad b\,db = a^2 \sin\chi \cos\chi \, d\chi \tag{24}$$

Finally, we integrate over $\mathbf{g}$. Although we omit all details, we thought it worthwhile to indicate the order of integration that we found most convenient. The final result is

$$\mathbf{D} = \frac{4\pi}{3}\frac{m}{M}\frac{kT}{M}\left(\frac{8kT}{\pi m}\right)^{1/2} a^2 \left[\frac{1}{2} + \frac{1}{2}\frac{T_{\mathrm{B}}}{T} + \frac{\pi}{8}\left(\frac{T_{\mathrm{B}}}{T}\right)^{1/2}\right]\mathbf{1} \tag{25}$$

Equation (25) agrees with the result of Slinn *et al.*[1] if we put $\alpha = 0$ ($f = 1$ in their notation; pure diffuse scattering).

In the diffuse case, we do not have the simple relationship between $\mathbf{D}$ and $\mathbf{B}$ given by Eq. (20), so $\mathbf{B}$ must be evaluated separately. The technique is exactly the same as for $\mathbf{D}$, and we get

$$\mathbf{B} = \frac{4\pi}{3}\frac{m}{M}\left(\frac{8kT}{\pi m}\right)^{1/2} a^2 \left[1 + \frac{\pi}{8}\left(\frac{T_{\mathrm{B}}}{T}\right)^{1/2}\right]\mathbf{1} \tag{26}$$

again in agreement with Ref. 1 for the case $\alpha = 0$. If one assumes, as is usual, that the elastic and diffuse w's are additive, with coefficients α and $1 - \alpha$, respectively, then the agreement with Ref. 1 is complete.

Thus our result properly reduces to the two known special cases.

As an additional example, we have computed $\mathbf{D}$ for the case of a particle in the shape of a thin, flat plate of arbitrary shape. This example is not completely academic, for one might expect such substances as graphite or mica to form platelets on fragmentation. For fixed orientation of the plate, $\mathbf{D}$ is obviously highly anisotropic, but we are compelled to average over all orientations, since our starting point, Eq. (2), refers to translational motion only, orientations having been already averaged. For the two scattering laws considered above, it turns out that the result is precisely that for a spherical particle of the same total area as that of the plate. The calculation neglects collisions with the narrow surfaces of the plate. It is tempting to conjecture that this result is general for any convex body, but it certainly cannot be true in general, e.g., for a B-particle with deep dimples.

5. DISCUSSION

In the kinetic theory of gases, scattering from a surface (of a B-particle, in our case) is often treated as a linear combination of elastic and perfectly diffuse scattering. Yet this is for reasons of simplicity rather than physical reality. For example, it is reasonable to expect that fast incident particles have a higher probability of elastic scattering than slow particles. This could be taken into account by weighting the elastic and diffuse kernels w by a function of incident velocity, rather than by a constant (Ref. 10, p. 470). For example, if we denote the elastic w [Eq. (17)] by w_e and the diffuse w [Eq. (23)] by w_d, we could assume

$$w = [1 - P(g)]w_e + P(g)w_d \tag{27}$$

where $P(g)$ is some weighting function that goes to zero for large g. Our formulas allow the frictional coefficients $\mathbf{B}$ and $\mathbf{D}$ to be evaluated in this case also.

If one again assumes the B-particle to be a sphere, then the angular integrations indicated in Section 4 are unchanged. The integrals over the magnitude of the velocities must, however, be redone, inserting P in the integrands. The simple Gaussian nature of the integrals is lost (unless one assumes P to be Gaussian, which has in fact been suggested[10,11]), but this is not a serious problem. For example, if one takes $P(g) \sim \exp(-\lambda g)$, the integrals are expressible in terms of Weber functions; alternatively, one can resort to numerical methods.

From the Fokker–Planck equation, one can go to a Smoluchowski equation for the B-particle positional distribution function. There are standard methods for doing this (see Ref. 2 for an example and references). We do not go into this problem here.

Equations (15) and (16) are the principal results of this paper. They show how to handle the problem of the stochastic motion of a heavy particle in a medium of light particles for arbitrary scattering law. Aside from the mathematical problem of computing the integrals and solving the equation, the remaining physical problem is the determination of the requisite scattering laws.

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