

Dynamical Conductivity of the Dilute Lorentz Gas with Spherically Symmetric Scatterers

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The dynamical conductivity of the Lorentz gas with spherically symmetric potentials is studied to lowest order in the density of scatterers. The frequency-dependent friction coefficient is calculated from the Fourier transform of the force-force time-correlation function determined by the dynamics of a single scattering process. The corresponding dynamical conductivity varies with frequency on the scale of the inverse collision time. As an example, the conductivity is calculated for a scattering potential of the Maxwell type.

KEY WORDS: Dynamical conductivity; Lorentz gas; friction; mobility.

1. INTRODUCTION

The Lorentz gas is a model of prime importance in the theory of transport of electrons, neutrons, and photons.^(1,2) In the following we use the language of electrical resistivity. Then the model describes the transport of electrons in a system of randomly distributed identical static scatterers. We shall consider only the classical model. Interactions between electrons are neglected, so that it suffices to consider the motion of a single electron in the potential of the scatterers. The object of our study is the frequency-dependent electrical conductivity, as given by linear response theory.

The Kubo-formula of linear response theory provides an exact expression for the dynamical conductivity.⁽³⁾ Although justification of its validity for the measured transport property may require subtle argument,^(4,5,6) we shall use it as a starting-point. The Lorentz-Boltzmann kinetic theory is

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based on an approximate equation for the velocity distribution of electrons,^(1,2) valid at low density of scatterers and on the time scale of relaxation of the mean velocity. Much effort has been devoted to the problem of its generalization to higher density.⁽⁷⁻¹⁰⁾ In the following we restrict ourselves to low density of scatterers, but consider the short time scale of single electron-ion collisions, corresponding to high-frequency behavior of the electrical conductivity.

By the Kubo-formula the electrical conductivity is expressed as the one-sided Fourier transform of a velocity-velocity time-correlation function. Our calculation is based on an exact rewriting of this expression in terms of a force-velocity and a force-force time-correlation function. At low density it suffices to consider the force-force time-correlation function of a single scatterer. In the derivation of the exact expression we largely follow the presentation of Huberman and Chester.⁽¹¹⁾ An alternative derivation based on Mori's projection operator formalism has been presented by Kubo.^(5, 12, 13) An essential additional step is the application of a cluster expansion, allowing a systematic expansion about the low density system. Related expansions have been proposed by Zwanzig,⁽¹⁴⁾ van Leeuwen and Weijland,⁽¹⁵⁾ and by Hauge and Cohen.⁽¹⁶⁾

We show that for spherical scatterers the low density dynamical conductivity can be calculated exactly. As an example we consider a power law central potential of the Maxwell type.⁽¹⁷⁾

2. MEAN VELOCITY

We consider a classical system of electrons of charge $-e$ and mass m , interacting with randomly distributed identical fixed scatterers. The interaction between electrons is neglected. For mathematical convenience periodic boundary conditions are imposed, with a simple cubic lattice dividing space into unit cells of volume Ω . Each cell contains N electrons and N_i scatterers. For brevity we shall refer to the scatterers as ions, although their charge may vanish. Since the electrons are independent, their dynamics in a unit cell may be described by the single electron Hamiltonian

$$H(\mathbf{r}, \mathbf{p}) = \frac{p^2}{2m} + \Phi(\mathbf{r}) \quad (2.1)$$

with potential

$$\Phi(\mathbf{r}) = \sum_{j=1}^{N_i} \phi(\mathbf{r} - \mathbf{R}_j) \quad (2.2)$$

It is assumed that the range is sufficiently short that at most N_i ions contribute to the potential at \mathbf{r} . The single electron distribution in phase space $f(\mathbf{r}, \mathbf{p}, t)$ is required to be periodic in \mathbf{r} . It is normalized to

$$\int_{\Omega} f(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p} = N \quad (2.3)$$

In the presence of an applied electric field $\mathbf{E}(\mathbf{r}, t)$, which is periodic in \mathbf{r} , the time evolution of the distribution function is given by

$$\frac{\partial f}{\partial t} + \frac{\mathbf{p}}{m} \cdot \frac{\partial f}{\partial \mathbf{r}} + \mathcal{F} \cdot \frac{\partial f}{\partial \mathbf{p}} = e \mathbf{E} \cdot \frac{\partial f}{\partial \mathbf{p}} \quad (2.4)$$

where \mathcal{F} is the total force exerted on an electron by the ions

$$\mathcal{F}(\mathbf{r}) = -\frac{\partial \Phi}{\partial \mathbf{r}} \quad (2.5)$$

For vanishing applied field any function $f_0(H)$ of the Hamiltonian is a stationary solution of Eq. (2.4). We assume that the distribution $f_0(H)$ is normalized as in Eq. (2.3). In linear response theory the distribution function is expanded in the applied field as

$$f(\mathbf{r}, \mathbf{p}, t) = f_0(H) + f_1(\mathbf{r}, \mathbf{p}, t) + o(E) \quad (2.6)$$

where $f_1(\mathbf{r}, \mathbf{p}, t)$ is linear in \mathbf{E} . It satisfies the equation

$$\frac{\partial f_1}{\partial t} + \mathcal{L} f_1 = e \mathbf{E} \cdot \frac{\partial f_0}{\partial \mathbf{p}} \quad (2.7)$$

where \mathcal{L} is the single electron Liouville operator defined by

$$\mathcal{L} A = \{A, H\} = \frac{\mathbf{p}}{m} \cdot \frac{\partial A}{\partial \mathbf{r}} + \mathcal{F} \cdot \frac{\partial A}{\partial \mathbf{p}} \quad (2.8)$$

For uniform oscillating field at frequency ω

$$\mathbf{E}(t) = \mathbf{E}_0 \exp(-i\omega t) \quad (2.9)$$

we consider the solution of Eq. (2.7) oscillating with the same time factor

$$f_1(\mathbf{r}, \mathbf{p}, t) = f_1(\mathbf{r}, \mathbf{p}) \exp(-i\omega t) \quad (2.10)$$

with $f_1(\mathbf{r}, \mathbf{p})$ satisfying

$$(-i\omega + \mathcal{L}) f_1 = \frac{e}{m} \mathbf{E}_0 \cdot \mathbf{p} f'_0 \quad (2.11)$$

where $f'_0(H) = df_0/dH$. The formal solution is

$$f_1 = \frac{e}{m} f'_0 (-i\omega + \mathcal{L})^{-1} \mathbf{p} \cdot \mathbf{E}_0 \quad (2.12)$$

where we have used the property $\mathcal{L} f'_0 = 0$.

We are interested in the mean electron velocity

$$\mathbf{V}(t) = \frac{1}{N} \int_{\Omega} \frac{\mathbf{p}}{m} f(\mathbf{r}, \mathbf{p}, t) d\mathbf{r} d\mathbf{p} \quad (2.13)$$

In the stationary state $\mathbf{V}_0 = 0$, by symmetry in the momentum. To first order in \mathbf{E}_0 , with omission of the exponential time factor,

$$\mathbf{V}_1 = \frac{1}{N} \int_{\Omega} \frac{\mathbf{p}}{m} f_1(\mathbf{r}, \mathbf{p}) d\mathbf{r} d\mathbf{p} \quad (2.14)$$

Substituting from Eq. (2.12) we find that the first order mean velocity is given by

$$\mathbf{V}_1 = -e \mathcal{Y}(\omega, X, \Omega) \cdot \mathbf{E}_0 \quad (2.15)$$

with admittance tensor

$$\mathcal{Y}(\omega, X, \Omega) = \frac{-1}{Nm^2} \int_{\Omega} f'_0 \mathbf{p} (-i\omega + \mathcal{L})^{-1} \mathbf{p} dx \quad (2.16)$$

We have abbreviated the phase space volume element as $dx = d\mathbf{r} d\mathbf{p}$. The tensor depends parametrically on the configuration X of ions in the unit cell. Later we shall perform an average over the probability distribution of configurations $W(X)$, which is assumed known.

3. FRICTION TENSOR

In Eq. (2.16) the admittance tensor for fixed ion configuration is expressed as the one-sided Fourier transform of a velocity correlation function. We show in the following that the corresponding friction tensor can

be expressed in terms of a mixed force-velocity and a force-force correlation function.⁽¹¹⁾

We consider the product $\mathbf{p}f'_0$ appearing on the right hand side of Eq. (2.11), and write it in the form

$$\mathbf{p}f'_0 = G(-i\omega + \mathcal{L})\mathbf{p}f'_0 \quad (3.1)$$

where G is the inverse operator $G(\omega) = (-i\omega + \mathcal{L})^{-1}$. Hence $\mathbf{p}f'_0$ can be expressed as the sum

$$\mathbf{p}f'_0 = -i\omega G\mathbf{p}f'_0 + G\mathcal{F}f'_0 \quad (3.2)$$

The integral over momentum of $\mathbf{p}f'_0$ vanishes, since the distribution $f_0(H)$ is even in \mathbf{p} . Multiplying Eq. (3.2) by \mathcal{F} and integrating over phase space we therefore find the identity

$$-i\omega \int_{\Omega} \mathcal{F}G\mathbf{p}f'_0 dx + \int_{\Omega} \mathcal{F}G\mathcal{F}f'_0 dx = 0 \quad (3.3)$$

We write this in the form

$$-i\omega m \mathbf{A}_{Fv}(\omega, X, \Omega) + \zeta_t(\omega, X, \Omega) = 0 \quad (3.4)$$

with the tensors \mathbf{A}_{Fv} and ζ_t defined by

$$\begin{aligned} \mathbf{A}_{Fv}(\omega, X, \Omega) &= \frac{-1}{Nm} \int_{\Omega} f'_0 \mathcal{F}G\mathbf{p} dx \\ \zeta_t(\omega, X, \Omega) &= \frac{-1}{N} \int_{\Omega} f'_0 \mathcal{F}G\mathcal{F} dx \end{aligned} \quad (3.5)$$

The tensor \mathbf{A}_{Fv} can be related directly to the admittance tensor \mathcal{Y} . Explicitly Eq. (2.11) reads

$$-i\omega f_1 + \frac{\mathbf{p}}{m} \cdot \frac{\partial f_1}{\partial \mathbf{r}} + \mathcal{F} \cdot \frac{\partial f_1}{\partial \mathbf{p}} = e \mathbf{E}_0 \cdot \frac{\partial f_0}{\partial \mathbf{p}} \quad (3.6)$$

Multiplying this by \mathbf{p}/Nm , using the definition (2.14), and integrating by parts on the right hand side we find

$$-i\omega \mathbf{V}_1 + \frac{1}{Nm^2} \int_{\Omega} \mathbf{p}\mathbf{p} \cdot \frac{\partial f_1}{\partial \mathbf{r}} d\mathbf{r} d\mathbf{p} + \frac{1}{Nm} \int_{\Omega} \mathbf{p}\mathcal{F} \cdot \frac{\partial f_1}{\partial \mathbf{p}} d\mathbf{r} d\mathbf{p} = -e \mathbf{E}_0 \quad (3.7)$$

The second term on the left vanishes by periodicity, and in the third we can integrate by parts. This yields

$$-i\omega \mathbf{V}_1 - \frac{1}{Nm} \int_{\Omega} \mathcal{F} f_1 d\mathbf{r} d\mathbf{p} = -e\mathbf{E}_0 \quad (3.8)$$

Substituting in solution f_1 from Eq. (2.12) and using the definition (2.15) we therefore find

$$\mathcal{Y}(\omega, X, \Omega) = \frac{-1}{i\omega m} [\mathbf{1} + \mathbf{A}_{Fv}(\omega, X, \Omega)] \quad (3.9)$$

With the friction tensor $\zeta(\omega, X, \Omega)$ defined by

$$\mathcal{Y}(\omega, X, \Omega) = [-i\omega m \mathbf{1} + \zeta(\omega, X, \Omega)]^{-1} \quad (3.10)$$

we find by comparison with Eq. (3.9)

$$\mathbf{A}_{Fv}(\omega, X, \Omega) = \zeta(\omega, X, \Omega) [i\omega m \mathbf{1} - \zeta(\omega, X, \Omega)]^{-1} \quad (3.11)$$

Substituting this into Eq. (3.4) we obtain

$$\zeta_r(\omega, X, \Omega) = i\omega m \zeta(\omega, X, \Omega) [i\omega m \mathbf{1} - \zeta(\omega, X, \Omega)]^{-1} \quad (3.12)$$

The inverse relation reads

$$\zeta(\omega, X, \Omega) = i\omega m \zeta_r(\omega, X, \Omega) [i\omega m \mathbf{1} + \zeta_r(\omega, X, \Omega)]^{-1} \quad (3.13)$$

Substituting here Eq. (3.4) again we finally find

$$\zeta(\omega, X, \Omega) = \zeta_r(\omega, X, \Omega) [\mathbf{1} + \mathbf{A}_{Fv}(\omega, X, \Omega)]^{-1} \quad (3.14)$$

Thus we have expressed the friction tensor in terms of the two correlation functions given by Eq. (3.5). In the language of Kubo *et al.*⁽⁵⁾ the tensor $\zeta_r(\omega, X, \Omega)$ is the one-sided Fourier transform of the true force correlation function, whereas the friction tensor $\zeta(\omega, X, \Omega)$ is the one-sided Fourier transform of the correlation function of Mori's random force.⁽¹⁸⁾

4. THERMODYNAMIC LIMIT AND CLUSTER EXPANSION

The relations derived so far are exact. They hold even for one electron and one ion per unit cell. In the following we consider the thermodynamic limit of a disordered system with many ions per unit cell. The probability

distribution $W(X)$ of configurations is assumed given. For simplicity we assume that the ions are spherically symmetric, so that the configuration X is completely specified by the positions of the ion centers $X = (\mathbf{R}_1, \dots, \mathbf{R}_{N_i})$ in the unit cell. The distribution $W(X)$ is assumed normalized to unity and symmetric in the labels $1, \dots, N_i$. The partial distribution functions

$$n_i(1, \dots, k) = \frac{N_i!}{(N_i - k)!} \int \cdots \int d\mathbf{R}_{k+1} \cdots d\mathbf{R}_{N_i} W(1, \dots, N_i) \quad (4.1)$$

give the probability of finding a configuration of k ions whatever the configuration of the remaining $N_i - k$ ions.

In the thermodynamic limit the cell volume Ω , the number of electrons N , and the number of ions N_i tend to infinity at constant densities $n = N/\Omega$ and $n_i = N_i/\Omega$. The system is assumed to become isotropic in the thermodynamic limit. Since in our case the electrons are independent, it would be sufficient to consider just a single electron. However, the system at constant density n corresponds closer to the physical situation. For the finite system the distribution of values of the tensors $\mathbf{A}_{Fv}(\omega, X, \Omega)$ and $\zeta_r(\omega, X, \Omega)$ is determined by the probability distribution $W(X)$. In the thermodynamic limit the tensors become isotropic and the distribution functions of the components become sharp.⁽¹⁹⁾ The mean values of the diagonal components can be identified with scalar coefficients $\mathcal{Y}(\omega)$, $\zeta(\omega)$ and $A_{Fv}(\omega)$. Because the distribution functions become sharp, the coefficients are related as in the preceding section. Thus we have

$$\mathcal{Y}(\omega) = \frac{1}{-i\omega m + \zeta(\omega)}, \quad \zeta(\omega) = \frac{\zeta_r(\omega)}{1 + A_{Fv}(\omega)} \quad (4.2)$$

with, for example,

$$\mathcal{Y}(\omega) = \lim_{\substack{N \rightarrow \infty \\ N_i \rightarrow \infty, \Omega \rightarrow \infty}} \frac{-1}{3Nm^2} \int_{\Omega} \langle f'_0 \mathbf{p} \cdot \mathbf{G} \mathbf{p} \rangle dx \quad (4.3)$$

where the angled brackets indicate the ensemble average over the distribution $W(X)$.

The three averaged correlation functions can be expanded formally in terms of an Ursell cluster expansion. For example, in the coefficient ζ_r ,

$$-\frac{1}{3} \langle f'_0 \mathcal{F} \cdot \mathbf{G} \mathcal{F} \rangle = \sum_{k=1}^{N_i} \frac{1}{k!} \int d\mathbf{R}_1 \cdots d\mathbf{R}_k n_i(\mathbf{R}_1, \dots, \mathbf{R}_k) C_{FFk}(\omega) \quad (4.4)$$

where $C_{FFk}(\omega)$ is the cluster term of order k corresponding to the product $-1/3f'_0 \mathcal{F} \cdot G \mathcal{F}$ on the left hand side. The friction coefficient $\zeta(\omega)$ in Eq. (4.2) is given by a ratio of two such expansions, taken in the thermodynamic limit. The ratio can be expanded and rearranged according to the number of ions involved, in the same way as for the dielectric cluster expansion.⁽²⁰⁾ This yields a cluster expansion for the friction coefficient of the form

$$\zeta(\omega) = \sum_{k=1}^{\infty} \zeta_k(\omega) \quad (4.5)$$

where k indicates the number of ion labels. The term for $k=1$ involves just a single ion, and is given by

$$\zeta_1(\omega) = -\frac{1}{3}n_i \int \varphi'_0(\varepsilon) \mathbf{F}(\mathbf{r}) \cdot G_1 \mathbf{F}(\mathbf{r}) dx \quad (4.6)$$

where $\varphi'_0(\varepsilon)$ is the derivative with respect to energy $\varepsilon(r, p)$ of the distribution

$$\varphi_0(\varepsilon) = \lim_{\Omega \rightarrow \infty} \Omega f_0(H_1) \quad (4.7)$$

normalized such that

$$\int \varphi_0(\varepsilon) d\mathbf{p} \rightarrow 1 \quad \text{as } r \rightarrow \infty \quad (4.8)$$

Here $\varepsilon(r, p)$ is the value of the Hamiltonian

$$H_1(\mathbf{r}, \mathbf{p}) = \frac{p^2}{2m} + \phi(r) \quad (4.9)$$

for an electron in the presence of a single ion centered at the origin. The force $\mathbf{F}(\mathbf{r})$ in Eq. (4.6) is simply the gradient $-\partial\phi/\partial\mathbf{r}$, and the operator G_1 is the inverse $(-i\omega + \mathcal{L}_1)^{-1}$ corresponding to the single-ion Hamiltonian (4.9). In practical applications the distribution $\varphi_0(\varepsilon)$ will be a Maxwell-Boltzmann or Fermi-Dirac distribution. In the following we shall consider a step function $\varphi_0(\varepsilon)$, for which the derivative $-\varphi'_0(\varepsilon)$ is proportional to a delta-function in the single particle energy.

As evident from Eq. (4.6), the first term in the cluster expansion of the friction coefficient is linear in the ion density n_i . The next term $\zeta_2(\omega)$ is not necessarily quadratic in n_i , since the pair distribution occurring in the pair cluster integral may involve higher powers of the density. A truncation of

the expansion Eq. (4.5) at the pair level leads to long range divergences in the low frequency limit,^(7, 8) and resummation is necessary to remove these. We shall be concerned only with the first order term, given by Eq. (4.6).

5. SINGLE-ION FRICTION COEFFICIENT

In the following we show how the single-ion friction coefficient, given by Eq. (4.6), can be calculated. We take advantage of the spherical symmetry of the ion. In order to calculate the effect of the inverse operator $G_1(\omega) = (-i\omega + \mathcal{L}_1)^{-1}$ we consider the solution of the vector equation

$$(-i\omega + \mathcal{L}_1) \mathbf{f} = \mathbf{F} \quad (5.1)$$

or, explicitly,

$$-i\omega \mathbf{f} + \left(\frac{\mathbf{p}}{m} \cdot \frac{\partial}{\partial \mathbf{r}} \right) \mathbf{f} + \left(\mathbf{F} \cdot \frac{\partial}{\partial \mathbf{p}} \right) \mathbf{f} = \mathbf{F} \quad (5.2)$$

with $\mathbf{F}(\mathbf{r}) = F(r) \hat{\mathbf{r}}$. We solve Eq. (5.2) by the Ansatz

$$\mathbf{f}(\mathbf{r}, \mathbf{p}) = \hat{g}_+(r, p, \xi) \mathbf{r} + \hat{h}_+(r, p, \xi) \mathbf{p} \quad (5.3)$$

where

$$\xi = \hat{\mathbf{r}} \cdot \hat{\mathbf{p}} \quad (5.4)$$

This leads to two coupled equations for the functions \hat{g}_+ and \hat{h}_+ , which can be cast in the form

$$\begin{aligned} -i\omega \hat{g}_+ + \frac{d\hat{g}_+}{ds} + \frac{F}{r} \hat{h}_+ &= \frac{F}{r} \\ -i\omega \hat{h}_+ + \frac{d\hat{h}_+}{ds} + \frac{1}{m} \hat{g}_+ &= 0 \end{aligned} \quad (5.5)$$

with the abbreviation

$$\frac{d}{ds} = \frac{1}{m} \xi p \frac{\partial}{\partial r} + \xi F \frac{\partial}{\partial p} + (1 - \xi^2) \left(\frac{p}{mr} + \frac{F}{p} \right) \frac{\partial}{\partial \xi} \quad (5.6)$$

This shows that we may define characteristics in (r, p, ξ) -space by the equations

$$\begin{aligned} \frac{dr}{ds} &= \frac{1}{m} \xi p, & \frac{dp}{ds} &= \xi F \\ \frac{d\xi}{ds} &= (1 - \xi^2) \left(\frac{p}{mr} + \frac{F}{p} \right) \end{aligned} \quad (5.7)$$

with scalar parameter s along the characteristics. The variables r and p are positive, whereas ξ takes values between -1 and 1 . It is convenient to choose $s=0$ for $\xi=0$. Then the solutions of Eq. (5.7) have the symmetry

$$r(s) = r(-s), \quad p(s) = p(-s), \quad \xi(s) = -\xi(-s) \quad (5.8)$$

From Eq. (5.7) one finds that the following quantities are constant along the characteristics

$$\begin{aligned} H_1(r, p) &= \frac{p^2}{2m} + \phi(r) \\ L(r, p, \xi) &= rp \sqrt{1 - \xi^2} \end{aligned} \quad (5.9)$$

Hence the characteristics are the intersections of the surfaces in (r, p, ξ) -space of constant $H_1(r, p) = \varepsilon$ and constant $L(r, p, \xi) = \lambda$. By use of these constants one can eliminate the variables p and ξ and reduce the problem to a first order ordinary differential equation for $r(s)$,

$$\frac{dr}{ds} = \sqrt{\frac{2}{m}} \left[\varepsilon - \phi(r) - \frac{\lambda^2}{2mr^2} \right]^{1/2} \quad (5.10)$$

We note parenthetically that the characteristics may be related to the radial motion in the central potential $\phi(r)$ by the introduction of the radial momentum p_r by

$$p_r^2 = p^2 - \frac{\lambda^2}{r^2} \quad (5.11)$$

Then the Hamiltonian becomes

$$H_1(r, p_r) = \frac{p_r^2}{2m} + \phi(r) + \frac{\lambda^2}{2mr^2} \quad (5.12)$$

and the solution $r(s)$, $p_r(s)$ of the equations

$$\frac{dr}{ds} = \frac{\partial H_1}{\partial p_r}, \quad \frac{dp_r}{ds} = -\frac{\partial H_1}{\partial r} \quad (5.13)$$

has $H_1(r, p_r) = \varepsilon$ as a constant of the motion. If $\zeta(s)$ is defined from

$$\zeta = \frac{p_r}{\sqrt{p_r^2 + \lambda^2/r^2}} \quad (5.14)$$

and $p(s)$ from Eq. (5.11), then L^2 defined by Eq. (5.9) takes the constant value λ^2 . Eliminating p_r from Eq. (5.13) we are again led to Eq. (5.10).

Once the solution $r(s)$ of Eq. (5.10) has been found we can define the function

$$U(s) = \frac{F(r(s))}{r(s)} \quad (5.15)$$

Then Eq. (5.5) becomes

$$\begin{aligned} -i\omega \hat{g}_+ + \frac{d\hat{g}_+}{ds} + U\hat{h}_+ &= U \\ -i\omega \hat{h}_+ + \frac{d\hat{h}_+}{ds} + \frac{1}{m} \hat{g}_+ &= 0 \end{aligned} \quad (5.16)$$

The solutions $\hat{g}_+(s, \omega)$, $\hat{h}_+(s, \omega)$ of this set of equations depend parametrically on ε and λ via the function $U(s)$. In our notation we shall usually not make the dependence on ε and λ explicit.

We can regard $\hat{g}_+(s, \omega)$ and $\hat{h}_+(s, \omega)$ as one-sided Fourier transforms of functions $g(s, t)$, $h(s, t)$,

$$\begin{aligned} \hat{g}_+(s, \omega) &= \int_0^\infty e^{i\omega t} g(s, t) dt \\ \hat{h}_+(s, \omega) &= \int_0^\infty e^{i\omega t} h(s, t) dt \end{aligned} \quad (5.17)$$

satisfying the partial differential equations

$$\begin{aligned} \frac{\partial g}{\partial t} + \frac{\partial g}{\partial s} + Uh &= 0 \\ \frac{\partial h}{\partial t} + \frac{\partial h}{\partial s} + \frac{1}{m} g &= 0 \end{aligned} \quad (5.18)$$

to be solved with initial conditions

$$g(s, 0) = U(s), \quad h(s, 0) = 0 \quad (5.19)$$

The solution of these equations for all t is given by

$$\begin{aligned} g(s, t) &= [-\psi'_1(s) \psi_2(s-t) + \psi'_2(s) \psi_1(s-t)] U(s-t) \\ h(s, t) &= \frac{1}{m} [\psi_1(s) \psi_2(s-t) - \psi_2(s) \psi_1(s-t)] U(s-t) \end{aligned} \quad (5.20)$$

where $\psi_1(s)$, $\psi_2(s)$ are solutions of the equation

$$m \frac{d^2\psi}{ds^2} - U\psi = 0 \quad (5.21)$$

satisfying the conditions

$$\begin{aligned} \psi_1(0) &= 1, & \psi'_1(0) &= 0 \\ \psi_2(0) &= 0, & \psi'_2(0) &= 1 \end{aligned} \quad (5.22)$$

The Fourier transforms of the functions $g(s, t)$, $h(s, t)$ are given by

$$\begin{aligned} \hat{g}(s, \omega) &= \int_{-\infty}^{\infty} e^{i\omega t} g(s, t) dt \\ \hat{h}(s, \omega) &= \int_{-\infty}^{\infty} e^{i\omega t} h(s, t) dt \end{aligned} \quad (5.23)$$

In the calculation of the friction coefficient $\zeta_1(\omega)$ in Eq. (4.6) we need the positive-frequency part $\hat{g}_+(s, \omega)$, $\hat{h}_+(s, \omega)$ of these transforms.

By use of spherical symmetry and the form Eq. (5.3) we find for the coefficient $\zeta_1(\omega)$

$$\zeta_1(\omega) = -\frac{8\pi^2}{3} n_i \int \varphi'_0(\varepsilon) F(r) [r\hat{g}_+(r, p, \xi) + \xi p\hat{h}_+(r, p, \xi)] r^2 p^2 dr dp d\xi \quad (5.24)$$

The three-dimensional integral is performed conveniently by transformation to the variables $(\varepsilon, \lambda^2, s)$. From the Jacobian of the transformation one finds for the new volume element

$$d\varepsilon d\lambda^2 ds = 2r^2 p^2 dr dp d\xi \quad (5.25)$$

In the Appendix we show how the Jacobian can be evaluated. The friction coefficient becomes

$$\zeta_1(\omega) = -\frac{4\pi^2}{3} n_i \int \varphi'_0 F[r\hat{g}_+ + \xi p\hat{h}_+] d\varepsilon d\lambda^2 ds \quad (5.26)$$

This can be written

$$\zeta_1(\omega) = -\frac{4\pi^2}{3} n_i \int \varphi'_0(\varepsilon) \hat{C}_+(\omega; \varepsilon) d\varepsilon \quad (5.27)$$

with frequency-dependent collision integral

$$\hat{C}_+(\omega; \varepsilon) = \iint F[r\hat{g}_+ + \xi p\hat{h}_+] d\lambda^2 ds \quad (5.28)$$

In our explicit evaluation we study the collision integral $\hat{C}_+(\omega; \varepsilon)$.

6. FREQUENCY-DEPENDENT COLLISION INTEGRAL

The frequency-dependent collision integral $\hat{C}_+(\omega; \varepsilon)$, defined by Eq. (5.28), can be decomposed into an integral over angular momentum values

$$\hat{C}_+(\omega; \varepsilon) = \int \hat{C}_+(\omega; \varepsilon, \lambda) d\lambda^2 \quad (6.1)$$

with

$$\hat{C}_+(\omega; \varepsilon, \lambda) = \int F[r\hat{g}_+ + \xi p\hat{h}_+] ds \quad (6.2)$$

given by an integral along the characteristic for energy ε and angular momentum λ . The range of the integral is determined by the range of the force $F(r)$. Clearly we can write

$$\hat{C}_+(\omega; \varepsilon, \lambda) = \int_0^\infty e^{i\omega t} C(t; \varepsilon, \lambda) dt \quad (6.3)$$

with the real function $C(t; \varepsilon, \lambda)$ defined for all t by

$$C(t; \varepsilon, \lambda) = \int F[rg(s, t) + \xi ph(s, t)] ds \quad (6.4)$$

From Eqs. (5.15) and (5.19) it follows that for $t = 0$,

$$rg(s, 0) + \xi ph(s, 0) = F \quad (6.5)$$

so that we may express Eq. (6.4) as

$$C(t; \varepsilon, \lambda) = \int FF(t) ds \quad (6.6)$$

with the time-dependent force along the characteristic (ε, λ) defined by

$$F(t) = rg(s, t) + \xi ph(s, t) \quad (6.7)$$

Thus $C(t; \varepsilon, \lambda)$ may be regarded as a force-force time-correlation function. It has the Fourier transform

$$\hat{C}(\omega; \varepsilon, \lambda) = \int_{-\infty}^{\infty} e^{i\omega t} C(t; \varepsilon, \lambda) dt \quad (6.8)$$

From Eq. (5.20) we find correspondingly

$$\begin{aligned} \hat{g}(s, \omega) &= [iQ(\omega) \psi_1'(s) + P(\omega) \psi_2'(s)] e^{i\omega s} \\ \hat{h}(s, \omega) &= \frac{1}{m} [-iQ(\omega) \psi_1(s) - P(\omega) \psi_2(s)] e^{i\omega s} \end{aligned} \quad (6.9)$$

with the functions

$$\begin{aligned} P(\omega) &= 2 \int_0^{\infty} \psi_1(s) U(s) \cos \omega s ds \\ Q(\omega) &= 2 \int_0^{\infty} \psi_2(s) U(s) \sin \omega s ds \end{aligned} \quad (6.10)$$

Here we have used the symmetries

$$\psi_1(s) = \psi_1(-s), \quad \psi_2(s) = -\psi_2(-s), \quad U(s) = U(-s) \quad (6.11)$$

Performing the integration over s in Eq. (6.4) and the Fourier transform in Eq. (6.8) we find

$$\hat{C}(\omega, \varepsilon, \lambda) = P(\omega) S(\omega) - Q(\omega) R(\omega) \quad (6.12)$$

with the functions

$$\begin{aligned} R(\omega) &= 2 \int_0^\infty F \left[r\psi'_1 - \frac{\xi p}{m} \psi_1 \right] \sin \omega s \, ds \\ S(\omega) &= 2 \int_0^\infty F \left[r\psi'_2 - \frac{\xi p}{m} \psi_2 \right] \cos \omega s \, ds \end{aligned} \quad (6.13)$$

Clearly the function $\hat{C}(\omega; \varepsilon, \lambda)$ is real for real ω and is even in ω . Hence the function $C(t, \varepsilon, \lambda)$ is invariant under time-reversal

$$C(t; \varepsilon, \lambda) = C(-t; \varepsilon, \lambda) \quad (6.14)$$

We must take the positive frequency part of the expression given in Eq. (6.12) to find the integrand $\hat{C}_+(\omega; \varepsilon, \lambda)$ in Eq. (6.1). This may be done via the general rule

$$\hat{C}_+(\omega; \varepsilon, \lambda) = \frac{1}{2\pi i} \int_{-\infty}^{\infty} \frac{\hat{C}(\omega'; \varepsilon, \lambda)}{\omega' - \omega} d\omega' \quad (\omega \in I_+) \quad (6.15)$$

for frequency ω in the upper half of the complex ω plane. In our numerical work we shall calculate the positive-frequency part $\hat{C}_+(\omega; \varepsilon)$ from the time-integral in Eq. (6.3) after integration over λ .

A typical time scale of decay of the force-force time-correlation function follows from the definition

$$\tau_M(\varepsilon, \lambda) = \frac{1}{2} \hat{C}(0; \varepsilon, \lambda) / C(0; \varepsilon, \lambda) \quad (6.16)$$

From Eq. (6.12) we find

$$\hat{C}(0; \varepsilon, \lambda) = P(0) S(0) \quad (6.17)$$

since $Q(0) = R(0) = 0$. The denominator in Eq. (6.16) follows from Eq. (6.6)

$$C(0; \varepsilon, \lambda) = \int F^2 \, ds \quad (6.18)$$

Hence the time scale $\tau_M(\varepsilon, \lambda)$ can be evaluated from simple quadratures, once the solutions $\psi_1(s)$ and $\psi_2(s)$ of Eq. (5.21) have been found. From the symmetry under time-reversal, Eq. (6.14), it follows that

$$\hat{C}_+(0; \varepsilon, \lambda) = \hat{C}_-(0; \varepsilon, \lambda) = \frac{1}{2} \hat{C}(0; \varepsilon, \lambda) \quad (6.19)$$

so that the numerator in Eq. (6.16) may be replaced by $\hat{C}_+(0; \varepsilon, \lambda)$.

Finally, we consider the relation to the Lorentz–Boltzmann equation by which the steady state friction coefficient $\zeta_1(0)$ is expressed in terms of the collision cross section.⁽²¹⁾ At a particular value of the energy

$$\zeta_1(0; \varepsilon) = n_i \sqrt{2m\varepsilon} \sigma_1(\varepsilon) \quad (6.20)$$

with $\sigma_1(\varepsilon)$ given by the integral

$$\sigma_1(\varepsilon) = 2\pi \int (1 - \cos \chi) b \, db \quad (6.21)$$

where χ is the scattering angle and b the impact parameter. This must be compared with the expression given by Eq. (5.27) with distribution

$$\varphi_0(\varepsilon) = A\theta(\varepsilon_0 - \varepsilon) \quad (6.22)$$

where the prefactor A follows from the normalization Eq. (4.8). Hence we find

$$A = \frac{3}{4\pi} (2m\varepsilon_0)^{-3/2} \quad (6.23)$$

Substituting into Eq. (5.27) we find at zero frequency for this distribution by comparison with Eq. (6.20)

$$\hat{C}_+(0; \varepsilon) = \frac{4}{\pi} m^2 \varepsilon^2 \sigma_1(\varepsilon) \quad (6.24)$$

The identity follows from Zwanzig's proof⁽¹⁴⁾ that at low density and zero frequency the admittance is given correctly by the Lorentz–Boltzmann equation.

7. EXAMPLE

In order to show that the frequency-dependent friction coefficient $\zeta_1(\omega)$ in Eq. (5.27) can be evaluated without great difficulty we study a specific example. We consider a power law central potential with radial force

$$F(r) = \frac{\kappa}{r^\nu} \quad (7.1)$$

This has the advantage that the solution of the equations of motion has a scaling property,⁽²²⁾ so that our numerical results, which will be given for

one particular value of the energy ε , actually have wider validity. For particular values of the exponent ν the equations of motion can be evaluated explicitly in terms of elliptic integrals,⁽²³⁾ but we shall not follow this route, since it is more useful to develop a numerical method which can be employed for a wide class of spherically symmetric potentials.

For a chosen characteristic with values ε, λ we must first solve Eq. (5.10) to find the distance $r(s)$ as a function of the parameter s . Then Eq. (5.15) yields the effective potential $U(s)$ as a function of the parameter. Next we find the fundamental solutions $\psi_1(s), \psi_2(s)$ of Eq. (5.21) satisfying the conditions (5.22). This allows us to calculate the functions $P(\omega), Q(\omega), R(\omega), S(\omega)$, given by Eqs. (6.10) and (6.13) as a function of positive frequency ω . By symmetry the functions are then also known for negative frequency. The frequency-dependent collision integral $\hat{C}(\omega; \varepsilon, \lambda)$ follows from Eq. (6.12). The calculation must be performed for a discrete, but dense set of values λ . The total frequency-dependent collision integral $\hat{C}(\omega; \varepsilon)$ for fixed energy ε follows by summation over the values λ^2 , as in Eq. (6.1). It is convenient to perform this summation before taking the positive frequency part as in Eq. (6.15). The two procedures can obviously be interchanged.

We have performed an explicit calculation along the above lines for the force law Eq. (7.1) with exponent $\nu=5$, corresponding to Maxwell molecules.⁽¹⁷⁾ We choose atomic units, so that we can put $m=1$, and consider energy $\varepsilon = \frac{1}{2}$ and force constant $\kappa=1$ in these units.

In Fig. 1 we plot the effective potential $U(s)$ as a function of the parameter s for $\varepsilon = \frac{1}{2}, \lambda = \frac{1}{2}$. The potential is well localized and has a range of order unity. In Fig. 2 we plot the total frequency-dependent collision integral $\hat{C}(\omega; \frac{1}{2})$ as a function of frequency. From the collision integral we calculate the time-correlation function $C(t; \varepsilon)$ by means of the Fourier transform

$$C(t; \varepsilon) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \hat{C}(\omega; \varepsilon) e^{-i\omega t} d\omega \quad (7.2)$$

We write the time-correlation function in the form

$$C(t; \varepsilon) = C(0; \varepsilon) \gamma(t; \varepsilon) \quad (7.3)$$

so that $\gamma(t; \varepsilon)$ has the initial value $\gamma(0; \varepsilon) = 1$. From Eq. (6.6) we find for the initial value $C(0; \varepsilon)$

$$C(0; \varepsilon) = \int F^2 ds d\lambda^2 \quad (7.4)$$

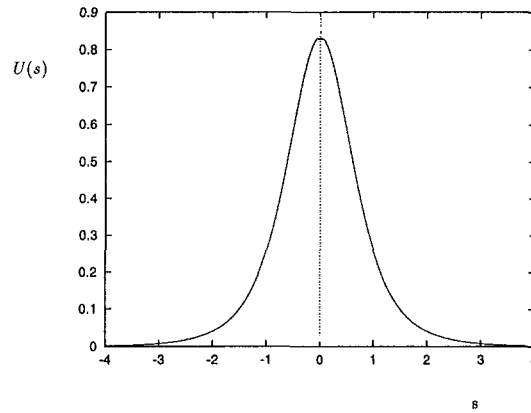


Fig. 1. Plot of the effective potential $U(s)$, defined by Eq. (5.15), for power law force (7.1) with $\nu=5$, as a function of the parameter s for potential strength $\kappa=1$, energy $\varepsilon=\frac{1}{2}$ and angular momentum $\lambda=\frac{1}{2}$ (in atomic units).

In Fig. 3 we plot the integrand

$$2\lambda C(0; \varepsilon, \lambda) = 2\lambda \int F^2 ds \quad (7.5)$$

as a function of λ for $\varepsilon=\frac{1}{2}$. This shows the relative contribution from different values of the angular momentum to the collision integral. For the initial value at energy $\varepsilon=\frac{1}{2}$ we find $C(0; \frac{1}{2})=1.087$. In Fig. 4 we plot the reduced time-correlation function $\gamma(t; \frac{1}{2})$ as a function of t . The positive-

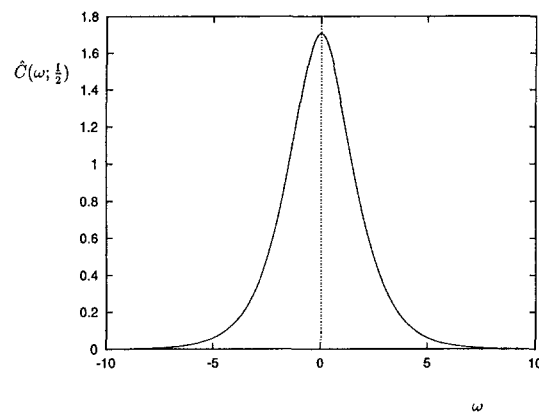


Fig. 2. Plot of the frequency-dependent collision integral $\hat{C}(\omega; \varepsilon)$ at energy $\varepsilon=\frac{1}{2}$ as a function of frequency.

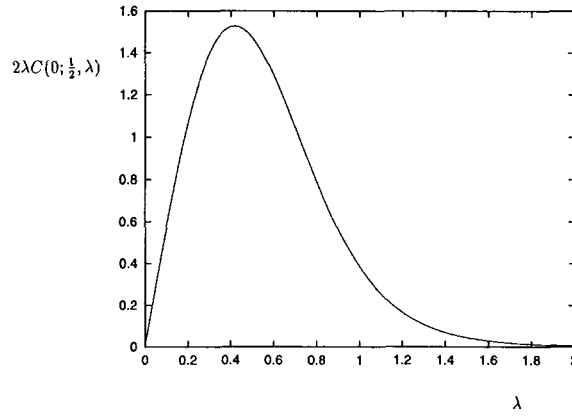


Fig. 3. Plot of the integrand $2\lambda C(0; \varepsilon, \lambda)$, given by Eq. (7.5), for $\varepsilon = \frac{1}{2}$ for the power law force with $\kappa = 1, \nu = 5$ as a function of angular momentum λ .

frequency part $\hat{C}_+(\omega; \varepsilon)$ follows from the one-sided Fourier-integral as in Eq. (6.3). We write the integral in the form

$$\hat{C}_+(\omega; \varepsilon) = C(0; \varepsilon) \tau_M(\varepsilon) \Gamma(-i\omega\tau_M(\varepsilon)) \tag{7.6}$$

with the mean relaxation time $\tau_M(\varepsilon)$ defined by

$$\tau_M(\varepsilon) = \hat{C}_+(0; \varepsilon)/C(0; \varepsilon) \tag{7.7}$$

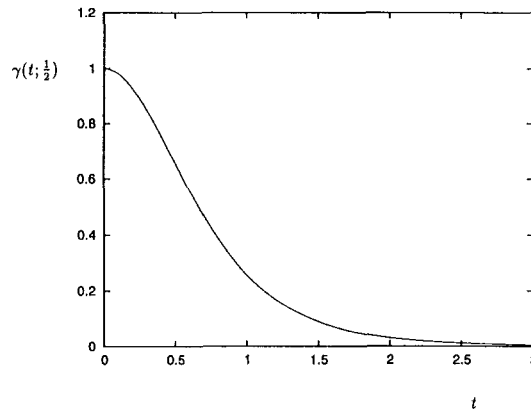


Fig. 4. Plot of the reduced time-correlation function $\gamma(t; \varepsilon)$ at energy $\varepsilon = \frac{1}{2}$ as a function of time t .

so that the function $\Gamma(-i\omega\tau_M(\varepsilon))$ is given by

$$\Gamma(-i\omega\tau_M(\varepsilon)) = \int_0^\infty e^{i\omega t} \gamma(t; \varepsilon) d(t/\tau_M(\varepsilon)) \quad (7.8)$$

The function $\Gamma(z)$ is dimensionless and has the properties

$$\Gamma(0) = 1, \quad \Gamma(z) \approx \frac{1}{z} \quad \text{as } z \rightarrow \infty \quad (7.9)$$

The mean relaxation time $\tau_M(\varepsilon)$ can be calculated from the collision integral $\hat{C}(0; \varepsilon)$, since $\hat{C}_+(0; \varepsilon) = \frac{1}{2}\hat{C}(0; \varepsilon)$. In our units we find at $\varepsilon = \frac{1}{2}$ for the relaxation time $\tau_M(\frac{1}{2}) = 0.786$. Assuming exponential time-dependence for the reduced correlation function $\gamma(t; \varepsilon)$ we find a Lorentzian lineshape for the function $\Gamma(z)$,

$$\Gamma_L(-i\omega\tau_M(\varepsilon)) = \frac{1}{1 - i\omega\tau_M(\varepsilon)} \quad (7.10)$$

centered at $\omega = 0$. In Fig. 5 we plot the real part $\text{Re } \Gamma(-i\omega\tau_M(\frac{1}{2}))$ as a function of $\omega\tau_M(\frac{1}{2})$ and compare with the Lorentzian approximation $\text{Re } \Gamma_L(-i\omega\tau_M(\frac{1}{2}))$. There are clear deviations from the Lorentzian, indicating a more complicated frequency-dependence.

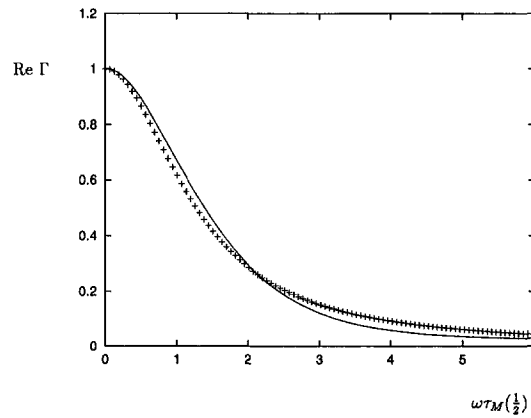


Fig. 5. Plot of $\text{Re } \Gamma(-i\omega\tau_M(\frac{1}{2}))$ as a function of reduced frequency $\omega\tau_M(\frac{1}{2})$ (solid line) compared with its Lorentzian approximation $\text{Re } \Gamma_L(-i\omega\tau_M(\frac{1}{2}))$, as given by Eq. (7.10) (crosses).

For the power law potential (7.1) the cross section defined in Eq. (6.21) is given by⁽¹⁷⁾

$$\sigma_1(\varepsilon) = 2\pi \left(\frac{\kappa}{2\varepsilon} \right)^{2/(\nu-1)} A_1(\nu) \quad (7.11)$$

with a numerical coefficient $A_1(\nu)$. The value $\hat{C}_+(0; \frac{1}{2}) = 0.854$ is in good agreement with the relation (6.24) and the numerical value⁽¹⁷⁾ $A_1(5) = 0.422$.

8. DISCUSSION

The preceding calculation shows that for a Lorentz gas of spherical scatterers the dynamical electrical conductivity can be calculated exactly to lowest order in density. It would be of interest to compare with molecular dynamics simulations at various densities. Such simulations have been performed by Joslin and Egelstaff for Lennard-Jones scatterers.⁽²⁴⁾ These authors focused on the dynamical scattering function. With minor changes our calculation can be made to apply in two dimensions. Baranyai *et al.*⁽²⁵⁾ have performed simulations for a triangular lattice of scatterers with soft potentials. It would be desirable to perform simulations also for a disordered two-dimensional Lorentz gas.

A challenging generalization of our theory would be the approximate inclusion of Coulomb interactions between electrons in terms of a self-consistent Vlasov field. Such a generalization would allow the calculation in classical approximation of photoabsorption by ions immersed in a plasma. The calculation would include single-particle effects and go beyond existing calculations based on a hydrodynamic model, accounting only for collective effects.^(26, 27)

The calculation performed here may suggest methods for the quantum Lorentz gas. As in the classical case, it would be of interest to include Coulomb interactions between electrons in a self-consistent manner. Such a theory would throw light on the intricate quantummechanical problem of photoabsorption in a plasma, with account of bound-bound, bound-free, and free-free transitions, as well as collective effects.

APPENDIX

In this Appendix we show how the Jacobian of the transformation from variables (r, p, ξ) to the variables $(\varepsilon, \lambda^2, s)$ can be evaluated. We denote $\mathbf{x} = (r, p, \xi)$ with components x^i , and $\mathbf{y} = (\varepsilon, \lambda^2, s)$ with components y^α . At

every point in \mathbf{x} -space with $-1 < \xi < 1$ one has the differential relations

$$dy^\alpha = \frac{\partial y^\alpha}{\partial x^i} dx^i, \quad dx^i = \frac{\partial x^i}{\partial y^\alpha} dy^\alpha \quad (\text{A.1})$$

where Einstein's summation convention is implied. This defines the transformation matrices \mathbf{T} and \mathbf{T}^{-1} with elements

$$T_{\alpha i} = \frac{\partial y^\alpha}{\partial x^i}, \quad (T^{-1})_{i\alpha} = \frac{\partial x^i}{\partial y^\alpha} \quad (\text{A.2})$$

The first and second rows of the matrix \mathbf{T} are known. Their elements are

$$\begin{aligned} T_{11} = -F, \quad T_{12} = \frac{p}{m}, \quad T_{13} = 0 \\ T_{21} = 2rp^2(1 - \xi^2), \quad T_{22} = 2r^2p(1 - \xi^2), \quad T_{23} = -2\xi r^2 p^2 \end{aligned} \quad (\text{A.3})$$

as follows from Eq. (5.9). The third column of the inverse matrix \mathbf{T}^{-1} is also known. Its elements are

$$(T^{-1})_{13} = \xi \frac{p}{m}, \quad (T^{-1})_{23} = \xi F, \quad (T^{-1})_{33} = (1 - \xi^2) \left(\frac{p}{mr} + \frac{F}{p} \right) \quad (\text{A.4})$$

as follows from Eq. (5.7). The identity $\mathbf{T}\mathbf{T}^{-1} = \mathbf{I}$ provides equations relating the remaining elements. These can be solved, leaving two elements unknown. In the calculation of the determinant of \mathbf{T} the unknown elements cancel. One finds

$$\det \mathbf{T} = -2r^2 p^2 \quad (\text{A.5})$$

This provides the value of the Jacobian given in Eq. (5.25). Remarkably, the Jacobian is independent of the central force $F(r)$.

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