Debye-Onsager relaxation effect in fully ionized plasmas

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We consider the virial expansion of the inverse conductivity in fully ionized plasmas $\sigma^{-1}=A(T)\ln n+B(T)+C(T)n^{1/2}\ln n+O(n^{1/2})$ and the coefficient $C(T)$, which is related to the Debye-Onsager relaxation effect, is evaluated in the nondegenerate case. In analogy to the Chapman-Enskog treatment of the Boltzmann equation, a moment expansion to evaluate $C(T)$ is presented. Including the two-particle distribution in the generalized linear response theory, the original Onsager result is recovered within a hydrodynamic approximation. Improved results for the relaxation effect beyond this approximation are obtained considering higher moments of the two-particle distribution function. [S1063-651X(98)09408-2]

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I. INTRODUCTION

One enduring problem in theoretical plasma physics is the evaluation of electrical conductivity. In the low-density limit, the conductivity of a fully ionized plasma is given by the Spitzer formula [1]. For dense plasmas, however, experimental values for different plasmas [2–4] show significant deviations from the Spitzer result. This discrepancy requires a quantum statistical approach for transport coefficients in nonideal, strongly coupled Coulomb systems that takes into account many-particle effects such as dynamical screening, dynamical self-energy, and the formation and medium modification of bound states; see [5] for a review.

A particular problem is the inclusion of nonequilibrium two-particle correlations that lead to modifications of electrical conductivity. In the theory of electrolytes, which can also be considered as Coulomb systems, the effect of a nonequilibrium two-particle distribution on the conductivity was obtained by Debye [6] and Onsager [7,8]. Different methods such as kinetic theory [9,10] and linear response theory [11] have been used to evaluate the Debye-Onsager relaxation effect in fully ionized plasmas. However, conflicting results for its contribution to electrical conductivity have been obtained.

In the present paper we calculate the electrical conductivity using a systematic quantum statistical approach. We consider a fully ionized hydrogen plasma consisting of the same number of electrons and protons to ensure charge neutrality. The electrical conductivity $\sigma(n,T)$ is given by the linear relation between the mean electrical current $\langle\mathbf{j}\rangle$ and the electrical field $\mathbf{E}$,

$$\langle\mathbf{j}\rangle=\left\langle\frac{1}{\Omega}\sum_{c,i}\frac{e_c}{m_c}\mathbf{p}_c^i\right\rangle=\sigma\mathbf{E}. \tag{1}$$

The index c identifies the species (for the electron $m_e=m$ and $e_e=-e$ and for the proton $m_p=M$ and $e_p=e$) and Ω is the system volume. In plasma physics, it is customary to introduce the dimensionless parameters

$$\Gamma=\frac{e^2}{4\pi\epsilon_0 k_BT}\left(\frac{4\pi n}{3}\right)^{1/3},\quad \Theta=\frac{2m_ek_BT}{\hbar^2}(3\pi^2n)^{-2/3}. \tag{2}$$

$\Gamma(n,T)$ describes the ratio between the mean potential energy and the kinetic energy and $\Theta(n,T)$ denotes the degree of degeneracy of an ideal electron gas. Using these quantities, we can express the electrical conductivity as

$$\sigma(n,T)=\frac{(k_BT)^{3/2}(4\pi\epsilon_0)^2}{m_e^{1/2}e^2}\sigma^*, \tag{3}$$

with a universal function $\sigma^*(\Gamma,\Theta)$ depending on only the characteristic plasma parameters. In the low-density limit ($\Gamma\ll 1$) the electrical conductivity of a fully ionized ideal plasma with statically screened interaction is given by the famous Spitzer formula [1]

$$\sigma^*_{\rm Sp}=0.591\left[\frac{1}{2}\ln\left(\frac{3}{2}\Gamma^{-3}\right)\right]^{-1}. \tag{4}$$

For nonideal plasmas, the effects of degeneracy and many-particle corrections have to be included. Interpolation formulas for the electrical conductivity that are valid in a wide region of temperatures and densities are given in [12–14] and can be both compared with experimental data [2–4] and Monte Carlo simulations [15]. Such interpolation formulas reproduce rigorous expressions for the conductivity such as the low-density limit as well as results obtained from the Ziman formula.

Our aim is the evaluation of electrical conductivity in the low-density limit, where rigorous results can be derived. To find a microscopic expression for the electrical conductivity we use the generalized Zubarev method of linear response theory, which can be found in detail, e.g., in [16]. This method can immediately be generalized to include nonideality effects such as degeneracy, the formation of bound states, and the ion structure factor, as shown in [17]. Using this approach, transport coefficients are expressed through equi-

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librium correlation functions and a systematical treatment of these many-particle effects is possible. Results obtained from kinetic theory in a perturbative treatment are reproduced in this method without partial summations, as it would be necessary in the Kubo approach.

A virial expansion for the electrical conductivity in the low-density, nondegenerate limit was given in [11],

$$\sigma^{-1}(n,T)=A(T)\ln n+B(T)+C(T)n^{1/2}\ln n+\cdots, \quad (5)$$

where $C(T)$ is related to the Debye-Onsager relaxation effect, which is known from the theory of electrolytes [7,8]. It describes the partial compensation of the electric field acting on a charged particle by the formation of an asymmetric screening cloud. Klimontovich and Ebeling [18] found the original Onsager result for the relaxation effect in *weakly* ionized plasmas, where the assumption of a local Maxwellian distribution is well founded. The treatment of the *fully* ionized plasma has been considered in [11], where also a local Maxwellian distribution is assumed. Again the Onsager result for $C(T)$ is found. Results for the coefficient $C(T)$, which are in disagreement with the Onsager result, are obtained in kinetic theory [9,10] when a pure one-particle picture is used. We show in our approach that nonequilibrium two-particle correlations have to be included to reproduce the Onsager result. Furthermore we find that $C(T)$ is modified in fully ionized plasmas if the assumption of a local Maxwellian distribution is dropped. In Sec. II we express the electrical conductivity in terms of equilibrium correlation functions. In analogy to the Chapman-Enskog approach in kinetic theory, a moment expansion of the single-particle and two-particle distribution function is presented in Sec. III. Starting from the lowest moment approximation for the single-particle as well as for the two-particle distribution function in Sec. IV, we consider in Sec. V the influence of higher moments of the single-particle distribution function. The inclusion of higher moments of the two-particle distribution function is presented in Sec. VI. The results are finally discussed in Sec. VII.

II. CONDUCTIVITY WITHIN GENERALIZED LINEAR RESPONSE THEORY

The system Hamiltonian of the fully ionized hydrogen plasma

$$H_S=\sum_{c,k} E_c(k)a_c^\dagger(k)a_c(k) + \frac{1}{2}\sum_{c,d,k,p,q} V_{cd}(q)a_c^\dagger(k-q)a_d^\dagger(p+q)a_d(p)a_c(k) \quad (6)$$

contains the kinetic energy $E_c(k)=\hbar^2k^2/2m_c$ and the Coulomb interaction $V_{cd}(q)=e_ce_d/\epsilon_0\Omega q^2$. The index k denotes the single-particle variable momentum and spin. The system is subjected to a static electric field $\mathbf{E}=E\hat{\mathbf{e}}_z$ and the total Hamiltonian reads

$$H_{\mathrm{tot}}=H_S-E\sum_{c,i} e_cr_{c,z}^i=H_S-ER. \quad (7)$$

A quantum statistical approach for the electrical conductivity σ (1) requires the knowledge of the statistical operator $\rho(t)$ to evaluate the average $\langle j\rangle=\mathrm{Tr}\{\rho(t)j\}$. The nonequilibrium statistical operator $\rho(t)$ is obtained within the Zubarev approach [20] from the solution of a modified Liouville–von Neumann equation

$$\frac{\partial\rho(t)}{\partial t}-\frac{1}{i\hbar}[H_{\mathrm{tot}},\rho(t)]=-\epsilon[\rho(t)-\rho_{\mathrm{rel}}(t)], \quad (8)$$

where the boundary condition, the weakening of initial correlations, is included in the form of a source term. The limit $\epsilon\rightarrow 0+$ has to be taken after the thermodynamic limit. The relevant statistical operator $\rho_{\mathrm{rel}}=Z_{\mathrm{rel}}^{-1}\exp(-\Sigma_n\lambda_nB_n)$ follows from the principle of maximizing the information entropy subject to constraints of given mean values of a set of relevant observables $\{B_n\}$. Starting from the solution of Eq. (8), it is possible to derive a generalized Boltzmann equation, which can directly be compared with expressions from standard kinetic theory if the set $\{B_n\}$ is given by the single-particle occupation numbers [20].

Using a finite set of relevant observables $\{B_n\}$ to characterize the nonequilibrium state, the electrical conductivity is found from linear response theory [5,17,20]

$$\sigma=-\frac{\beta}{\Omega}\frac{1}{|D[B_{n'};B_n]|}\begin{vmatrix}0 & N[B_n]\\ Q[B_n] & D[B_{n'};B_n]\end{vmatrix}. \quad (9)$$

This expression relates the electrical conductivity to equilibrium correlation functions, which can be considered as a special form of the fluctuation-dissipation theorem. The elements of the determinants are

$$N[B_n]=(\dot{R};B_n),$$
$$Q[B_n]=(\dot{R};B_n)+\langle\dot{R};\dot{B}_n\rangle, \quad (10)$$
$$D[B_{n'};B_n]=(B_{n'};\dot{B}_n)+\langle\dot{B}_{n'};\dot{B}_n\rangle,$$

with $\dot{R}=-e[(m+M)/mM]P$. In Eq. (10) we use Kubo's scalar product [19]

$$(A;B)=\frac{1}{\beta}\int_0^\beta d\tau\ \mathrm{Tr}[\rho_0A(-i\hbar\tau)B], \quad (11)$$

the equilibrium correlation function

$$\langle A;B\rangle=\int_{-\infty}^0 dt\ e^{\epsilon t}(A(t);B), \quad (12)$$

and the equilibrium statistical operator $\rho_0=Z_0^{-1}\exp[-\beta(H_S-\Sigma_c\mu_cN_c)]$. In the Heisenberg picture the time evolution is $A(t)=\exp(iH_St/\hbar)A\exp(-iH_St/\hbar)$ and $\dot{A}=(i/\hbar)[H_S,A]$.

The evaluation of the equilibrium correlation function can be performed using the formalism of thermodynamic Green's functions [5]. This allows for a perturbative expansion with respect to the interaction. Only the lowest orders are considered in the present work. A very important question is the choice of the set of relevant observables $\{B_n\}$, which will be discussed in the following section. In connection with expression (9) we note that this expression corresponds to the Chapman-Enskog or Grad approach of kinetic theory if an appropriate set of relevant observables is used; see [5,11].

III. CHOICE OF RELEVANT OBSERVABLES

For small densities it is sufficient to take fluctuations of the single-particle occupation number $\delta n_c(k)=a_c^\dagger(k)a_c(k)-\langle a_c^\dagger(k)a_c(k)\rangle_0$ as relevant observables B_n [11]. The mean value $\langle \delta n_k\rangle = f_k - f_k^0$ describes the deviation of the single-particle distribution function f_k from its equilibrium value f_k^0. The response equations derived from Eq. (8) are equivalent to the Boltzmann equation for the single-particle distribution function [20], where the collision integral is expressed in terms of correlation functions. The evaluation of the corresponding correlation function (10) in the ladder approximation and considering screening effects has been performed in [11], where results for the coefficients $A(T)$ and $B(T)$ in Eq. (5) were obtained. Furthermore, it was shown that no contributions to the coefficient $C(T)$ occur within the single-particle description.

In order to evaluate the electrical conductivity to order $n^{1/2}\ln n$ with respect to the density we have to include the two-particle distribution function. In the homogeneous case this means that we have to enlarge the set of relevant observables B_n by inclusion of the two-particle observables

$$\delta n_{cd}(pkq)=a_c^\dagger(k-q/2)a_d^\dagger(p+q/2)a_d(p-q/2)a_c(k+q/2) \tag{13}$$

for $q\neq 0$. Hence the response equations that lead to Eq. (9) have the form of a coupled system of equations for the single- and two-particle distribution functions.

The full solution of the response equations for the single- and two-particle distribution functions is equivalent to the solution of a coupled system of integral equations if we consider the indices k and kpq, respectively, as continuous variables. We will find an approximate solution of the response equations by taking into account only a finite number of moments of the distribution functions. In the adiabatic limit we consider only the electron distribution. In particular, instead of solving the response equations for the single-particle distribution we consider a finite number of moments

$$P_n=\sum_k \hbar k_z\left[\beta\frac{\hbar^2k^2}{2m_e}\right]^{(n/2)} a_e^\dagger(k)a_e(k). \tag{14}$$

This means that instead of the single-particle occupation number only a finite number of moments P_n are included in the set of relevant observables B_n. In the simplest case only the total momentum of the electron system P_0 is considered as a relevant observable. This leads to the well known force-force correlation approach to the conductivity.

For a sufficient high number of moments, the single-particle distribution function is approximated well and the conductivity is obtained by the algebraic solution of the system of response equations. This procedure corresponds to the Grad or the Chapman-Enskog approach for solving the linearized Boltzmann equation. As it is well known from the Kohler variational principle [21], the inclusion of higher moments will increase the conductivity. It has been shown [11] that the inclusion of a finite number of low-order moments results in a converging expression for the term A in the virial expansion (5).

A similar approach can be applied for the treatment of the two-particle distribution function. Obviously the dependence on three momenta $(\mathbf{k},\mathbf{p},\mathbf{q})$ requires a larger variety of moments. Conserving the dependence on the relative distance $\mathbf{r}$ in the form of the Fourier transform $\mathbf{q}$, moments with respect to the momenta $\mathbf{k}$ and $\mathbf{p}$ of species c and d, respectively, are introduced

$$\delta n_{c,d}^{m,m'}(q)=\sum_{k,p} f_{c,d}^{m,m'}(\mathbf{k},\mathbf{p},\mathbf{q})a_c^\dagger(k-q/2) \times a_d^\dagger(p+q/2)a_d(p-q/2)a_c(k+q/2). \tag{15}$$

The functions $f_{c,d}^{m,m'}(\mathbf{k},\mathbf{p},\mathbf{q})$, which will be discussed in detail in Secs. IV and V, are of mth order in $\mathbf{k}$ and of m'th order in $\mathbf{p}$. In this way, the moments $\delta n_{c,d}^{m,m'}(q)$ refer to the momentum distributions of both particles of species c and d.

We will proceed as follows. In the first step, we consider only the moment $\delta n_{c,d}^{0,0}(q)$ in combination with the moment P_0 [Eq. (14)] of the single-particle distribution to demonstrate the formalism. We will extend the number of relevant observables by including further moments P_n. In the second step we will confine ourselves to P_0,P_2 and will improve the two-particle distribution by including the second moments.

IV. ONSAGER'S RESULT

In the first step, we rederive Onsager's result using lowest moments only. As argued by Onsager [7], the two-particle correlation function of the electron-electron and proton-proton density [$\delta n_{ee}^{0,0}(q)$ and $\delta n_{pp}^{0,0}(q)$] does not affect the relaxation effect in the linear regime because of symmetry properties. Therefore, the set of relevant observables $\{B_n\}$ is confined to the first moments of the single-particle and two-particle correlation function

$$P_0=\sum_k \hbar k_z a_e^\dagger(k)a_e^\dagger(k),$$

$$\delta n_{ep}^{0,0}(q)=\sum_{k,p} a_e^\dagger(k-q/2)a_p^\dagger(p+q/2) \times a_p(p-q/2)a_e(k+q/2). \tag{16}$$

This set corresponds to a hydrodynamic, local Maxwellian approximation of the distribution functions. For the correlation functions in expression (9), we consider thermodynamic Green's functions. Using a diagrammatic representation, they can be treated within perturbation theory and a systematical inclusion of many-particle effects is possible. In the present work the correlation functions are evaluated in the Born approximation with a statical screened Coulomb interaction $V_S(q)=e^2/\epsilon_0\Omega(q^2+\kappa^2)$. Here $\kappa^2=2n\beta e^2/\epsilon_0$ is the screening parameter for a two-component plasma in the nondegenerate limit. A more detailed treatment of the collision term (dynamic screening and ladder summations) would result in corrections of $O(n^{1/2})$ in the density expansion (5).

This was shown for the single-particle distribution function in [11]. We find with the two-particle distribution $n_2(q) = N^2\beta e^2/\epsilon_0\Omega(q^2+\kappa^2)$ for the matrix elements

$$N[P_0]=Q[P_0]=-\frac{eN}{\beta},$$

$$N[\delta n_{ep}^{0,0}(q)]=0,$$

$$Q[\delta n_{ep}^{0,0}(q)]=\frac{ie}{\epsilon\beta}\frac{M+m}{Mm}q_z n_2(q),$$

$$D[P_0;P_0]=\frac{4\sqrt{2\pi}}{3}\frac{N^2}{\Omega}\sqrt{m\beta}\frac{e^4}{(4\pi\epsilon_0)^2}L_{\rm BH}, \tag{17}$$

$$D[P_0;\delta n_{ep}^{0,0}(q)]=-D[\delta n_{ep}^{0,0}(q),P_0]=\frac{i}{\beta}q_z n_2(q),$$

$$D[\delta n_{ep}^{0,0}(q');\delta n_{ep}^{0,0}(q)]=\frac{1}{\epsilon\beta}\frac{M+m}{Mm}[N^2-Nn_2(q)]q^2\delta_{q',q}.$$

Here $L_{\rm BH}=\frac{1}{2}\int_0^\infty x(x+\kappa^2)^{-2}\exp(-\beta\hbar^2x/8m)dx$ is the Brooks-Herring–type Coulomb logarithm. For a detailed evaluation of the correlation functions see the Appendix.

We can now calculate the determinants in Eq. (9). Note that the continuous variable q should be replaced by a finite-set q_i of discrete wave numbers to have a finite-dimensional matrix and the transition to an infinite number of q can be performed. The ratio of two finite matrices, which are constructed with indices P_0 and $\delta n_{ep}^{0,0}(q_i)$, can be evaluated due to the diagonality in $\delta n_{ep}^{0,0}(q_i)$. We are able to rearrange also the numerator determinant into a diagonal structure with respect to the indices q_i. This can be done successively by standard manipulations producing zeros instead of $Q[\delta n_{ep}^{(0,0)}(q_i)]$. The diagonal part connected with the q_i cancels and we find for the electrical conductivity

$$\sigma=-\frac{e^2N}{\Omega}\frac{1}{D[P_0;P_0]}\left[\frac{N}{\beta}+\sum_q\frac{[iq^zn_2(q)]^2}{[N^2-Nn_2(q)]\beta q^2}\right]$$
$$=\frac{e^2N^2}{\beta\Omega}\frac{1}{D[P_0;P_0]}\left[1-\frac{1}{3(2+\sqrt{2})}\beta\kappa\frac{e^2}{4\pi\epsilon_0}\right], \tag{18}$$

or using the reduced conductivity of Eq. (3)

$$\sigma^*=\frac{3}{4\sqrt{2\pi}}L_{\rm BH}^{-1}\left[1-\frac{1}{3(2+\sqrt{2})}\beta\kappa\frac{e^2}{4\pi\epsilon_0}\right]. \tag{19}$$

This result was already found in [11] within linear response theory. The contribution of the relaxation effect is identical to the original Debye-Onsager result [7]

$$C_{\rm DO}=\frac{1}{3(2+\sqrt{2})}\beta\kappa\frac{e^2}{4\pi\epsilon_0}, \tag{20}$$

obtained in the theory of electrolytes.

However, the assumption of a local Maxwellian distribution cannot be expected in fully ionized plasmas. This will be demonstrated in the following section. Using the Born approximation, the correct low-density expression (5) can be obtained by including higher moments of the single-particle and two-particle distribution functions.

V. VARIATION OF THE SINGLE-PARTICLE DISTRIBUTION FUNCTION

In the first step we extend the set of relevant observables by including higher moments of the single-particle distribution function P_n [Eq. (14)] for the electrons and the lowest moment of the pair-distribution function. Correspondingly, the determinants occurring in Eq. (9) contain additional matrix elements compared to the simplest case, discussed in the preceding section, in particular

$$N[P_n]=Q[P_n]=(\dot{R};P_n)=-\frac{\Gamma\left(\frac{5+n}{2}\right)}{\Gamma\left(\frac{5}{2}\right)}\frac{eN}{\beta}. \tag{21}$$

The additional term $\langle\dot{R};\dot{P}_n\rangle$ occurring in $Q[P_n]$ vanishes in the Born approximation. This results from symmetry properties of the distribution function; see also (A15). Furthermore, we have to calculate the matrix elements $D[P_{n'};P_n] = \langle\dot{P}_{n'}(\epsilon);\dot{P}_n\rangle$. For the first moment P_0 the result was already given by

$$\langle\dot{P}_0(\epsilon);\dot{P}_0\rangle=\frac{4}{3}(2\pi)^{1/2}N^2\frac{1}{\Omega}m^{1/2}\beta^{1/2}\frac{e^4}{(4\pi\epsilon_0)^2}L_{\rm BH}. \tag{22}$$

For higher-order moments P_n, according to $\dot{P}_n = (i/\hbar)[H_S,P_n]$, also the electron-electron interaction contributes to $\langle\dot{P}_{n'}(\epsilon);\dot{P}_n\rangle$ and we obtain with $a_{n'n} = \langle\dot{P}_{2n'}(\epsilon);\dot{P}_{2n}\rangle/\langle\dot{P}_0(\epsilon);\dot{P}_0\rangle$

$$a_{10}=a_{01}=1,\ a_{20}=a_{02}=2,\ a_{11}=2+\sqrt{2},$$
$$a_{21}=a_{12}=6+11/\sqrt{2},\ \ a_{22}=24+157/\sqrt{8}, \tag{23}$$

which can be found, e.g., in [11]. A correlation function between the moments of the single-particle distribution and the pair-distribution function appears as a new term

$$D[\delta n_{ep}^{0,0}(q);P_n]=-D[\delta n_{ep}^{0,0}(q);P_n]=(\delta\dot{n}_{ep}^{0,0}(q);P_n). \tag{24}$$

It can be evaluated in the nondegenerate case and we find [see Eq. (A12)]

$$(\delta\dot{n}_{ep}^{0,0}(q);P_n)=-\frac{i}{\hbar}\left[\frac{\beta\hbar^2}{2m}\right]\left(\frac{3+n}{n}\right)$$
$$\times\sum_{k,p}q_z\langle a_e^\dagger(k-q/2)a_p^\dagger(p+q/2)$$
$$\times a_p(p-q/2)a_e(k+q/2)\rangle$$
$$=-\frac{\Gamma\left(\frac{5+m}{2}\right)}{\beta\Gamma\left(\frac{5}{2}\right)}iq_zn_2(q), \tag{25}$$

TABLE I. The prefactor of Eq. (28) using different orders of single-particle moments P_n is compared with the Spitzer value.

Order of single-particle moments P_n	Prefactor in Eq. (28)
P_0	0.299
P_0,P_2	0.578
P_0,P_2,P_4	0.583
Spitzer	0.591

The correlation function $\langle \delta\dot{n}_{ep}^{0,0}(q);\dot{P}_n\rangle$ vanishes in the Born approximation.

Having the full set of correlation functions at our disposal, the determinants occurring in Eq. (9) have to be evaluated. The procedure explained in connection with Eq. (18) can be applied again, yielding

$$\sigma=-\frac{\beta}{\Omega}\frac{1}{|D[P_n;P_{n'}]|}\begin{vmatrix}0 & N[P_n]\\ \overline{N}[P_{n'}] & D[P_n;P_{n'}]\end{vmatrix}, \quad (26)$$

with

$$\overline{N}[P_{n'}]=-\frac{\Gamma\left(\frac{5+n'}{2}\right)}{\Gamma\left(\frac{5}{2}\right)}\frac{eN}{\beta}\left[1-\frac{1}{3(2+\sqrt{2})}\frac{\beta\kappa e^2}{4\pi\epsilon_0}\right]. \quad (27)$$

Taking the first three moments $\{P_0,P_2,P_4\}$ of the single-particle distribution function, we obtain for the conductivity

$$\sigma^*=0.583L_{\mathrm{BH}}^{-1}\left[1-\frac{1}{3(2+\sqrt{2})}\frac{\beta e^2\kappa}{4\pi\epsilon_0}\right]. \quad (28)$$

In Table I we compare the Spitzer value with the prefactors of Eq. (28), which were found from the successive inclusion of higher single-particle moments P_n; see also [17]. The Spitzer result for the electrical conductivity of an ideal plasma can be derived only if we drop the assumption of a local Maxwellian distribution for the single-particle distribution. The relaxation effect is not affected by the improved single-particle description. We find that the Debye-Onsager correction term to the conductivity is obtained if the two-particle distribution function is considered in hydrodynamic approximation, i.e., only $\delta n_{ep}^{0,0}(q)$ is taken into account.

VI. VARIATION OF THE PAIR-DISTRIBUTION FUNCTION

To derive a rigorous result for the coefficient $C(T)$ in Eq. (5), we want to investigate to what extent higher moments of the pair-distribution function influence the Debye-Onsager correction factor. The inclusion of the second moment of the single-particle distribution (the energy current) yields the main contribution to the coefficients A and C in the virial expansion of the electrical conductivity; see Table I. Therefore, we will also focus on the second moments of the pair-distribution function. The set of relevant observables now consists of the variables P_0,P_2, and $\delta n_{ep}^{m,n}(q)$ with $\{m,n\leqslant 2\}$. In general, according to Eq. (15), the two-particle density $\delta n_{ep}^{m,n}(q)$ contains the function $f_{e,p}(\mathbf{k},\mathbf{p},\mathbf{q})$ depending not only on the momentum $\mathbf{q}$ but also on the electron momentum $\mathbf{p}$ and the proton momentum $\mathbf{k}$. In the adiabatic limit $(m\ll M)$ the proton momenta can be neglected. Then we define the moments of the pair distribution function up to second order, writing

$$\delta n_{ep}^{m,0}(q)=\sum_{k,p} f_e^{m,0}(\mathbf{k})a_e^\dagger(k-q/2)a_p^\dagger(p+q/2)\times a_p(p-q/2)a_e(k+q/2) \quad (29)$$

with

$$f_e^{m,0}(\mathbf{k})=\begin{cases}1, & m=0\\ k,\mathbf{kq}, & m=1\\ k^2,\mathbf{k}(\mathbf{kq}),(\mathbf{kq}),^2 & m=2.\end{cases}$$

If $\delta n_{ep}^{0,0}(q)$ is included in the set of relevant observables it is not necessary to consider $\delta\dot{n}_{ep}^{0,0}(q)$ too, as shown in [23]. Thus we can restrict ourselves to $\delta n_{ep}^{0,0}(q)$ and

$$\delta n_{ep}^{1,0}(q)=\sum_{k,p}\mathbf{k}a_e^\dagger(k-q/2)a_p^\dagger(p+q/2)a_p(p-q/2)\times a_e(k+q/2),$$

$$\delta n_{ep}^{2,0}(q)=\sum_{k,p}k^2a_e^\dagger(k-q/2)a_p^\dagger(p+q/2)a_p(p-q/2)\times a_e(k+q/2) \quad (30)$$

for the moments of the pair-distribution function. The correlation functions containing $\delta n_{ep}^{1,0}(q)$ as a relevant observable lead to contributions that are of higher order in the density expansion (5) than $O(n^{1/2}\ln n)$ and thus do not affect the Debye-Onsager relaxation effect. Therefore, we consider only $\delta n_{ep}^{m,0}(q)$ with $m=\{0,2\}$. New contributions to the determinants in Eq. (9) are the matrix elements

$$N[\delta n_{ep}^{m,0}(q)]=0,$$

$$Q[\delta n_{ep}^{m,0}(q)]=-\frac{e}{m}(P;\delta\dot{n}_{ep}^{m,0}(q))=\frac{2\Gamma\left(\frac{3+m}{2}\right)}{\epsilon\beta\sqrt{\pi}}iq_z\frac{e}{m}n_2(q),$$

$$D[\delta n_{ep}^{m,0}(q);P_n]=-D[P_n;\delta n_{ep}^{m,0}(q)]=(\delta\dot{n}_{ep}^{m,0}(q);P_n)=-2\left(\frac{3+n}{3}\right)\frac{\Gamma\left(\frac{3+m+n}{2}\right)}{\beta\sqrt{\pi}}iq_zn_2(q). \quad (31)$$

The last two results can be found in analogy to Eq. (25). Furthermore, we have to evaluate the collision term $D[\delta n_{ep}^{m',0}(q');\delta n_{ep}^{m,0}(q)]$. The correlation functions $\langle \delta\dot{n}_{ep}^{m',0}(q');\delta\ddot{n}_{ep}^{m,0}(q)\rangle$ are of $O(q^3/\epsilon)$ and can therefore be omitted in the quasiclassical limit $(q\to 0)$. Consequently, the contributions to the collision terms are

$$\begin{aligned}
D[\delta n_{ep}^{m',0}(q');\delta n_{ep}^{m,0}(q)] &= \frac{1}{\epsilon}(\delta\dot{n}_{ep}^{m',0}(q');\delta\dot{n}_{ep}^{m,0}(q))\\
&= \left(\frac{m+m'+3}{3}\right)\frac{q^2\delta_{q,q'}}{\epsilon m\beta}\sum_{k,p} k^{m+m'}\langle a_e^\dagger(k)a_p^\dagger(p)a_p(p)a_e(l)\rangle\\
&\quad+\left(\frac{m+m'+3}{3}\right)\frac{q^2\delta_{q,q'}}{\epsilon m\beta}\sum_{k,p,h} k^{m+m'}\langle a_e^\dagger(k)a_p^\dagger(p-q/2)a_p^\dagger(h+q/2)a_p(h-q/2)a_p(p+q/2)a_e(k)\rangle\\
&= \frac{\Gamma\left(\frac{5+m+m'}{2}\right)}{\Gamma\left(\frac{5}{2}\right)}\frac{q^2\delta_{q,q'}}{\epsilon m\beta}[N^2-Nn_2(q)]. \qquad (32)
\end{aligned}$$

Again we evaluate the determinants in Eq. (9) and find for the electrical conductivity

$$\sigma^* = 0.578L_{\rm BH}^{-1}\left[1-\frac{1}{3(2+\sqrt{2})}(1.078)\frac{\beta e^2\kappa}{4\pi\epsilon_0}\right]. \qquad (33)$$

This expression contains a numerical correction to the Onsager result $C_{\rm DO}(T)$. In fully ionized plasmas we improve the Onsager correction term to the conductivity by including the second moment of the pair-distribution function.

VII. DISCUSSION

In this paper a systematic treatment of the Debye-Onsager relaxation effect in fully ionized plasmas is performed. We treat the nonideality corrections to the electrical conductivity in the low-density limit. A virial expansion of the inverse conductivity is found in the nondegenerate case $(\Theta \gg 1)$ according to

$$\sigma^{-1} = a(\ln\Gamma^{-3/2}+b+c\Gamma^{3/2}\ln\Gamma^{-3/2}). \qquad (34)$$

This virial expansion coincides with expression (5) in the nondegenerate case where the density dependence is condensed in the plasma parameter $\Gamma \propto n^{1/3}/T$ and the additional dependence on the temperature is contained in the prefactor a, which yields the dimension of the conductivity. The dimensionless parameters b and c cannot depend on temperature or density. More explicitly, for hydrogen plasmas we have

$$\begin{aligned}
\sigma &= 0.591\frac{(4\pi\epsilon_0)^2(k_BT)^{3/2}}{e^2m^{1/2}}\\
&\quad\times\left[\ln\Gamma^{-3/2}+1.124+\frac{1.078}{\sqrt{6}+\sqrt{3}}\Gamma^{3/2}\ln\Gamma^{-3/2}\right]^{-1}\\
&= 1.530\times 10^{-2}T^{3/2}[\ln\Gamma^{-3/2}+1.124\\
&\quad+0.258\Gamma^{3/2}\ln\Gamma^{-3/2}]^{-1}\ \ (\Omega\ \mathrm{m\ K}^{3/2})^{-1}. \qquad (35)
\end{aligned}$$

The main result of the present paper is the evaluation of the coefficient c in Eq. (34), which is related to the Debye-Onsager relaxation effect. The results derived in our quantum statistical approach are compared with other results found in the literature in Table II.

Within the kinetic theory a different value from the Onsager result for c is obtained if only the medium modification of the single-particle propagator is considered. We have shown that nonequilibrium correlations are responsible for the relaxation effect. Therefore, it is expected that, in contrast to recent claims [9,10], the correct Debye-Onsager result can be derived in a kinetic theory only if the two-particle distribution function is included.

The original Onsager result is reproduced if a Maxwellian form for the momentum distribution of the two-particle distribution function is assumed. The hydrodynamic approximation, where also the single-particle distribution function has a Maxwellian form, is justified for electrolytes or weakly ionized plasmas. We have shown that a modification of the Onsager value is obtained for the fully ionized plasma. The exact value of the coefficient can be approximated successively by including higher moments. The value, given in Table II, corresponds to the inclusion of the second moments of the nonequilibrium two-particle momentum distribution.

TABLE II. Comparison of the coefficient c in the virial expansion (34) for the electrical conductivity obtained from different methods.

Methods	c
kinetic theory[a]	0.408
hydrodynamic approximation[b]	0.239
linear response theory	
lowest moments[c]	0.239
higher moments[d]	0.258

[a]References [9,10].
[b]Reference [18].
[c]Reference [11].
[d]Present work.

TABLE III. The resulting values of the electrical conductivity including the relaxation effect (35) are compared with both experimental data and the Spitzer result.

Plasma	T (10^3 K)	n_e (10^{25} m^{-3})	Γ	Θ	$\sigma_{\rm expt}$ (10^2 Ω^{-1} m^{-1})	σ Eq. (36)	$\sigma_{\rm Sp}$
Ar[a]	22.2	2.8	0.368	56.9	190	187	193
	20.3	5.5	0.505	33.2	155	197	206
	19.3	8.1	0.604	24.4	170	208	218
	19.0	14	0.736	16.7	255	242	253
	17.8	17	0.838	13.7	245	252	262
Xe[a]	30.1	25	0.564	17.9	450	385	403
Ne[a]	19.8	1.1	0.303	94.6	130	142	146
	19.6	1.9	0.367	65.0	165	155	160
air[a]	11.0	0.13	0.267	218	60	56	57
Ar[b]	16.4	0.06	0.128	551	83	76	76
		0.1	0.165	385	79	83	84
		0.13	0.18	324	76	86	87
		0.15	0.19	291	64	88	89
Xe[b]	12.4	0.06	0.185	403	46	57	58
		0.12	0.234	252	41	63	64
	12.6	0.07	0.192	371	48	59	60
		0.14	0.239	238	44	65	66
H[c]	15.4	0.1	0.175	364	62	77	78
	18.7	0.15	0.165	337	91	101	102
	21.5	0.25	0.170	276	114	126	128

[a]Reference [2].
[b]Reference [3].
[c]Reference [4].

The inclusion of the higher moments of the distribution function leads to a further reduction of the conductivity compared to the Onsager result.

The relaxation effect describes the nonideality correction to the conductivity in the low-density case. The applicability of the conductivity formula (35) is restricted to the region $\Gamma < 1$. Corrections are small in this region. To demonstrate the influence of the Debye-Onsager relaxation effect on the electrical conductivity we compare in Table III different experimental values of the conductivity with the Spitzer result and with the result of Eq. (35), where the relaxation effect is taken into account. On the average, the Spitzer values of the conductivity are larger than the corresponding experimental values. The virial coefficient c in Eq. (34) yields a reduction of the Spitzer conductivity. The resulting values for the conductivity, shown in Table III, lie within the experimental error bars of about 30%.

In order to describe the electrical conductivity in a large region of temperature and density further effects have to be included. In particular, effects of degeneracy and the formation of bound states contribute in higher orders of the density expansion. Experimental results for the electrical conductivity of nonideal plasmas should be compared with interpolation formulas that are valid in an appropriate region of density and temperature. The low-density expansion of Eq. (35) may serve as an input to construct an expression for the conductivity for nonideal plasmas. The derivation of interpolation formulas for the electrical conductivity, which are valid also in the strongly coupled case $\Gamma \geqslant 1$ and in the Born limit $\Gamma^2\Theta \ll 1$, should contain Eq. (35) as a limiting case. We plan to do this in the future.

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APPENDIX: CORRELATION FUNCTIONS

In this appendix we outline the calculation of the correlation function. We use relations that result from a partial integration of the equilibrium correlation function (12)

$$\langle A;B\rangle = \frac{1}{\epsilon}(A;B) + \frac{1}{\epsilon}\langle A;B\rangle, \tag{A1}$$

from Kubo's identity

$$(\dot{A};B) = \frac{i}{\hbar\beta}\langle [B,A]\rangle, \tag{A2}$$

and from the definition of the scalar product (11)

$$(\dot{A};B) = -(A;\dot{B}). \tag{A3}$$

Starting with the matrix element $N[P_0]$, we find with Eq. (A2)

$$N[P_0] = (\dot{R};P_0) = \frac{i}{\hbar\beta}\langle [R,P_0]\rangle = -\frac{eN}{\beta}. \tag{A4}$$

For the evaluation of $N[\delta n_{ep}^{0,0}(q)]$ in the Born approximation we start with the integration over τ. Applying Wick's theorem we find that this term

$$N[\delta n_{ep}^{0,0}(q)]=(\dot{R};\delta n_{ep}^{0,0}(q)) =-\frac{e}{\beta}\frac{m+M}{mM}\int_0^\beta d\tau\,\mathrm{Tr}[\rho_0 P_0(-i\hbar\tau)\delta n_{ep}^{0,0}(q)] =-e\frac{m+M}{mM}\sum_{l,k,p}\hbar l_z\langle a_e^\dagger(l)a_e(l)a_e^\dagger(k-q/2) \times a_p^\dagger(p+q/2)a_p(p-q/2)a_e(k+q/2)\rangle \quad (A5)$$

gives no contribution due the symmetry ($l_z\rightarrow -l_z$) of the distribution function f_l.

In the matrix element

$$D[P_0;\delta n_{ep}^{0,0}(q)]=(P_0;\delta\dot{n}_{ep}^{0,0}(q))+\langle\dot{P}_0,\delta\dot{n}_{ep}^{0,0}(q)\rangle \quad (A6)$$

we have to calculate two correlation functions. Again, the second term vanishes in the Born approximation for symmetry reasons. For the first we use Eq. (A2) and arrive at

$$(P_0;\delta\dot{n}_{ep}^{0,0}(q))=-\frac{i}{\beta\hbar}\langle[P_0,\delta n_{ep}^{0,0}(q)]\rangle =\frac{i}{\beta}\sum_{k,p}q_z\langle a_e^\dagger(k-q/2)a_p^\dagger(p+q/2) \times a_p(p-q/2)a_e(k+q/2)\rangle. \quad (A7)$$

According to [22], this correlation function

$$\langle a_e^\dagger(k-q/2)a_p^\dagger(p+q/2)a_p(p-q/2)a_e(k+q/2)\rangle =\int\frac{d\omega}{\pi}\frac{1}{e^{\beta\omega}-1}\,\mathrm{Im}G(k,p,q,\omega+i\epsilon) \quad (A8)$$

is associated with a Green's function $G(k,p,q,\omega)$, which can be evaluated in the ladder approximation. In the Born approximation for the two-particle Green function only one interaction line is included

$$G(12,1'2')=G_0(11')G_0(22')+G_0(13)G_0(24) \times V(34,3'4')G_0(3'1')G_0(4'2'). \quad (A9)$$

Applying the Matsubara technique of standard quantum statistics [22], the Fourier transformation of Eq. (A9) $G(k,p,q,\omega_\lambda)$ is given in the limit of small densities ($f_k \ll 1$) by

$$G(k,p,q,\omega_\lambda)=\frac{V(q)}{\Delta E_{kpq}}\left[\frac{1}{\omega_\lambda-E_{p+q/2}-E_{k-q/2}}-\frac{1}{\omega_\lambda-E_{p-q/2}-E_{k+q/2}}\right], \quad (A10)$$

with $\Delta E_{kpq}=E_{p+q/2}+E_{k-q/2}-E_{p-q/2}-E_{k+q/2}$. Exploiting the Dirac identity, we find with Eq. (A8) for the correlation function

$$\langle a_{l-q/2}^\dagger a_{h+q/2}^\dagger a_{h-q/2}a_{l+q/2}\rangle =\frac{V_S(q)}{\Delta E_{kpq}}[n_B(E_{p+q/2}+E_{k-q/2})-n_B(E_{p-q/2}+E_{k+q/2})] =-\beta V_S(q)n^2\frac{(2\pi\beta\hbar^2)^3}{m^{3/2}M^{3/2}}\exp[-\beta(E_{p-q/2}+E_{k+q/2})], \quad (A11)$$

with the Bose distribution function $n_B(\omega)=[\exp(\beta\omega)-1]^{-1}$. Now we find for Eq. (A7)

$$(P_0;\delta\dot{n}_{ep}^{0,0}(q))=iq_zN^2\frac{e^2}{\epsilon_0\Omega(q^2+\kappa^2)}=i\frac{1}{\beta}q_zn_2(q). \quad (A12)$$

The result of the first correlation function for the matrix element

$$Q[P_0]=(\dot{R};P_0)+\langle\dot{R};\dot{P}_0\rangle \quad (A13)$$

is already known from Eq. (A4). This yields in the Born approximation

$$\langle\dot{R};\dot{P}_0\rangle=\frac{-ie\hbar\beta}{\epsilon}\frac{M+m}{Mm}\sum_{l,h,k,q}k_zq_z\langle a_e^\dagger(l)a_e(l)a_e^\dagger(k-q/2) \times a_p^\dagger(p+q/2)a_p(p-q/2)a_e(k+q/2)\rangle,$$

which gives no contribution because of the same symmetry arguments as in Eq. (A5).

The first term of the matrix element $Q[\delta n_{ep}^{0,0}(q)] =(\dot{R};\delta n_{ep}^{0,0}(q))+\langle\dot{R};\delta\dot{n}_{ep}^{0,0}(q)\rangle$ is known from Eq. (A5). According to Eq. (A1) we apply in the second term a partial integration and find two new correlation functions. The first expression was calculated in Eq. (A3), whereas the second term vanishes in the Born approximation:

$$\langle\dot{R};\delta\dot{n}_{ep}^{0,0}(q)\rangle=\frac{1}{\epsilon}(\dot{R};\delta\dot{n}_{ep}^{0,0}(q))+\frac{1}{\epsilon}\langle\dot{R};\delta\ddot{n}_{ep}^{0,0}(q)\rangle =-\frac{e}{\epsilon}\frac{M+m}{Mm}(P_0;\delta\dot{n}_{ep}^{0,0}(q)) =\frac{ie}{\epsilon\beta}\frac{M+m}{Mm}q_zn_2(q). \quad (A14)$$

In the collision term of matrix element $D[P_0;P_0]$, we have to consider $\dot{P}_0=(i/\hbar)[H_S,P_0]$. Because of momentum conservation in the electron system the electron-electron interaction does not contribute. Applying Wick's theorem, we get for the low-density limit ($f_k \ll 1$)

$$D[P_0;P_0]=(P_0;\dot{P}_0)+\langle\dot{P}_0;\dot{P}_0\rangle$$
$$=-\frac{i}{\beta\hbar}[P_0;P_0]+\frac{1}{\beta}\int_{-\infty}^{0}dt\,e^{\epsilon t}$$
$$\times\int_0^\beta d\tau\ \mathrm{Tr}(\rho_0 e^{(it/\hbar+\tau)H_S}\dot{P}_0 e^{-(it/\hbar+\tau)H_S}\dot{P}_0)$$
$$=\frac{1}{\beta}\int_{-\infty}^{0}dt\ e^{\epsilon t}\int_0^\beta d\tau\sum_{k,p,q}V_S^2(q)$$
$$\times e^{(it/\hbar+\tau)\Delta E_{kpq}}q_z^2 f_{k-q}f_{p+q}, \qquad (A15)$$

with the notation $\Delta E_{kpq}=E_{k-q}+E_{p+q}-E_k-E_p$. Performing the λ and τ integration, we find with the help of Dirac's identity

$$D[P_0;P_0]=-\pi\hbar\sum_{k,p,q}V_S^2(q)q_z^2 f_{k-q}f_{p+q}\delta(\Delta E_{kpq})$$
$$=\frac{4\sqrt{2\pi}}{3}\frac{N^2}{\Omega}\sqrt{m\beta}\frac{e^4}{(4\pi\epsilon_0)^2}L_{\mathrm{BH}}, \qquad (A16)$$

which results in a final expression with the Brooks-Herring–type Coulomb logarithm

$$L_{\mathrm{BH}}=\frac{1}{2}\int_0^\infty dx\ x(x+\kappa^2)^{-2}\exp(-\beta\hbar^2 x/8m). \qquad (A17)$$

Using the identity (A2), the term $D[\delta n_{ep}^{0,0}(q),P_0]$ is equivalent to the matrix element (A6) and yields the result

$$D[\delta n_{ep}^{0,0}(q);P_0]=(\delta n_{ep}^{0,0}(q);\dot{P}_0)+\langle\delta\dot{n}_{ep}^{0,0}(q);\dot{P}_0\rangle$$
$$=-\frac{i}{\beta}q_z n_2(q). \qquad (A18)$$

Finally, we calculate the contribution of $D[\delta n_{ep}^{0,0}(q');\delta n_{ep}^{0,0}(q)]=(\delta n_{ep}^{0,0}(q');\delta\dot{n}_{ep}^{0,0}(q))+\langle\delta\dot{n}_{ep}^{0,0}(q');\delta\dot{n}_{ep}^{0,0}(q)\rangle$ to the determinants in Eq. (9). The first correlation function vanishes due to the identity (A2). The second term is transformed via a partial integration into

$$D[\delta n_{ep}^{0,0}(q');\delta n_{ep}^{0,0}(q)]=\frac{1}{\epsilon}(\delta\dot{n}_{ep}^{0,0}(q');\delta\dot{n}_{ep}^{0,0}(q))$$
$$+\frac{1}{\epsilon}\langle\delta\dot{n}_{ep}^{0,0}(q');\delta\ddot{n}_{ep}^{0,0}(q)\rangle. \qquad (A19)$$

Whereas the second term is found to be of $\mathcal{O}(q^3)$ and can be neglected in the quasiclassical limit $q\to 0$, the calculation of the first term yields with the help of Eq. (A2) the result

$$D[\delta n_{ep}^{0,0}(q');\delta n_{ep}^{0,0}(q)]$$
$$=\frac{1}{\epsilon\beta}\frac{m+M}{mM}qq'\delta_{q,q'}\sum_{k,p}\langle a_e^\dagger(k)a_p^\dagger(p)a_p(p)a_e(k)\rangle$$
$$+\frac{1}{\epsilon\beta}\frac{m+M}{mM}qq'\delta_{q,q'}\sum_{k,p,h}\langle a_e^\dagger(k)a_p^\dagger(p-q'/2)$$
$$\times a_p^\dagger(h+q'/2)a_p(h-q'/2)a_p(p+q'/2)a_e(k)\rangle. \qquad (A20)$$

In the leading order of the interaction we truncate the correlation functions in Eq. (A20) and use Eq. (A11) with the result

$$D[\delta n_{ep}^{0,0}(q');\delta n_{ep}^{0,0}(q)]$$
$$=\frac{1}{\epsilon\beta}\frac{m+M}{mM}qq'\delta_{q,q'}\sum_{l,h}\langle a_e^\dagger(k)a_p^\dagger(p)a_p(p)a_e(k)\rangle$$
$$+\frac{1}{\epsilon\beta}\frac{m+M}{mM}qq'\delta_{q,q'}\sum_{k,p,h}\langle a_e^\dagger(k)a_e(k)\rangle$$
$$\times\langle a_p^\dagger(p-q'/2)a_p^\dagger(h+q'/2)$$
$$\times a_p(h-q'/2)a_p(p+q'/2)\rangle$$
$$=\frac{1}{\epsilon\beta}\frac{m+M}{mM}[N^2-Nn_2(q)]qq'\delta_{q',q}. \qquad (A21)$$

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