

Dielectric function beyond the random-phase approximation: Kinetic theory versus linear response theory

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Calculating the frequency-dependent dielectric function for strongly coupled plasmas, the relations within kinetic theory and linear response theory are derived and discussed in comparison. In this context, we give a proof that the Kohler variational principle can be extended to arbitrary frequencies. It is shown to be a special case of the Zubarev method for the construction of a nonequilibrium statistical operator from the principle of the extremum of entropy production. Within kinetic theory, the commonly used energy-dependent relaxation time approach is strictly valid only for the Lorentz plasma in the static case. It is compared with the result from linear response theory that includes electron-electron interactions and applies for arbitrary frequencies, including bremsstrahlung emission. It is shown how a general approach to linear response encompasses the different approximations and opens options for systematic improvements.

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I. INTRODUCTION

Different approaches have been elaborated to evaluate the response of a plasma to external time- and space-dependent electric fields. This applies, e.g., to absorption and emission of radiation (in particular, bremsstrahlung), Thomson scattering, and stopping power. The dielectric function $\epsilon(\vec{k}, \omega)$, depending on the wave number \vec{k} and frequency ω as the central quantity, is related to the polarization function, the dynamical conductivity, or the dynamical structure factor. The random-phase approximation (RPA) is improved if collisions are taken into account. In this context, a nonlocal dynamical collision frequency is introduced [1–4]. Alternatively, the concept of a local-field factor [5] can be extended to dynamical local-field corrections [2,6–8]. In the present work, we focus on the dynamical conductivity and restrict ourselves to the long-wavelength limit $k \rightarrow 0$, i.e., the response of a charged particle system to a homogeneous, time-dependent electrical field.

A well-known expression for the dc conductivity of a fully ionized plasma in the classical, low-density limit has been given by Spitzer and Härn [9] within kinetic theory (KT). Further approaches by Lee and More [10], Stygar [11], and others improved the electron-ion interaction using the relaxation time approach. However, to recover the Spitzer result for the conductivity, electron-electron collisions have to be taken into account. This is not consistently possible within the relaxation time approach [12]. We discuss a general approach that allows also for a systematic treatment of electron-electron collisions.

The investigation of time-dependent fields is somehow difficult in KT, too. Often, a combination of the collisionless kinetic equation with the relaxation time ansatz is used;

see Landau and Lifshits [13], Dharma-wardana [14], or Kurilenko *et al.* [1,15]. It has been emphasized by Landau and Lifshits [13] that such an approach is only applicable in the low-frequency limit. The high-frequency region, where bremsstrahlung is relevant, has to be treated in another way. In this work, we present general expressions applicable to arbitrary frequencies of the external field.

In linear response theory (LRT), the Kubo formula [16] was considered as a promising approach to the dynamical conductivity in dense, strongly interacting systems at arbitrary degeneracy. A generalized approach to nonequilibrium processes was then given by Zubarev *et al.* [17], which will be applied here. It relates transport properties to equilibrium correlation functions, such as current-current or force-force correlation functions. Different methods can be applied to evaluate these correlation functions, such as numerical simulations, density functional approaches [14,18], or analytical expressions derived from perturbation theory [19–21]. Note that also strict results such as sum rules can be employed to construct the dynamical structure factor; see [22–24]. We will show how consistent approximations are obtained from a general scheme of nonequilibrium statistical physics and systematic improvements can be given.

In the present work, we will restrict ourselves to homogeneous systems and therefore do not consider any dependence on the position \vec{r} in space, e.g., due to external potentials, in addition to the homogeneous, time-dependent electrical field that is treated as a perturbation. The focus is on the generalization of relations which were originally derived in KT; see Sec. II. Starting from LRT (see Sec. III), a generalized Boltzmann equation with a frequency-dependent collision term is derived. In Sec. IV, a variational approach is applied for the solution of the generalized linear Boltzmann equation. Similar to the use of polynomials [12,25,26] to solve the static Boltzmann equation, we consider moments of the single-particle distribution function to find approximate solutions.

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Furthermore, in Sec. II, different limiting cases, such as the dc conductivity and the high-frequency limit of the absorption coefficient, are considered. The dynamical conductivity from KT using an energy-dependent relaxation time, which has often been used in the literature, is compared with the rigorous treatment within LRT. Conclusions are drawn in Sec. V.

II. KINETIC EQUATIONS

A. Single-particle distribution function

We consider neutral Coulomb systems that consist of charged particles such as electrons and ions. The response to an electromagnetic field is described by the dielectric function, taken in the long-wavelength limit here,

$$\lim_{k \rightarrow \infty} \epsilon(\vec{k}, \omega) = 1 + \frac{i}{\epsilon_0 \omega} \sigma(\omega), \quad (1)$$

or the dynamical conductivity $\sigma(\omega)$. Treating the Coulomb interaction in the mean-field approximation, the random-phase approximation (RPA) is obtained. To improve RPA, one has to include collisions. A standard way to treat collisions is the Boltzmann equation where the interaction between the constituents leads to the collision term. As a consequence, a dynamical collision frequency $\nu(\omega)$ can be introduced according to a generalized Drude formula,

$$\sigma(\omega) = \frac{\epsilon_0 \omega_{\text{pl}}^2}{-i\omega + \nu(\omega)}, \quad (2)$$

with the plasmon frequency $\omega_{\text{pl}} = \sqrt{e^2 n / (\epsilon_0 m)}$, where n is the electron density and m is the (reduced) mass. The collision frequency $\nu(\omega)$ should be a complex, frequency-dependent quantity in order to satisfy sum rules of the dielectric function. This is in contrast to a static relaxation time $\tau = 1/\nu$, as used in the kinetic approach, and will be explained in more detail below.

Taking the adiabatic approximation, N electrons interact with singly charged heavy ions that are considered as external potential. The Hamiltonian with the electronic degrees of freedom only is

$$\hat{H} = \sum_p E_p \hat{a}_p^\dagger \hat{a}_p + \sum_{pq} V_{ei}(q) \hat{a}_{p+q}^\dagger \hat{a}_p + \frac{1}{2} \sum_{p_1 p_2 q} V_{ee}(q) \hat{a}_{p_1+q}^\dagger \hat{a}_{p_2-q}^\dagger \hat{a}_{p_2} \hat{a}_{p_1}, \quad (3)$$

with $E_p = \hbar^2 p^2 / (2m)$. The interaction with the ions, $V_{ei}(\vec{q}) = -V(q) \sum_j^N \exp[i\vec{q} \cdot \vec{R}_j]$, describes Coulomb potentials $V(q) = e^2 / (\epsilon_0 \Omega_0 q^2)$ due to various ion sites \vec{R}_j , which leads to the structure factor $S(\vec{q}) = (1/N) \langle \sum_{i,j} \exp[i\vec{q} \cdot (\vec{R}_i - \vec{R}_j)] \rangle$. Ω_0 is the normalization volume. The electron-electron interaction is given by the Coulomb interaction $V_{ee}(q) = V(q)$. The account of the ion dynamics is straightforwardly taken into account within a two-component plasma [3], but the notations become more complex and will not be given here.

For the derivation of kinetic equations, in particular the Boltzmann equation, we consider the electron single-particle distribution function $f(\vec{p}, t) = \text{Tr}\{\hat{n}_p \hat{\rho}(t)\} = \langle \hat{n}_p \rangle^t$ that is the quantum statistical average, taken with the nonequilibrium

statistical operator $\hat{\rho}(t)$, of the single-particle occupation number operator $\hat{n}_p = \hat{a}_p^\dagger \hat{a}_p$ of momentum $\hbar \vec{p}$. Considering homogeneous systems, the density matrix is diagonal with respect to the wave vector \vec{p} . Spin variables are not explicitly given unless it is pointed out. Subsequently, the single-particle distribution function does not depend on the position \vec{r} either.

In thermal equilibrium, the single-particle distribution function $f_0(\vec{p}) = \text{Tr}\{\hat{n}_p \hat{\rho}_0\}$ is calculated with the grand canonical statistical operator $\hat{\rho}_0 = \exp[-\beta(\hat{H} - \mu \hat{N})] / \text{Tr}\{\exp[-\beta(\hat{H} - \mu \hat{N})]\}$. Neglecting the interaction term, we find the ideal Fermi gas with distribution $f_p = \{\exp[\beta(E_p - \mu)] + 1\}^{-1}$. Under the influence of an external perturbation \hat{H}_{ext}^t , the single-particle distribution function $f(\vec{p}, t)$ is modified. Its deviation

$$\delta f(\vec{p}, t) = f(\vec{p}, t) - f_0(\vec{p}) = \text{Tr}\{\delta \hat{n}_p \hat{\rho}(t)\} \quad (4)$$

from the equilibrium distribution $f_0(\vec{p})$ is the average of the fluctuations of the single-particle occupation number $\delta \hat{n}_p = \hat{n}_p - f_0(\vec{p})$. The time dependence of the single-particle distribution function $f(\vec{p}, t)$ is determined by the nonequilibrium statistical operator $\hat{\rho}(t)$, as shown in the following section.

Alternatively, the dynamics of the single-particle distribution function can be determined from a hierarchy of equations of motion for the many-particle distribution functions. By truncating the hierarchy, a kinetic equation [13] is obtained with the structure

$$\frac{\partial}{\partial t} f(\vec{p}, t) = D[f(\vec{p}, t)] + C[f(\vec{p}, t)] \quad (5)$$

describing drift in the single-particle phase space via the drift term $D[f(\vec{p}, t)]$, and collisions that are caused by the interaction between the particles. The collision term $C[f(\vec{p}, t)]$ is related to the higher-order distribution functions due to the interaction mechanisms within the system. To obtain closed kinetic equations, the higher distribution functions are expressed in terms of $f(\vec{p}, t)$.

In the following, we consider a homogeneous system under the influence of an external time-dependent electric field $\vec{E}(t)$. The total Hamiltonian $\hat{H}_{\text{tot}}^t = \hat{H} + \hat{H}_{\text{ext}}^t$ contains the interaction with the external field, $\hat{H}_{\text{ext}}^t = -e \vec{E}(t) \cdot \sum_i \hat{r}_i$, for the electron position operators \hat{r}_i . From the respective external force $e \vec{E}(t)$, the drift term follows as

$$\begin{aligned} D[f(\vec{p}, t)] &= -\frac{e}{\hbar} \vec{E}(t) \cdot \frac{\partial}{\partial \vec{p}} f(\vec{p}, t) \\ &\approx \frac{e \hbar}{m} \beta f_p (1 - f_p) \vec{E}(t) \cdot \vec{p} \end{aligned} \quad (6)$$

in the first order with respect to the external field $\vec{E}(t)$, with $\beta = 1/(k_{\text{B}} T)$. Expressions for the collision term $C[f(\vec{p}, t)]$ will be given below.

With the distribution function $f(\vec{p}, t)$, the current density is given by

$$\vec{j}(t) = \frac{e}{m \Omega_0} \sum_p \hbar \vec{p} f(\vec{p}, t) = \frac{e}{m \Omega_0} \vec{P}_1(t). \quad (7)$$

The total momentum $\vec{P}_1(t)$ is the first moment of the distribution function. In the following, we also consider the operators of arbitrary moments,

$$\hat{P}_v = \sum_p \hbar p_E (\beta E_p)^{(v-1)/2} \hat{n}_p, \quad (8)$$

where $p_E = \vec{p} \cdot \vec{E} / |\vec{E}|$ denotes the component of \vec{p} in the direction of \vec{E} .

The arbitrary time dependence of an electric field can be expressed by the superposition of harmonic time dependences. Within the linear response, each component $\vec{E}(t) = \frac{1}{2} \vec{E}(\omega) \exp(-i\omega t) + \text{c.c.}$ causes an induced single-particle distribution function,

$$\delta f(\vec{p}, t) = \frac{1}{2} \delta \tilde{f}(\vec{p}, \omega) \exp(-i\omega t) + \text{c.c.} \quad (9)$$

with the same time dependence. The dynamical conductivity follows from $\tilde{j}(\omega) = \sigma(\omega) \vec{E}$ as

$$\sigma(\omega) = \frac{e}{m \vec{E}} \frac{1}{\Omega_0} \sum_p \hbar p_E \delta \tilde{f}(\vec{p}, \omega). \quad (10)$$

Note that all Fourier components marked with a tilde, e.g., \tilde{F}_p , are frequency dependent in general. The dependence on ω will be omitted in some of the following expressions for them to be more compact.

B. Relaxation time approximation and dynamical conductivity

To start with an analytically solvable example, we first discuss the solution of the kinetic equation (5) for the Lorentz model where the electron-electron interaction in the Hamiltonian (3) is neglected. Considering a constant electric field, the distribution function $f(\vec{p}, t) = f(\vec{p})$ is static. In the standard treatment (see [27]), the collision term reads

$$C_{\text{Lorentz}}[f(\vec{p})] = \sum_{p'} \{ f(\vec{p}') w_{ei}(\vec{p}, \vec{p}') [1 - f(\vec{p})] - f(\vec{p}) w_{ei}(\vec{p}', \vec{p}) [1 - f(\vec{p}')] \}. \quad (11)$$

The transition rates can be determined in the Born approximation from the golden rule, $w_{ei}(\vec{p}, \vec{p}') = (2\pi/\hbar) |V_{ei}(|\vec{p} - \vec{p}'|)|^2 \delta(E_p - E_{p'})$. Since the energy of electrons is conserved in adiabatic approximation, a relaxation time τ_p is introduced via an ansatz for the linear term of the expansion of the distribution function, $f(\vec{p}) = f_p - F_p \frac{1}{\beta} \frac{\partial}{\partial E_p} f_p$. In analogy to the drift term (6), we assume

$$\delta f(\vec{p}) = \frac{e\hbar}{m} \beta \tau_p \vec{E} \cdot \vec{p}, \quad (12)$$

which realizes the linearity with respect to the external field \vec{E} . For isotropic systems, τ_p is a scalar depending only on the modulus of \vec{p} . By inserting Eq. (12) into the collision term (11) and taking into account the detailed balance in equilibrium $w_{ei}(\vec{p}, \vec{p}') f_{p'} (1 - f_p) = w_{ei}(\vec{p}', \vec{p}) f_p (1 - f_{p'})$, as well as the energy balance of the transition rates, the collision term (11) is

$$\begin{aligned} C_{\text{Lorentz}}[f(\vec{p})] &= - \sum_{p'} w_{ei}(\vec{p}, \vec{p}') f_{p'} (1 - f_p) (F_p - F_{p'}) \\ &= -\delta f(\vec{p}) / \tau_p. \end{aligned} \quad (13)$$

For the kinetic equation (5) with the drift term (6), we then find

$$\begin{aligned} \vec{E} \cdot \vec{p} &= - \sum_{p'} w_{ei}(\vec{p}, \vec{p}') \frac{f_{p'}}{f_p} \vec{E} \cdot (\tau_{p'} \vec{p}' - \tau_p \vec{p}) \\ &= -\tau_p \sum_q w_{ei}(\vec{p}, \vec{p} + \vec{q}) \vec{E} \cdot \vec{q}, \end{aligned} \quad (14)$$

with $\vec{q} = \vec{p}' - \vec{p}$. With the golden rule for the transition rates given above and $S(q) \approx 1$, $|V_{ei}(q)|^2 \approx NV^2(q)$, the energy-dependent relaxation time can be calculated as

$$\frac{1}{\tau_p} = -\frac{2\pi}{\hbar} \sum_q NV^2(q) \delta(E_p - E_{p+q}) \frac{\vec{E} \cdot \vec{q}}{\vec{E} \cdot \vec{p}}. \quad (15)$$

The \vec{q} integral in Eq. (15) can be performed using spherical coordinates where \vec{p} is in the z direction, and \vec{E} is in the x - z plane. It is convergent only in the case of a screened Coulomb potential. Using the statically screened Debye potential

$$V_D(q) = \frac{e^2}{\epsilon_0 \Omega_0 (q^2 + \kappa_D^2)}, \quad \kappa_D^2 = \beta n e^2 / \epsilon_0, \quad (16)$$

we find the energy-dependent collision frequency

$$\nu_p = \tau_p^{-1} = n \frac{e^4}{4\pi \epsilon_0^2 \hbar^3 p^3} \left(\ln \sqrt{1+b} - \frac{1}{2} \frac{b}{1+b} \right), \quad (17)$$

with $b = 4p^2/\kappa_D^2$ in the Coulomb logarithm. The static conductivity is determined from Eq. (10), $\omega = 0$, as

$$\begin{aligned} \sigma_{\text{dc, Lorentz}} &= \frac{e^2 \hbar^2}{m^2} \beta \frac{1}{\Omega_0} \sum_p p_E^2 \tau_p f_p (1 - f_p) \\ &= \epsilon_0 \omega_{\text{pl}}^2 \tau_{\text{Lorentz}} = \frac{e^2 n}{m \nu_{\text{Lorentz}}}. \end{aligned} \quad (18)$$

We introduce the average relaxation time τ_{Lorentz} and the static collision frequency $\nu_{\text{Lorentz}} = 1/\tau_{\text{Lorentz}}$.

We are now interested in extending the static case (18) by evaluating the permittivity $\epsilon(\omega)$, given by Eq. (1), or the dynamical conductivity, given by Eq. (10). From the kinetic equation (5) with the drift term (6), we derive the frequency-dependent Boltzmann equation

$$\begin{aligned} -i\omega \delta \tilde{f}(\vec{p}, \omega) &= \frac{e\hbar}{m} \beta \vec{E}(\omega) \cdot \vec{p} f_p (1 - f_p) \\ &+ C_{\text{Lorentz}}[\delta \tilde{f}(\vec{p}, \omega)]. \end{aligned} \quad (19)$$

In a standard approach (see, e.g., Landau and Lifshits [27]), it is proposed to extend the static case to the dynamic case assuming that the relaxation time is the same as in the static case; see Eq. (13). Subsequently, the following relation is derived:

$$-\left(i\omega - \frac{1}{\tau_p}\right) \delta \tilde{f}(\vec{p}, \omega) = \frac{e\hbar}{m} \beta \vec{E}(\omega) \cdot \vec{p} f_p (1 - f_p), \quad (20)$$

so that for the dynamical conductivity (10), we get (spin factor 2, $p_E^2 \rightarrow p^2/3$ for isotropic systems)

$$\sigma_{\text{KT}}(\omega) = \frac{2}{3} \frac{e^2 \hbar^2 \beta}{m^2} \int \frac{d^3 \vec{p}}{(2\pi)^3} \frac{p^2 f_p (1 - f_p)}{-i\omega + 1/\tau_p}. \quad (21)$$

This result can be interpreted as a Vlassov approach where the frequency ω is replaced by a complex frequency $\omega + i/\tau_p$.

However, the introduction of an energy-dependent, static relaxation time is an approximation that cannot be applied, in particular, at high frequencies, where bremsstrahlung emission is expected. Note that it is not possible to give an explicit expression for a frequency-dependent collision frequency as desired for a generalized Drude formula according to Eq. (2). Furthermore, inelastic collisions such as electron-electron interactions are not taken into account by a collision time ansatz. Further evaluation of Eq. (21) is given in Appendix E; results are shown in Fig. 2 and discussed below.

III. LINEAR RESPONSE EQUATIONS

A. Linear response theory

To evaluate the response (4) to an external perturbation \hat{H}_{ext}^t , we determine the nonequilibrium statistical operator $\rho(t)$ within a generalized linear response theory. The conceptual ideas and main expressions relevant for the further analysis of the single-particle distribution function will be given here according to [4,28–30].

We introduce the relevant statistical operator

$$\begin{aligned}\hat{\rho}_{\text{rel}}(t) &= \frac{1}{Z_{\text{rel}}(t)} e^{-\beta(\hat{H}-\mu\hat{N})+\sum_n F_n(t)\hat{B}_n}, \\ Z_{\text{rel}}(t) &= \text{Tr}\{e^{-\beta(\hat{H}-\mu\hat{N})+\sum_n F_n(t)\hat{B}_n}\}\end{aligned}\quad (22)$$

as a generalized Gibbs ensemble, which is derived from the principle of maximum of the entropy,

$$S(t) = -k_B \text{Tr}\{\hat{\rho}_{\text{rel}}(t) \ln[\hat{\rho}_{\text{rel}}(t)]\}, \quad (23)$$

where the Lagrange parameters β , μ , and $F_n(t)$, which are real valued numbers, are introduced to fix the given averages

$$\text{Tr}\{\hat{B}_n \hat{\rho}(t)\} = \langle \hat{B}_n \rangle^t = \text{Tr}\{\hat{B}_n \hat{\rho}_{\text{rel}}(t)\}. \quad (24)$$

These self-consistency conditions mean that the observed averages $\langle \hat{B}_n \rangle^t$ are correctly reproduced by the Hermitian $\hat{\rho}_{\text{rel}}(t)$. Similar relations are used in equilibrium to eliminate the Lagrange parameters β and μ . In the linear response, the response parameters $F_n(t)$ are considered to be small so that we can solve the implicit relation (24) expanding up to the first order,

$$\hat{\rho}_{\text{rel}}(t) = \left[1 + \sum_n F_n(t) \int_0^1 d\lambda e^{-\beta\lambda(\hat{H}-\mu\hat{N})} \delta\hat{B}_n e^{\beta\lambda(\hat{H}-\mu\hat{N})} \right] \hat{\rho}_0. \quad (25)$$

Note that the expansion of $Z_{\text{rel}}(t)$ in Eq. (22) leads to the subtraction of the equilibrium average in $\delta\hat{B}_n = \hat{B}_n - \langle \hat{B}_n \rangle_0$. The average fluctuations can now be explicitly calculated by inserting Eq. (25) in Eq. (24),

$$\langle \delta\hat{B}_n \rangle^t = \sum_m \langle \delta\hat{B}_n, \delta\hat{B}_m \rangle F_m(t), \quad (26)$$

where we introduced the Kubo scalar product

$$\langle \hat{A}, \hat{B} \rangle = \int_0^1 d\lambda \text{Tr}\{\hat{A} \hat{B}^\dagger(i\hbar\beta\lambda)\hat{\rho}_0\}. \quad (27)$$

The time dependence $\hat{A}(t) = e^{i\hat{H}t/\hbar} \hat{A} e^{-i\hat{H}t/\hbar}$ is given by the Heisenberg picture with respect to the system Hamiltonian \hat{H} , and $\hat{A} = i[\hat{H}, \hat{A}]/\hbar$.

A statistical operator for the nonequilibrium is constructed with the help of the relevant statistical operator (22); see Appendix A. Expanding up to the first order with respect to the external field \vec{E} and the response parameters \tilde{F}_n , where $F_n(t) = \text{Re}\{\tilde{F}_n(\omega)e^{-i\omega t}\}$, we arrive at the response equations,

$$\begin{aligned}\sum_m w[\langle \hat{B}_n; \hat{B}_m \rangle + \langle \hat{B}_n; \hat{B}_m \rangle_z - i\omega\{\langle \hat{B}_n; \hat{B}_m \rangle + \langle \hat{B}_n; \delta\hat{B}_m \rangle_z}] \tilde{F}_m \\ = \beta \frac{e}{m} \{ \langle \hat{B}_n; \hat{P} \rangle + \langle \hat{B}_n; \hat{P} \rangle_z \} \cdot \vec{E},\end{aligned}\quad (28)$$

where $z = \omega + i\epsilon$ is the total momentum of electrons $\hat{P} = \sum_p \hbar \hat{p} \hat{n}_p$, and the Laplace transform of the correlation functions is

$$\begin{aligned}\langle \hat{A}; \hat{B} \rangle_z &= \int_0^\infty dt e^{izt} \langle \hat{A}(t), \hat{B} \rangle \\ &= \int_0^\infty dt e^{izt} \int_0^1 d\lambda \text{Tr}\{\hat{A}(t - i\hbar\beta\lambda) \hat{B}^\dagger \hat{\rho}_0\}.\end{aligned}\quad (29)$$

Considering N_B relevant observables \hat{B}_n , Eq. (28) is a system of N_B linear equations to determine the response parameters \tilde{F}_n for a given external field \vec{E} . It is the most general form of LRT, allowing for the arbitrary choice of relevant observables \hat{B}_n and corresponding response parameters F_n . We show below that with respect to kinetic theory, the first two terms on the left-hand side of Eq. (28) can be identified as a collision term, while the right-hand side represents the drift term due to the external perturbing field.

B. Generalized linear Boltzmann equations

In kinetic theory, the nonequilibrium state is characterized by the single-particle distribution function $f(\vec{p}, t)$. In order to derive expressions in parallel to the kinetic theory, we choose the fluctuations $\delta\hat{n}_p$ of the single-particle occupation number [see Eq. (4)] as relevant observables B_n . The modification of the single-particle distribution function can then be calculated straightforwardly according to Eq. (26),

$$\text{Tr}\{\hat{\rho}_{\text{rel}}(t) \delta\hat{n}_p\} = \sum_{p'} \langle \delta\hat{n}_p, \delta\hat{n}_{p'} \rangle F_{p'}(t) = \delta f(\vec{p}, t). \quad (30)$$

The Lagrange multipliers $F_p(t) = \tilde{F}_p(\omega) \exp(-i\omega t)/2 + \text{c.c.}$ are determined from the response equations (28). We arrive at the generalized linear Boltzmann equations ($\delta\hat{n}_p = \hat{n}_p$),

$$\begin{aligned}\sum_{p'} [\langle \delta\hat{n}_p, \hat{n}_{p'} \rangle + \langle \hat{n}_p; \hat{n}_{p'} \rangle_z - i\omega\{ \langle \delta\hat{n}_p, \delta\hat{n}_{p'} \rangle + \langle \hat{n}_p; \delta\hat{n}_{p'} \rangle_z \}] \tilde{F}_{p'} \\ = \frac{e\hbar}{m} \beta \sum_{p''} [\langle \delta\hat{n}_p, \hat{n}_{p''} \rangle + \langle \hat{n}_p; \hat{n}_{p''} \rangle_z] \vec{p}'' \cdot \vec{E}.\end{aligned}\quad (31)$$

The time derivative of the position operator in \hat{H}_{ext}^t leads to the total momentum $\sum_i \hbar \vec{p}_i = m \sum_i \vec{r}_i$, and subsequently to the right-hand side of Eq. (31). We analyze the different terms of Eq. (31) below and compare with the kinetic equation (5), considering the Born approximation. Notice that this result can be extended by introducing stochastic forces [29] if we go beyond the Born approximation. Further relevant observables beyond the single-particle occupation numbers can be included in order to characterize the nonequilibrium state, such as long-living correlations and the formation of bound states.

It is possible to go beyond the Boltzmann equation if higher correlations such as bound-state formation are included in the set of relevant observables.

We give the entropy as obtained from Eq. (23),

$$S(t) = -k_B \text{Tr} \left\{ \hat{\rho}_{\text{rel}}(t) \left[-\ln[Z_{\text{rel}}(t)] - \beta(\hat{H} - \mu\hat{N}) + \sum_p F_p(t)\hat{n}_p \right] \right\} = S_0(\beta, \mu) - k_B \sum_p F_p(t) \delta f(\vec{p}, t) \quad (32)$$

in the first order of $F_p(t)$. The entropy in the thermodynamic equilibrium is denoted by $S_0(\beta, \mu)$. With Eq. (30), we find that the entropy decreases in nonequilibrium because $\delta S(t) = -\sum_{pp'} F_{p'}(t)(\delta\hat{n}_{p'}, \delta\hat{n}_p)F_p(t) \leq 0$. The proof is given using the spectral density for $\hat{F}(t) = \sum_p F_p(t)\delta\hat{n}_p$; see [17]. With the eigenstates $(\hat{H} - \mu\hat{N})|n\rangle = E_n|n\rangle$ of the system Hamiltonian, we have

$$\delta S(t) = -(\hat{F}(t), \hat{F}(t)) = \frac{1}{Z_0\beta} \sum_{nm} \frac{e^{-\beta E_n} - e^{-\beta E_m}}{E_n - E_m} |\langle n|\hat{F}(t)|m\rangle|^2 \leq 0. \quad (33)$$

This result corresponds to the second law of thermodynamics in which the entropy of the many-particle system exhibits its maximum in the equilibrium state.

C. Evaluation of equilibrium correlation functions and Born approximation

Quantum statistics provides us with different methods to calculate correlation functions in thermal equilibrium, such as perturbation theory and diagram techniques. By applying perturbation theory with respect to the interaction, Wick's theorem can be used. We find in the lowest order for the Kubo scalar product (27),

$$(\hat{n}_p, \hat{n}_{p'}) = \text{Tr}\{\rho_0 \hat{a}_p^\dagger \hat{a}_{p'}^\dagger \hat{a}_p \hat{a}_{p'}\} = f_{p'} f_p + f_p(1 - f_{p'})\delta_{pp'}, \quad (34)$$

so that $(\delta\hat{n}_p, \delta\hat{n}_{p'}) = (\delta\hat{n}_p, \hat{n}_{p'}) = f_p(1 - f_{p'})\delta_{pp'}$. The remaining Kubo scalar product vanishes, $(\delta\hat{n}_p, \hat{n}_{p'}) = 0$, as shown

from the Kubo identity (A3), with $\hat{C} = \delta\hat{n}_p$, and $\langle [n_{p'}, n_p] \rangle_0 = 0$ after cyclic invariance of the trace.

For the deviation of the single-particle occupation numbers from equilibrium, we find from Eq. (30) that $\delta f(\vec{p}, t) = F_p(t)f_p(1 - f_p)$, which is equivalent to the expansion (12) in kinetic theory. Thus, we solved the self-consistency condition (24) to eliminate the Lagrange parameters $F_p(t)$. According to (9), the Fourier components

$$\delta \tilde{f}(\vec{p}, \omega) = f_p(1 - f_p)\tilde{F}_p(\omega) \quad (35)$$

are complex amplitudes, containing in general a phase factor.

The equation of motion that leads to the generalized linear Boltzmann equation (31) allows one to relate the response to the external field. The right-hand side is the drift term that contains the external field. In the Born approximation, we can neglect the correlation function $\langle \hat{n}_p; \hat{n}_{p'} \rangle_z$ because it is of a higher order of interaction compared with $(\delta\hat{n}_p, \hat{n}_{p'})$. Then, the right-hand side of Eq. (31) reads

$$D_p = \frac{e\hbar}{m} \beta f_p(1 - f_p) \vec{p} \cdot \vec{E}, \quad (36)$$

in agreement with Eq. (6). By the same argument, we have the term due to the explicit time dependence,

$$-\sum_{p'} i\omega[(\delta\hat{n}_p, \delta\hat{n}_{p'}) + \langle \hat{n}_p; \delta\hat{n}_{p'} \rangle_z] \tilde{F}_{p'} = -i\omega \delta \tilde{f}(\vec{p}, \omega) = -i\Omega_p \tilde{F}_p, \quad (37)$$

with $\Omega_p = \omega f_p(1 - f_p)$. Note that the correlation function $\langle \hat{n}_p; \hat{n}_{p'} \rangle_z$ is eliminated, introducing stochastic forces [17,29], so that the result $-i\omega \delta \tilde{f}(\vec{p}, \omega)$ holds also beyond the Born approximation.

The remaining term in Eq. (31) describes the collision integral,

$$C_p = -\sum_{p'} \langle \hat{n}_p; \hat{n}_{p'} \rangle_{\omega+i\epsilon} \tilde{F}_{p'} = -\sum_{p'} \mathcal{L}_{pp'}(\omega) \tilde{F}_{p'}. \quad (38)$$

It is evaluated in the Born approximation (see Appendix B), with the generalized Onsager coefficients $\mathcal{L}_{pp'}(\omega) = \mathcal{L}_{pp'}^{ei}(\omega) + \mathcal{L}_{pp'}^{ee}(\omega)$, leading to

$$\begin{aligned} \mathcal{L}_{pp'}^{ei}(\omega) &= -\frac{1}{\hbar^2} \sum_q |V_{ei}(q)|^2 \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} \left\{ \pi\delta \left[\omega + \frac{1}{\hbar}(E_p - E_{p+q}) \right] + \pi\delta \left[\omega - \frac{1}{\hbar}(E_p - E_{p+q}) \right] \right. \\ &\quad \left. + i\text{P} \frac{1}{\omega + (E_p - E_{p+q})/\hbar} + i\text{P} \frac{1}{\omega - (E_p - E_{p+q})/\hbar} \right\} [\delta_{p', p+q} - \delta_{p', p}], \\ \mathcal{L}_{pp'}^{ee}(\omega) &= -\frac{1}{\hbar^2} \sum_{p_1, q} |V_{ee}(q)|^2 \frac{f_p f_{p_1} (1 - f_{p_1-q} - f_{p+q}) - f_{p+q} f_{p_1-q} (1 - f_{p_1} - f_p)}{\beta(E_{p+q} + E_{p_1-q} - E_{p_1} - E_p)} \\ &\quad \left[\frac{i}{\omega + i\epsilon + \Delta_{p, p_1, q}} + \frac{i}{\omega + i\epsilon - \Delta_{p, p_1, q}} \right] [\delta_{p', p+q} + \delta_{p', p_1-q} - \delta_{p', p_1} - \delta_{p', p}], \end{aligned} \quad (39)$$

where $\Delta_{p, p_1, q} = (E_{p+q} + E_{p_1-q} - E_{p_1} - E_p)/\hbar$ and P denotes the principal value. Exchange contributions have been discarded; see Appendix B. The decomposition of $\mathcal{L}_{pp'}^{ee}(\omega)$ into the real and imaginary parts is analogous to $\mathcal{L}_{pp'}^{ei}(\omega)$.

In conclusion, the generalized linearized Boltzmann equation (31) can be given in the same way as assumed in the relaxation time approach [see Eq. (19)],

$$\begin{aligned} -i\omega\delta\tilde{f}(\vec{p},\omega) &= \frac{e\hbar}{m}\beta f_p(1-f_p)\vec{p}\cdot\tilde{\vec{E}} - \sum_{p'}\mathcal{L}_{pp'}(\omega)\tilde{F}_{p'} \\ &= D_p + C_p[\delta\tilde{f}(\vec{p},\omega)], \end{aligned} \quad (41)$$

with Eq. (37) and the drift term (36), after replacing the response parameters \tilde{F}_p in the collision term (38) by the single-particle distribution according to Eq. (35). This holds for arbitrary frequencies ω and degeneracy; see Appendix A. At zero frequency, the collision integral (11) of the Lorentz plasma is recovered if calculations are taken in the Born approximation and restricted to the electron-ion interaction only. At arbitrary frequencies, the collision integral becomes a complex quantity in contrast to the scalar relaxation time. The real and imaginary parts are connected via Kramers-Kronig relations. The Born approximation can be improved in a systematic way if the correlation functions are evaluated in higher orders with respect to the interaction. A Kubo-

Greenwood formula can be derived that expresses the collision term by T matrices [29,30].

IV. SOLUTION OF THE GENERALIZED LINEAR BOLTZMANN EQUATION

A. Variational principle

Having derived an explicit expression for the Onsager coefficients $\mathcal{L}_{pp'}$ in the Born approximation, given by Eqs. (39) and (40), we can now determine the response parameters by solving the generalized linear Boltzmann equation (41) given as

$$-i\Omega_p\tilde{F}_p(\omega) = D_p - \sum_{p'}\mathcal{L}_{pp'}(\omega)\tilde{F}_{p'}(\omega). \quad (42)$$

As a further constraint on the response parameters \tilde{F}_p , we consider the entropy leading to a variational problem as follows.

We determine the time derivative of the entropy (32). The time-dependent term reads

$$\begin{aligned} \frac{d}{dt}S(t) &= -2\sum_p\frac{1}{f_p(1-f_p)}\delta f(\vec{p},t)\delta\dot{f}(\vec{p},t) = -\frac{1}{2}\sum_p\frac{1}{f_p(1-f_p)}[\delta\tilde{f}(\vec{p})e^{-i\omega t} + \text{c.c.}][-i\omega\delta\tilde{f}(\vec{p})e^{-i\omega t} + \text{c.c.}] \\ &= -\frac{1}{2}\sum_p[\tilde{F}_pe^{-i\omega t} + \tilde{F}_p^*e^{i\omega t}]\left[D_p[\tilde{\vec{E}}](e^{-i\omega t} + e^{i\omega t}) - \sum_{p'}\mathcal{L}_{pp'}(\omega)\tilde{F}_{p'}e^{-i\omega t} - \sum_{p'}\mathcal{L}_{pp'}^*(\omega)\tilde{F}_{p'}^*e^{i\omega t}\right] \end{aligned} \quad (43)$$

if we insert the Boltzmann equation (41) for $-i\omega\delta\tilde{f}(\vec{p})$ for the last line. Oscillating terms $\propto e^{2i\omega t}, e^{-2i\omega t}$ arise that disappear in the time average. The remaining terms cancel, which can be directly seen, if replacing $\delta\tilde{f}(\vec{p})$ by the Lagrange multipliers \tilde{F}_p using Eq. (35). Thus the total entropy is constant in the average over a period of time, $d\bar{S}(t)/dt = 0$. However, even in the time average, there is an entropy production which is dissipated as entropy export due to the external field in the drift term. We have

$$\begin{aligned} \frac{d\bar{S}(t)}{dt} &= \dot{S}_{\text{ext}} + \dot{S}_{\text{int}} \\ &= -\frac{e\hbar}{2m}\beta\sum_p\tilde{F}_p^*f_p(1-f_p)\vec{p}\cdot\tilde{\vec{E}} \\ &\quad + \frac{1}{2}\sum_{pp'}\tilde{F}_p^*\mathcal{L}_{pp'}(\omega)\tilde{F}_{p'} + \text{c.c.} = 0. \end{aligned} \quad (44)$$

Therefore, let us consider the functional

$$\dot{S}_{\text{int}}[\tilde{G}_p] = \sum_{pp'}\tilde{G}_p^*\mathcal{L}_{pp'}(\omega)\tilde{G}_{p'} + \text{c.c.} \quad (45)$$

for any function \tilde{G}_p that obeys the constraint

$$\sum_p\tilde{G}_p^*\left[-D_p - i\Omega_p\tilde{G}_p + \sum_{p'}\mathcal{L}_{pp'}(\omega)\tilde{G}_{p'}\right] = 0 \quad (46)$$

that can be considered as an integral over the Boltzmann equation (42). It is easily shown that the time-averaged change

of entropy (43) vanishes for arbitrary functions \tilde{G}_p that obey the constraint (46). The maximum of the functional $\dot{S}_{\text{int}}[\tilde{G}_p]$ occurs at $\tilde{G}_p = \tilde{F}_p$, which is the solution of the linear Boltzmann equation (42); see Appendix C for the proof.

This is a generalization of the Kohler variational principle [31,32] for arbitrary frequencies ω . It can be related to the principle of extremum of entropy production given by Prigogine and Glandsdorff [33]. The static case $\omega = 0$ has been considered in Refs. [12,31,32,34]. Some attempts to extent this to arbitrary frequencies can be found in [35], but, to our knowledge, a consistent approach has not been given until now.

In order to apply the variational principle given here, one can consider a class of trial functions $\tilde{G}^{(N_v)}(\Phi_v; \vec{p}) = \sum_{v=1}^{N_v}\Phi_v g_v(\vec{p})$ with respect to an arbitrary but finite (N_v) set of linear independent functions $g_v(\vec{p})$. Determining the extremum of $\dot{S}_{\text{int}}[\Phi_v]$ leads to an optimal set of parameters, $\Phi_v^{\text{opt}} = F_v^{(N_v)}$. The extension of the class of trial functions to an infinite number of functions then gives the exact result, $\tilde{F}_p = \lim_{N_v \rightarrow \infty} \sum_{v=1}^{N_v} F_v^{(N_v)} g_v(\vec{p})$.

Alternatively, the relevant observables \hat{n}_p are replaced by a reduced set of N_v relevant observables, $\hat{B}_v = \sum_p g_v(\vec{p})\hat{n}_p$. The solution of the finite system of linear equations (A4) then gives the Lagrange multipliers F_v , which can be expressed in terms of determinants. This leads to identical results as for the variational principle. In previous papers, we used a finite number of moments, $g_v(\vec{p}) = \hbar p_E(\beta E_p)^{(v-1)/2}$, according to the general moments (8). An alternative basis set would be the Sonine polynomials [25] that are appropriate in the static,

nondegenerate limit. It has been shown that within perturbation expansion [36,37], results are converging with an increasing number of moments used.

B. One-moment Born approximation

In the lowest approximation, we choose, with $\tilde{G}_p = F_1 g_1(p) = F_1 \hbar p_E$, the first moment of the distribution function (8) as the trial function. The variational parameter F_1 is fixed by the auxiliary condition (46) where we insert Eq. (36) and Ω_p from Eq. (37), and we find

$$\begin{aligned} & \sum_p F_1 \hbar p_E \frac{e\hbar}{m} \beta f_p (1 - f_p) p_E \tilde{E} \\ &= -i\omega \sum_p (F_1 \hbar p_E)^2 f_p (1 - f_p) - \sum_{p,p'} F_1 \hbar p_E \mathcal{L}_{pp'}^{ei}(\omega) F_1 \hbar p'_E. \end{aligned} \quad (47)$$

The electron-electron collisions do not contribute in the one-moment approach because of conservation of total momentum. We assume the general structure of the variational parameter,

$$F_1 = \frac{e\beta}{m} \frac{1}{[-i\omega + \nu_D(\omega)]} \tilde{E}. \quad (48)$$

After some calculations, given in Appendix D, we find the collision frequency for the case of the statically screened Coulomb potential (16), and $S(q) \approx 1$,

$$\begin{aligned} \nu_D(\omega) &= i g_{\text{degen}} \int_0^\infty dy \frac{y^3}{(y^2 + \bar{n})^2} \int_{-\infty}^\infty \frac{dx}{x} \frac{1}{w + i\varepsilon - x} \\ &\times \ln \left[\frac{1 + e^{-(x/y - y)^2 + \beta\mu}}{1 + e^{-(x/y + y)^2 + \beta\mu}} \right], \end{aligned} \quad (49)$$

with

$$g_{\text{degen}} = \frac{1}{48\pi^4} \frac{e^4 m}{\epsilon_0^2 \hbar^3}, \quad w = \frac{\beta \hbar \omega}{4}, \quad \bar{n} = \frac{\beta \hbar^2 \kappa_D^2}{8m}, \quad (50)$$

which is valid for any degeneracy. In the nondegenerate limit $\beta\mu \ll 1$, we can expand the logarithm. With $e^{\beta\mu} = n(2\pi\beta\hbar^2/m)^{3/2}/2 = n\Lambda^3/2$ and spin factor 2, we find

$$\begin{aligned} \nu_D(\omega) &= i g n \int_0^\infty dy \frac{y^4}{(y^2 + \bar{n})^2} \\ &\times \int_{-\infty}^\infty dx \frac{1 - e^{-4xy}}{xy(w - xy + i\varepsilon)} e^{-(x-y)^2}, \end{aligned} \quad (51)$$

with $g = \Lambda^3 g_{\text{degen}}/2$.

The dynamical conductivity (10) can now be calculated with Eq. (35) and the optimized Lagrange parameter (48) so that $\tilde{F}_p = F_1 \hbar p_E$. We find

$$\sigma_D(\omega) = \frac{e}{m\tilde{E}} F_1 \frac{1}{\Omega_0} \sum_p (\hbar p_E)^2 f_p (1 - f_p). \quad (52)$$

For isotropic systems, the sum is evaluated as $\sum_p (\hbar p_E)^2 f_p (1 - f_p) = Nm/\beta$; see Appendix D. By inserting the derived expression (48), we obtain a generalized Drude-type expression (2),

$$\sigma_D(\omega) = \frac{\epsilon_0 \omega_{\text{pl}}^2}{-i\omega + \nu_D(\omega)} \quad (53)$$

for the dynamical conductivity. The comparison with σ_{KT} (21) will be performed in the following section.

It is instructive to investigate the alternative approach where only moments of the distribution function \hat{P}_v (8) are taken as relevant observables \hat{B}_n , instead of the fluctuations $\delta\hat{n}_p$ of the single-particle occupation operator as originally introduced in Sec. III B. Taking the component of the total momentum of the electrons, $\hat{P}_1 = \sum_p \hbar p_E \hat{n}_p$, in the direction of \tilde{E} as a one-moment approach, we have, with Eqs. (7) and (26),

$$\tilde{j} = \frac{e}{m\Omega_0} \langle \hat{P}_1 \rangle F_1 = \frac{e}{m\Omega_0} \langle \hat{P}_1, \hat{P}_1 \rangle F_1. \quad (54)$$

The generalized linear Boltzmann equation (31) is now reduced to a single equation that reads, in the Born approximation ($\langle \hat{P}_1 \rangle_0 = 0$ in thermal equilibrium),

$$[\langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\varepsilon} - i\omega \langle \hat{P}_1, \hat{P}_1 \rangle] F_1 = \langle \hat{P}_1, \hat{P}_1 \rangle \frac{e}{m} \beta \tilde{E}, \quad (55)$$

containing force-force correlation functions as the collision term. With $\langle \hat{P}_1, \hat{P}_1 \rangle = Nm/\beta$ [see Appendix D and the statically screened interaction (16)], the expressions for the dynamical conductivity (53) and the corresponding dynamical collision frequency

$$\nu_D^{(P_1)}(\omega) = \frac{\beta}{m n \Omega_0} \langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\varepsilon} \quad (56)$$

are obtained that coincide with the results (49) and (51) above. This is a preliminary result of the LRT based on the one-moment Born approximation. Going beyond the Born approximation, we denote $\nu^{(P_1)}(\omega) = \beta/(mN) \langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\varepsilon}$ as the collision frequency of the one-moment approach. Systematic treatments of the perturbation expansions are performed with the help of Green's function techniques. In particular, the Gould-DeWitt approximation for $\nu^{(P_1)}(\omega)$ has been performed that accounts for the correction of the long-range interaction by dynamical screening and considers strong collisions at short ranges [3,30].

C. Higher moment approaches

An improvement of the dynamical conductivity (53) can be achieved by extending the set of trial functions or relevant observables within the variational approach or the relevant statistical operator, respectively. Using higher-order moments \hat{P}_v (8) of the distribution function, converging expressions are obtained for the transport coefficients [37,38]. In particular, higher moments are needed in order to take into account electron-electron collisions. Taking higher-order moments into account, the change of the dynamical conductivity can be represented by a complex function $r(\omega)$ so that $\nu(\omega) = r(\omega)\nu^{(P_1)}(\omega)$ [3,4,39],

$$\sigma(\omega) = \frac{\epsilon_0 \omega_{\text{pl}}^2}{-i\omega + r(\omega)\nu^{(P_1)}(\omega)}. \quad (57)$$

As a special case, we discuss the two-moment approach with \hat{P}_1, \hat{P}_3 as relevant observables (i.e., particle current and energy current). The account of these two functions in p space allows for a better variational approach to the single-particle distribution function. For the electrical current density, we

have, with Eqs. (7) and (26),

$$\tilde{j} = \frac{e}{m\Omega_0} \langle \tilde{P}_1 \rangle = \frac{e}{m\Omega_0} \{ (\hat{P}_1, \hat{P}_1) F_1 + (\hat{P}_1, \hat{P}_3) F_3 \} = \sigma(\omega) \tilde{E}. \quad (58)$$

According to the response equations (28) [see also Eq. (55)], the Lagrange parameters F_1, F_2 are determined via the generalized linear Boltzmann equations, taken in the Born approximation,

$$\begin{aligned} [\langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\epsilon} - i\omega(\hat{P}_1, \hat{P}_1)] F_1 + [\langle \hat{P}_3; \hat{P}_3 \rangle_{\omega+i\epsilon} - i\omega(\hat{P}_1, \hat{P}_3)] F_3 &= (\hat{P}_1, \hat{P}_1) \frac{e}{m} \beta \tilde{E}, \\ [\langle \hat{P}_3; \hat{P}_1 \rangle_{\omega+i\epsilon} - i\omega(\hat{P}_3, \hat{P}_1)] F_1 + [\langle \hat{P}_3; \hat{P}_3 \rangle_{\omega+i\epsilon} - i\omega(\hat{P}_3, \hat{P}_3)] F_3 &= (\hat{P}_3, \hat{P}_1) \frac{e}{m} \beta \tilde{E}. \end{aligned} \quad (59)$$

As shown in Appendix D, we have $(\hat{P}_1, \hat{P}_1) = Nm/\beta$, $(\hat{P}_1, \hat{P}_3) = (\hat{P}_3, \hat{P}_1) = \frac{5}{2} Nm/\beta$, $(\hat{P}_3, \hat{P}_3) = \frac{5}{2} \frac{7}{2} Nm/\beta$. Using Cramer's rule, the response parameters F_1, F_2 are expressed in terms of the electrical field \tilde{E} and correlation functions. For the dynamical conductivity (58), we find, after algebraic manipulations, the expression (57) with

$$r(\omega) = \frac{\frac{5}{2} i\omega N \frac{m}{\beta} - \langle \hat{P}_3; \hat{P}_3 \rangle_{\omega+i\epsilon} + \frac{\langle \hat{P}_1; \hat{P}_3 \rangle_{\omega+i\epsilon} \langle \hat{P}_3; \hat{P}_1 \rangle_{\omega+i\epsilon}}{\langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\epsilon}}}{\frac{5}{2} i\omega N \frac{m}{\beta} - \frac{25}{4} \langle \hat{P}_1; \hat{P}_1 \rangle_{\omega+i\epsilon} + \frac{5}{2} \langle \hat{P}_1; \hat{P}_3 \rangle_{\omega+i\epsilon} + \frac{5}{2} \langle \hat{P}_3; \hat{P}_1 \rangle_{\omega+i\epsilon} - \langle \hat{P}_3; \hat{P}_3 \rangle_{\omega+i\epsilon}}. \quad (60)$$

An evaluation of the correlation functions occurring in the renormalization factor $r(\omega)$ in the Born approximation is given in Appendix E.

Results for the renormalization factor at solar core conditions and lower densities are shown in Fig. 1. At solar core conditions ($T = 573 \text{ eV} = 42.13 \text{ Ry}$, $n = 1.51 \times 10^{25} \text{ cm}^{-3} = 2.22 a_B^{-3}$), we have a weakly interacting [plasma parameter $\Gamma = e^2/(4\pi\epsilon_0 k_B T) (4\pi n/3)^{1/3} = 0.1$] and nearly degenerate [degeneration parameter $\Theta = 2mk_B T/\hbar^2 (3\pi^2 n)^{-2/3} = 1.3$] plasma. At the lower densities, the plasma becomes more classical. At high frequencies (i.e., large compared with the inverse relaxation time), $r(\omega)$ approaches 1, and higher moments of the momentum distribution that describe the deformation from a shifted Fermi distribution are not relevant. In the static case, the real part $\text{Re } r(0)$ shows the effect of e - e collisions according to the Spitzer result [4,36]. Since the Coulomb logarithm (17) depends on the density, in addition to the correct prefactor, the density dependence of the Coulomb

logarithm occurring in the different moments is also seen. Only in the very low-density limit, the different Coulomb logarithms cancel.

So far we have evaluated the equilibrium correlation functions occurring in the generalized linear Boltzmann equation (31) with the help of perturbation theory. Thus we solved a kinetic equation using a variational approach or a reduced set of relevant observables. Note that one can go beyond the kinetic equation that treats the single-particle distribution function by considering fluctuations in the two-particle states as additional relevant observables in the generalized LRT [19,40].

D. Limiting cases

1. Zero-frequency limit: Static conductivity

We rewrite the dynamical collision frequency (49) in a symmetric form by transforming $x \rightarrow -x$ in half of the

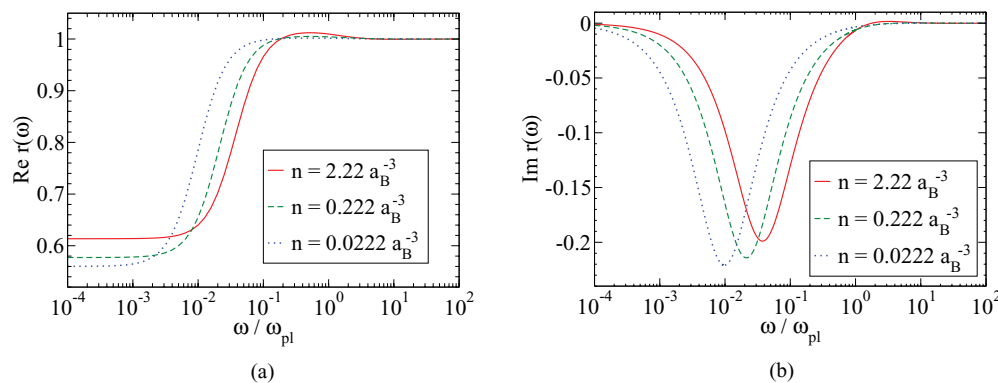


FIG. 1. (Color online) Frequency dependence of (a) the real part and (b) the imaginary part of the renormalization factor (60). Hydrogen plasmas at temperature $T = 42.13 \text{ Ry} = 573 \text{ eV}$ (solar core) and three different electron densities n are considered.

expression and using the Dirac identity,

$$\begin{aligned} \nu_D(\omega) = & \frac{g_{\text{degen}}}{2} \int_0^\infty dy \frac{y^3}{(y^2 + \bar{n})^2} \int_{-\infty}^\infty \frac{dx}{x} \left\{ \pi \delta(x - w) \right. \\ & \left. + \pi \delta(x + w) - iP \frac{1}{x - w} + iP \frac{1}{x + w} \right\} \\ & \times \ln \left[\frac{1 + e^{-(x/y-y)^2 + \beta\mu}}{1 + e^{-(x/y+y)^2 + \beta\mu}} \right]. \end{aligned} \quad (61)$$

The principal values compensate in the static case $w = 0$. After expanding for small x , $e^{-(x/y-y)^2 + \beta\mu} \approx e^{-y^2 + \beta\mu} [1 + 2x]$, the integral over x can be performed with the result

$$\lim_{\omega \rightarrow 0} \nu_D(\omega) = 2\pi g_{\text{degen}} \int_0^\infty dy \frac{y^3}{(y^2 + \bar{n})^2} \frac{1}{e^{y^2 - \beta\mu} + 1}. \quad (62)$$

Note that only e - i collisions contribute to the one-moment Born approximation.

First we discuss the Lorentz model. It is solved for the static case in KT using an energy-dependent relaxation time. The dc conductivity in the Born approximation for the one-moment approach (53), $\sigma_D(0) = \epsilon_0 \omega_{\text{pl}}^2 / \nu_D(0)$, is not identical with σ_{dc} obtained from Eq. (18) with the Coulomb logarithm (17) because $1/\nu_D(0) \neq \tau_{\text{Lorentz}}$. This difference stems from the fact that in the one-moment approach with the variational parameter F_1 , the p dependence is specified as $g_1(p) = \hbar p_E$. The p dependence necessary for the Lorentz model to reproduce the result for the relaxation time approach is given by $g_4(p)$ [see Eq. (8)], and is only roughly approximated by $g_1(p)$ within the interval of relevance. However, if we add further moments $g_v(p)$, not necessarily including $g_4(p)$, the approximation of the exact p dependence is improving. This has already been extensively investigated; see Refs. [37,41]. The dc conductivity within LRT follows from Eq. (57) as $\sigma(0) = \epsilon_0 \omega_{\text{pl}}^2 / [r(0) \nu^{(P_1)}(0)]$, with the static renormalization factor $r(0)$. The collision frequency $\nu^{(P_1)}(0)$ improves the Born approximation $\nu_D(0)$ if further effects like dynamical screening and strong collisions are included.

The equivalence of the KT and LRT for the Lorentz plasma in the static case $\omega = 0$ can be shown rigorously by inspection of the kinetic equation. Taking the linearized Boltzmann equation (41) with the collision term (38) and (39) in the static limit,

$$\begin{aligned} -\frac{e\hbar}{m} \beta f_p (1 - f_p) \vec{p} \cdot \vec{E} &= - \sum_{p'} \mathcal{L}_{pp'}^{ei}(\omega) F_{p'}, \quad (63) \\ &= \frac{2\pi}{\hbar} \sum_q |V_{ei}(q)|^2 \delta(E_{p+q} - E_p) \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} [F_{p+q} - F_p] \\ &= \frac{2\pi}{\hbar} \sum_q |V_{ei}(q)|^2 \delta(E_{p+q} - E_p) [\delta \tilde{f}(\vec{p} + \vec{q}) - \delta \tilde{f}(\vec{p})], \end{aligned} \quad (64)$$

where the expression (35) is used to insert the change of the single-particle distribution function $\delta \tilde{f}(\vec{p})$ after expanding $f_{p+q} - f_p \approx (\partial/\partial\beta E_p) f_p = -\beta(E_{p+q} - E_p) f_p (1 - f_p)$. This equation coincides with the equation of motion for the single-particle distribution function (11), which is obtained in the static case from KT and is solved using the relaxation time ansatz.

Considering the electron-ion plasma, it should be pointed out that the relaxation time approximation is not applicable if electron-electron collisions are relevant. In contrast, $\sigma(0)$ obtained from LRT contains also the contribution of electron-electron collisions as given by (40) in the static limit. For this, the static renormalization factor $r(0)$ can be evaluated from Eq. (60). In particular, it gives the correct Spitzer result if strong collisions are included [19,37,41]; see also Sec. IV C.

2. High-frequency limit: inverse bremsstrahlung absorption

The dielectric function $\epsilon(\omega) = [n_r(\omega) + ic/(2\omega)\alpha(\omega)]^{1/2}$ determines the refraction index $n_r(\omega)$ as well as the absorption coefficient $\alpha(\omega)$. We consider the long-wavelength limit where the transversal and longitudinal dielectric function coincide. The dielectric function or the optical conductivity $\sigma(\omega)$ can be used to calculate the inverse bremsstrahlung absorption. In the high-frequency limit, where $n_r(\omega) \approx 1$ and $\omega \gg \nu$, we have

$$\alpha(\omega) = \frac{\omega}{c n_r(\omega)} \text{Im} \epsilon(\omega) \approx \frac{\omega_{\text{pl}}^2}{\omega^2 c} \text{Re} \nu(\omega), \quad (65)$$

so that the inverse bremsstrahlung absorption coefficient is directly related to the dynamical collision frequency obtained above from the solution of the Boltzmann equation.

Bremsstrahlung radiation is described by the Bethe-Heitler expression resulting from QED in the second order of the interaction [42,43]. In the nonrelativistic limit and for soft photons, the absorption coefficient for a hydrogen plasma ($Z_i = 1$) is given by [44,45]

$$\begin{aligned} \alpha^{\text{Born}}(\omega) &= \frac{64\pi^{3/2} n^2 \sqrt{\beta}}{3\sqrt{2} m^{3/2} \hbar c \omega^3} \left(\frac{e^2}{4\pi\epsilon_0} \right)^3 \\ &\times \sinh\left(\frac{1}{2}\beta\hbar\omega\right) K_0\left(\frac{1}{2}\beta\hbar\omega\right), \end{aligned} \quad (66)$$

where $K_0(x) = \int_0^\infty dt \exp[-x \cosh(t)] = \int_0^\infty dy \exp[-y^2 - x^2/(4y^2)]/y$ is the modified Bessel function of the zeroth order.

Generalized LRT gives the same result. We use the collision frequency (51) in the nondegenerate case. At finite frequencies ω , the integral with $\bar{n} = 0$ is no longer divergent at $y = 0$. Therefore, the screening of the Coulomb potential can be neglected ($\bar{n} = 0$). We find [4,44]

$$\alpha^{\text{Born}}(\omega) = \frac{16\sqrt{2} \pi^{7/2} n^2 \sqrt{\beta}}{(3m)^{3/2} \hbar c \omega^3} \left(\frac{e^2}{4\pi\epsilon_0} \right)^3 (1 - e^{-\beta\hbar\omega}) g_{ff}^{\text{Born}}(\omega), \quad (67)$$

with the free-free Gaunt factor in the Born approximation,

$$g_{ff}^{\text{Born}}(\omega) = \frac{\sqrt{3}}{\pi^2} e^{\beta\hbar\omega/2} K_0\left(\frac{1}{2}\beta\hbar\omega\right). \quad (68)$$

The well-known Kramers formula for the inverse bremsstrahlung absorption [46] results with the Gaunt factor, $g_{ff}^{\text{Kramers}}(\omega) = 1$.

The one-moment Born approximation can be improved taking into account dynamical screening, strong collisions, and higher moments of the distribution function, as discussed earlier. However, in the high-frequency limit, the dynamical screening is not of relevance. The frequency dependence of

the renormalization factor has been discussed in [4] (see also Fig. 1), and converges to 1 in the high-frequency limit. Strong collisions have been considered and lead to the famous Sommerfeld result for the Gaunt factor [47,48]. For dense plasmas, the account of ion correlation $S(\vec{q})$ [see Eq. (3)] has a major effect and can directly be included in the Born approximation [49].

The standard treatment of the kinetic equation using a relaxation time ansatz (see Sec. II B) fails to describe the inverse bremsstrahlung absorption. The frequently used expression (21) for the dynamical conductivity, or the corresponding expression for the dielectric function, are restricted to the low-frequency region since a static but energy-dependent relaxation time cannot be applied to the high-frequency region. Different approaches using Fermi's golden rule have been used [13] to derive expressions for the emission of radiation. A common treatment unifying both limiting cases, $\omega \rightarrow 0$ and $\omega \rightarrow \infty$, is missing in KT within the relaxation time approximation.

In contrast, our approach within LRT covers the entire frequency regime consistently. Note that it can also be applied to the degenerate case and to the relativistic regime; see [50]. An important feature of LRT is the possibility to include medium effects in dense plasmas, such as the Landau-Pomeranchuk-Migdal effect [51].

E. Dimensionless dynamical conductivity

In the following, we use Rydberg units where $\hbar = 1$, $a_B = 1$, $m = 1/2$, $e^2/(4\pi\epsilon_0) = 2$, and $k_B = 1$. The temperature T is then given in Ry = 13.6 eV and the electron density n in a_B^{-3} . We introduce dimensionless quantities $\omega^* = \omega/\omega_{pl} \equiv \omega T/\sqrt{\pi n}$ and

$$\sigma^*(\omega) = \frac{e^2 \beta^{3/2} m^{1/2}}{(4\pi\epsilon_0)^2} \sigma(\omega). \quad (69)$$

In Fig. 2(a), the ratio of the kinetic theory to the linear response theory is shown for the real part of the dynamical conductivity at various parameter values. The one-moment approximation is used, corresponding to the force-force correlation function. In Fig. 2(b), the renormalization factor is included. In the low-frequency limit, deviations are shown

that are due to the inclusion of $e-e$ contributions. We give the limits of the expressions (E5) and (E4), given in Appendix E, in the static case,

$$\begin{aligned} \sigma_{KT}^*(\omega = 0) &= \frac{2^{5/2}}{\pi^{3/2}} \frac{1}{\Lambda_{KT}}, \\ \sigma_{LRT,1}^*(\omega = 0) &= \frac{3}{2^{5/2}\pi^{1/2}} \frac{1}{\Lambda_{LRT,1}}. \end{aligned} \quad (70)$$

In both approaches, the Coulomb logarithm behaves like $\lim_{n \rightarrow 0} \Lambda \sim -\frac{1}{2} \ln n$ in the low-density limit. At finite densities, different expressions are observed. The prefactor of the inverse Coulomb logarithm takes the value 1.015 for the Lorentz model that corresponds to KT in the relaxation time approximation. The Spitzer value 0.591 is approached in LRT considering the Born approximation (0.2992 in the one-moment case, 0.5781 in the two-moment case). This quick convergence is known from the literature; see [37]. The inclusion of the third moment of the momentum distribution takes the electron-electron interaction as well as transport of heat into account.

In the high-frequency limit, we find, from (E5) and (E4), the asymptotic expansions

$$\begin{aligned} \text{Re } \sigma_{KT}^*(\omega \rightarrow \infty) &= \frac{16\sqrt{2}n}{3\sqrt{\pi}T^3} \Lambda_{KT} \frac{1}{\omega^2}, \\ \text{Re } \sigma_{LRT,1}^*(\omega \rightarrow \infty) &= \frac{\sqrt{2}n^{1/4}}{3\pi^{5/4}T^{3/2}} \frac{1}{\omega^{7/2}}. \end{aligned} \quad (71)$$

The ratio between KT and LRT behaves as $\omega^{3/2}$. Thus, in the high-frequency limit, the ratio diverges; see Fig. 2. In conclusion, above the plasma frequency, the kinetic approach becomes essentially wrong.

V. CONCLUSION

Considering the interaction of radiation with matter, often a dielectric function or dynamical conductivity is used that is derived from kinetic theory using an energy-dependent relaxation time; see (21) and (E5). However, this expression is valid only for elastic collisions of electrons, so that electron-electron collisions cannot be included. Furthermore, the frequency dependence is not correctly described. In particular,

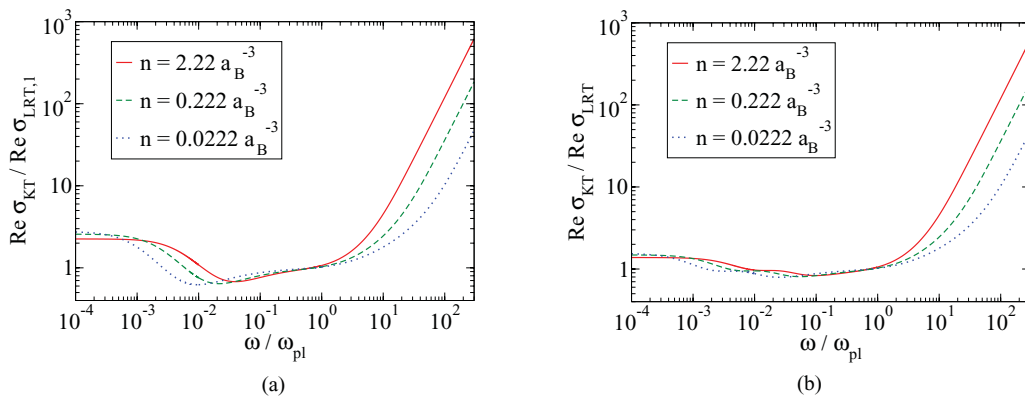


FIG. 2. (Color online) Ratio of the real part of the dynamical conductivity, calculated within the relaxation time ansatz (E5), in comparison to the generalized linear Boltzmann equation (LRT) in the (a) one-moment approximation (E4) and (b) two-moment approximation (E1). Hydrogen plasmas at temperature $T = 42.13$ Ry = 573 eV (solar core) and three different electron densities n are considered.

the high-frequency behavior has a wrong dependence on ω and fails to describe inverse bremsstrahlung. We developed an alternative approach that is free from these shortcomings.

We have derived a generalized linear Boltzmann equation (31) that is valid for any frequencies and at arbitrary degeneracy. Besides electron-ion interaction, also electron-electron interaction is included. The drift term and the collision term are expressed in terms of equilibrium correlation functions that are, in general, complex quantities. In order to apply this approach consistently, one has to deal with two problems as follows.

First, the correlation functions can be evaluated numerically or, using quantum statistical methods, in perturbation theory. As the simplest approximation, we considered the Born approximation; see Eq. (53) with Eqs. (49), (51), and (E1). This leads to analytic expressions that are tractable to be used for simple evaluations.

Second, solving the generalized linear Boltzmann equation, a variational principle has been applied that optimizes the single-particle distribution function within a subspace of trial functions. In particular, we considered a finite number of moments of the distribution function. The single-moment treatment gives a result for the dynamical conductivity that is improved if higher moments of the distribution functions are taken into account. The contribution of higher moments is represented by the renormalization factor $r(\omega)$ that is, in general, a complex quantity. The high-frequency limit is not modified by the inclusion of higher moments and reproduces the well-known results for bremsstrahlung. The static limit converges to the Spitzer result for the conductivity with the inclusion of higher moments that describe also the contribution of electron-electron interaction.

We compared both approaches for different plasma properties. In the case of the Lorentz plasma that takes into account only elastic scattering of electrons by the ions, the correct static conductivity is obtained in KT using an energy-dependent relaxation time. To get this result in LRT, the variational solution with only the lowest moment P_1 is not sufficient, and higher moments should be considered. In particular, the inclusion of the fourth moment P_4 alone gives the exact result for the static conductivity. The solution of KT with an energy-dependent relaxation time becomes increasingly inappropriate with higher frequencies. In contrast, the expressions obtained from LRT are applicable at any frequency.

Considering the more realistic case of the electron-ion plasma, the relaxation time ansatz to solve the kinetic equation breaks down. The inclusion of electron-electron collisions where the single-particle energy is not conserved represents no problem in LRT. The exact results for the transport coefficients in the low-density limit given by the Spitzer formula are reproduced by LRT, in contrast to KT. The correct treatment of inverse bremsstrahlung shows that LRT is valid in the entire frequency domain, in contrast to KT using the energy-dependent relaxation time that cannot reproduce the correct frequency dependence of the optical conductivity.

Starting from a general LRT, a linearized Boltzmann kinetic equation has been obtained, and the relation to the results of the relaxation time approach in the KT have been discussed. We restricted ourselves to a two-moment Born approximation. Possible improvements, as pointed out

throughout the paper, are summarized here again as an outlook to further considerations and calculations.

(a) Taking the single-particle occupation number n_p as relevant observables B_n , the deviations from equilibrium $\langle \hat{n}_p \rangle^t - f_0(\vec{p})$ describe the nonequilibrium state. The set of relevant observables can be extended by including initial state correlations, in particular the formation of bound states. This is straightforward in a general version of LRT; see, e.g., [41,52]. Sophisticated approaches have been worked out to show conservation of total energy and the systematic inclusion of correlations and bound-state formation, using nonequilibrium Green's function theory [53,54] or within generalized linear response theory [19,55]. This is of relevance to investigate partially ionized plasmas, but also allows for the treatment of quasiparticle formation and the Debye-Onsager relaxation effect.

(b) In linear response theory, the drift term and the collision term are expressed in terms of equilibrium correlation functions. They can be evaluated numerically or within perturbation theory if we expand with respect to the interaction. The Born approximation is improved if higher orders with respect to the interaction are taken into account. The technique of thermodynamic Green's functions has been used for the evaluation of equilibrium correlation functions [3,4]. The binary collision approximation is obtained if ladder diagrams are summed up. Dynamical screening results from the summation of ring diagrams. Perturbation expansions are more efficient if correlations are already included in the set of relevant observables so that they do not have to be generated by a dynamical treatment, i.e., by considering higher-order perturbation expansions. As an example, we refer to the formation of bound states discussed above. Instead of finding their influence using higher orders of perturbation theory, we can treat them as new degrees of freedom introducing the corresponding relevant observables, e.g., their distribution function or a finite number of moments. Then, memory effects become less important, and the Markov approximation can be used, e.g., introducing stochastic forces [17].

Equilibrium correlation functions that determine the transport coefficients can be calculated for arbitrary frequencies, degeneracy, electron-electron collisions, and including collective excitations. The frequency dependence and further aspects are disregarded if a relaxation time is introduced. The relaxation time approach is exact only in the case of elastic scattering, for instance of electrons by ions in the adiabatic limit. Electron-electron scattering as well as finite frequencies of the electric field cannot be treated by the relaxation time ansatz. Thus, the generalized linear Boltzmann equation obtained from linear response theory reproduces some well-known benchmarks such as the Spitzer result for the static conductivity of the fully ionized plasma or the Kramers formula for the bremsstrahlung.

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APPENDIX A: DERIVATION OF THE RESPONSE EQUATIONS

The Hermitian observables \hat{B}_n are assumed to conserve the total particle number so that the entropy operator $\hat{H} - \mu\hat{N}$ is replaced by the system's Hamiltonian \hat{H} in the λ dependence of the relevant statistical operator (25). Note that the averages are calculated with the equilibrium statistical operator that is known to us, and quantum statistical methods can be applied, such as Green's function techniques or numerical simulations, to evaluate it. Thus, in linear response theory, the Lagrange multipliers $F_n(t)$ can be eliminated using equilibrium correlation functions.

The relevant statistical operator serves as the initial condition to determine the nonequilibrium statistical operator $\rho(t)$. Further correlations are built up by the dynamical evolution

[17] with the total Hamiltonian $\hat{H}_{\text{tot}}^t = \hat{H} + \hat{H}_{\text{ext}}^t$,

$$\hat{\rho}(t) = \lim_{\epsilon \rightarrow 0} \epsilon \int_{-\infty}^t dt' e^{-\epsilon(t-t')} \hat{U}(t, t') \hat{\rho}_{\text{rel}}(t') \hat{U}^\dagger(t, t'), \quad (\text{A1})$$

with the time evolution operator $\hat{U}(t, t')$ given by $i\hbar(\partial/\partial t)\hat{U}(t, t') = \hat{H}_{\text{tot}}^t \hat{U}(t, t')$ and $\hat{U}(t, t) = 1$. The external perturbation to the system's Hamiltonian \hat{H} shall have the general form $\hat{H}_{\text{ext}}^t = \sum_j h_j(t) \hat{A}_j$. Decomposition of the time dependence of the field into Fourier components $h_j(t) = \tilde{h}_j(\omega) e^{-i\omega t} / 2 + \text{c.c.} = \text{Re}\{\tilde{h}_j(\omega) e^{-i\omega t}\}$ is particularly convenient in the linear response since the reaction of the system is the superposition of the reaction to different spectral components of the external perturbation. Subsequently, the time dependence of the response to each component will have the same frequency in the stationary case, i.e., $F_n(t) = \text{Re}\{\tilde{F}_n(\omega) e^{-i\omega t}\}$. In the following, we consider a fixed value ω for the frequency of the external perturbation.

We now perform a partial integration of the statistical operator (A1) and linearize with respect to the external fields \tilde{h}_j and the response parameters \tilde{F}_n ,

$$\begin{aligned} \hat{\rho}_{\text{irrel}}(t) &= \hat{\rho}(t) - \hat{\rho}_{\text{rel}}(t) = - \lim_{\epsilon \rightarrow 0} \int_{-\infty}^t dt' e^{-\epsilon(t-t')} e^{-i\hat{H}(t-t')/\hbar} \left\{ \frac{i}{\hbar} [\hat{H}_{\text{ext}}^{t'}, \hat{\rho}_0] \right. \\ &\quad \left. + \sum_n \int_0^1 d\lambda e^{-\beta\lambda(\hat{H} - \mu\hat{N})} \left(\frac{i}{\hbar} [\hat{H}, \delta\hat{B}_n] F_n(t') + \delta\hat{B}_n \frac{\partial}{\partial t'} F_n(t') \right) e^{\beta\lambda(\hat{H} - \mu\hat{N})} \right\} e^{i\hat{H}(t-t')/\hbar} \hat{\rho}_0. \end{aligned} \quad (\text{A2})$$

According to Eq. (24), we have $\text{Tr}\{B_n \hat{\rho}_{\text{irrel}}(t)\} = 0$; for details, see [4,29]. Finally, applying the Kubo identity

$$\beta \int_0^1 d\lambda e^{-\lambda\beta\hat{H}} [\hat{C}, \hat{H}] e^{\lambda\beta\hat{H}} \hat{\rho}_0 = \int_0^1 d\lambda \frac{d}{d\lambda} \hat{C}(-i\hbar\beta\lambda) \hat{\rho}_0 = [\hat{C}, \hat{\rho}_0], \quad (\text{A3})$$

with $\hat{C} = \hat{H}_{\text{ext}}^{t'}$, we find an expression that relates the response parameters \tilde{F}_n to the external fields \tilde{h}_j ,

$$\sum_m [\langle \hat{B}_n; \hat{B}_m \rangle_z - i\omega \langle \hat{B}_n; \delta\hat{B}_m \rangle_z] \tilde{F}_m = -\beta \sum_j \langle \hat{B}_n; \hat{A}_j \rangle_z \tilde{h}_j, \quad (\text{A4})$$

where the Laplace transform of the correlation functions (29) has been introduced. After partial integration, $-iz \langle \hat{A}; \hat{B} \rangle_z = (\hat{A}, \hat{B}) - \langle \hat{A}; \hat{B} \rangle_z = (\hat{A}, \hat{B}) + \langle \hat{A}; \hat{B} \rangle_z$, we arrive at the response equations (28) with the external perturbation $\hat{H}_{\text{ext}}^t = -e\hat{R} \cdot \vec{E}(t)$, $\hat{R} = \sum_i \hat{r}_i$, and $\hat{R} = \hat{P}/m$.

APPENDIX B: EVALUATION OF THE COLLISION TERM

We evaluate the Onsager coefficient $\mathcal{L}_{pp'}(\omega) = \langle \hat{n}_{p'}; \hat{n}_p \rangle_{\omega+i\epsilon}$, which occurs in the collision term (38) of the linearized equation of motion for the single-particle distribution function (31). By inserting the time derivative of the occupation number [using $V^*(-q) = V(q)$],

$$\hat{n}_p = \frac{i}{\hbar} [\hat{H}, \hat{n}_p] = \frac{i}{\hbar} \sum_q V_{ei}(q) [\hat{a}_{p+q}^\dagger \hat{a}_p - \hat{a}_p^\dagger \hat{a}_{p+q}] + \frac{i}{\hbar} \sum_{p'/q} V_{ee}(q) [\hat{a}_{p+q}^\dagger \hat{a}_{p'-q}^\dagger \hat{a}_{p'} \hat{a}_p - \hat{a}_p^\dagger \hat{a}_{p'}^\dagger \hat{a}_{p'-q} \hat{a}_{p+q}], \quad (\text{B1})$$

into Eqs. (27) and (29), we evaluate the correlation functions for the electron-ion contribution in the Born approximation:

$$\begin{aligned} \langle \hat{n}_p; \hat{n}_{p'} \rangle_{\omega+i\epsilon}^{ei} &= -\frac{1}{\hbar^2} \sum_{q,q'} V_{ei}(q) V_{ei}(q') \int_0^\infty dt e^{i(\omega+i\epsilon)t} \int_0^1 d\lambda \{ [\text{Tr}\{\rho_0 \hat{a}_{p+q}^\dagger \hat{a}_p \hat{a}_{p'+q}^\dagger \hat{a}_{p'}\} - \text{Tr}\{\rho_0 \hat{a}_{p+q}^\dagger \hat{a}_p \hat{a}_{p'}^\dagger \hat{a}_{p'+q}\}] e^{\frac{i}{\hbar}(E_{p+q}-E_p)(t-i\hbar\beta\lambda)} \\ &\quad - [\text{Tr}\{\rho_0 \hat{a}_p^\dagger \hat{a}_{p+q} \hat{a}_{p'+q}^\dagger \hat{a}_{p'}\} - \text{Tr}\{\rho_0 \hat{a}_p^\dagger \hat{a}_{p+q} \hat{a}_{p'}^\dagger \hat{a}_{p'+q}\}] e^{\frac{i}{\hbar}(E_p-E_{p+q})(t-i\hbar\beta\lambda)} \}. \end{aligned} \quad (\text{B2})$$

The λ integral can be executed. The application of the Wick theorem to the quantum statistical averages $\text{Tr}\{\rho_0 \dots\}$ leads to δ functions, in particular, $q = -q'$. Contributions with $q = 0$ cancel. We assume isotropic interaction $V(\vec{q}) = V(-\vec{q})$ and obtain

$$\begin{aligned} \mathcal{L}_{pp'}^{ei}(\omega) &= -\frac{1}{\hbar^2} \sum_q |V_{ei}(q)|^2 \frac{e^{\beta(E_{p+q}-E_p)} - 1}{\beta(E_{p+q} - E_p)} f_{p+q}(1 - f_p) \frac{-1}{i(\omega + i\epsilon) + i(E_{p+q} - E_p)/\hbar} [\delta_{p',p+q} - \delta_{p',p}] \\ &\quad + \frac{1}{\hbar^2} \sum_q |V_{ei}(q)|^2 \frac{e^{\beta(E_p - E_{p+q})} - 1}{\beta(E_p - E_{p+q})} f_p(1 - f_{p+q}) \frac{-1}{i(\omega + i\epsilon) + i(E_p - E_{p+q})/\hbar} [\delta_{p',p} - \delta_{p',p+q}] \\ &= -\frac{1}{\hbar^2} \sum_q |V_{ei}(q)|^2 \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} \left\{ \frac{i}{\omega + i\epsilon + (E_{p+q} - E_p)/\hbar} + \frac{i}{\omega + i\epsilon - (E_{p+q} - E_p)/\hbar} \right\} [\delta_{p',p+q} - \delta_{p',p}], \quad (\text{B3}) \end{aligned}$$

using $(e^{\beta(E_{p'}-E_p)} - 1)f_{p'}(1 - f_p) = f_p - f_{p'}$. Subsequently, the Onsager coefficient can be given as Eq. (39).

With this result, the collision term (38) for the Lorentz plasma reads

$$C_p^{ei} = \frac{1}{\hbar^2} \sum_q |V_{ei}(q)|^2 \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} \left\{ \frac{i}{\omega + i\epsilon + (E_p - E_{p-q})/\hbar} + \frac{i}{\omega + i\epsilon - (E_p - E_{p-q})/\hbar} \right\} (\tilde{F}_{p+q} - \tilde{F}_p), \quad (\text{B4})$$

which is now a frequency-dependent and complex quantity. We can eliminate the Lagrange multiplier \tilde{F}_p according to Eq. (35) in order to express the collision integral in terms of the single-particle distribution function.

A similar calculation gives the electron-electron contribution in the Born approximation:

$$\begin{aligned} \mathcal{L}_{pp'}^{ee}(\omega) &= -\frac{1}{\hbar^2} \sum_{p_1, q} V_{ee}(q) V_{ee,ex}(q; p, p_1) \\ &\quad \times \left\{ \frac{e^{\beta(E_{p+q} + E_{p_1-q} - E_{p_1} - E_p)} - 1}{\beta(E_{p+q} + E_{p_1-q} - E_{p_1} - E_p)} \frac{i}{\omega + i\epsilon - (E_{p+q} + E_{p_1-q} - E_{p_1} - E_p)/\hbar} f_{p+q} f_{p_1-q} (1 - f_{p_1})(1 - f_p) \right. \\ &\quad \left. + \frac{e^{\beta(E_p + E_{p_1} - E_{p_1-q} - E_{p+q})} - 1}{\beta(E_p + E_{p_1} - E_{p_1-q} - E_{p+q})} \frac{i}{\omega + i\epsilon - (E_p + E_{p_1} - E_{p_1-q} - E_{p+q})/\hbar} f_p f_{p_1} (1 - f_{p_1-q})(1 - f_{p+q}) \right\} \\ &\quad \times [\delta_{p',p+q} + \delta_{p',p_1-q} - \delta_{p',p_1} - \delta_{p',p}], \quad (\text{B5}) \end{aligned}$$

where $V_{ee,ex}(q; p, p_1) = V_{ee}(q) - \delta_{\sigma_1, \sigma_2} V_{ee}(|\vec{p}_1 - \vec{p} - \vec{q}|)$ is the exchange interaction with σ_i denoting the spin explicitly. The respective Onsager coefficient can be given as Eq. (40). It is easily seen from the final expressions (39) and (40) that the real part of the Onsager coefficient, $\mathcal{L}_{pp'}(\omega) = \mathcal{L}_{pp'}^{ei}(\omega) + \mathcal{L}_{pp'}^{ee}(\omega)$, is non-negative, $\text{Re } \mathcal{L}_{pp'}(\omega) \geq 0$.

APPENDIX C: PROOF OF THE VARIATIONAL SOLUTION

To begin with, we show that the entropy production (45),

$$\dot{S}_{\text{int}}[\tilde{G}_p] = \sum_{pp'} \tilde{G}_p^* [\mathcal{L}_{pp'}(\omega) + \mathcal{L}_{p'p}^*(\omega)] \tilde{G}_{p'} = \sum_{pp'} \tilde{G}_p^* \langle \hat{n}_p; \hat{n}_{p'} \rangle_{\omega+i\epsilon} \tilde{G}_{p'}, \quad (\text{C1})$$

as a functional of an arbitrary \tilde{G}_p , is positive definite. Using the spectral density of the operator $\hat{G} = \sum_p \tilde{G}_p \hat{n}_p$, we find

$$\dot{S}_{\text{int}}[\tilde{G}_p] = \langle \hat{G}; \hat{G} \rangle_{\omega+i\epsilon} = \frac{1}{Z_0} \sum_{nm} \frac{e^{-\beta E_m} - e^{-\beta E_n}}{\beta(E_n - E_m)} \pi \delta \left[\omega + \frac{1}{\hbar} (E_n - E_m) \right] | \langle n | \hat{G} | m \rangle |^2 \geq 0. \quad (\text{C2})$$

Now we consider the functional (C1) for the function $(\tilde{G}_p - \tilde{F}_p)$ and decompose

$$\dot{S}_{\text{int}}[(\tilde{G}_p - \tilde{F}_p)] = \dot{S}_{\text{int}}[\tilde{G}_p] - \sum_{pp'} [\tilde{G}_p^* \mathcal{L}_{pp'}(\omega) \tilde{F}_{p'} + \text{c.c.}] - \sum_{pp'} [\tilde{F}_p^* \mathcal{L}_{pp'}(\omega) \tilde{G}_{p'} + \text{c.c.}] + \dot{S}_{\text{int}}[\tilde{F}_p]. \quad (\text{C3})$$

Making use of the constraint (46), the first contribution is expressed as

$$\dot{S}_{\text{int}}[\tilde{G}_p] = \sum_p [\tilde{G}_p^* + \tilde{G}_p] D_p, \quad (\text{C4})$$

which is the terms with $i\Omega_p$ compensate. Since \tilde{F}_p solves the linear Boltzmann equation (42), the second contribution is transformed into

$$\sum_{pp'} \tilde{G}_p^* \mathcal{L}_{pp'}(\omega) \tilde{F}_{p'} + \text{c.c.} = \sum_p \tilde{G}_p^* D_p + \sum_p i\Omega_p \tilde{G}_p^* \tilde{F}_p + \text{c.c.} \quad (\text{C5})$$

For the transformation of the third term, we use the symmetry $\mathcal{L}_{pp'}(\omega) = \mathcal{L}_{p'p}(\omega)$ due to detailed balance, which can be seen easily from the explicit expressions (B3) and (B5). Furthermore, the proof of the reciprocity condition $\mathcal{L}_{pp'}(\omega) = \mathcal{L}_{pp'}^*(-\omega)$ can be shown generally using the eigenstates $|n\rangle$ of the system Hamiltonian,

$$\mathcal{L}_{pp'}(\omega) = \frac{1}{\hbar^2} \frac{1}{Z_0 \beta} \sum_{nm} \frac{e^{-\beta E_n} - e^{-\beta E_m}}{E_n - E_m} \frac{(E_n - E_m)^2}{i\omega - \epsilon - (i/\hbar)(E_n - E_m)} \langle n | \hat{n}_p | m \rangle \langle m | \hat{n}_{p'} | n \rangle, \quad (\text{C6})$$

interchanging n and m . Finally, we find

$$\sum_{pp'} \tilde{F}_p^* \mathcal{L}_{pp'}(\omega) \tilde{G}_{p'} = \sum_{pp'} [\tilde{G}_{p'}^* \mathcal{L}_{pp'}^*(\omega) \tilde{F}_p]^* = \sum_{pp'} [\tilde{G}_p^* \mathcal{L}_{pp'}(-\omega) \tilde{F}_{p'}]^* = \sum_p [D_p + i\Omega_p \tilde{F}_p^*] \tilde{G}_p. \quad (\text{C7})$$

We sum up all contributions in Eq. (C3) using Eqs. (C4), (C5), and (C7),

$$\dot{S}_{\text{int}}[(\tilde{G}_p - \tilde{F}_p)] = \dot{S}_{\text{int}}[\tilde{F}_p] - \dot{S}_{\text{int}}[\tilde{G}_p] \geq 0. \quad (\text{C8})$$

This is a positive definite expression due to Eq. (C2). Thus we find that the entropy production is maximal if the trial function \tilde{G}_p is the solution \tilde{F}_p of the Boltzmann equation.

APPENDIX D: EVALUATION OF EQUATION (47)

We execute the \vec{p} integration on the left-hand side of Eq. (47) with $p_E^2 = p^2/3$,

$$\begin{aligned} \frac{1}{3} \sum_p \hbar^2 p^2 f_p (1 - f_p) &= \frac{8\pi m}{3} \frac{\Omega_0}{(2\pi)^3} \int E_p \left(-\frac{\partial f_p}{\partial \beta E_p} \right) p^2 dp = -\frac{4\pi m}{3\beta} \frac{(2m)^{3/2}}{\hbar^3} \frac{\Omega_0}{(2\pi)^3} \int \frac{\partial f_p}{\partial E_p} E_p^{3/2} dE_p \\ &= \frac{2\pi m}{\beta} \frac{(2m)^{3/2}}{\hbar^3} \frac{\Omega_0}{(2\pi)^3} \int f_p E_p^{1/2} dE_p = \frac{m}{\beta} \frac{4\pi \Omega_0}{(2\pi)^3} \int f_p p^2 dp = \frac{m}{\beta} \sum_p f_p = \frac{Nm}{\beta}, \end{aligned} \quad (\text{D1})$$

after integration by parts. This is also identical to (\hat{P}_1, \hat{P}_1) , which is the Kubo scalar product (27) of the first moment (8).

In the collision term, which is the second term on the right-hand side of Eq. (47), we insert the expression (39). The sum over p' is immediately executed and gives q_E . The first contribution (from the δ function) as well as the third contribution (from the first principal part) are considered together and can be transformed by $\vec{q} \rightarrow -\vec{q}$, then $\vec{p} \rightarrow \vec{p} + \vec{q}$, so that they coincide with the second and fourth contributions, respectively. We find, after canceling some common factors,

$$e\hbar^2 N \tilde{E} = -i\omega \frac{m}{\beta} \hbar^2 N F_1 - \sum_q |V_{ei}(q)|^2 q_E^2 \sum_p \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} \frac{i}{\omega + i\epsilon + (E_{p+q} - E_p)/\hbar} F_1. \quad (\text{D2})$$

From Eq. (48), we find

$$v_D(\omega) = -\frac{\beta}{mN} \sum_{p,q} q_E^2 |V_{ei}(q)|^2 \frac{f_p - f_{p+q}}{\beta(E_{p+q} - E_p)} \frac{i}{\omega + i\epsilon + (E_{p+q} - E_p)/\hbar}. \quad (\text{D3})$$

We shift $\vec{p} \rightarrow \vec{p} - \vec{q}/2$ so that $E_{p+q/2} - E_{p-q/2} = \hbar^2 \vec{p} \cdot \vec{q}/m$ and with spin factor 2,

$$\begin{aligned} v_D(\omega) &= \frac{\beta}{mN} \sum_q q_E^2 |V_{ei}(q)|^2 \frac{m}{\beta \hbar^2 q} \frac{2\Omega_0}{(2\pi)^2} \int_{-\infty}^{\infty} ds \frac{1}{s} \frac{i}{\omega + i\epsilon + \hbar q s/m} \\ &\times \int_0^{\infty} r dr \frac{e^{\beta(\hbar^2/2m)(r^2+s^2+sq+q^2/4)-\beta\mu} - e^{\beta(\hbar^2/2m)(r^2+s^2-sq+q^2/4)-\beta\mu}}{(e^{\beta(\hbar^2/2m)(r^2+s^2+sq+q^2/4)-\beta\mu} + 1)(e^{\beta(\hbar^2/2m)(r^2+s^2-sq+q^2/4)-\beta\mu} + 1)}, \end{aligned} \quad (\text{D4})$$

where cylindrical coordinates with respect to the \vec{q} direction have been introduced. s is the component of \vec{p} in the \vec{q} direction, and r is the component orthogonal to this axis. The integral over r can be performed,

$$\frac{1}{2} \int_0^{\infty} dr^2 \frac{1}{e^{\beta(\hbar^2/2m)(r^2+s^2+sq+q^2/4)-\beta\mu} + 1} \frac{1}{e^{-\beta(\hbar^2/2m)(r^2+s^2-sq+q^2/4)+\beta\mu} + 1} = \frac{m}{\beta \hbar^2} \frac{1}{e^{\beta(\hbar^2/2m)sq} - 1} \ln \left[\frac{1 + e^{-\beta(\hbar^2/2m)(s-q/2)^2 + \beta\mu}}{1 + e^{-\beta(\hbar^2/2m)(s+q/2)^2 + \beta\mu}} \right]. \quad (\text{D5})$$

Furthermore, we neglect the ion correlation so that $S(\vec{q}) = 1$ for the structure factor. Note that the Born approximation (D4) is divergent at zero frequency. As is well known, this problem is solved if we go beyond the Born approximation and take into account higher-order contributions due to dynamical screening and strong collisions. This was already discussed by Landau and Lifshitz [56] and has been shown to be consistent using Green's function techniques; see [3,4,29]. In this way, the correct zero-frequency limit of the collision frequency is obtained. As shown in Ref. [57], alternatively, the Coulomb potential in

Eq. (D4) can be replaced by a statically screened potential, namely, the Debye potential (16), so that $|V_{ei}(q)|^2 \approx NV_D^2$. With $s = \sqrt{\frac{2m}{\beta\hbar^2}} \frac{x}{y}$, $q = \sqrt{\frac{8m}{\beta\hbar^2}} y$, expression (49) follows.

APPENDIX E: RENORMALIZATION FACTOR AND DYNAMICAL CONDUCTIVITY

We use Rydberg units, as introduced at the beginning of Sec. IV E and in Eq. (69). In LRT, the conductivity (57) within the one-moment Born approximation in the nondegenerate limit (51) gives ($w = \omega^* \sqrt{\pi n/T}$)

$$\sigma_{\text{LRT}}^* = -\sqrt{\frac{2n}{\pi T^3}} \left[i\omega^* - i \frac{2}{3\pi} \sqrt{\frac{n}{T^3}} r(w) \int_0^\infty dy \frac{y^4}{(y^2 + 2\pi n/T^2)^2} \int_{-\infty}^\infty dx \frac{1 - e^{-4xy}}{xy(w - xy + i\varepsilon)} e^{-(x-y)^2} \right]^{-1}. \quad (\text{E1})$$

The renormalization factor $r(w)$ is taken with the first and third moment of the distribution function (i.e., particle current and energy current). According to Eq. (60), generalized force-force correlation functions have to be calculated after decomposition: $\langle \dot{P}_l; \dot{P}_m \rangle_{\omega+i\varepsilon} = \langle \dot{P}_l^{ei}; \dot{P}_m^{ei} \rangle_{\omega+i\varepsilon} + \langle \dot{P}_l^{ee}; \dot{P}_m^{ee} \rangle_{\omega+i\varepsilon}$. Considering the nondegenerate limit of the Born approximation again, we have, from the electron-ion interaction,

$$\langle \dot{P}_l^{ei}; \dot{P}_m^{ei} \rangle_{\omega+i\varepsilon} = i \frac{4}{3\sqrt{\pi}} \frac{Nn}{\sqrt{T}} \int_0^\infty dy \frac{y^4}{(y^2 + 2\pi n/T^2)^2} \int_{-\infty}^\infty dx \frac{1 - e^{-4xy}}{xy(w - xy + i\varepsilon)} e^{-(x-y)^2} \{x, y\}_{lm}^{ei}, \quad (\text{E2})$$

where $\{x, y\}_{11}^{ei} = 1$, $\{x, y\}_{31}^{ei} = 1 + 3x^2 + y^2$, and $\{x, y\}_{33}^{ei} = 2 + 10x^2 + 9x^4 + 2y^2 + 6x^2y^2 + y^4$.

For the electron-electron interaction, we find

$$\langle \dot{P}_l^{ee}; \dot{P}_m^{ee} \rangle_{\omega+i\varepsilon} = -i \frac{4}{3\sqrt{2\pi}} \frac{Nn}{\sqrt{T}} \int_0^\infty dy \frac{y^4}{(y^2 + 4\pi n/T^2)^2} \int_{-\infty}^\infty dx \frac{1 - e^{-4xy}}{xy(w - xy + i\varepsilon)} e^{-(x-y)^2} \{x, y\}_{lm}^{ee}, \quad (\text{E3})$$

where due to momentum conservation ($\dot{P}_1^{ee} = 0$) we have $\{x, y\}_{11}^{ee} = \{x, y\}_{31}^{ee} = 0$ and $\{x, y\}_{33}^{ee} = 1 + (19/4)x^2$.

For the evaluation, we use $\frac{1}{xy-w-i\varepsilon} = \text{P} \frac{1}{xy-w} + i\pi \delta(xy-w)$. The δ function allows one to perform the integral over x to obtain the real part of the correlation functions, $\langle \dot{P}_l; \dot{P}_m \rangle_{\omega+i\varepsilon}$. For the imaginary part, we also can perform the x integral after partial fraction decomposition and using $\text{P} \int_{-\infty}^\infty dx \frac{e^{-x^2}}{x+a} = \pi e^{-a^2} \text{erfi}(a)$.

In particular, we have, for the single moment approximation where $r(w) = 1$,

$$\sigma_{\text{LRT},1}^* = -\sqrt{\frac{2n}{\pi T^3}} \left(i\omega^* - \frac{2}{3w} \sqrt{\frac{n}{T^3}} \int_0^\infty dy \frac{y^3}{(y^2 + \frac{2\pi n}{T^2})^2} \left\{ e^{-(y-\frac{w}{y})^2} - e^{-(y+\frac{w}{y})^2} - 2i \left[e^{-(y-\frac{w}{y})^2} \text{erfi}\left(y - \frac{w}{y}\right) - e^{-y^2} \text{erfi}(y) \right] \right\} \right)^{-1}. \quad (\text{E4})$$

For direct comparison, we give explicitly the dynamical conductivity from KT (21) with the energy-dependent relaxation time for the Lorentz plasma (17),

$$\sigma_{\text{KT}}^* = -\frac{8}{3\sqrt{\pi}} \sqrt{\frac{2n}{\pi T^3}} \frac{1}{T} \int_0^\infty dx \frac{x^4 e^{-x^2/T}}{i\omega^* - \sqrt{\pi n} [\ln(1+b) - b/(1+b)]/x^3}, \quad (\text{E5})$$

with $b = x^2 T / (2\pi n)$.

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