Kramers-Kronig relations for the dielectric function and the static Coulomb systems

V. B. Bobrov\(^{(a)}\), S. A. Trigger\(^{(b)}\), G. J. F. van Heijst\(^{2}\) and P. P. J. M. Schram\(^{2}\)

\(^{1}\) Joint Institute for High Temperatures, Russian Academy of Sciences - 13/19, Izhorskaia Str., Moscow 125412, Russia
\(^{2}\) Eindhoven University of Technology - P.O. Box 513, MB 5600 Eindhoven, The Netherlands, EU

received 10 March 2010; accepted in final form 30 March 2010
published online 19 April 2010
PACS 05.20.Dd – Kinetic theory
PACS 05.30.Fk – Fermion systems and electron gas
PACS 52.25.Mq – Dielectric properties

Abstract – The mutual influence of singularities of the dielectric permittivity \(\varepsilon(q, \omega)\) in a Coulomb system in two limiting cases \(\omega \rightarrow 0, q \rightarrow 0\) and \(q \rightarrow 0, \omega \rightarrow 0\) is established. It is shown that the dielectric permittivity \(\varepsilon(q \rightarrow 0, \omega)\) satisfies the Kramers-Kronig relations, which possesses the singularity due to a finite value of the static conductivity. This singularity is associated with the “long tails” of the time correlation functions.

Copyright © EPLA, 2010

Introduction. – The dielectric function \(\varepsilon(q, \omega)\) of a Coulomb system is one of the most important characteristics of matter [1–3]. Exact relations for the dielectric function (dielectric permittivity, abbreviated below as DP), which determine the general behavior of the DP as a function of the wave vector \(q\) (in the present paper we consider only the isotropic Coulomb system, therefore the vector \(q\) is replaced by its modulus \(q\)) and the frequency \(\omega\), have a special significance in the theory of Coulomb systems (CS). At first, it is necessary to mention the Kramers-Kronig relations (KKR). According to the theory of linear response \([2,3]\), the DP \(\varepsilon(q, \omega)\) of a homogeneous and isotropic Coulomb system in volume \(V\) at temperature \(T\) is determined for \(q \neq 0\) by the expression

\[
\frac{1}{\varepsilon(q, \omega)} = 1 + \frac{4\pi}{q^2} \chi^R(q, \omega + i0),
\]

where \(\chi^R(q, z)\) is the retarded “charge-charge” correlation function, analytical in the upper half-plane of the complex variable \(z (\text{Im} z > 0)\):

\[
\chi^R(q, z) = -\frac{i}{\hbar V} \int_0^\infty \text{dt} \exp(izt) \langle |p_q(t), \rho_{-q}(0)| \rangle = \frac{1}{V} \langle \rho_q | \rho_{-q} \rangle z.
\]

Here \(\rho_q(t)\) is the Fourier component of the charge density in the Heisenberg representation, and the angle brackets (\(\ldots\)) denoting the averaging in the grand canonical ensemble, containing charge particles of various species \(a\) with charge \(e_a\), mass \(m_a\), chemical potential \(\mu_a\) and average density \(n_a\). We assume that the quasi-neutrality condition

\[
\sum_a e_a n_a = 0
\]

is satisfied. The equality (2) has to be treated in the thermodynamic limit \(V \rightarrow \infty, \langle N_a \rangle \rightarrow \infty, n_a = \langle N_a \rangle / V \rightarrow \text{const}\), with \(N_a\) the operator of the total number of particles of species \(a\). On the basis of eqs. (1), (2) and the spectral representation for the function \(\chi^R(q, \omega + i0)\) it is easy to check the validity of the relations [1–3] for the charge-charge correlation function and the dielectric permittivity

\[
\begin{align*}
\Re \varepsilon(q, \omega) &= \Re \varepsilon(q, -\omega), \\
\Im \varepsilon(q, \omega) &= -\Im \varepsilon(q, -\omega),
\end{align*}
\]

(4)
the KKR can be written in the form (7), (8) on the condition that the functions $\varepsilon^{-1}(q, \omega)$ and $\varepsilon(q, \omega)$ have no singularities on the real axis $\omega$. In particular, if the conditions
\[ \lim_{\omega \to +0} \text{Im} \varepsilon^{-1}(q, \omega) = 0, \quad \lim_{\omega \to -0} \text{Im} \varepsilon(q, \omega) = 0 \] (9)
are satisfied, it is easy to derive from (4), (5) and (7) the inequality (6). The KKR for CS at arbitrary thermodynamic parameters are valid only for the inverse DP $\varepsilon^{-1}(q, \omega)$ [4]. The DP $\varepsilon(q, \omega)$ for arbitrary non-zero wave vectors does not obey the KKR without violation of the causality and stability conditions [4]. If the KKR are valid for the DP $\varepsilon(q, \omega)$ the value of the static DP $\varepsilon(q, 0)$ is restricted by the condition [1, 2]
\[ \varepsilon(q, 0) > 1. \] (10)
At the same time, according to (7), from the KKR for the inverse DP $\varepsilon^{-1}(q, \omega)$ negative values of the static DP $\varepsilon(q, 0)$ are also possible [4]:
\[ \varepsilon(q, 0) < 0. \] (11)
Moreover, the inequality (10) is the necessary and sufficient condition of validity of the KKR for the DP [4].

According to [4], eq. (10) can be violated in the following cases.

a) For the model of one-component plasma (OCP) (in the classical limit, as well as in the completely degenerated case). In the model under consideration, the violation is associated with the negativity of the compressibility $K_T$ and the pressure $P$ and with the well-known problem of the thermodynamic instability of the OCP. Evidently, in this case, the violation is formal and non-physical (in sense of an experimental realization of such equilibrium system).

b) For the model of classical two-component plasma (CTCP), the violation is caused by the instability of the classical system of charges of opposite signs in the case of the “pure” Coulomb interaction [3]. To eliminate this instability, various types of the model pseudo-potentials, with non-Coulomb short-range part are used (see, e.g., [3]). Therefore, the model of CTCP is not an example of the “pure” Coulomb system, which is under consideration in the present paper. This means that, in general, the condition (10) can be violated. However, this violation also is also formal and non-physical.

c) For the Coulomb system in solid state, which is an anisotropic system and, therefore, cannot be studied under conditions considered in this paper. For the solid state, instead of the function $\varepsilon(q, 0)$ a matrix arises in the space of the reciprocal vectors of the crystallized lattice. For such a system the condition of isotropy, considered in this paper, is not fulfilled.

The static DP, determined by the limit
\[ \varepsilon(q, 0) = \lim_{\omega \to 0} \text{Re} \varepsilon(q, \omega), \] (12)
characterizes (taking into account condition (9)) the equilibrium state of a Coulomb system in a weak electric field. The frequency dependence of the DP characterizes non-equilibrium processes in CS, connected, at first, with the existence of an electric current. These processes are described by the conductivity $\sigma(q, \omega)$, connected with the DP $\varepsilon(q, \omega)$ by the relation [1, 2]
\[ \varepsilon(q, \omega) = 1 + \frac{4\pi i}{\omega + i0} \sigma(q, \omega). \] (13)
As is well known, the conductivity characterizes the relation between the current density $J(q, \omega)$ and the electric field $E(q, \omega)$ in a material: $J(q, \omega) = \sigma(q, \omega)E(q, \omega)$.

The most interesting case is the action of a weakly inhomogeneous electric field ($q \to 0$). It is easy to see, on the basis of physical arguments [1], that the static conductivity is determined by the limit
\[ \sigma_{st} = \lim_{\omega \to 0} \lim_{q \to 0} \sigma(q, \omega) \geq 0. \] (14)

Take now into account that for all known substances, for non-zero temperatures, the static conductivity $\sigma_{st}$ has a finite non-zero value. Then from (13) and (14) it follows that
\[ \lim_{q \to 0} \lim_{\omega \to 0} \varepsilon(q, \omega) \neq \lim_{\omega \to 0} \lim_{q \to 0} \varepsilon(q, \omega). \] (15)

**Positivity** $\varepsilon(q \to 0, 0)$ and KKR for the DP. – Using the methods of quantum field theory and the diagram technique for the temperature Green functions [3, 5], the exact relations for the DP and the response function $\chi^R(q, z)$ can be established. In particular, it was shown in [3] that
\[ \chi^R(q, z) = \frac{\Pi(q, z)}{\varepsilon(q, z)}, \quad \varepsilon(q, z) = 1 - u(q)\Pi(q \cdot z), \] (16)
where the function $\Pi(q, z)$ is the so-called “charge-charge” polarization operator—the irreducible (in the $q$-channel on one line of interaction $u(q) = 4\pi/q^2$) part of the temperature “charge-charge” Green function. Therefore, the DP $\varepsilon(q, \omega)$ of a homogeneous and isotropic CS can be represented as
\[ \varepsilon(q, \omega) = 1 - u(q)\Pi(q, \omega + i0). \] (17)
It should be mentioned that the polarization operator $\Pi(q, \omega + i0)$ can be introduced without use the perturbation theory of the temperature diagram technique. In contrast with the Green function $\chi^R(q, \omega + i0)$, which determines the response of CS on a weak external field, the polarization operator $\Pi(q, \omega + i0)$ characterizes the response on the full field in a medium. It means in
Kramers-Kronig relations for the dielectric function and the static conductivity of Coulomb systems

general that the polarization operator is not an analytical function in the upper half-plane of the complex variable \( z \). Therefore, as follows from (16) and (17), the KKR for the DP \( \varepsilon(q, \omega) \) can be violated [4]. However, the polarization operator \( \Pi(q, z) \), in contrast with the Green function \( \chi_R(q, z) \), has no singularities for small values of wave vector \( q \), connected with the form of the interaction potential \( u(q) = 4\pi \rho^2(q) \) (in general this statement is true for normal CS). Owing to this circumstance the DP \( \varepsilon(q, \omega) \) for small wave vectors satisfies the following limiting relations [6–8]:

\[
\lim_{q \to 0} q^2 \varepsilon(q, 0) = 4\pi \sum_{a, b} z_a z_b \epsilon^2 \left( \frac{\partial n_a}{\partial \mu_b} \right)_{\mu_c, T},
\]

(18) \( \varepsilon(\omega) = \lim_{q \to 0} \varepsilon(q, \omega) = 1 - \frac{\omega^2}{(\omega + i0)^2} - \frac{\varphi(\omega + i0)}{\omega + i0}^2, \)

(19) \( \varphi(z) = \frac{4\pi}{3V} \left( \langle I^3 \rangle \right)_z, \text{ Im } z > 0, \)

(20) \( I^\beta = \sum_a z_a I^\beta_a, \quad I^\beta_a = \sum_p \frac{\hbar p_a}{m_a} a_p^+ a_p, \)

(21) \( \omega_p = \left( \sum_a 4\pi z_a^2 e^2 n_a/m_a \right)^{1/2}. \)

Here \( I^\beta \) is the operator of the total current, \( \omega_p \) is the plasma frequency. Using the grand canonical ensemble it is easy to see [9] that

\[
T \left( \frac{\partial n_a}{\partial \mu_b} \right)_{\mu_c, T} = \frac{1}{V} (\delta N_a \delta N_b), \quad \delta N_a = N_a - \langle N_a \rangle.
\]

(22)

Substitution of (22) into (18) leads for arbitrary thermodynamic parameters (in a normal CS) to the following result [10]:

\[
\lim_{q \to 0} q^2 \varepsilon(q, 0) = \kappa^2 = \frac{4\pi}{T} \left( \frac{Z^2}{V} \right) \geq 0, \quad Z = \sum_a z_a n_a.
\]

(23)

It is evident that the value \( \kappa^2 \) in the appropriate limiting cases of weakly interacting classical and weakly interacting degenerate CS corresponds to the inverse Debye and the inverse Thomas-Fermi screening length, respectively [3]. Therefore, for small wave vectors \( q \) the static DP of CS satisfies the inequality (10) \( \varepsilon(q \to 0, 0) > 0 \). On this basis we conclude that the function \( \varepsilon(\omega) \) (19) should satisfy the KKR.

Actually, by integrating (20) by parts, one can rewrite (19) in the form

\[
\varepsilon(\omega) = 1 + \frac{4\pi i \sigma(\omega)}{\omega + i0}, \quad \sigma(\omega) = \lim_{q \to 0} \sigma(q, \omega), \quad \varphi(\omega) = \lim_{z \to 0} \varphi(z),
\]

(24) \( \sigma(z) = -\frac{1}{3V} \left( \langle I^3 \rangle \right)_z, \quad P^\beta = \int r^3 \rho(r) \mathrm{d}r. \)

Here the operators \( \rho(r) \) and \( P^\beta \) are the charge density operator and the charge dipole moment operators, respectively, with

\[
\frac{dP^\beta(t)}{dt} = I^\beta(t), \quad \frac{4\pi i}{3HV} \left( \langle I^3 \rangle, P^\beta(0) \right) = \omega_p^2.
\]

(26)

Therefore, we show that the conductivity \( \sigma(\omega) \) (24)–(26) is determined by the Kubo formula [11] (see also [6,7]). The functions \( z(\varepsilon(z) - 1)/4\pi \nu \) and \( z^2(\varepsilon(z) - 1)/4\pi \nu \) are, according to eqs. (19), (20), (24) and (25), analytical functions in the upper half-plane of the complex variable \( z \) and satisfy the KKR in the traditional form, similar to (7), (8). The function \( \varepsilon(z) \) is also an analytical function in the upper half-plane of the complex variable \( z \). However, the notation of the KKR for this function is different from the traditional form. The reason for this difference is the singularity \( 4\pi i \sigma_{st}/\omega \) of the function \( \varepsilon(\omega) \), which lies on the real axis at small \( \omega \). This implies that the second condition (9) in the long-wavelength limit \( q \to 0 \) is violated. Therefore [9], the KKR for the function \( \varepsilon(\omega) \) takes the form

\[
\text{Re } \varepsilon(\omega) = 1 + P \int_{-\infty}^{\infty} \frac{\text{Im } \varepsilon(\xi)}{\pi} \frac{d\xi}{\xi - \omega},
\]

(27) \( \text{Im } \varepsilon(\omega) = -P \int_{-\infty}^{\infty} \left[ \text{Re } \varepsilon(\xi) - 1 \right] \frac{d\xi}{\pi} \frac{1}{\xi - \omega} + \frac{4\pi i \sigma_{st}}{\omega}. \)

(28)

Therefore, in the static limit \( \omega \to 0 \) we cannot put \( \omega = 0 \) in the integrals in (27), (28) and we arrive at restrictions similar to (6), (10) and (11). Moreover, the function \( \text{Re } \varepsilon(\omega) \) may also possess singularities in the limit \( \omega \to 0 \).

**Asymptotic behavior of time-dependent correlation functions and the static conductivity.** – To analyze the mentioned singularities, let us generalize the relations (19), (24) and (25), introducing the functions \( \varepsilon^{(0)}(\omega) \), \( \sigma^{(0)}(\omega) \) and \( \varphi^{(0)}(\omega) \), determined on the real axis \( \omega \)

\[
\varepsilon^{(0)}(\omega) = 1 + \frac{4\pi i \sigma^{(0)}(\omega)}{\omega} = 1 - \frac{\omega_p^2}{\omega^2} \frac{\varphi^{(0)}(\omega)}{\omega^2},
\]

(29) \( \sigma^{(0)}(\omega) = \int_{-\infty}^{\infty} \exp(i\omega t) \sigma(t) \mathrm{d}t, \quad \varphi^{(0)}(\omega) = \int_{-\infty}^{\infty} \exp(i\omega t) \varphi(t) \mathrm{d}t, \)

(30) \( \sigma(t) = \frac{i}{3HV} \left( \langle I^3(t) \rangle, P^0(0) \right), \quad \varphi(t) = \frac{i}{3HV} \left( \langle I^3(t) \rangle, I^3(0) \right). \)

(31)

Taking into account eq. (26) it is easy to check that \( \sigma(t) \) and \( \varphi(t) \) are real functions. The presence of singularities in the function \( \varepsilon^{(0)}(\omega) \) is a consequence of behavior of the time correlation functions \( \sigma(t) \) and \( \varphi(t) \) on a large time scale (in the limit \( t \to \infty \)). If the condition

\[
\lim_{t \to \infty} \int_{0}^{t} \sigma(t) \mathrm{d}t = 0
\]

(32)

is fulfilled, the function \( \varepsilon^{(0)}(\omega) \) has no singularities for \( \omega \to 0 \). However, actually the condition (32) is violated,
since the conductivity $\sigma(\omega \to 0)$ always has a finite value in a normal CS. Moreover, according to (29) and taking into account (26) we arrive at the equality
\[
\lim_{t \to \infty} \frac{1}{3TV} \int_0^t \sigma(t) dt - \sigma_{st} = \lim_{t \to \infty} \frac{i}{3TV} \langle [P^3(t), P^3(0)] \rangle. \tag{33}
\]

It should be stressed that in the classical limit ($\hbar \to 0$) eq. (33) can be represented in a form equivalent to the Einstein relation \cite{12} for the diffusion coefficient of a Brownian particle
\[
\sigma_{st} = \lim_{t \to \infty} \frac{1}{3TV} \langle [P^3(0), P^3(t)] \rangle. \tag{34}
\]

Therefore, the low-frequency expansion ($\omega \to 0$) of the functions $\sigma^{(0)}(\omega)$ and $\varphi^{(0)}(\omega)$ has an asymptotic character. This kind of behavior is possible only if the functions $\sigma(t)$ and $\varphi(t)$ tend to zero at large values of $t$ ($t \to \infty$), following some non-exponential law. This means that these functions possess so-called “long tails”. It should be noted that such long tails of correlation functions have also been observed in other systems \cite{12,13} than Coulomb systems.

Conclusions. – In the present paper it is shown that the DP $\varepsilon(q, 0)$ of a homogeneous and isotropic Coulomb system has a singularity $\kappa^2 q^2$ \cite{23} for small values $q$ ($q \to 0$) and satisfies the inequality $\varepsilon(q \to 0, 0) > 1$ for arbitrary thermodynamic parameters. It is shown that this inequality leads to the KKR relations for the DP $\varepsilon(\omega)$, which take the form \cite{27,28}. For low frequencies the function $\varepsilon(\omega) = \lim_{\omega \to 0} \varepsilon(q, \omega)$ has a singularity $4\pi i \sigma_{st}/\omega$, which is taken into account in the KKR.

One can assert that there is one-to-one correspondence between these two singularities if the respective coefficients $\kappa$ and $\sigma_{st}$ are non-zero
\[
\kappa \neq 0, \quad \sigma_{st} \neq 0. \tag{35}
\]

The value $\kappa^{-1}$ in \cite{23} characterizes the penetration depth of the electric field in a medium. As in the case of the “metal–dielectric” transition, on the basis of a conductivity analysis \cite{14}, one can assert that the difference between “metals” and “dielectrics” has a relative character, since all known dielectrics have a non-zero static conductivity at $T \neq 0$.

The analogous statement is true with respect to the penetration depth $\kappa^{-1} \sim \sqrt{TV/(Z^2)}$ \cite{15} of the electric field in a medium. In “metals”, the penetration depth is very small, while for “dielectrics” the penetration depth can be of the order of the macroscopic size of the system.

According to our consideration one can introduce the concept of the “true” dielectric when the conditions
\[
\kappa \to 0, \quad \sigma_{st} \to 0, \tag{36}
\]
are satisfied. According to \cite{23}, a homogeneous and isotropic Coulomb system is in a “true” dielectric state if the limiting relation
\[
\frac{4\pi(Z^2)}{TV} \to 0 \tag{37}
\]
is fulfilled. On the basis of the above analysis we establish that the static conductivity of a homogeneous and isotropic CS is determined by the “long tails” of the time-dependent correlation functions \cite{33,34}. According to eqs. (33) and (36), if the condition (37) is fulfilled and the CS is in the state of a “true” dielectric, the following limiting relation
\[
\lim_{t \to \infty} \frac{i}{3TV} \langle [P^3(t), P^3(0)] \rangle \to 0 \tag{38}
\]
has to be satisfied.

***

The authors (VBB and SAT) are thankful to the Netherlands Organization for Scientific Research (NWO) for support of this work in the framework of the grant 047.017.2006.007.

REFERENCES

1. Silin V. P. and Rukhadze A. A., Electromagnetic Properties of Plasma and Plasma-like Media (Gosatomiizdat, Moscow) 1961 (in Russian).
2. Pines D. and Nozières P., The Theory of Quantum Liquids, Normal Fermi Liquids (Benjamin, New York-Amsterdam) 1966.
3. Kraeft W.-D., Kremp D., Ebeling W. and Ropke G., Quantum Statistics of Charged Particle Systems (Akademie-Verlag, Berlin) 1986.
4. Dolgov O. V., Kirzhnits D. A. and Maksimov E. G., Rev. Mod. Phys., 53 (1981) 81.
5. Abrkosev A. A., Gorkov L. P. and Dzyaloshinski E. I., Methods of Quantum Field Theory in Statistical Physics (Doover Publications, Inc., New York) 1975.
6. Bobrov V. B., Ozrin V. D. and Trigger S. A., Physica A, 164 (1990) 453.
7. Bobrov V. B., Klyuchnikov N. I. and Trigger S. A., Theor. Math. Phys., 89 (1991) 263 (in Russian).
8. Bobrov V. B., Zh. Eksp. Teor. Fiz., 102 (1992) 1808 (in Russian).
9. Landau L. D. and Lifshitz E. M., Statistical Physics, part I (Nauka, Moscow) 1976 (in Russian).
10. Bobrov V. B. and Trigger S. A., Bulletin of the Lebedev Physical Institute, No. 2 (Lebedev P. N. Physics Institute of the Russian Academy of Sciences) 2010, p. 35; On the static dielectric function for Coulomb system in the long wavelength limit, arXiv:0911.1519v1 [cond-mat.stat-mech] November 2009.
11. Kubo R., J. Phys. Soc. Jpn., 12 (1957) 570.
12. Resibois P. and de Leener M., Classical Kinetic Theory of Fluids (Wiley, New York) 1977.
13. Balescu R., Equilibrium and Nonequilibrium Statistical Mechanics, Vol. 1 (Wiley, New York, London, Sydney, Toronto) 1975.
14. Alekseev V. A., Maksimov E. G., Ponomarev Ja. G and Khomskii D. I., Usp. Fiz. Nauk, 112 (1974) 173 (in Russian).