Charge Density Fluctuation in Ionic Mixtures

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SYNOPSIS

The total charge fluctuation spectrum of ionic mixtures is analyzed on the basis of the solution of the hypernetted-chain equation for the binary ionic mixtures. A simple but sufficiently accurate scaling law in the domain of intermediate coupling is obtained and the result is applied to the calculation of the bremsstrahlung emission and absorption rates in high density plasmas composed of ions of many species.

1. INTRODUCTION

The knowledge on the properties of the gas composed of charged particles is indispensable to understand the behavior of high temperature or high density matter: In these materials, atoms and molecules are either temperature ionized or pressure ionized and we have mixtures of classical ions and classical or degenerate electrons.

The correlations between ions are closely related to some of important properties of ionized matter. For example, the efficiency of electronic transport of momentum or energy is mainly determined by the rate of electron-ion scattering and thus related to the total charge density fluctuation due to ions.

The simplest model of such a charged particle system of is the classical one-component plasma (OCP), the system of classical ions of

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one species embedded in the Fermi sea of completely degenerate
electrons which provides a uniform neutralizing background. This
model system is characterized by one dimensionless parameter and
various quantities have already been analyzed. In this case, the
charge density fluctuation spectrum is simply proportional to the
ionic structure factor and accurate values of the latter have been
accumulated by numerical experiments and analytical approaches based
on integral equations.[1]

As a model of ionized matter, the system of multicomponent ions
in the uniform background may be closer to reality than OCP. This
system is called ionic mixture. In the case of ionic mixture,
however, physical properties cannot be characterized by a single
parameter, and it is almost impossible, even for binary mixtures, to
perform numerical experiments over many combinations of the parameters
and tabulate the results. It may therefore be useful to express
various quantities of ionic mixtures by known quantities of OCP.

For thermodynamic quantities of binary ionic mixtures, the so-
called linear law is known to give very accurate values in the domain
of strong coupling:[2]   Results of numerical experiments are
reproduced by this scaling within experimental errors. For the charge
density fluctuation spectrum, however, the scaling properties are not
known except for the case of weak coupling and their analyses are the
purpose of this paper. Since experimental data over wide range of
parameters are not available, we limit ourselves within the domain of
weak or intermediate coupling where approximate data can be obtained
by the hypernetted-chain (HNC) equation.

2.   CHARGE DENSITY FLUCTUATION SPECTRUM

We consider a mixture of ions (nuclei) in uniform neutralizing
background of electrons in thermal equilibrium at the temperature T.
We denote the charge, the number, and the number density of ions of
species α by $Z_\alpha e$, N_α, and n_α, respectively. The total number density
of ions is given by

$$n = \Sigma_\alpha n_\alpha .$$ (1)

We define the mean distance a between ions by

$$4\pi n a^3 / 3 = 1 .$$ (2)

The density fluctuation spectrum $S^{\alpha\beta}(k)$ of the species α and β is defined by

$$S^{\alpha\beta}(k) = \langle \rho^{\alpha}_{k} \rho^{\beta}_{-k} \rangle / (N_{\alpha} N_{\beta})^{1/2}. \tag{3}$$

Here

$$\rho^{\alpha}_{k} = \sum_{i=1}^{N_{\alpha}} \exp(-i\vec{k} \cdot \vec{r}_{(i,\alpha)}), \tag{4}$$

$\vec{r}_{(i,\alpha)}$ is the position of the i-th particle of species α, and $\langle \ \rangle$ denotes the statistical average. The pair correlation function $h^{\alpha\beta}(r)$ is related to $S^{\alpha\beta}(k)$ as

$$(n_{\alpha} n_{\beta})^{1/2} h^{\alpha\beta}(r) = (2\pi)^{-3} \int d\vec{k} [S^{\alpha\beta}(k) - \delta_{\alpha\beta}] \exp(i\vec{k} \cdot \vec{r}). \tag{5}$$

The fluctuation spectrum of the total charge density $e^2 S_c(k)$ is given by

$$e^2 S_c(k) = \Sigma_{\alpha\beta} Z_{\alpha} Z_{\beta} e^2 (n_{\alpha} n_{\beta}/n^2)^{1/2} S^{\alpha\beta}(k). \tag{6}$$

When the wave number is sufficiently large, the density fluctuation spectrum reduces to the values given by self correlation as

$$S^{\alpha\beta}(k) \simeq \delta_{\alpha\beta}. \tag{7}$$

The total charge fluctuation spectrum is thus given by

$$S_c(k) \simeq \langle Z^2 \rangle. \tag{8}$$

For small values of the wave number, the behavior of the total charge fluctuation is determined by the perfect screening sum rule[3] as

$$S_c(k) \simeq \langle Z^2 \rangle (k^2/k_D^2). \tag{9}$$

Here k_D is the Debye wave number defined by

$$k_D^2 = \langle Z^2 \rangle 4\pi n e^2 / k_B T \tag{10}$$

and k_B is the Boltzmann constant.

Since the repulsive interaction energy between ions becomes infinite when they approach to each other, the pair correlation function $h^{\alpha\beta}(r)$ has the property

$$h^{\alpha\beta}(r=0) = -1. \tag{11}$$

This leads to sum rules for the density fluctuation $S^{\alpha\beta}(k)$ and the total charge density fluctuation spectrum $e^2 S_c(k)$ as

$$(2\pi)^{-3}\int d(\vec{ka})[S^{\alpha\beta}(k)-\delta_{\alpha\beta}]=-(3/4\pi)(n_\alpha n_\beta/n^2)^{1/2}, \qquad (12)$$

$$(2\pi)^{-3}\int d(\vec{ka})[S_c(k)/\langle Z^2\rangle-1]=-(3/4\pi)\langle Z\rangle^2/\langle Z^2\rangle. \qquad (13)$$

The correlation energy e_c (per ion) is expressed by $S_c(k)$ as

$$e_c=(1/2)(2\pi)^{-3}\int d\vec{k}(4\pi e^2/k^2)[S_c(k)-\langle Z^2\rangle]. \qquad (14)$$

Some exact properties of this spectrum have been analyzed in relation to the correlation energy.[4]

3.   RANDOM PHASE APPROXIMATION

When the Coulomb coupling is sufficiently weak, the fluctuation spectrum is given by the random phase approximation (RPA) as

$$S_c(k)=\langle Z^2\rangle k^2/(k^2+k_D^2)=\langle Z^2\rangle(ka)^2/[(ka)^2+3\langle Z^2\rangle(e^2/k_BTa)] \qquad (15)$$

and the correlation energy is given by

$$e_c/k_BT=-(3^{1/2}/2)[\langle Z^2\rangle e^2/k_BTa]^{3/2}. \qquad (16)$$

These RPA values are written as

$$S_c(k)=\langle Z^2\rangle S^{OCP}(ka, \Gamma_{eff}), \qquad (17)$$

where $S^{OCP}(k)$ is the OCP structure factor and the effective coupling constant is given by

$$\Gamma_{eff}=\langle Z^2\rangle(e^2/k_BTa). \qquad (18)$$

The RPA value (15) is one of exact lower bounds for the charge density fluctuation spectrum and the correlation energy is exactly larger than (16).[4] Since the RPA correlation functions diverge to negative infinity at r=0 and do not satisfy (11), the property (12) is not satisfied. When the conditions $h^{\alpha\beta}(r)\geq-1$ are taken into account,

we have much improved lower bounds for thermodynamic quantities.[4]

4. STRONGLY COUPLED BINARY IONIC MIXTURES

We now consider a mixture of two kinds of ions. This system is characterized by three independent parameters which may be taken, for example, as

$$\Gamma = e^2/k_B Ta, \tag{19}$$

$\langle Z \rangle$, and $\langle Z^2 \rangle$.

In the domain of strong coupling, it is known that the thermodynamic quantities are accurately reproduced by the so-called linear law[2] as

$$e_c/k_B T \simeq (n_1/n)E[Z_1^{5/3}\langle Z\rangle^{1/3}\Gamma] + (n_2/n)E[Z_2^{5/3}\langle Z\rangle^{1/3}\Gamma]. \tag{20}$$

Here $E[\Gamma]$ is the OCP value for the coupling constant Γ. Since the OCP value is approximately given by[1]

$$E[\Gamma] \simeq -0.9\Gamma, \tag{21}$$

(20) suggests the scaling of the effective coupling constant

$$\Gamma_{eff} = \langle Z^{5/3}\rangle\langle Z\rangle^{1/3}\Gamma. \tag{22}$$

5. SCALING FOR INTERMEDIATE COUPLING

In the domain of intermediate coupling, the scaling of the effective coupling constant may show a crossover from (18) to (22). In order to analyze the behavior of charge density fluctuation spectrum, we solve the hypernetted-chain (HNC) equations for binary ionic mixtures.

The HNC equations are obtained from exact equations by neglecting the contributions of the elementary diagrams and it is known that they work as a good approximation for systems with the long range Coulomb interaction at least in the domain of weak or intermediate coupling.[1] We solve the HNC equations by separating the interaction potential into the long range and the short range parts following Ng.[5]

Obtaining the total charge density fluctuation spectrum, we

observe the behavior of the height and the position of the first peak
for various combinations of the parameters. Analyses are made on the
domain where $2 \leq Z_2/Z_1 \leq 10$, $0.1 \leq n_1/n \leq 0.9$, and the second and higher peaks
are not remarkable.

As a result, we find that the height of the first peak of
$S_c(k)/\langle z^2 \rangle$ approximately corresponds to that of OCP structure factor
with the coupling parameter

$$\Gamma_{eff} = \langle Z^2 \rangle \Gamma \qquad (23)$$

and the position k_1 approximately satisfies

$$[\langle Z^2 \rangle / \langle Z \rangle^2]^{1/3} k_1 a = const. \qquad (24)$$

We thus expect the charge density fluctuation spectrum follow the
scaling

$$S_c = \langle Z^2 \rangle S^{OCP}[(\langle Z^2 \rangle / \langle Z \rangle^2)^{1/3} ka; \ \langle Z^2 \rangle \Gamma]. \qquad (25)$$

In Figs.1a-1g', the results of the HNC equations are compared
with the above scaling. We see that the scaling (25) is satisfied
with sufficient accuracy except for the case where there exist small
number of ions with much larger Z compared with host ions. In the
latter case, the structure of the first peak is not simple and is not
reproduced by the OCP structure factor.

According to (25), the correlation energy of binary ionic mixture
is given by that of OCP as

$$e_c/k_B T = [\langle Z \rangle^2 / \langle Z^2 \rangle]^{1/3} E[\langle Z^2 \rangle \Gamma]. \qquad (26)$$

Values given by (16) and scaling (26) are compared with experiments[2]
in Table 1. We see that (26) reproduces experimental values with
sufficient accuracy.

The scaling (23) is identical with (18): The mixture is still in
the domain where effective coupling parameter follows the scaling in
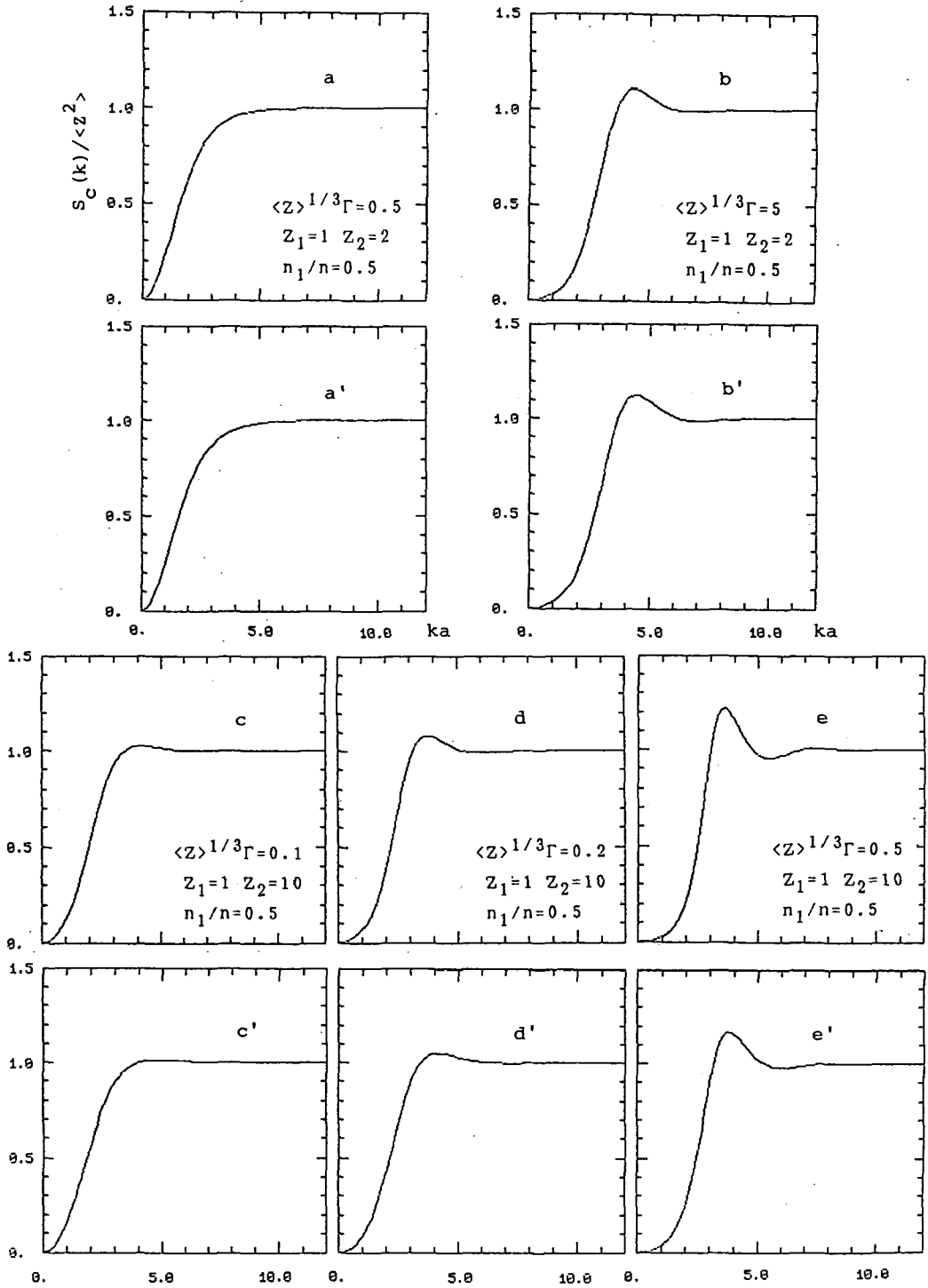the domain of weak coupling.

The scaling (24) is a result of the sum rule (13). Since the OCP
structure factor itself satisfies the sum rule (12) as

$$(2\pi)^{-3} \int d(\vec{k}a) [S^{OCP}(ka; \ \Gamma) - \delta_{\alpha\beta}] = -(3/4\pi), \qquad (27)$$

the wave number should necessarily be scaled by (24) in order to
satisfy the sum rule (13):

$$(2\pi)^{-3} \int d(\vec{k}a) \{S^{OCP}[(\langle Z^2 \rangle / \langle Z \rangle^2)^{1/3} ka; \ \langle Z^2 \rangle \Gamma] - 1\} = -(3/4\pi)\langle Z \rangle^2 / \langle Z^2 \rangle. \qquad (28)$$

Fig.1. Charge density fluctuation spectrum $S_c(k)/\langle z^2 \rangle$ (a-g) vs.
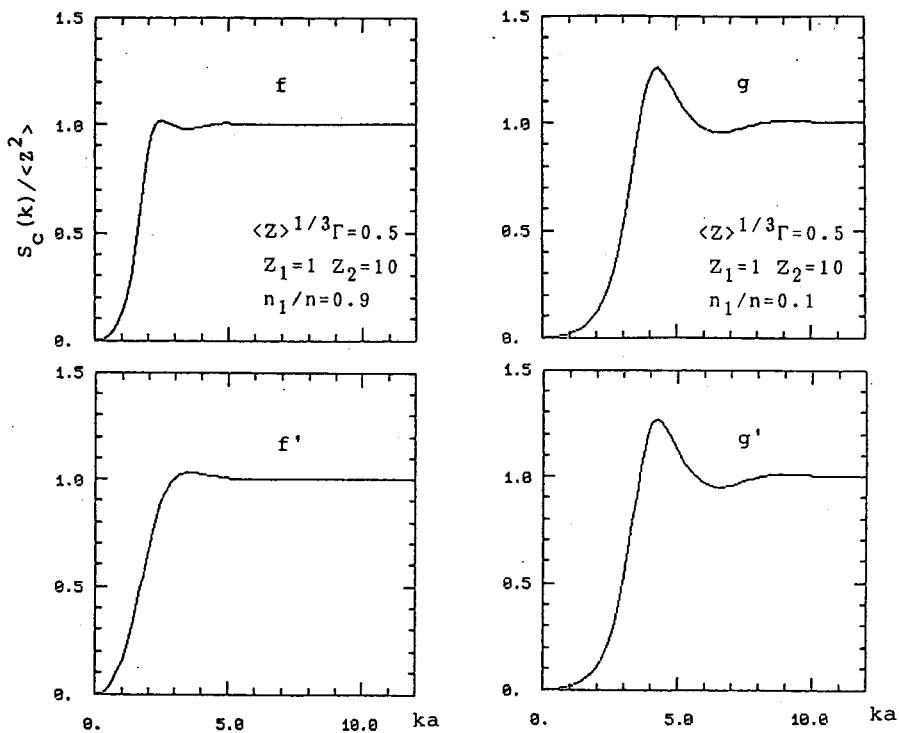interpolation formula (25) (a'-g').

Table 1. Normalized correlation energy $e_c/k_B T$. Experiments vs. interpolation formula (26) ($Z_1=1$, $Z_2=2$).

Γ	n_1/n	$\langle Z^2 \rangle$	$\langle Z \rangle$	exp.	(26)	RPA(16)
1	1/2	2.5	1.5	−1.645	−1.654	−3.423
1	5/6	1.5	7/6	−0.907	−0.908	−1.591
5	1/2	2.5	1.5	−9.715	−9.796	−38.27
5	5/6	1.5	7/6	−5.601	−5.672	−17.79
10	1/2	2.5	1.5	−20.13	−20.32	
10	5/6	1.5	7/6	−11.73	−11.92	

Since (25) is based on the weak coupling scaling (18) and the sum rule (13) neither of which are special to the case of binary mixtures, we expect it to hold also in the case of more complicated mixtures.

6. APPLICATION

As an application of the scaling of the total charge density fluctuation, we consider the effect of ion correlation on the bremsstrahlung in high density plasmas.

When an electron is scattered by an ion (nucleus) and changes its momentum, the emission or absorption of photon occurs with some probability. This is called bremsstrahlung or inverse bremsstrahlung and is one of important processes which determine the energy transport in plasmas. When the electron temperature is about 1keV, for example, the main part of the emission or absorption spectrum is in the range of soft X-ray.

In low density plasmas, the total emission from unit volume of plasma is obtained by simply multiplying the emission (integrated over distribution of electron momentum) by the number density of ions; the emission is proportional to the number of target ions.

In high density plasmas, however, the ions are correlated by mutual Coulomb interactions. As a result, the number of targets is effectively decreased or increased compared with the case where the same number of ions are distributed at random.[6,7]

For one-component plasmas, the ion correlation modifies the bremsstrahlung emission or absorption rate through the factor R^{OCP} which includes the ionic structure factor $S(k)$ as[7]

$$R^{OCP} = \int dk [S(k)F(k)/k]/\int dk[F(k)/k], \qquad (29)$$

$$F(k) \simeq \ln\{1+\exp[\mu/k_B T_e - (\hbar^2/2mk_B T_e)(k/2 - m\omega/\hbar k)^2]\}$$
$$- \ln\{1+\exp[\mu/k_B T_e - (\hbar^2/2mk_B T_e)(k/2 + m\omega/\hbar k)^2]\}. \qquad (30)$$

Here $\hbar$ is the Planck constant, m the electronic mass, T_e the electron temperature, μ the chemical potential of electrons, and ω the frequency of photon. When the Coulomb coupling between ions is weak

or intermediate, the effect of ion correlation usually reduces the probability of emission. The parameter dependence of the factor R^{OCP} is approximately given by

$$R^{OCP} \simeq R^{OCP}[\hbar\omega/k_B T; \ Z^2\Gamma, \ Z^{2/3}k_B T_e/E_F], \tag{31}$$

where E_F is the Fermi energy of electrons.

In plasmas composed of multi-component ions, the rate of bremsstrahlung emission or absorption is determined by the total charge density of ions. When ions are not correlated, the rate is the same as if we have ions of a single species with the charge number $\langle Z^2 \rangle^{1/2}$ is distributed at random with the total number density n. Ion correlations modify the rate by a factor R compared with the case of random ions. When we apply the scaling (25) for the total charge density of ions, this factor is expressed by that of OCP as

$$R \simeq R^{OCP}[\hbar\omega/k_B T; \ \langle Z^2 \rangle\Gamma, \ (\langle Z^2 \rangle/\langle Z \rangle)^{2/3}k_B T_e/E_F] \tag{32}$$

This formula gives the factor R as a function of frequency with parameters Γ, the average charge number $\langle Z \rangle$, and the average of the square of the charge number $\langle Z^2 \rangle$.

In order to realize the controlled thermonuclear fusion, experimental investigations have been made on the possibility of compression and heating of the inertially confined fuel (deuteron and triton) by intense laser beam or particle beam. The target includes layers of heavy materials as well as the fuel and we have high density and high temperature mixture of ions in the process of compression. To analyze the transfer of energy by radiation in such a mixture, our results may be useful.

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