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Viscoelastic description of electron subsystem of a semi-bounded metal within generalized "jellium" model

P.P. Kostrobij[1], B.M. Markovych[1], A.I. Vasylenko[2], M.V. Tokarchuk[1,2]

[1] Lviv Polytechnic National University, 12 Bandera Str., 79013 Lviv, Ukraine
[2] Institute for Condensed Matter Physics of the National Academy of Sciences of Ukraine, 1 Svientsitskii Str., 79011 Lviv, Ukraine

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Viscoelastic description of the electron subsystem of a semi-bounded metal on the basis of the generalized "jellium" model using the method of nonequilibrium statistical Zubarev operator is proposed. The nonequilibrium statistical operator and the quasi-equilibrium partition function calculated by means of the functional integration method are obtained. Transport equations for nonequilibrium mean values of electron density and momentum are received in the Gaussian approximation and in the following higher approximation that corresponds to the third-order cumulant averages in calculation of the quasi-equilibrium partition function.

Key words: *generalized "jellium" model, nonequilibrium statistical Zubarev operator, semi-bounded metal, transport equation, quasi-equilibrium partition function*

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1. Introduction

Today, the studies of diffusion mechanism and catalytic reactions of adsorbed atoms, formation of nanostructures on a metal surface are quite topical in surface physics [1–9]. In the processes of adsorption, desorption, and surface diffusion, a metal surface undergoes a reconstruction accompanied by a variation of nonequilibrium properties of both electron and ion subsystems. In this case, the electro-diffusive, viscothermal, and electromagnetic properties of the electron subsystem change in the field of metal surface ions. Studies of transport processes and their particularities in the electron subsystem of semi-bounded metals are of great importance for nanotechnologies and catalytic technologies. Generally, the electron subsystem stays in the states far from equilibrium during transport processes. This fact significantly complicates the electron subsystem description. To discover transport processes, various theoretical approaches are developed for the spatially inhomogeneous electrons-atoms systems. Particularly, the time-dependent density functional theory (TDDFT) is widely used [10–22]. Another theoretical approach is related to the hydrodynamic model of surface plasmons for a spatially inhomogeneous electron gas proposed in [23–25] with the use of the response theory [26] based on the Boltzmann kinetic equation. The quantum statistical theory for the description of nonequilibrium processes in the "metal–adsorbate–gas" systems was developed in the works [8, 27, 28] using the Zubarev method of nonequilibrium statistical operator (NSO) [29, 30]. In particular, a self-consistent description of nonequilibrium processes in the atomic and the electron subsystems was presented in [27] at the kinetic level of the description of electron processes. To study the ionic and electron structures of a semi-bounded metal, a generalized approach that takes the effect of discreteness of the ion subsystem into account and is based upon the model of a semi-bounded "jellium" [31, 32] was proposed in [8, 28]. It is worth noting that the effect of discreteness of ionic density on the characteristics of a semi-bounded "jellium" was considered in [33–35] by means of constructing a perturbation theory with respect to the electron-ion interaction pseudopotential. However, the linear response of the electron subsystem to the lattice potential does not take into account the effects of inhomogeneity of the electron subsystem. The approach described in [28, 31, 32] allows one to model the formation of a surface potential and to calculate the partition function for the generalized model in terms of the cumulant averages of the "jellium" model. In [28], the generalized "jellium" model is a basis for the statistical description of electro-diffusive processes for the electron subsystem of a semi-bounded metal with the use of the NSO method, where the single parameter of the reduced description is the nonequilibrium average value of the electron density operator. For such a system, the quasi-equilibrium statistical sum was calculated by means of the functional integration method for the case of an electron-ion interaction pseudopotential. In principle, it enables us to obtain expressions for the nonequilibrium statistical operator in the Gaussian and in the higher approximations with respect to the dynamic electron correlations. In [28], the nonequilibrium statistical operator and the generalized transport equation of inhomogeneous diffusion were obtained for weakly nonequilibrium processes (linear approximation with respect to the gradient of the electrochemical potential). The same approximation is used to find the equation for the "density-density" time correlation function that determines the dynamic structural factor of the electron subsystem of a semi-bounded metal and to demonstrate the connection of this electro-diffusive model in the linear approximation with the TDDFT [10–13]. The expressions of a nonequilibrium statistical operator in the Gaussian and higher approximations with respect to the dynamic electron correlations with the quasi-equilibrium partition function calculated by means of the functional integration method were obtained in [36]. It enables us to go beyond the linear approximation with respect to the gradient of electrochemical potential. The generalized transport equations for a nonequilibrium average of electron density operator for strongly nonequilibrium processes in an electron subsystem of a semi-bounded metal were presented for the corresponding approximations of the nonequilibrium statistical operator.

In this paper we perform a viscoelastic description of the electron subsystem of a semi-bounded metal on the basis of the generalized "jellium" model, when the nonequilibrium mean values of the electron density operator and the electron momentum density operator are chosen for reduced description parameters. These parameters also play an important role in TDDFT [15, 21]. In the second section we find the nonequilibrium statistical operator of a viscoelastic model for an electron subsystem of a semi-bounded metal. The general calculation of a quasi-equilibrium partition function for a quasi-equilibrium statistical operator of an electron subsystem of a semi-bounded metal is obtained using a functional integration method. The found quasi-equilibrium statistical operator of the electron subsystem is used for the construction of a nonequilibrium statistical operator of the system by means of the Zubarev method [29, 30]. In the third section we work with the Gaussian approximation for a quasi-equilibrium partition function, when the operators of electron density and electron momentum density do not correlate as pair correlation functions. Moreover, we receive a nonequilibrium statistical operator and the corresponding equations of generalized dynamics of the viscoelastic model for nonequilibrium averages of the operators of electron density and their momentum density in the Gaussian approximation. In the section 4 we use the following higher approximation for a quasi-equilibrium partition function, when static correlations between the operators of electron density and electron momentum density occur with the third-order cumulant averages. In this approximation, the nonequilibrium statistical operator and the non-linear dynamics equations of the viscoelastic model of an electron subsystem of a semi-bounded metal is obtained.

2. Nonequilibrium statistical operator for the electron subsystem of a semi-bounded metal within the viscoelastic model

We start with the generalized "jellium" model of a semi-bounded metal that considers the effect of the ionic subsystem discreteness. The Hamiltonian of the system could be written as follows:

$$H = \sum_{\mathbf{p},\alpha} E_\alpha(\mathbf{p}) a^\dagger_\alpha(\mathbf{p}) a_\alpha(\mathbf{p}) + \frac{1}{2SL}{\sum_{\mathbf{q}}}' \sum_{k} \nu_k(\mathbf{q}) \rho_k(\mathbf{q}) \rho_{-k}(-\mathbf{q})$$

$$
\begin{aligned}
&-\frac{\mathcal{Z}N_{\rm ion}}{SL}\sum_{\mathbf{q}}\sum_{k}\nu_k(\mathbf{q})S_k(\mathbf{q})\rho_k(\mathbf{q})+\frac{eN_{\rm ion}}{SL}\sum_{\mathbf{q}}\sum_{k}S_k(\mathbf{q})f_k(\mathbf{q})\rho_k(\mathbf{q})\\
&-\frac{N}{2S}\sum_{\mathbf{q}}{}'\nu(\mathbf{q}|0)+\frac{1}{2}\sum_{i\neq j=1}^{N_{\rm ion}}\frac{1}{S}\sum_{\mathbf{q}}{}'\mathcal{Z}^2\nu(\mathbf{q}|Z_i-Z_j)\,\mathrm{e}^{\mathrm{i}\mathbf{q}(\mathbf{R}_{\|i}-\mathbf{R}_{\|j})},
\end{aligned}
\tag{2.1}
$$

where $-e$ is the charge of an electron, m stands for the electron mass, $\mathbf{r}_i$, $i=1,2,\ldots,N$ is the electron coordinate, $N_{\rm ion}$ means the number of ions in a metal with relevant charges $\mathcal{Z}e$ and coordinates $\mathbf{R}_j$; $(-\infty<X_j,Y_j<+\infty$, $Z_j\leqslant Z_0$, $Z_0=\text{const})$, $z=Z_0$ is a dividing surface, $V=SL$ is a volume of the system, S means a surface square of a semi-bounded metal, L denotes an area of a normal coordinate changing: $z\in(-L/2,+L/2)$, $S\to\infty$, $L\to\infty$. $E_\alpha(\mathbf{p})=\frac{\hbar^2p^2}{2m}+\varepsilon_\alpha$ is the electron energy in the state $(\mathbf{p},\alpha)$. The hachure at sum denotes the absence of terms with $\mathbf{q}=0$ (two-dimension wavevector in a semi-bounded metal surface) due to the electro-neutrality condition $\mathcal{Z}N_{\rm ion}=N$. $\nu_k(\mathbf{q})=4\pi e^2/(q^2+k^2)$ is the Fourier-image of interaction potential between electrons, $f_k(\mathbf{q})$ is the Fourier-image of the model pseudopotential of interaction between ions and electrons $w(\mathbf{r}_i-\mathbf{R}_j)=-\frac{\mathcal{Z}e}{|\mathbf{r}_i-\mathbf{R}_j|}+f(\mathbf{r}_i-\mathbf{R}_j)$, $\mathbf{R}_{\|j}=(X_j,Y_j)$, $\nu(\mathbf{q}|z)=2\pi e^2\,\mathrm{e}^{-q|z|}/q$ means the two-dimension Fourier-image of the Coulomb potential,

$$
S_k(\mathbf{q})=\frac{1}{N_{\rm ion}}\sum_{j=1}^{N_{\rm ion}}\mathrm{e}^{-\mathrm{i}\mathbf{q}\mathbf{R}_{\|j}-\mathrm{i}kZ_j}
\tag{2.2}
$$

is the structure factor of the ionic subsystem, the Fourier-components of electrons density, the "collective" variable is

$$
\rho_k(\mathbf{q})=\sum_{\mathbf{p},\alpha_1,\alpha_2}\langle\alpha_1|\,\mathrm{e}^{\mathrm{i}kz}|\alpha_2\rangle a^\dagger_{\alpha_1}(\mathbf{p})a_{\alpha_2}(\mathbf{p}-\mathbf{q}),
\tag{2.3}
$$

where

$$
\langle\alpha_1|\ldots|\alpha_2\rangle=\int\!\mathrm{d}z\,\varphi^*_{\alpha_1}(z)\ldots\varphi_{\alpha_2}(z),
$$

$\varphi_\alpha(z)$ and ε_α are the eigenfunctions and the eigenvalues of the Schrodinger equation

$$
\left[-\frac{\hbar^2}{2m}\frac{\mathrm{d}^2}{\mathrm{d}z^2}+V(z)\right]\varphi_\alpha(z)=\varepsilon_\alpha\varphi_\alpha(z),
$$

$V(\mathbf{r})=V(z)$ is a surface potential that is a function of only normal electron coordinate.

The averages of the electrons density operator $\langle\rho(\mathbf{r})\rangle^t$ and their momentum density $\langle\mathbf{p}(\mathbf{r})\rangle^t$ could be chosen for the main parameters of the reduced description for the study of viscoelastic processes within the formulated model. These parameters are connected with the relevant inhomogeneous electric and magnetic fields:

$$
\begin{aligned}
&\boldsymbol{\nabla}\cdot\langle\mathbf{E}(\mathbf{r})\rangle^t=e\langle\rho(\mathbf{r})\rangle^t,\\
&\boldsymbol{\nabla}\times\langle\mathbf{H}(\mathbf{r})\rangle^t=-\frac{1}{c}\frac{\partial}{\partial t}\langle\mathbf{E}(\mathbf{r})\rangle^t+\frac{4\pi}{c}\frac{e}{m}\langle\mathbf{p}(\mathbf{r})\rangle^t,
\end{aligned}
\tag{2.4}
$$

where c is the speed of light, $\langle\ldots\rangle^t=\mathrm{Sp}[\ldots\rho(t)]$, $\rho(t)$ means the nonequilibrium statistical operator of the generalized "jellium" model, which satisfies the Liouville equation with the Hamiltonian (2.1). The solution of the Liouville equation for $\rho(t)$ in the Zubarev method with taking into account a projecting technique can be presented in a general form:

$$
\rho(t)=\rho_{\rm q}(t)-\int_{-\infty}^{t}\mathrm{e}^{\varepsilon(t-t')}T_{\rm q}(t,t')\big[1-\mathcal{P}_{\rm q}(t')\big]\mathrm{i}L_N\rho_{\rm q}(t')\mathrm{d}t',
\tag{2.5}
$$

where

$$
T_{\rm q}(t,t')=\exp_+\left\{-\int_{t'}^{t}\big[1-\mathcal{P}_{\rm q}(t'')\big]\mathrm{i}L_N\mathrm{d}t''\right\}
$$

denotes the generalized evolution operator with taking into account a projecting technique, $\mathcal{P}_{\mathrm{q}}(t')$ is the generalized Kawasaki-Gunton projection operator. Its structure depends on the quasi-equilibrium statistical operator $\rho_{\mathrm{q}}(t)$. In our case $\mathcal{P}_{\mathrm{q}}(t)$ could be written down as:

$$\begin{aligned}\mathcal{P}_{\mathrm{q}}(t)\rho' &= \left(\rho_{\mathrm{q}}(t)-\sum_{\mathbf{q},k}\frac{\delta\rho_{\mathrm{q}}(t)}{\delta\langle\rho_k(\mathbf{q})\rangle^t}\langle\rho_k(\mathbf{q})\rangle^t-\sum_{\mathbf{q},k}\frac{\delta\rho_{\mathrm{q}}(t)}{\delta\langle\mathbf{p}_k(\mathbf{q})\rangle^t}\cdot\langle\mathbf{p}_k(\mathbf{q})\rangle^t\right)\mathrm{Sp}\,\rho' \\ &+ \sum_{\mathbf{q},k}\frac{\delta\rho_{\mathrm{q}}(t)}{\delta\langle\rho_k(\mathbf{q})\rangle^t}\mathrm{Sp}\,\rho_k(\mathbf{q})\rho'+\sum_{\mathbf{q},k}\frac{\delta\rho_{\mathrm{q}}(t)}{\delta\langle\mathbf{p}_k(\mathbf{q})\rangle^t}\cdot\mathrm{Sp}\,\mathbf{p}_k(\mathbf{q})\rho' \qquad (2.6)\end{aligned}$$

and it has the following properties

$$\mathcal{P}_{\mathrm{q}}(t)\rho(t)=\rho_{\mathrm{q}}(t),\qquad \mathcal{P}_{\mathrm{q}}(t)\rho_{\mathrm{q}}(t')=\rho_{\mathrm{q}}(t),\qquad \mathcal{P}_{\mathrm{q}}(t)\mathcal{P}_{\mathrm{q}}(t')=\mathcal{P}_{\mathrm{q}}(t),\qquad \big(1-\mathcal{P}_{\mathrm{q}}(t)\big)\mathcal{P}_{\mathrm{q}}(t)=0.$$

We define the quasi-equilibrium statistical operator $\rho_{\mathrm{q}}(t)$ with the principle of the Gibbs entropy maximum when the parameters of the reduced description $\langle\rho(\mathbf{r})\rangle^t$ and $\langle\mathbf{p}(\mathbf{r})\rangle^t$ are fixed and the normalization condition $\mathrm{Sp}\,\rho_{\mathrm{q}}(t)=1$ is satisfied. In our case we find

$$\rho_{\mathrm{q}}(t)=\exp\left(-\Phi(t)-\beta\left\{H-\frac{1}{SL}\sum_{\mathbf{q},k}\Big[\overline{\mu}_k(\mathbf{q};t)\rho_k(\mathbf{q};t)+\mathbf{A}_k(\mathbf{q};t)\cdot\mathbf{p}_k(\mathbf{q};t)\Big]\right\}\right), \qquad (2.7)$$

$$\Phi(t)=\ln\mathrm{Sp}\exp\left(-\beta\left\{H-\frac{1}{SL}\sum_{\mathbf{q},k}\Big[\overline{\mu}_k(\mathbf{q};t)\rho_k(\mathbf{q};t)+\mathbf{A}_k(\mathbf{q};t)\cdot\mathbf{p}_k(\mathbf{q};t)\Big]\right\}\right), \qquad (2.8)$$

where $\overline{\mu}_k(\mathbf{q};t)=\mu_k(\mathbf{q};t)+e\varphi_k(\mathbf{q};t)$ stands for the Fourier-component of the electrochemical potential of electrons, $\mu_k(\mathbf{q};t)$ is the Fourier-component of the chemical potential, $\varphi_k(\mathbf{q};t)$ is the Fourier-component of the local electrical potential, $\mathbf{A}_k(\mathbf{q};t)=\mathbf{v}_k(\mathbf{q};t)-c^{-1}\mathbf{a}_k(\mathbf{q};t)$, $\mathbf{v}_k(\mathbf{q};t)$ denotes the Fourier-component of the average velocity of electrons, $\mathbf{a}_k(\mathbf{q};t)$ is the Fourier-component of the vector potential $\mathbf{a}(\mathbf{r};t)$ of electromagnetic field:

$$\langle\mathbf{H}(\mathbf{r})\rangle^t=\boldsymbol{\nabla}\times\mathbf{a}(\mathbf{r},t),\qquad \langle\mathbf{E}^t(\mathbf{r})\rangle^t=-\frac{1}{c}\frac{\partial}{\partial t}\mathbf{a}(\mathbf{r},t), \qquad (2.9)$$

where $\langle\mathbf{E}^t(\mathbf{r})\rangle^t$ is a tangential part of electrical field, $\overline{\mu}_k(\mathbf{q};t)$ is defined with the self-consistent condition:

$$\langle\rho_k(\mathbf{q})\rangle^t=\langle\rho_k(\mathbf{q})\rangle^t_{\mathrm{q}} \qquad (2.10)$$

and the thermodynamical relations:

$$\frac{\delta\Phi(t)}{\delta\frac{\beta}{SL}\overline{\mu}_k(\mathbf{q};t)}=\langle\rho_k(\mathbf{q})\rangle^t, \qquad (2.11)$$

$$\frac{\delta S(t)}{\delta\langle\rho_k(\mathbf{q};t)\rangle^t}=-\frac{\beta}{SL}\mu_k(\mathbf{q};t),\qquad \frac{\delta S(t)}{\delta\langle e\rho_k(\mathbf{q};t)\rangle^t}=-\frac{\beta}{SL}\varphi_k(\mathbf{q};t), \qquad (2.12)$$

when $\langle\mathbf{p}_k(\mathbf{q};t)\rangle^t$ are fixed, where $S(t)$ is the nonequilibrium entropy defined using the Gibbs method:

$$\begin{aligned}S(t) &= -\mathrm{Sp}\,\big[\ln\rho_{\mathrm{q}}(t)\big]\rho_{\mathrm{q}}(t) \\ &= \Phi(t)+\beta\left\{\langle H\rangle^t-\frac{1}{SL}\sum_{\mathbf{q}}\sum_k\Big[\overline{\mu}_k(\mathbf{q};t)\langle\rho_k(\mathbf{q})\rangle^t_{\mathrm{q}}+\mathbf{A}_k(\mathbf{q};t)\cdot\langle\mathbf{p}_k(\mathbf{q})\rangle^t_{\mathrm{q}}\Big]\right\} \\ &= \ln Z(t)+\beta\left\{\langle H\rangle^t-\frac{1}{SL}\sum_{\mathbf{q}}\sum_k\Big[\overline{\mu}_k(\mathbf{q};t)\langle\rho_k(\mathbf{q})\rangle^t+\mathbf{A}_k(\mathbf{q};t)\cdot\langle\mathbf{p}_k(\mathbf{q})\rangle^t\Big]\right\}, \qquad (2.13)\end{aligned}$$

and besides

$$\frac{\delta\Phi(t)}{\delta\frac{\beta}{SL}\mathbf{A}_k(\mathbf{q};t)} = \langle\mathbf{p}_k(\mathbf{q})\rangle^t, \tag{2.14}$$

$$\frac{\delta S(t)}{\delta\langle\mathbf{p}_k(\mathbf{q})\rangle^t} = -\frac{\beta}{SL}\langle\mathbf{v}_k(\mathbf{q})\rangle^t, \qquad \frac{\delta S(t)}{\delta\langle\frac{1}{c}\mathbf{p}_k(\mathbf{q})\rangle^t} = -\frac{\beta}{SL}\mathbf{a}_k(\mathbf{q};t). \tag{2.15}$$

$Z(t)$ is the partition function of the quasi-equilibrium statistical operator:

$$Z(t) = \mathrm{Sp}\exp\left(-\beta\left\{H - \frac{1}{SL}\sum_{\mathbf{q},k}\left[\overline{\mu}_k(\mathbf{q};t)\rho_k(\mathbf{q}) + \mathbf{A}_k(\mathbf{q};t)\cdot\mathbf{p}_k(\mathbf{q})\right]\right\}\right). \tag{2.16}$$

One should calculate the quasi-equilibrium partition function (2.16) to define the structure of the Kawasaki-Gunton projection operator, to find the nonequilibrium parameters $\overline{\mu}_k(\mathbf{q};t)$, $\mathbf{A}_k(\mathbf{q};t)$, and thus, to obtain the nonequilibrium statistical operator $\rho(t)$. The approach based on the “jellium” model is proposed in [28, 36] in the diffusion description case, where the average value of the electron density operator $\langle\rho(\mathbf{r})\rangle^t$ is chosen for the reduced description parameter. We use this approach to calculate $Z(t)$ (2.16). We apply the functional integration method to present $Z(t)$ as follows:

$$Z(t) = \exp\left[\beta\frac{N}{2S}{\sum_{\mathbf{q}}}'\nu(\mathbf{q}|0)\right]Z_{\text{jell}}\Delta Z(t). \tag{2.17}$$

Here,

$$Z_{\text{jell}} = \mathrm{Sp}\left[\exp(-\beta H_0)\mathrm{T}S_1(\beta)\right] \tag{2.18}$$

is the partition function of the “jellium” model of the electron subsystem that corresponds to the equilibrium state, found in [8, 28, 31, 37].

$$S_1(\beta) = \exp\left[-\frac{1}{SL}\int_0^\beta \mathrm{d}\beta'{\sum_{\mathbf{q}}}'\sum_k \nu_k(\mathbf{q})\rho_k(\mathbf{q}|\beta')\rho_{-k}(-\mathbf{q}|\beta')\right] \tag{2.19}$$

denotes the contribution of electron interaction and $\rho_k(\mathbf{q}|\beta') = \mathrm{e}^{\beta' H_0}\rho_k(\mathbf{q})\,\mathrm{e}^{-\beta' H_0}$,

$$\Delta Z(t) = \frac{1}{Z_{\text{jell}}}\mathrm{Sp}\left[\exp(-\beta H_0)\mathrm{T}S_1(\beta)S_2(\beta;t)\right] = \langle S_2(\beta;t)\rangle_{\text{jell}}\,, \tag{2.20}$$

where

$$\langle(\ldots)\rangle_{\text{jell}} = \frac{1}{Z_{\text{jell}}}\mathrm{Sp}\left[\exp(-\beta H_0)\mathrm{T}S_1(\beta)(\ldots)\right],$$

$$S_2(\beta;t) = \mathrm{T}\exp\left[-\frac{1}{SL}\int_0^\beta \mathrm{d}\beta'\sum_{k,\mathbf{q}}\widetilde{B}(\mathbf{q},k;t)\widetilde{W}_k^{(+)}(\mathbf{q};\beta')\right], \tag{2.21}$$

and

$$\widetilde{C}(\mathbf{q},k;t) = \mathrm{col}\left[B_k(\mathbf{q};t),\mathbf{A}_k(\mathbf{q};t)\right]$$

is a column vector, $B_k(\mathbf{q};t) = N_{\text{ion}}S_k(\mathbf{q})w_k(\mathbf{q}) - \overline{\mu}_k(\mathbf{q};t)$,

$$\widetilde{W}_k^{(+)}(\mathbf{q};\beta) = \left[\rho_k(\mathbf{q};\beta),\ \mathbf{p}_k(\mathbf{q};\beta)\right]$$

is a row vector. One can write down $\Delta Z(t)$ after applying the cumulant representation:

$$\begin{aligned}\Delta Z(t) \;&=\; \exp\left[\sum_{n=1}^{\infty}\frac{\mathrm{i}^n}{n!}\left(\frac{\beta}{SL}\right)^n\sum_{k_1,\ldots,k_n}\sum_{\mathbf{q}_1,\ldots,\mathbf{q}_n}\widetilde{C}(\mathbf{q}_1,k_1;t)\ldots\right.\\ &\times\;\left.\widetilde{C}(\mathbf{q}_n,k_n;t)\widetilde{\mathfrak{M}}_{-k_1,\ldots,-k_n}(-\mathbf{q}_1,\ldots,-\mathbf{q}_n;\beta)\right],\end{aligned} \tag{2.22}$$

where

$$\widetilde{\mathfrak{M}}_{k_1,\dots,k_n}(\mathbf{q}_1,\dots,\mathbf{q}_n;\beta) = \mathrm{i}^n \left\langle \mathrm{T}\widetilde{W}_{k_1}(\mathbf{q}_1;\beta)\dots\widetilde{W}_{k_n}(\mathbf{q}_n;\beta)\right\rangle^{\mathrm{c}}_{\mathrm{jell}} \tag{2.23}$$

denote matrices of fluctuations of the cumulant averages of electron density and electron momentum, which are obtained with the nonequilibrium statistical operator of the "jellium" model of the electron subsystem [8, 28, 31, 32]. Particularly, the matrix of the second cumulant has the following structure:

$$\widetilde{\mathfrak{M}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) = \begin{pmatrix} \mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) & \boldsymbol{\mathfrak{M}}^{\rho\mathbf{p}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) \\ \boldsymbol{\mathfrak{M}}^{\mathbf{p}\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) & \boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) \end{pmatrix}, \tag{2.24}$$

where

$$\mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) = \langle\rho_{k_1}(\mathbf{q}_1;\beta)\rho_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}} - \langle\rho_{k_1}(\mathbf{q}_1;\beta)\rangle_{\mathrm{jell}}\langle\rho_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}}\,, \tag{2.25}$$

$$\boldsymbol{\mathfrak{M}}^{\rho\mathbf{p}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) = \langle\rho_{k_1}(\mathbf{q}_1;\beta)\mathbf{p}_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}} - \langle\rho_{k_1}(\mathbf{q}_1;\beta)\rangle_{\mathrm{jell}}\langle\mathbf{p}_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}}\,, \tag{2.26}$$

$$\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) = \langle\mathbf{p}_{k_1}(\mathbf{q}_1;\beta)\mathbf{p}_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}} - \langle\mathbf{p}_{k_1}(\mathbf{q}_1;\beta)\rangle_{\mathrm{jell}}\langle\mathbf{p}_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}}\,, \tag{2.27}$$

and $\langle\mathbf{p}_{k_i}(\mathbf{q}_i;\beta)\rangle_{\mathrm{jell}} = 0$, since averaging of the momentum density operator proceeds with the equilibrium statistical operator. For the same reason, the averages

$$\langle\rho_{k_1}(\mathbf{q}_1;\beta)\mathbf{p}_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}} = \langle\mathbf{p}_{k_1}(\mathbf{q}_1;\beta)\rho_{k_2}(\mathbf{q}_2;\beta)\rangle_{\mathrm{jell}} = 0,$$

therefore, matrix (2.24) is a diagonal one:

$$\widetilde{\mathfrak{M}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) = \begin{pmatrix} \mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) & 0 \\ 0 & \boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) \end{pmatrix}. \tag{2.28}$$

Considering the above, one can present $\Delta Z(t)$ in the Gaussian approximation as

$$\begin{aligned}\Delta Z^{\mathrm{G}}(t) \quad &= \quad \exp\Bigg[-\frac{1}{2}\left(\frac{\beta}{SL}\right)^2 \sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\Big(B_{k_1}(\mathbf{q}_1;t)B_{k_2}(\mathbf{q}_2;t)\mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta) \\ &+ \quad \mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2;\beta)\cdot\mathbf{A}_{k_2}(\mathbf{q}_2;t)\Big)\Bigg].\end{aligned} \tag{2.29}$$

In the next section we find the nonequilibrium statistical operator in the Gaussian approximation, where the operators of electron density and electron momentum density do not correlate as a pair correlation function.

3. The Gaussian approximation

Let us present the relevant statistical operator $\rho_{\mathrm{q}}(t)$ to find the nonequilibrium statistical operator in the Gaussian approximation. Considering (2.29) one can write down:

$$\rho^{(\mathrm{G})}_{\mathrm{q}}(t) = \exp\left[-\widehat{S}^{\mathrm{G}}(t)\right], \tag{3.1}$$

where

$$\begin{aligned}\widehat{S}^{\mathrm{G}}(t) \quad &= \quad \beta\frac{N}{2S}\sum_{\mathbf{q}}{}'\nu(\mathbf{q}|0) + \ln Z_{\mathrm{jell}} \\ &- \quad \frac{1}{2}\left(\frac{\beta}{SL}\right)^2\sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\Big[B_{k_1}(\mathbf{q}_1;t)B_{k_2}(\mathbf{q}_2;t)\mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2) \\ &+ \quad \mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2)\cdot\mathbf{A}_{k_2}(\mathbf{q}_2;t)\Big] \\ &+ \quad \beta\Big\{H - \frac{1}{SL}\sum_{k,\mathbf{q}}\Big[\overline{\mu}_k(\mathbf{q};t)\rho_k(\mathbf{q}) + \mathbf{A}_k(\mathbf{q};t)\cdot\mathbf{p}_k(\mathbf{q})\Big]\Big\}\end{aligned} \tag{3.2}$$

is the entropy operator. In order to exclude the parameters $\overline{\mu}_k(\mathbf{q};t)$ we use the following thermodynamical relation:

$$\frac{\delta\Phi^{(\mathrm{G})}(t)}{\delta\frac{\beta}{SL}\overline{\mu}_k(\mathbf{q};t)} = \langle\rho_k(\mathbf{q})\rangle^t,$$

from which one can find:

$$\langle\rho_k(\mathbf{q})\rangle^t = \left(\frac{\beta}{SL}\right)\sum_{k',\mathbf{q}'}\left[\overline{S}_{k'}(\mathbf{q}') - \overline{\mu}_{k'}(\mathbf{q}';t)\right]\mathfrak{M}^{\rho\rho}_{-k',-k}(-\mathbf{q}',-\mathbf{q}), \tag{3.3}$$

where $\overline{S}_k(\mathbf{q}) = N_{\mathrm{ion}}S_k(\mathbf{q})w_k(\mathbf{q})$.

Denoting $[\mathfrak{M}^{\rho\rho}]^{-1}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2)$ as the inverse function of $\mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2)$:

$$\sum_{k'',\mathbf{q}''}[\mathfrak{M}^{\rho\rho}]^{-1}_{k,k''}(\mathbf{q},\mathbf{q}'')\mathfrak{M}^{\rho\rho}_{k'',k'}(\mathbf{q}'',\mathbf{q}') = \delta_{k,k'}\delta_{\mathbf{q},\mathbf{q}'} \tag{3.4}$$

from the Fourier-component of the electrons one can find the electrochemical potential:

$$\overline{\mu}_k(\mathbf{q};t) = \overline{S}_k(\mathbf{q}) - \left(\frac{\beta}{SL}\right)^{-1}\sum_{k',\mathbf{q}'}\langle\rho_{k'}(\mathbf{q}')\rangle^t\,[\mathfrak{M}^{\rho\rho}]^{-1}_{k',k}(-\mathbf{q}',-\mathbf{q}). \tag{3.5}$$

As we can see, the Fourier-component of the electrochemical potential in the Gaussian approximation is expressed in terms of the structure factor of the ionic subsystem and the Fourier-component of the local pseudopotential of interaction between electrons and ions. Time-dependence is described with the average equilibrium value of electrons density, renormalized in terms of the structure factor of the ionic subsystem, the pseudopotential $w_k(\mathbf{q})$ and the inverse function $[\mathfrak{M}^{\rho\rho}]^{-1}_{k',k}(-\mathbf{q}',-\mathbf{q})$ of the pair irreducible cumulant average of electrons density fluctuations. Similarly we exclude the parameters $\mathbf{A}_k(\mathbf{q};t)$ from (3.2) using the thermodynamical relation in the Gaussian approximation:

$$\frac{\delta\Phi^{(\mathrm{G})}(t)}{\delta\frac{\beta}{SL}\mathbf{A}_k(\mathbf{q};t)} = \langle\mathbf{p}_k(\mathbf{q})\rangle^t,$$

hence, one can find

$$\langle\mathbf{p}_k(\mathbf{q})\rangle^t = -\left(\frac{\beta}{SL}\right)\sum_{k',\mathbf{q}'}\mathbf{A}_{k'}(\mathbf{q}')\cdot\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k',k}(-\mathbf{q}',-\mathbf{q}). \tag{3.6}$$

Defining $[\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}]^{-1}_{-k',-k''}(-\mathbf{q}',-\mathbf{q}'')$ as the inverse function of $\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{-k',-k''}(-\mathbf{q}',-\mathbf{q}'')$:

$$\sum_{k'',\mathbf{q}''}[\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}]^{-1}_{k,k''}(\mathbf{q},\mathbf{q}'')\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}_{k'',k'}(\mathbf{q}'',\mathbf{q}') = \delta_{k,k'}\delta_{\mathbf{q},\mathbf{q}'}$$

for the Fourier-component of $\mathbf{A}_k(\mathbf{q};t)$ one can obtain from (3.6):

$$\mathbf{A}_k(\mathbf{q};t) = -\left(\frac{\beta}{SL}\right)^{-1}\sum_{k',\mathbf{q}'}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t\cdot[\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q}), \tag{3.7}$$

the time-dependence of which is described with the average value of a density momentum operator, renormalized via function $[\boldsymbol{\mathfrak{M}}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})$, which is the inverse function of the pair irreducible cumulant average value of the momentum density fluctuation of electrons. In consideration of (3.5) and (3.7), the entropy operator could be written down as follows:

$$\begin{aligned}
\widehat{S}^{\mathrm{G}}(t) &= \beta\frac{N}{2S}{\sum_{\mathbf{q}}}'\nu(\mathbf{q}|0)+\ln Z_{\mathrm{jell}}-\sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\Big[\langle\rho_{k_1}(\mathbf{q}_1)\rangle^t\,[\mathfrak{M}^{\rho\rho}]^{-1}_{-k_1,-k_2}(-\mathbf{q}_1,-\mathbf{q}_2)\,\langle\rho_{k_2}(\mathbf{q}_2)\rangle^t\\
&+ \langle\mathbf{p}_{k_1}(\mathbf{q}_1)\rangle^t\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k_1,-k_2}(-\mathbf{q}_1,-\mathbf{q}_2)\cdot\langle\mathbf{p}_{k_2}(\mathbf{q}_2)\rangle^t\Big]\\
&+ \beta\Big\{H-\frac{1}{SL}\sum_{k,\mathbf{q}}\Big[\overline{S}_k(\mathbf{q})-\Big(\frac{\beta}{SL}\Big)^{-1}\sum_{k',\mathbf{q}'}\langle\rho_{k'}(\mathbf{q}')\rangle^t\,[\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\Big]\rho_k(\mathbf{q})\\
&+ \frac{1}{SL}\Big(\frac{\beta}{SL}\Big)^{-1}\sum_{k,k'}\sum_{\mathbf{q},\mathbf{q}'}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\cdot\mathbf{p}_k(\mathbf{q})\Big\}. \qquad (3.8)
\end{aligned}$$

Then, in order to calculate the nonequilibrium statistical operator (2.5) in the Gaussian approximation (3.1), (3.8) one should reveal the structure of the Kawasaki-Gunton projection operator (2.6), and its effect and the effect of the Liouville operator on $\rho_{\mathrm{q}}^{(\mathrm{G})}$ as it is shown in appendix A. Then, considering (A.3), (A.4) for the nonequilibrium statistical operator one can get:

$$\begin{aligned}
\rho(t) &= \rho_{\mathrm{q}}^{(\mathrm{G})}(t)-\sum_{k,\mathbf{q}}\int_{-\infty}^{t}\mathrm{e}^{\varepsilon(t'-t)}T_{\mathrm{q}}^{\mathrm{G}}(t,t')\\
&\times \Bigg\{\int_0^1\mathrm{d}\tau\Big[\rho_{\mathrm{q}}^{(\mathrm{G})}(t')\Big]^{\tau}I_{\rho}^{(\mathrm{G})}(k,\mathbf{q};t')\Big[\rho_{\mathrm{q}}^{(\mathrm{G})}(t')\Big]^{1-\tau}W_{\rho\rho}^{(\mathrm{G})}(k,\mathbf{q};t')\\
&+ \int_0^1\mathrm{d}\tau\Big[\rho_{\mathrm{q}}^{(\mathrm{G})}(t')\Big]^{\tau}\mathbf{I}_{\mathbf{p}}^{(\mathrm{G})}(k,\mathbf{q};t')\Big[\rho_{\mathrm{q}}^{(\mathrm{G})}(t')\Big]^{1-\tau}\cdot\mathbf{W}_{\mathbf{pp}}^{(\mathrm{G})}(k,\mathbf{q};t')\Bigg\}\mathrm{d}t', \qquad (3.9)
\end{aligned}$$

where

$$I_{\rho}^{(\mathrm{G})}(k,\mathbf{q};t')=\Big[1-\mathcal{P}^{(\mathrm{G})}(t')\Big]\mathrm{i}L\rho_k(\mathbf{q}),\qquad \mathbf{I}_{\mathbf{p}}^{(\mathrm{G})}(k,\mathbf{q};t')=\Big[1-\mathcal{P}^{(\mathrm{G})}(t')\Big]\mathrm{i}L\mathbf{p}_k(\mathbf{q}) \qquad (3.10)$$

are the generalized fluxes, $\mathcal{P}^{(\mathrm{G})}(t)$ is a Mori-like projection operator, which effects as follows:

$$\begin{aligned}
\mathcal{P}^{(\mathrm{G})}(t)\hat{A} &= \sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\Big[\delta\rho_{k'}(\mathbf{q}';t)[\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k''}(-\mathbf{q}',-\mathbf{q}'')\Big\langle\rho_{k''}(\mathbf{q}'')\hat{A}\Big\rangle_{\mathrm{G}}^{t}\\
&+ \delta\mathbf{p}_{k'}(\mathbf{q}';t)\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k''}(-\mathbf{q}',-\mathbf{q}'')\cdot\Big\langle\mathbf{p}_{k''}(\mathbf{q}'')\hat{A}\Big\rangle_{\mathrm{G}}^{t}\Big], \qquad (3.11)
\end{aligned}$$

where

$$\begin{aligned}
\langle\ldots\rangle_{\mathrm{G}}^{t} &= \mathrm{Sp}\left[\ldots\rho_{\mathrm{q}}^{\mathrm{G}}(t)\right],\\
\delta\rho_{k'}(\mathbf{q}';t)=\rho_{k'}(\mathbf{q}')-\langle\rho_{k'}(\mathbf{q}')\rangle^t,&\qquad \delta\mathbf{p}_{k'}(\mathbf{q}';t)=\mathbf{p}_{k'}(\mathbf{q}')-\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t.
\end{aligned}$$

Using the effect of operator $\mathcal{P}^{(\mathrm{G})}(t')$ on $\mathrm{i}L\rho_k(\mathbf{q})$, one can show that $I_{\rho}^{(\mathrm{G})}(k,\mathbf{q};t')=0$. According to (3.9) the nonequilibrium statistical operator in the Gaussian approximation is the functional of the observable values $\langle\rho_k(\mathbf{q})\rangle^t$, $\langle\mathbf{p}_k(\mathbf{q})\rangle^t$ and the generalized fluxes of momentum density $\mathbf{I}_{\mathbf{p}}^{(\mathrm{G})}(k,\mathbf{q};t')$. One can obtain the relevant transport equations for $\langle\rho_k(\mathbf{q})\rangle^t$ and $\langle\mathbf{p}_k(\mathbf{q})\rangle^t$ with the nonequilibrium statistical operator as follows:

$$\frac{\partial}{\partial t}\langle\rho_k(\mathbf{q})\rangle^t + \mathrm{i}k\mathbf{q}\frac{1}{m}\cdot\langle\mathbf{p}_k(\mathbf{q})\rangle^t = 0, \tag{3.12}$$

$$\begin{aligned}\frac{\partial}{\partial t}\langle\mathbf{p}_k(\mathbf{q})\rangle^t &+ \sum_{k',\mathbf{q}'}\left\langle\mathbf{p}_k(\mathbf{q})\dot{\rho}_{k'}(\mathbf{q}')\right\rangle^t_{\mathrm{G}} W^{(\mathrm{G})}_{\rho\rho}(k',\mathbf{q}';t)\\ &- \sum_{k,\mathbf{q}}\int\limits_{-\infty}^{t}\mathrm{e}^{\varepsilon(t'-t)}\varphi^{(\mathrm{G})}_{\mathbf{pp}}(k,\mathbf{q};k',\mathbf{q}';t,t')\mathbf{W}^{(\mathrm{G})}_{\mathbf{pp}}(k',\mathbf{q}';t')\mathrm{d}t' = 0,\end{aligned} \tag{3.13}$$

where

$$\varphi^{(\mathrm{G})}_{\mathbf{pp}}(k,\mathbf{q};k',\mathbf{q}';t,t') = \mathrm{Sp}\left\{\mathbf{I}^{(\mathrm{G})}_{\mathbf{p}}(k,\mathbf{q};t)\,T^{\mathrm{G}}_{\mathrm{q}}(t,t')\int\limits_0^1\mathrm{d}\tau\left[\rho^{(\mathrm{G})}_{\mathrm{q}}(t')\right]^{\tau}\mathbf{I}^{(\mathrm{G})}_{\mathbf{p}}(k',\mathbf{q}';t')\left[\rho^{(\mathrm{G})}_{\mathrm{q}}(t')\right]^{1-\tau}\right\} \tag{3.14}$$

is the generalized memory function that describes dissipative processes and takes into account

$$\left\langle\dot{\mathbf{p}}_k(\mathbf{q})\right\rangle^t_{\mathrm{G}} = -\sum_{k',\mathbf{q}'}\left\langle\mathbf{p}_k(\mathbf{q})\dot{\rho}_{k'}(\mathbf{q}')\right\rangle^t_{\mathrm{G}}W^{(\mathrm{G})}_{\rho\rho}(k',\mathbf{q}';t). \tag{3.15}$$

It is the time correlation function of the generalized fluxes of momentum density, averaged with the quasi-equilibrium statistical operator $\rho^{(\mathrm{G})}_{\mathrm{q}}(t)$ in the Gaussian approximation. $\dot{\mathbf{p}}_k(\mathbf{q}) = \mathrm{i}L_N\mathbf{p}_k(\mathbf{q}) = \mathrm{i}k\mathbf{q} : \overset{\leftrightarrow}{\mathbf{T}}_k(\mathbf{q})$ denotes the tensor operator of a viscous stress of the electron subsystem. This means that the generalized memory function (3.14) defines the generalized viscosity coefficient of the electron subsystem:

$$\varphi^{(\mathrm{G})}_{\mathbf{pp}}(k,\mathbf{q};k',\mathbf{q}';t,t') = -k\mathbf{q} : \eta^{(\mathrm{G})}(k,\mathbf{q};k',\mathbf{q}';t,t'') : k'\mathbf{q}'. \tag{3.16}$$

It is notable that the system of equations (3.12), (3.13) has the same structure as in the case of weakly nonequilibrium processes with the only difference in the relevant averages: $\rho^{(\mathrm{G})}_{\mathrm{q}}(t) \to \rho^0_{\mathrm{q}}(t)$, where

$$\begin{aligned}\rho^0_{\mathrm{q}}(t) = \rho_0\Bigg\{1 + \sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\Bigg[&\delta\rho_{k'}(\mathbf{q}',t)[\widetilde{\mathfrak{M}}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q},-\mathbf{q}')\int\limits_0^1\mathrm{d}\tau\rho_0^{\tau}\rho_k(\mathbf{q})\rho_0^{-\tau}\\ + &\delta\mathbf{p}_{k'}(\mathbf{q}',t)\cdot[\widetilde{\mathfrak{M}}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q},-\mathbf{q}')\int\limits_0^1\mathrm{d}\tau\rho_0^{\tau}\cdot\mathbf{p}_k(\mathbf{q})\rho_0^{-\tau}\Bigg]\Bigg\},\end{aligned}$$

where ρ_0 is the equilibrium statistical operator of the system.

In the next section we work with the following higher approximation for the quasi-equilibrium partition function, when the static correlations between the operators of electron density and electron momentum density occur with the third-order cumulant averages.

4. The quasi-equilibrium partition function in the approximation of the third-order cumulant averages

According to (2.17), (2.22) the relevant statistical operator $\rho_{\mathrm{q}}(t)$ in this approximation is

$$\rho^{\mathrm{G}+1}_{\mathrm{q}}(t) = \exp\left[-\hat{S}^{(\mathrm{G}+1)}(t)\right], \tag{4.1}$$

$$\hat{S}^{(\mathrm{G+1})}(t) = \beta\frac{N}{2S}{\sum_{\mathbf{q}}}'\nu(\mathbf{q}|0) + \ln Z_{\mathrm{jell}} + \beta\left\{H - \frac{1}{SL}\sum_{k,\mathbf{q}}\left[\overline{\mu}_k(\mathbf{q};t)\rho_k(\mathbf{q}) + \mathbf{A}_k(\mathbf{q};t)\cdot\mathbf{p}_k(\mathbf{q})\right]\right\} - \Phi^{(\mathrm{G+1})}(t), \tag{4.2}$$

$$\begin{aligned}\Phi^{(\mathrm{G+1})}(t) &= \frac{1}{2}\left(\frac{\beta}{SL}\right)^2\sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\Big[B_{k_1}(\mathbf{q}_1;t)\mathfrak{M}^{\rho\rho}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2)B_{k_2}(\mathbf{q}_2;t)\\ &+ \mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\mathfrak{M}^{\mathbf{pp}}_{k_1,k_2}(\mathbf{q}_1,\mathbf{q}_2)\cdot\mathbf{A}_{k_2}(\mathbf{q}_2;t)\Big]\\ &+ \frac{\mathrm{i}}{3!}\left(\frac{\beta}{SL}\right)^3\sum_{k_1,k_2,k_3}\sum_{\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3}\Big[B_{k_1}(\mathbf{q}_1;t)B_{k_2}(\mathbf{q}_2;t)B_{k_3}(\mathbf{q}_3;t)\mathfrak{M}^{\rho\rho\rho}_{k_1,k_2,k_3}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3)\\ &+ 3\mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\mathfrak{M}^{\mathbf{pp}\rho}_{k_1,k_2,k_3}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}_3)\cdot\mathbf{A}_{k_2}(\mathbf{q}_2;t)B_{k_3}(\mathbf{q}_3;t)\Big].\end{aligned} \tag{4.3}$$

In order to obtain an explicit form of the nonequilibrium statistical operator and the transport equations within the approximation (4.1) one should exclude parameters $\overline{\mu}_k(\mathbf{q};t)$ and $\mathbf{A}_k(\mathbf{q};t)$ from (4.2). In the same way as in the Gaussian approximation case we use the thermodynamical relations:

$$\frac{\delta\Phi^{(\mathrm{G+1})}(t)}{\delta\frac{\beta}{SL}\overline{\mu}_k(\mathbf{q};t)} = \langle\rho_k(\mathbf{q})\rangle^t, \tag{4.4}$$

$$\frac{\delta\Phi^{(\mathrm{G+1})}(t)}{\delta\frac{\beta}{SL}\mathbf{A}_k(\mathbf{q};t)} = \langle\mathbf{p}_k(\mathbf{q})\rangle^t. \tag{4.5}$$

Considering the structure of $\Phi^{(\mathrm{G+1})}(t)$ in $\hat{S}^{(\mathrm{G+1})}(t)$ from (4.4) one can get equations to define the electrochemical potential of the electron subsystem of a semi-bounded metal within the generalized "jellium" model:

$$\begin{aligned}\langle\rho_k(\mathbf{q})\rangle^t &= \frac{\beta}{SL}\sum_{k_1,\mathbf{q}_1}B_{k_1}(\mathbf{q}_1;t)\mathfrak{M}^{\rho\rho}_{k_1,k}(\mathbf{q}_1,\mathbf{q})\\ &+ \frac{\mathrm{i}}{2!}\left(\frac{\beta}{SL}\right)^2\sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\Big[B_{k_1}(\mathbf{q}_1;t)\mathfrak{M}^{\rho\rho\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})B_{k_2}(\mathbf{q}_2;t)\\ &- 3\mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\mathfrak{M}^{\mathbf{pp}\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})\cdot\mathbf{A}_{k_2}(\mathbf{q}_2;t)\Big],\end{aligned} \tag{4.6}$$

and to define $\mathbf{A}_k(\mathbf{q};t)$ from (4.5) one can find:

$$\begin{aligned}\langle\mathbf{p}_k(\mathbf{q})\rangle^t &= -\frac{\beta}{SL}\sum_{k_1,\mathbf{q}_1}\mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\mathfrak{M}^{\mathbf{pp}}_{k_1,k}(\mathbf{q}_1,\mathbf{q})\\ &- \mathrm{i}\left(\frac{\beta}{SL}\right)^2\sum_{k_1,k_2}\sum_{\mathbf{q}_1,\mathbf{q}_2}\mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot\mathfrak{M}^{\mathbf{pp}\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})B_{k_2}(\mathbf{q}_2;t).\end{aligned} \tag{4.7}$$

The detailed calculations are presented in the appendix B. Considering (B.12) the nonequilibrium statistical operator of the electron subsystem of a semi-bounded metal within the generalized "jellium" model, according to (2.5) we present as follows:

$$\begin{aligned}\rho(t) &= \rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t) - \sum_{k,\mathbf{q}} \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t'-t)} T_{\mathrm{q}}^{(\mathrm{G}+1)}(t,t') \left[1 - \mathcal{P}_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right] \\ &\times \left\{ \int_0^1 \mathrm{d}\tau \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{\tau} \dot{\rho}_k(\mathbf{q}) \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{1-\tau} W_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q};t') \right. \\ &+ \left. \int_0^1 \mathrm{d}\tau \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{\tau} \dot{\mathbf{p}}_k(\mathbf{q}) \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{1-\tau} \cdot \mathbf{W}_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q};t) \right\} \mathrm{d}t', \end{aligned} \tag{4.8}$$

where

$$\dot{\rho}_k(\mathbf{q}) = \mathrm{i}L_N \rho_k(\mathbf{q}), \qquad \dot{\mathbf{p}}_k(\mathbf{q}) = \mathrm{i}L_N \mathbf{p}_k(\mathbf{q}). \tag{4.9}$$

Then, taking into account the above found we obtain the transport equations for the reduced description parameters $\langle \rho_k(\mathbf{q}) \rangle^t$, $\langle \mathbf{p}_k(\mathbf{q}) \rangle^t$:

$$\begin{aligned}\frac{\partial}{\partial t}\langle \rho_k(\mathbf{q}) \rangle^t &= \langle \dot{\rho}_k(\mathbf{q}) \rangle^t_{(\mathrm{G}+1)} - \sum_{k',\mathbf{q}'} \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t'-t)} \Phi_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') W_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q};t')\mathrm{d}t' \\ &- \sum_{k',\mathbf{q}'} \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t'-t)} \Phi_{\rho\,\mathbf{p}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') \cdot \mathbf{W}_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q};t')\mathrm{d}t', \end{aligned} \tag{4.10}$$

$$\begin{aligned}\frac{\partial}{\partial t}\langle \mathbf{p}_k(\mathbf{q}) \rangle^t &= \langle \dot{\mathbf{p}}_k(\mathbf{q}) \rangle^t_{(\mathrm{G}+1)} - \sum_{k',\mathbf{q}'} \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t'-t)} \Phi_{\mathbf{p}\,\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') W_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q};t')\mathrm{d}t' \\ &- \sum_{k',\mathbf{q}'} \int_{-\infty}^{t} \mathrm{e}^{\varepsilon(t'-t)} \Phi_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') : \mathbf{W}_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q};t')\mathrm{d}t', \end{aligned} \tag{4.11}$$

where

$$\langle (\ldots) \rangle^t_{(\mathrm{G}+1)} = \mathrm{Sp}(\ldots)\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t),$$

$\Phi_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$, $\Phi_{\rho\,\mathbf{p}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$, $\Phi_{\mathbf{p}\,\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$, $\Phi_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$ are the generalized transport kernels that describe diffusive, visco-diffusive and viscous processes of the electron subsystem of a semi-bounded metal within the generalized "jellium" model. Particularly, $\Phi_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$ and $\Phi_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$ have the following structure:

$$\begin{aligned}\Phi_{\rho\rho}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') &= \Big\langle \dot{\rho}_k(\mathbf{q}) T_{\mathrm{q}}^{(\mathrm{G}+1)}(t,t') \left[1 - \mathcal{P}_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right] \\ &\times \int_0^1 \mathrm{d}\tau \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{\tau} \dot{\rho}_{k'}(\mathbf{q}') \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{-\tau} \Big\rangle^{t'}_{(\mathrm{G}+1)} \\ &= k\,\mathbf{q} \cdot D^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') \cdot k'\,\mathbf{q}', \end{aligned} \tag{4.12}$$

$$\begin{aligned}\Phi_{\mathbf{pp}}^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') &= \Big\langle \dot{\mathbf{p}}_k(\mathbf{q}) T_{\mathrm{q}}^{(\mathrm{G}+1)}(t,t') \left[1 - \mathcal{P}_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right] \\ &\times \int_0^1 \mathrm{d}\tau \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{\tau} \dot{\mathbf{p}}_{k'}(\mathbf{q}') \left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t')\right]^{-\tau} \Big\rangle^{t'}_{(\mathrm{G}+1)} \\ &= k\,\mathbf{q} : \eta^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t') : k'\,\mathbf{q}', \end{aligned} \tag{4.13}$$

$D^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$, $\eta^{(\mathrm{G}+1)}(k,\mathbf{q},k',\mathbf{q}';t,t')$ denote the coefficients of nonlinear diffusion and nonlinear viscosity of the electron subsystem of a semi-bounded metal within the "jellium" model. The generalized transport equations (4.10), (4.11) and the nonequilibrium statistical operator are strongly nonlinear ones in comparison with the transport equations (3.12), (3.13), which correspond to the Gaussian approximation for $\rho_{\mathrm{q}}^{(\mathrm{G})}(t)$.

5. Conclusion

Viscoelastic processes in the electron subsystem of a semi-bounded metal are described on the basis of the generalized "jellium" model with the use of the NSO method, where the parameters of the reduced description are the nonequilibrium average values of electron density and electron momentum. Applying the functional integration technique, we have calculated the quasi-equilibrium partition function for such a system in the case of the model pseudopotential of electron-ion interaction in a metal in the Gaussian approximation and in the following higher approximation, where the static correlations between the operators of electron density and electron momentum density are taken into account with the third-order cumulant averages.

We have also obtained the expressions for a nonequilibrium statistical operator, which enables us to go beyond the linear approximation with respect to the gradients of electrochemical potential and average electron density. In the respective approximations for a nonequilibrium statistical operator we have derived the generalized transport equations for a nonequilibrium average values of the electron density operator and the electron momentum density operator that can be applied to the description of strongly nonequilibrium processes for an electron subsystem of a semi-bounded metal. The generalized transport coefficients (that related, for example, with the generalized viscosity coefficient of the electron subsystem of a semi-bounded metal) which are contained in the corresponding transport equations are calculated using the quasi-equilibrium statistical operator in the respective approximations: the Gaussian one (3.1) and the following higher approximation (4.1). An important point in such an approach is that the time correlation functions and the generalized transport coefficients are calculated with the quasi-equilibrium statistical operator in the corresponding approximation and represent the functionals of the observable quantities $\langle\rho_k(\vec{q})\rangle^t$, $\langle\mathbf{p}_k(\mathbf{q})\rangle^t$ of a certain order. Of special interest in this approach are the investigations of the dynamic structure factor for the nonequilibrium electron subsystem of a semi-bounded metal.

A. Kawasaki-Gunton projection operator in the Gaussian approximation for $\rho_{\mathbf{q}}^{(\mathrm{G})}(t)$

Considering (3.1), (3.8) and

$$\frac{\delta\rho_{\mathrm{q}}^{(\mathrm{G})}(t)}{\delta\left\langle\rho_k(\mathbf{q})\right\rangle^t} = -\sum_{k',\mathbf{q}'}\left[\rho_{k'}(\mathbf{q}';\tau;t)-\left\langle\rho_{k'}(\mathbf{q}')\right\rangle^t\right][\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\rho_{\mathrm{q}}^{(\mathrm{G})}(t), \tag{A.1}$$

$$\frac{\delta\rho_{\mathrm{q}}^{(\mathrm{G})}(t)}{\delta\left\langle\mathbf{p}_k(\mathbf{q})\right\rangle^t} = -\sum_{k',\mathbf{q}'}\left[\mathbf{p}_{k'}(\mathbf{q}';\tau;t)-\left\langle\mathbf{p}_{k'}(\mathbf{q}')\right\rangle^t\right]\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\rho_{\mathrm{q}}^{(\mathrm{G})}(t), \tag{A.2}$$

where

$$\rho_{k'}(\mathbf{q}';\tau;t)=\int_0^1\mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{\tau}\rho_{k'}(\mathbf{q}')\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{-\tau},$$

$$\mathbf{p}_{k'}(\mathbf{q}';\tau;t)=\int_0^1\mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{\tau}\mathbf{p}_{k'}(\mathbf{q}')\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{-\tau},$$

we find the Kawasaki-Gunton projection operator:

$$P_{\mathrm{q}}^{(\mathrm{G})}(t)\rho' = \Bigg\{\rho_{\mathrm{q}}^{(\mathrm{G})}(t) + \sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\left[\rho_{k'}(\mathbf{q}';\tau,t) - \langle\rho_{k'}(\mathbf{q}')\rangle^{t}\right]$$
$$\times\,[\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\,\langle\rho_k(\mathbf{q})\rangle^{t}\,\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\,\mathrm{Sp}(\rho')$$
$$+\sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\left(\mathbf{p}_{k'}(\mathbf{q}';\tau,t) - \langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\right)\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\cdot\langle\mathbf{p}_k(\mathbf{q})\rangle^{t}\,\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\,\mathrm{Sp}(\rho')$$
$$-\sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\left(\rho_{k'}(\mathbf{q}';\tau,t) - \langle\rho_{k'}(\mathbf{q}')\rangle^{t}\right)[\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\,\mathrm{Sp}\left(\rho_k(\mathbf{q})\rho'\right)\rho_{\mathrm{q}}^{(\mathrm{G})}(t)$$
$$-\sum_{k,\mathbf{q}}\sum_{k',\mathbf{q}'}\left(\mathbf{p}_{k'}(\mathbf{q}';\tau,t) - \langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\right)\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\cdot\mathrm{Sp}(\mathbf{p}_k(\mathbf{q})\rho')\Bigg\}\rho_{\mathrm{q}}^{(\mathrm{G})}(t) \quad (\mathrm{A}.3)$$

Taking into account:

$$\mathrm{i}L_N\rho_{\mathrm{q}}^{(\mathrm{G})}(t) = \sum_{k,\mathbf{q}} W_{\rho\rho}^{(\mathrm{G})}(k,\mathbf{q};t)\int_0^1 \mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{\tau}\dot{\rho}_k(\mathbf{q})\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{-\tau}$$
$$- \sum_{k,\mathbf{q}} \mathbf{W}_{\mathbf{pp}}^{(\mathrm{G})}(k,\mathbf{q};t)\int_0^1 \mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{\tau}\cdot\dot{\mathbf{p}}_k(\mathbf{q})\left[\rho_{\mathrm{q}}^{(\mathrm{G})}(t)\right]^{-\tau}, \quad (\mathrm{A}.4)$$

where

$$W_{\rho\rho}^{(\mathrm{G})}(k,\mathbf{q};t) = \frac{\beta}{SL}\overline{\mu}_k(\mathbf{q};t)$$
$$= \frac{\beta}{SL}\left[\overline{S}_k(\mathbf{q}) - \left(\frac{\beta}{SL}\right)^{-1}\sum_{k',\mathbf{q}'}\langle\rho_{k'}(\mathbf{q}')\rangle^{t}\,[\mathfrak{M}^{\rho\rho}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q})\right], \quad (\mathrm{A}.5)$$
$$\mathbf{W}_{\mathbf{pp}}^{(\mathrm{G})}(k,\mathbf{q};t) = \sum_{k',\mathbf{q}'}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\cdot[\mathfrak{M}^{\mathbf{pp}}]^{-1}_{-k',-k}(-\mathbf{q}',-\mathbf{q}). \quad (\mathrm{A}.6)$$

B. Approximate determination of $\bar{\mu}_k(\mathrm{q};t)$ in $\rho_{\mathbf{q}}^{(\mathbf{G})}(t)$

Then, we apply the approximation [36] that linearize the equations (4.6), (4.7) with values $\overline{\mu}_k(\mathbf{q};t)$ (3.6) and $\mathbf{A}_k(\mathbf{q};t)$ (3.8) in the Gaussian approximation. So in (4.7) one can obtain:

$$\langle\mathbf{p}_k(\mathbf{q})\rangle^{t} = -\frac{\beta}{SL}\sum_{k_1,\mathbf{q}_1}\mathbf{A}_{k_1}(\mathbf{q}_1;t)\cdot G^{(3)}_{k_1,k}(\mathbf{q}_1,\mathbf{q};t), \quad (\mathrm{B}.1)$$
$$\mathbf{G}^{(3)}_{k_1,k}(\mathbf{q}_1,\mathbf{q};t) = \mathfrak{M}^{\mathbf{pp}}_{k_1,k}(\mathbf{q}_1,\mathbf{q})$$
$$+ \mathrm{i}\sum_{k',k_2}\sum_{\mathbf{q}',\mathbf{q}_2}\langle\rho_{k'}(\mathbf{q}';t)\rangle^{t}\,[\mathfrak{M}^{\rho\rho}]^{-1}_{k',k_2}(\mathbf{q}',\mathbf{q}_2)\mathfrak{M}^{\mathbf{pp}\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}). \quad (\mathrm{B}.2)$$

Defining the inverse function $\left[\mathbf{G}^{(3)}\right]^{-1}_{k_1,k}(\mathbf{q}_1,\mathbf{q};t)$ of $\mathbf{G}^{(3)}_{k_1,k}(\mathbf{q}_1,\mathbf{q};t)$:

$$\sum_{k'',\mathbf{q}''}\left[\mathbf{G}^{(3)}\right]^{-1}_{k',k''}(\mathbf{q}',\mathbf{q}'';t)\mathbf{G}^{(3)}_{k'',k_1}(\mathbf{q}'',\mathbf{q}_1;t) = \delta_{k',k_1}\delta_{\mathbf{q}',\mathbf{q}_1},$$

we find from (B.1) the following:

$$\mathbf{A}_{k_1}(\mathbf{q}_1;t) = -\left(\frac{\beta}{SL}\right)^{-1}\sum_{k',\mathbf{q}'}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\cdot\left[\mathbf{G}^{(3)}\right]^{-1}_{k',k_1}(\mathbf{q}',\mathbf{q}_1;t). \quad (\mathrm{B}.3)$$

Considering (B.3) with the linearization (4.6), we obtain the equation for defining $\overline{\mu}_k(\mathbf{q};t)$:

$$\begin{aligned}\langle\rho_k(\mathbf{q})\rangle^t &= \frac{\beta}{SL}\sum_{k',\mathbf{q}'}\left(\overline{S}_{k'}(\mathbf{q}')-\overline{\mu}_{k'}(\mathbf{q}';t)\right)\mathfrak{M}^{\rho\rho}_{k',k}(\mathbf{q}',\mathbf{q})\\ &+\mathrm{i}\frac{\beta}{SL}\sum_{k_1,k_2,k'}\sum_{\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}'}\langle\rho_{k'}(\mathbf{q}')\rangle^t\left[\mathfrak{M}^{\rho\rho}\right]^{-1}_{k',k_1}(\mathbf{q}',\mathbf{q}_1)\left(\overline{S}_{k_2}(\mathbf{q}_2)-\overline{\mu}_{k_2}(\mathbf{q}_2;t)\right)\mathfrak{M}^{\rho\rho\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})\\ &-\frac{3\mathrm{i}}{2!}\sum_{k',k''}\sum_{\mathbf{q}',\mathbf{q}''}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t\cdot\mathbf{K}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q})\cdot\langle\mathbf{p}_{k''}(\mathbf{q}'')\rangle^t,\end{aligned} \tag{B.4}$$

where

$$\mathbf{K}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q})=\sum_{k_1,\mathbf{q}_1}\sum_{k_2,\mathbf{q}_2}\left[\mathbf{G}^{(3)}\right]^{-1}_{k',k_1}(\mathbf{q}',\mathbf{q}_1;t)\boldsymbol{\mathfrak{M}}^{\mathbf{pp}\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})\left[\mathbf{G}^{(3)}\right]^{-1}_{k_2,k''}(\mathbf{q}_2,\mathbf{q}'';t) \tag{B.5}$$

is the function that takes into account the complicated renormalization of the cumulant average "momentum–momentum–density". Then, selecting terms from $\overline{\mu}_k(\mathbf{q};t)$, we rewrite (B.4):

$$\begin{aligned}\langle\rho_k(\mathbf{q})\rangle^t &= \frac{\beta}{SL}G^{(1)}_k(\mathbf{q})-\frac{\beta}{SL}\sum_{k',\mathbf{q}'}\overline{\mu}_{k'}(\mathbf{q}';t)G^{(2)}_{k',k}(\mathbf{q}',\mathbf{q};t)\\ &- \frac{3\mathrm{i}}{2!}\sum_{k',k''}\sum_{\mathbf{q}',\mathbf{q}''}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t\cdot\mathbf{K}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q})\cdot\langle\mathbf{p}_{k''}(\mathbf{q}'')\rangle^t,\end{aligned} \tag{B.6}$$

where

$$\begin{aligned}G^{(1)}_k(\mathbf{q}) &= \sum_{k',\mathbf{q}'}\Big[\overline{S}_{k'}(\mathbf{q}')\mathfrak{M}^{\rho\rho}_{k',k}(\mathbf{q}',\mathbf{q})\\ &+ \mathrm{i}\sum_{k_1,\mathbf{q}_1}\sum_{k_2,\mathbf{q}_2}\langle\rho_{k'}(\mathbf{q}')\rangle^t\left[\mathfrak{M}^{\rho\rho}\right]^{-1}_{k',k_1}(\mathbf{q}',\mathbf{q}_1)\overline{S}_{k_2}(\mathbf{q}_2)\mathfrak{M}^{\rho\rho\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q})\Big],\end{aligned} \tag{B.7}$$

$$\begin{aligned}G^{(2)}_{k',k}(\mathbf{q}',\mathbf{q};t) &= \mathfrak{M}^{\rho\rho}_{k',k}(\mathbf{q}',\mathbf{q})\\ &+ \sum_{k_1,\mathbf{q}_1}\sum_{k_2,\mathbf{q}_2}\langle\rho_{k'}(\mathbf{q}')\rangle^t\left[\mathfrak{M}^{\rho\rho}\right]^{-1}_{k',k_1}(\mathbf{q}',\mathbf{q}_1)\mathfrak{M}^{\rho\rho\rho}_{k_1,k_2,k}(\mathbf{q}_1,\mathbf{q}_2,\mathbf{q}).\end{aligned} \tag{B.8}$$

Defining $\left[G^{(2)}\right]^{-1}_{k_2,k}(\mathbf{q}_2,\mathbf{q};t)$, the inverse function of $G^{(2)}_{k',k}(\mathbf{q}',\mathbf{q};t)$:

$$\sum_{k'',\mathbf{q}''}\left[G^{(2)}\right]^{-1}_{k',k''}(\mathbf{q}',\mathbf{q}'';t)G^{(2)}_{k'',k_1}(\mathbf{q}'',\mathbf{q}_1;t)=\delta_{k',k_1}\delta_{\mathbf{q}',\mathbf{q}_1},$$

one can get from (B.6):

$$\begin{aligned}\overline{\mu}_k(\mathbf{q};t) &= -\left(\frac{\beta}{SL}\right)^{-1}\sum_{k',\mathbf{q}'}\left[\langle\rho_{k'}(\mathbf{q}')\rangle^t-\frac{\beta}{SL}G^{(1)}_{k'}(\mathbf{q}')\right]\left[G^{(2)}\right]^{-1}_{k',k}(\mathbf{q}',\mathbf{q};t)\\ &+ \frac{3\mathrm{i}}{2!}\left(\frac{\beta}{SL}\right)^{-1}\sum_{k',k''}\sum_{\mathbf{q}',\mathbf{q}''}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^t\cdot\overline{\mathbf{K}}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q};t)\cdot\langle\mathbf{p}_{k''}(\mathbf{q}'')\rangle^t,\end{aligned} \tag{B.9}$$

where

$$\overline{\mathbf{K}}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q};t)=\sum_{k_1,\mathbf{q}_1}\mathbf{K}^{\mathbf{pp}\rho}_{k',k'',k_1}(\mathbf{q}',\mathbf{q}'',\mathbf{q}_1;t)\left[G^{(2)}\right]^{-1}_{k_1,k}(\mathbf{q}_1,\mathbf{q};t). \tag{B.10}$$

Now considering (B.9), (B.3) we can write down the entropy operator $\hat{S}^{(\mathrm{G}+1)}(t)$ (4.2) as follows:

$$\hat{S}^{(\mathrm{G}+1)}(t)=\beta\frac{N}{2S}{\sum_{\mathbf{q}}}'\nu(\mathbf{q}|0)+\ln Z_{\mathrm{jell}}+\beta H$$
$$+\sum_{k,\mathbf{q}}\Bigg\{\sum_{k',\mathbf{q}'}\left[\langle\rho_{k'}(\mathbf{q}')\rangle^{t}-\frac{\beta}{SL}G^{(1)}_{k'}(\mathbf{q}')\right]\left[G^{(2)}\right]^{-1}_{k',k}(\mathbf{q}',\mathbf{q};t)\rho_{k}(\mathbf{q})$$
$$-\frac{3\mathrm{i}}{2!}\sum_{k',k''}\sum_{\mathbf{q}',\mathbf{q}''}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\cdot\overline{\mathbf{K}}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q};t)\cdot\langle\mathbf{p}_{k''}(\mathbf{q}'')\rangle^{t}\rho_{k}(\mathbf{q})\Bigg\}-\Phi^{(\mathrm{G}+1)}(t),\quad\text{(B.11)}$$

where $\Phi^{(\mathrm{G}+1)}(t)$ according to (4.3) and (B.3), (B.9) is a function of a higher order with respect to the parameters of the reduced description $\langle\rho_k(\mathbf{q})\rangle^t$, $\langle\mathbf{p}_k(\mathbf{q})\rangle^t$, including the sixth with respect to $\langle\mathbf{p}_k(\mathbf{q})\rangle^t$ and its structure is important in calculating the Kawasaki-Gunton projection operator (2.6) in terms of $\dfrac{\delta\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)}{\delta\langle\rho_k(\mathbf{q})\rangle^t}$, $\dfrac{\delta\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)}{\delta\langle\mathbf{p}_k(\mathbf{q})\rangle^t}$. Considering the above, we get

$$\begin{aligned}\mathrm{i}L_N\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t) &= \sum_{k,\mathbf{q}}\int_0^1\mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)\right]^{\tau}\dot{\rho}_k(\mathbf{q})\left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)\right]^{1-\tau}W^{(\mathrm{G}+1)}_{\rho\rho}(k,\mathbf{q};t)\\ &+ \sum_{k,\mathbf{q}}\int_0^1\mathrm{d}\tau\left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)\right]^{\tau}\dot{\mathbf{p}}_k(\mathbf{q})\left[\rho_{\mathrm{q}}^{(\mathrm{G}+1)}(t)\right]^{1-\tau}\cdot\mathbf{W}^{(\mathrm{G}+1)}_{\mathbf{pp}}(k,\mathbf{q};t),\end{aligned}\quad\text{(B.12)}$$

where

$$\begin{aligned}W^{(\mathrm{G}+1)}_{\rho\rho}(k,\mathbf{q};t) &= \sum_{k',\mathbf{q}'}\left[\langle\rho_{k'}(\mathbf{q}')\rangle^{t}-\frac{\beta}{SL}G^{(1)}_{k'}(\mathbf{q}')\right]\left[G^{(2)}\right]^{-1}_{k',k}(\mathbf{q}',\mathbf{q};t)\\ &- \frac{3\mathrm{i}}{2!}\sum_{k',k''}\sum_{\mathbf{q}',\mathbf{q}''}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\cdot\overline{\mathbf{K}}^{\mathbf{pp}\rho}_{k',k'',k}(\mathbf{q}',\mathbf{q}'',\mathbf{q};t)\cdot\langle\mathbf{p}_{k''}(\mathbf{q}'')\rangle^{t},\end{aligned}\quad\text{(B.13)}$$

$$\mathbf{W}^{(\mathrm{G}+1)}_{\mathbf{pp}}(k,\mathbf{q};t)=\sum_{k',\mathbf{q}'}\langle\mathbf{p}_{k'}(\mathbf{q}')\rangle^{t}\cdot\left[\mathbf{G}^{(3)}\right]^{-1}_{k',k}(\mathbf{q}',\mathbf{q};t).\quad\text{(B.14)}$$

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В’язко-еластичний опис нерівноважної електронної підсистеми напівобмеженого металу в узагальненій моделі “желе”

П. П. Костробій[1], Б. М. Маркович[1], А. І. Василенко[2], М. В. Токарчук[1,2]

[1] Національний університет “Львівська політехніка”, вул. С. Бандери, 79013 Львів, Україна

[2] Інститут фізики конденсованих систем НАН України, вул. І. Свєнціцького, 1, 79011 Львів, Україна

Запропоновано в’язко-еластичний опис електронної підсистеми напівобмеженого металу на основі узагальненої моделі “желе” із застосуванням методу нерівноважного статистичного оператора Зубарєва. Отримано нерівноважний статистичний оператор та відповідні узагальнені рівняння переносу для нерівноважних середніх значень операторів густин числа електронів та їх імпульсу у гаусовому та вищому за ним наближені, що відповідає кумулянтним середнім третього порядку при розрахунку квазірівноважної статистичної суми методом функціонального інтегрування.

Ключові слова: *узагальнена модель “желе”, нерівноважний статистичний оператор Зубарєва, напівобмежений метал, рівняння переносу, квазірівноважна статистична сума*