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Fluctuation-dissipation theorem and frequency moments of response functions of a dense plasma to an electromagnetic field

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The fluctuation-dissipative theorem and frequency moments for quadratic functions of the reaction of a dense plasma in a constant magnetic field to an electromagnetic field are considered. The frequency moments of the corresponding correlation functions are studied. A model approach is proposed to calculate quadratic reaction functions that determine nonlinear phenomena caused by the quadratic interaction of electromagnetic waves in a dense charged medium (Coulomb systems, plasma) in a constant magnetic field.

Keywords: dense plasma, nonlinear fluctuation-dissipation theorem, quadratic response functions, nonlinear phenomena.

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Introduction

Recently, nonlinear phenomena in various media (crystals, graphene, various mixtures, composites, weakly ionized gases, suspensions etc.) are being actively studied; they are related to nonlinear interaction of electromagnetic (EM) waves (see, for instance, [1-4]). At the same time, less attention has been paid to nonlinear phenomena in dense charged systems (Coulombic systems and plasma), caused by a quadratic response to an electromagnetic field — second harmonic generation (SHG), parametric generation of radiation (PGR) (see [5] and references therein), or by thermal perturbations — multiplicity of thermal conditions in media with volumetric heat release [6,7]. SHG and PGR in dense plasma in laboratory conditions can be implemented in the presence of stationary relatively strong magnetic fields [5]. Studied have shown (see, for instance, [5,7,8–11]) that successive analytic calculation of precise formal expressions (obtained according to the response theory [12,13]) for quadratic response functions (QRF) of dense charged media with a strong interparticle interaction under perturbations is impossible, computer modeling of precise expressions for QRFs even for model Coulombic systems is difficult, therefore, model approaches should be used to determine these functions. One of the model variants can be the application (for the response function) of an explicit approximation with adjustment parameters determined from precise frequency moments of QRF and the corresponding correlators [5–7]. In the present paper we will consider the quadratic fluctuation-dissipative theorem and frequency moments of quadratic response functions under the action of an electromagnetic field of dense charged media in a constant magnetic field, on which the suggested model is based.

1. Quadratic fluctuation-dissipative theorem

Let us define precise expressions for the quadratic response function of a charged medium (dense plasma) in a constant magnetic field with vector potential $\mathbf{A}(\mathbf{r})$ under the action of an electromagnetic field according to the nonlinear response theory. In doing so, we will use the complete Hamiltonians of the system ($\widehat{H}$), medium (H_0) and the perturbation Hamiltonian H^{ext} related to exposure of the medium to an external field $\mathbf{D}(\mathbf{r}, t)$, in the known form (exclusive of the particle spin; see, for instance, [12]).

$$\widehat{H} = H_0 + H^{\text{ext}},$$

$$H_0 = \Sigma_\mu \Sigma_i \frac{1}{2m_i} \left[p_{\mu i} - \frac{e_i}{c} A_\mu(\mathbf{r}_i)\right]^2 + U\{\mathbf{r}_i\},$$

$$H^{\text{ext}} = -\Sigma_i e_i \mathbf{r}_i \cdot \mathbf{D}(\mathbf{r}, t) e^{\eta t}. \quad (1)$$

Here c, m_i, e_i, $p_{\mu i}$, A_μ, $U\{\mathbf{r}_i\}$ are respectively the speed of light, mass, charge, μ-component of impulse of the i-th particle, μ — component of the vector potential of the constant external magnetic field and energy of interaction of the medium (plasma) particles with each other, $\Sigma_i e_i \mathbf{r}_i$ — dipole moment of the medium, η — small positive quantity that ensures adiabaticity of inclusion of perturbation (and causality — see [13]). Let us write H^{ext} for generality as

$$H^{\text{ext}} = -\sum_j \int d\mathbf{r} B_j(\mathbf{r}) b_j^{\text{ext}}(\mathbf{r}, t). \quad (2)$$

Here $B(\mathbf{r})$ is a certain observable system property (e.g., space charge density, electric current density, dipole moment), $b^{\text{ext}}(\mathbf{r}, t)$ is the corresponding generalized force (e.g., external electric field). Let us write out an expression for response of a certain observable system property $B(\mathbf{r})$ to perturbation (1) [13]

$$\langle B\rangle = \langle B\rangle_0 + \sum_{n=1}^{\infty} \frac{1}{(i\hbar)^n} \int_{-\infty}^{t}\int_{-\infty}^{t_1} \ldots \int_{-\infty}^{t_{n-1}} Sp\{B(\mathbf{r})[H^{\text{ext}}(t_1)$$

$$\times [H^{\text{ext}}(t_2)\ldots[H^{\text{ext}}(t_n), \rho_e]\ldots]\}dt_1\ldots dt_n. \quad (3)$$

Here $\langle\ldots\rangle$, $\langle\ldots\rangle_0$ denotes averaging by ρ and ρ_e (equilibrium matrix of density) respectively, [..,..] — commutator. The quadratic response has the form

$$\langle B\rangle^{(2)} = \frac{1}{(i\hbar)^2} \int_{-\infty}^{t}\int_{-\infty}^{t_1} Sp\{B(\mathbf{r})[H^{\text{ext}}(t_1)[H^{\text{ext}}(t_2, \rho_e)]]\}dt_1 dt_2,$$

$$\langle B_i\rangle^{(2)} = \sum_{j,k} \int_{-\infty}^{t}\int_{-\infty}^{t_1} \hat{\chi}^{(2)}_{ijk}(t-t_1, t-t_2; \mathbf{r}-\mathbf{r}_1, \mathbf{r}-\mathbf{r}_2)$$

$$\times b_j^{\text{ext}}(\mathbf{r}_1, t_1) b_k^{\text{ext}}(\mathbf{r}_2, t_2) dt_1 dt_2 d\mathbf{r}_1 d\mathbf{r}_2, \quad (4)$$

$B(\mathbf{r}, t)$, $H^{\text{ext}}(t)$ in these expressions are operators in Heisenberg representation ($H^{\text{ext}}(t) = e^{iH_0t}H^{\text{ext}}e^{-iH_0t}$, here H_0 is the Hamiltonian of an unperturbed system, $\{b_j, b_k$ are perturbations. Let us write out, having introduced the θ-functions, the second-order response function in a symmetrized form (according to the last two indices) (see, for instance, [7]; V is system volume):

$$\hat{\chi}^{(2)}_{ijk}(\mathbf{r}-\mathbf{r}_1, t-t_1, \mathbf{r}-\mathbf{r}_2, t-t_2) = -\frac{\theta(t-t_1)\theta(t-t_2)}{2\hbar^2 V}$$

$$\times \{\theta(t_1-t_2)\langle[[B_i(\mathbf{r}, t), B_j(\mathbf{r}_1, t)_1], B_k(\mathbf{r}_2, t_2)]\rangle_0$$

$$+ \theta(t_2-t_1)\langle[[B_i(\mathbf{r}, t), B_k(\mathbf{r}_2, t_2)], B_j(\mathbf{r}_1, t_1)]\rangle_0\}. \quad (5)$$

Thanks to this definition of QRF, the time integration limits in (4) can be extended to infinity. Let us substitute (5) in (4) and go to a Fourier representation

$$\langle B_i(\mathbf{k}, \omega)\rangle^{(2)} = \frac{1}{V(2\pi)^2} \sum_{k_1,k_2} \int d\omega_1 d\omega_2 \hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)$$

$$\times b_j^{\text{ext}}(\mathbf{k}_1, \omega_1) b_k^{\text{ext}}(\mathbf{k}_2, \omega_2). \quad (6)$$

The expressions for $\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)$ and $\langle B_i(-\mathbf{k}, -\omega)B_j(\mathbf{k}_1, \omega_1)B_k(\mathbf{k}_2, \omega_2)\rangle_0$ are

$$\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2) = -\frac{N}{2\hbar^2 V}\left\{\int \frac{d\omega_1'}{2\pi}\int \frac{d\omega_2'}{2\pi}\right.$$

$$\times [1/i(\omega_1' - \omega_1)]\,[1/i(\omega_1' + \omega_2' - \omega_1 - \omega_2)]$$

$$\times [S(012) + S(210) - S(102) - S(201)]$$

$$+ \int \frac{d\omega_1'}{2\pi}\int \frac{d\omega_2'}{2\pi}[1/i(\omega_2' - \omega_2)][1/i(\omega_1' + \omega_2' - \omega_1 - \omega_2)]$$

$$\left.\times [S(021) + S(120) - S(201) - S(102)]\right\}, \quad (7)$$

$$\langle B_i(-\mathbf{k}, -\omega)B_j(\mathbf{k}_1, \omega_1)B_k(\mathbf{k}_2, \omega_2)\rangle_0$$

$$= 2\pi N\delta(\omega - \omega_1 - \omega_2)\delta(\mathbf{k} - \mathbf{k}_1 - \mathbf{k}_2)S(012). \quad (8)$$

Set $\{a, b, c\}$ in correlator $S(abc)$ denotes combination k, ω with the corresponding indices. Symmetry of response function $\hat{\chi}^{(2)}_{ijk}$ in relation to the two last indices is evident from definition (5). For the Fourier transform of response function $\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)$ the ratios are

$$\hat{\chi}^{(2)}_{ikj}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2) = \hat{\chi}^{(2)}_{ijk}(\mathbf{k}_2, \omega_2; \mathbf{k}_1, \omega_1),$$

$$\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2) = \hat{\chi}^{(2)^*}_{ijk}(-\mathbf{k}_1, -\omega_1; -\mathbf{k}_2, -\omega_2), \quad (9)$$

since $\hat{\chi}^{(2)}_{ijk}(\mathbf{r}_1, \tau_1; \mathbf{r}_2, \tau_2)$ is a real function, which follows from its phenomenological definition. The real and imaginary parts $\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)$ are interrelated similarly to the Kramers–Kronig relations for linear response functions [6,7]. It should be noted than (5), (7) can be considered as one of the simplest forms of the non-linear fluctuation-dissipative theorem (NFDT). By applying the Sokhotski formula $(1/(x \pm i\delta) = P(1/x) \mp i\pi\delta(x))$ [13], we find relations between the real and imaginary parts of the response function and $S(abc)$ correlators. By making combinations from $\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)$ and by considering the properties of a cyclic rearrangement of operators in $S(abc)$ $(\exp(-\beta\omega_a\hbar)S(abc) = S(bca))$, we obtain the ratios between $\operatorname{Re}\hat{\chi}^{(2)}_{ijk}$ and the $S(abc)$ correlators, which represent one of the variants of a non-linear FDT (see [7] and references therein):

$$-\frac{\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1, \omega_1; \mathbf{k}_2, \omega_2)}{\omega_1\omega_2} - \frac{\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_2, \omega_2; \mathbf{k}, \omega)}{\omega\omega_2}$$

$$-\frac{\operatorname{Re}\hat{\chi}^{(2)}_{ijk}(\mathbf{k}, \omega; \mathbf{k}_1, \omega_1)}{\omega_1\omega} = \frac{-n}{4\hbar^2}\left\{\frac{S(102) + S(201)}{\omega_1\omega_2}\right.$$

$$\left.-\frac{S(012) + S(210)}{\omega\omega_2} - \frac{S(021) + S(120)}{\omega_1\omega}\right\}. \quad (10)$$

These expressions can be in principle applied to calculate frequency moments of the real part of quadratic response functions, since they relate frequency moments of response functions to frequency moments of correlators. Calculation of frequency moments of correlators is described in the next section. By setting a QRF, according to the suggested model (see Introduction), as an explicit approximation with adjustment parameters, we can determine these parameters by comparing the frequency moments in the right and left members (10). Adjustment parameters for QRF depend on thermophysical characteristics of the charged medium (dense plasma, Coulombic system) and magnetic field. It should be noted that correlators in the linear case are related to the imaginary part of the response function and the FDT contains one response function and one correlator (see, for instance, [6]). The quadratic FDT looks the simplest in the classical limit. Taking into account $\hbar \to 0$, (10) is substituted by

$$-\omega \mathrm{Re}\,\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2) - \omega_1 \mathrm{Re}\,\hat{\chi}^{(2)}_{ijk}(\mathbf{k}_2,\omega_2;\mathbf{k},\omega)$$
$$-\omega_2 \mathrm{Re}\,\hat{\chi}^{(2)}_{ijk}(\mathbf{k},\omega;\mathbf{k}_1,\omega_1) = \frac{n\beta^2}{8}\left[S(210)\omega_2\omega_1(\omega_1+\omega_2)\right.$$
$$\left.+S(201)\omega_2\omega_1(\omega_2+\omega_1)\right] = \frac{n\beta^2}{4}S(012)\omega\omega_1\omega_2, \qquad (11)$$

since correlators in the classical limit are invariant in relation to the rearrangement of arguments. Formula (11) is used below since laboratory dense plasma is often non-degenerate [5].

Let us consider QRF $\hat{\chi}^{(2)}_{p,ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ and the corresponding correlators. These QRFs determine the quadratic contribution to polarization of charged media $\mathbf{P}^{(2)}$ ($\mathbf{P} = \Sigma_i e_i \mathbf{r}_i/V$), i.e. SHG and PGR (see, for instance, [7] and references therein). From the ratio (see, for instance, [14])

$$\dot{\mathbf{P}} = 4\pi\mathbf{j} \qquad (12)$$

QRF $\hat{\chi}^{(2)}_{J,ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$, related to $\hat{\chi}^{(2)}_{p,ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$, can be assigned in a form similar to (7). Tensor $\hat{\chi}^{(2)}_{J,ijk}$ describes the quadratic contribution $j^{(2)}_i(k,\omega)$ to the electric current density of the charged medium under the EM-field action and, naturally, determines the quadratic electrical conductivity $\hat{\sigma}^{(2)}_{ijk}(\mathbf{k}_1,\omega_1;\mathbf{k}_2,\omega_2)$ (cf. [7]). In $\hat{\chi}^{(2)}_{J,ijk}$ (and $\hat{\sigma}^{(2)}_{ijk}$), $S_j(abc)$ corresponds to the correlator of current densities.

At the same time, the NDFT in form (11) is inconvenient in implementation of a model approach that consists in the application of an explicit approximation for response functions $\hat{\chi}^{(2)}_{p,ijk}$ (e.g., in the form from [5]) or $\hat{\chi}^{(2)}_{J,ijk}$ with adjustment parameters and precise values of frequency moments of the corresponding correlators, because tensors in (11) in this case have 18 components each and are not invariant in relation to the representations. Therefore, we will perform a convolution (according to wave vectors $(\mathbf{k},\mathbf{k}_1,\mathbf{k}_2)$) for $\hat{\chi}^{(2)}_{J,ijk}$ and the correlator in the right member of (11) and use the charge conservation equation (13) (see, for instance, [6])

$$\partial\rho/\partial t = -\mathrm{div}\,\mathbf{J}. \qquad (13)$$

In other words, by making a longitudinal projection of tensors $\hat{\chi}^{(2)}_{J,ijk}$ and correlator in (11), we get an NFDT in the form of an invariant ratio for scalars: QRF $\hat{\chi}^{(2)}$ of plasma in a constant magnetic field to a longitudinal field in form (11), which describes the contribution of $\rho^{(2)}(k,\omega)$ to the total charge density, and a correlator of charge densities. Let us write out the longitudinal NFDT

$$-\omega \mathrm{Re}\,\hat{\chi}^{(2)}(12) - \omega_1 \mathrm{Re}\,\hat{\chi}^{(2)}(20) - \omega_2 \mathrm{Re}\,\hat{\chi}^{(2)}(01)$$
$$= \frac{n\beta^2}{4}S_\rho(012)\omega\omega_1\omega_2. \qquad (14)$$

In (14) $\hat{\chi}^{(2)}$ is the QRF of charges [7] to an external field, set (a,b) denotes $k\omega$ combination with the corresponding indices.

Thus, (14) can be used to determine the „scalar“ adjustment parameters for the $\hat{\chi}^{(2)}$ assigned according to the suggested model in the form of an explicit approximation with adjustment parameters — see Introduction), which depend on thermophysical characteristics of a charged medium (dense plasma, Coulombic system) and magnetic field. These parameters should be used for the whole range of QRFs ($\hat{\chi}^{(2)}_{p,ijk}$, $\hat{\chi}^{(2)}_{J,ijk}$, $\hat{\chi}^{(2)}$) interrelated with each other.

2. Frequency moments of correlators for a charged medium in a constant magnetic field

Let us consider a procedure for calculation of frequency moments of density correlator $S_\rho(abc)$ for equilibrium dense plasma in a constant magnetic field, taking into account the parity of density operators in relation to time reversal. Frequency moments $S_\rho(012)$ in the classical limit are determined according to the relation (see, for instance, [6,7])

$$S^{r_1,r_2}_\rho(012) = \int\limits_{-\infty}^{\infty} d\omega_1 \int\limits_{-\infty}^{\infty} d\omega_2 \omega_1^{r_1}\omega_2^{r_2} S_{cl}(012) = (i)^{r_1+r_2}$$
$$\times \left.\frac{\partial^{r_1+r_2}S_{cl}(012)}{\partial t_1^{r_1}\partial t_2^{r_2}}\right|_{t_1-t_2=0} = (i)^{r_1+r_2}\frac{\partial^{r_1}}{\partial t_1^{r_1}}\frac{\partial^{r_2}}{\partial t_2^{r_2}}$$
$$\times \frac{1}{N}\langle\rho(k,0)\rho(k_1,t_1)\rho(k_2,t_2)\rangle_0\big|_{t_1=t_2=0}. \qquad (15)$$

Densities in $S^{r_1,r_2}_\rho(012)$ ($\mathbf{k}\parallel\mathbf{z}$) have the form

$$\rho(\mathbf{k},0) = \sum_{t=1}^{N} ez_i e^{-ikz_i},$$
$$\rho(\mathbf{k}_1,t_1) = \sum_{j=1}^{N} ez_j e^{-i\left(k_1^z z_j(t_1) + k_1^x x_j(t_1)\right)},$$
$$\rho(\mathbf{k}_2,t_2) = \sum_{k=1}^{N} ez_k e^{-i\left(k_2^z z_k(t_2) + k_2^x x_k(t_2)\right)}. \qquad (16)$$

N in (16) is the total number of particles in the system. We can consider moments $S_\rho^{r_1,r_2}(012)$ at $r_1, r_2 = 0.0$; 0.2; 2.2; 1.3 etc. according to (15), (16). Let us write out the first time derivatives from densities (v_j^z is the z-coordinate of speed of the j-th particle)

$$\dot\rho_j(\mathbf{k}_1, t_1) = \sum_{j=1}^N ez_j\left[-ik_1^z v_j^z(t_1) - ik_1^x v_j^x(t_1)\right] \times e^{-i\left(k_1^z z_j(t_1)+k_1^x x_j(t_1)\right)},$$

$$\dot\rho_k(\mathbf{k}_2, t_2) = \sum_{k=1}^N ez_k\left[-ik_2^z v_k^z(t_2) - ik_2^x v_k^x(t_2)\right] \times e^{-i\left(k_2^z z_k(t_2)+k_2^x x_k(t_2)\right)},$$

$$\ddot\rho_j(\mathbf{k}_1, t_1) = \sum_{j=1}^N ez_j\left[-ik_1^z \dot v_j^z(t_1) - ik_1^x \dot v_j^x(t_1)\right] \times e^{-i\left(k_1^z z_j(t_1)+k_1^x x_j(t_1)\right)} + \sum_{j=1}^N ez_j\left[-ik_1^x v_j^x(t_1) - ik_1^z v_j^z(t_1)\right]^2 \times e^{-i\left(k_1^z z_j(t_1)+k_1^x x_j(t_1)\right)},$$

$$\ddot\rho_k(\mathbf{k}_2, t_2) = \sum_{k=1}^N ez_k\left[-ik_2^z \dot v_k^z(t_2) - ik_2^x \dot v_k^x(t_2)\right] \times e^{-i\left(k_2^z z_k(t_2)+k_2^x x_k(t_2)\right)} + \sum_{k=1}^N ez_k\left[-ik_2^x v_k^x(t_2) - ik_2^z v_k^z(t_2)\right]^2 \times e^{-i\left(k_2^z z_k(t_2)+k_2^x x_k(t_2)\right)}.$$

Let us write an expression which is a frequency integral $S_\rho^{r_1,r_2}(012)$ (see (14)) at $r_1,\ r_2 = 2.2$:

$$(i)^{r_1+r_2}\frac{\partial^{r_1}}{\partial t_1^{r_1}}\frac{\partial^{r_2}}{\partial t_2^{r_2}}\frac{1}{N}\langle\rho(\mathbf{k},0)\rho(\mathbf{k}_1,t_1)\rho(\mathbf{k}_2,t_2)\rangle_0\Big|_{t_1-t_2=0} = (i)^4\frac{1}{N}\langle\rho(\mathbf{k},0)\ddot\rho(\mathbf{k}_1,t_1)\ddot\rho(\mathbf{k}_2,t_2)\rangle_0\Big|_{t_1-t_2=0} = (i^4/N)$$
$$\times\Big\langle\sum_{i=1}^N ez_i e^{-ikz_i}\Big\{\sum_{j=1}^N ez_j\left[-ik_1^z\dot v_j^z(t_1) - ik_1^x\dot v_j^x(t_1)\right] \times e^{-i\left(k_1^z z_j(t_1)+k_1^x x_j(t_1)\right)} + \sum_{j=1}^N ez_j\left[-ik_1^x v_j^x(t_1) - ik_1^z v_j^z(t_1)\right]^2$$
$$\times e^{-i\left(k_1^z z_j(t_1)+k_1^x x_j(t_1)\right)}\Big\}\Big\{\sum_{k=1}^N ez_k\left[-ik_2^z\dot v_k^z(t_2) - ik_2^x\dot v_k^x(t_2)\right] \times e^{-i\left(k_2^z z_k(t_2)+k_2^x x_k(t_2)\right)} + \sum_{k=1}^N ez_k\left[-ik_2^x v_k^x(t_2) - ik_2^z v_k^z(t_2)\right]^2$$
$$\times e^{-i\left(k_2^z z_k(t_2)+k_2^x x_k(t_2)\right)}\Big\}\Big\rangle_0. \quad (17a)$$

Calculations as per (15)−(17a) are performed using the following definitions for the averaging of sums and products (see [6] and references therein, V is system volume, F_s is s-partial correlation function):

$$\overline{M_s} = \frac{N(N-1)\dots(N-s+1)}{s!V^s}\int_V\dots\int_V f(r_1\dots r_s) \times F_s(r_1\dots r_s)dr_1\dots dr_s,$$

$$M_s = \sum_{1\le i_1\dots i_s\le N} f(r_{i_1}\dots r_{i_s}),$$

$$\left\langle\frac{\partial U}{\partial x_i}F(r_1\dots r_N)\right\rangle_0 = \beta^{-1}\langle\partial F(r_1\dots r_N)/\partial x_i\rangle_0. \quad (17b)$$

Similarly to [6], we find the following for $S_\rho^{2,2}(012)$ (signs of exponent sums and averaging are omitted in formulas 1−4 for brevity — cf. (17a); H — magnetic field):

1. $$[-ik_1^z\dot v_j^z - ik_1^x\dot v_j^x][-ik_2^z\dot v_k^z - ik_2^x\dot v_k^x] = -k_1^z\dot v_j^z k_2^z\dot v_k^z - k_1^x\dot v_j^x k_2^z\dot v_k^z - k_1^z\dot v_j^z k_2^x\dot v_k^x - k_1^x\dot v_j^x k_2^x\dot v_k^x = -k_1^z k_2^z\frac{1}{m_j m_k}\frac{\partial U}{\partial z_j}\frac{\partial U}{\partial z_k} + k_1^x k_2^z\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j} + \frac{ez_j}{cm_j}\left(v_j^y H\right)^x\right)\frac{1}{m_k}\frac{\partial U}{\partial z_k}$$
$$+ k_1^z k_2^x\frac{1}{m_j}\frac{\partial U}{\partial z_j}\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k} + \frac{ez_k}{cm_k}\left(v_k^y H\right)^x\right) \times -k_2^x\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j} + \frac{ez_j}{cm_j}\left(v_j^y H\right)^x\right) \times k_1^x\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k} + \frac{ez_k}{cm_k}\left(v_k^y H\right)^x\right);$$

2. $$[-ik_1^z\dot v_j^z - ik_1^x\dot v_j^x][-ik_2^x v_k^x - ik_2^z v_k^z]^2 = \Big[ik_1^z \times\left(\frac{1}{m_j}\frac{\partial U}{\partial z_j}\right) - ik_1^x\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j} + \frac{ez_j}{cm_j}\left(v_j^y H\right)^x\right)\Big]$$
$$\times[-(k_2^x v_k^x)^2 - (k_2^z v_k^z)^2 - 2k_2^x v_k^x k_2^z v_k^z] = -ik_1^z\left(\frac{1}{m_j}\frac{\partial U}{\partial z_j}\right) \times[(k_2^x v_k^x)^2 + (k_2^z v_k^z)^2 + 2k_2^x v_k^x k_2^z v_k^z] - ik_1^x\Big(-\frac{1}{m_j}\frac{\partial U}{\partial x_j}$$
$$+\frac{ez_j}{cm_j}\left(v_j^y H\right)^x\Big)][-(k_2^x v_k^x)^2 - (k_2^z v_k^z)^2 - 2k_2^x v_k^x k_2^z v_k^z];$$

3. $$[-ik_1^x v_j^x - ik_1^z v_j^z]^2[-ik_2^z\dot v_k^z - ik_2^x\dot v_k^x] = -[(k_1^x v_j^x)^2 + (k_2^z v_j^z)^2 + 2(k_1^x v_j^x)(k_1^z v_j^z]\Big[ik_2^z\left(\frac{1}{m_k}\frac{\partial U}{\partial z_k}\right) - ik_2^x\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k} + \frac{ez_k}{cm_k}\left(v_k^y H\right)^x\right)\Big];$$

4. $[-ik_1^x v_j^x - ik_1^z v_j^z]^2[-ik_2^x \dot{v}_k^x - ik_2^z \dot{v}_k^z]^2 = [(k_1^x v_j^x)^2 + (k_1^z v_j^z)^2 + 2(k_1^x v_j^x)(k_1^z v_j^z]\,[(k_2^x v_k^x)^2 + (k_2^z v_k^z)^2 + 2k_2^x v_k^x k_2^z v_k^z]$.

Averaging is performed according to the Gibbs distribution (in the classical limit of Hamiltonian H_0) in (17a), using expressions 1−4. After the selection according to the speed integration results, the term with H remains only in item 1, the other summands in moment $S_\rho^{2,2}(012)$ do not depend on magnetic field. Let us write out the expressions corresponding to items 1−4:

$$1.\quad -k_1^z k_2^z \frac{1}{m_j m_k}\frac{\partial U}{\partial z_j}\frac{\partial U}{\partial z_k} + k_1^x k_2^z\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j} + \frac{ez_j}{cm_j}\left(v_j^y H\right)^x\right)\frac{1}{m_k}\frac{\partial U}{\partial z_k} + k_1^z k_2^x \frac{1}{m_j}\frac{\partial U}{\partial z_j}\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k} + \frac{ez_k}{cm_k}\left(v_k^y H\right)^x\right) - k_2^x\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j} + \frac{ez_j}{cm_j}\left(v_j^y H\right)^x\right) \times k_1^x\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k} + \frac{ez_k}{cm_k}\left(v_k^y H\right)^x\right) \to (i^4/N) \times \Big\langle \sum_{i=1}^N ez_i e^{-ikz_i}\sum_{j=1}^N ez_j e^{-i(k_1^z z_j + k_1^x x_j)}\sum_{k=1}^N ez_k e^{-i(k_2^z z_k + k_2^x x_k)} \times \left\{-k_1^z k_2^z \frac{1}{m_j m_k}\frac{\partial U}{\partial z_j}\frac{\partial U}{\partial z_k} + k_1^x k_2^z\left(-\frac{1}{m_j}\frac{\partial U}{\partial x_j}\right)\frac{1}{m_k}\frac{\partial U}{\partial z_k} + k_1^z k_2^x \frac{1}{m_j}\frac{\partial U}{\partial z_j}\left(-\frac{1}{m_k}\frac{\partial U}{\partial x_k}\right) - k_1^x k_2^x \frac{1}{m_j}\frac{\partial U}{\partial x_j}\frac{1}{m_k}\frac{\partial U}{\partial x_k} - k_1^x k_2^x \frac{ez_j}{cm_j}(v_j^y H)^x \frac{ez_k}{cm_k}(v_k^y H)^x\right\}\Big\rangle_0;$$

$$2.\quad (i^4/N)\Big\langle \sum_{i=1}^N ez_i e^{-ikz_i}\sum_{j=1}^N ez_j e^{-i(k_1^z z_j + k_1^x x_j)} \times \sum_{k=1}^N ez_k e^{-i(k_2^z z_k + k_2^x x_k)}\left[-ik_1^z\left(\frac{1}{m_j}\frac{\partial U}{\partial z_j}\right) - ik_1^x\left(\frac{1}{m_j}\frac{\partial U}{\partial x_j}\right)\right][(k_2^x v_k^x)^2 + (k_2^z v_k^z)^2]\Big\rangle_0;$$

$$3.\quad (i^4/N)\Big\langle \sum_{i=1}^N ez_i e^{-ikz_i}\sum_{j=1}^N ez_j e^{-i(k_1^z z_j + k_1^x x_j)} \times \sum_{k=1}^N ez_k e^{-i(k_2^z z_k + k_2^x x_k)}[-(k_1^x v_j^x)^2 - (k_1^z v_j^z)^2] \times \left[-ik_2^z\left(\frac{1}{m_k}\frac{\partial U}{\partial z_k}\right) - ik_2^x\left(\frac{1}{m_k}\frac{\partial U}{\partial x_k}\right)\right]\Big\rangle_0;$$

$$4.\quad [(k_1^x v_j^x)^2 + (k_1^z v_j^z)^2 + 2(k_1^x v_j^x)(k_1^z v_j^z]\,[(k_2^x v_k^x)^2 + (k_2^z v_k^z)^2 + 2k_2^x v_k^x k_2^z v_k^z] \to (i^4 N)\Big\langle \sum_{i=1}^N ez_i e^{-ikz_i} \times \sum_{j=1}^N ez_j e^{-i(k_1^z z_j + k_1^x x_j)}\sum_{k=1}^N ez_k e^{-i(k_2^z z_k + k_2^x x_k)}\{(k_1^x v_j^x)^2 \times (k_2^x v_k^x)^2 + (k_1^x v_j^x)^2(k_2^z v_k^z)^2 + (k_1^z v_j^z)^2(k_2^x v_k^x)^2 + (k_1^z v_j^z)^2(k_2^z v_k^z)^2 + 4k_1^x v_j^x k_1^z v_j^z k_2^x v_k^x k_2^z v_k^z\}\Big\rangle_0. \tag{18a}$$

Let us perform averaging in the expression for $S_\rho^{2,2}$, using (17b), for two-component fully ionized hydrogen plasma with the classical statistics. Let us consider the summands from items 1-4 term by term. The first summand from item 1. has the form

$$1.1.\quad (i^4/N)\Big\langle \sum_{i,k,j=1}^N e^3 z_i z_j z_k e^{-ikz_i} e^{-i(k_1^z z_j + k_1^x x_j)} e^{-i(k_2^z z_k + k_2^x x_k)} (-)k_1^z k_2^z \frac{1}{m_j m_k}\frac{\partial U}{\partial z_j}\frac{\partial U}{\partial z_k}\Big\rangle_0 = \beta^{-1}\sum_{i,k,j=1}^N e^3 z_i z_j z_k (-)k_1^z k_2^z \times \frac{1}{m_j m_k}\left[\frac{\partial U}{\partial z_j}(-ik_2^z) + \frac{\partial^2 U}{\partial z_j \partial z_k}\right] e^{-i(kz_i + k_1^z z_j + k_1^x x_j + k_2^z z_k + k_2^x x_k)}$$
$$= (i^4/N)\beta^{-1}\Big\langle \sum_{i,k,j=1}^N e^3 z_i z_j z_k (-)k_1^z k_2^z \frac{1}{m_j m_k}\frac{\partial^2 U}{\partial z_j \partial z_k} \times e^{-i(kz_i + k_1^z z_j + k_1^x x_j + k_2^z z_k + k_2^x x_k)}\Big\rangle_0 + (i^4/N)\beta^{-2}\Big\langle \sum_{i,k,j=1}^N e^3 z_i z_j z_k \times (k_1^z k_2^z)^2 \frac{1}{m_j m_k} e^{-i(kz_i + k_1^z z_j + k_1^x x_j + k_2^z z_k + k_2^x x_k)}\Big\rangle_0. \tag{18b}$$

In the thermodynamic limit, we define

$$U = \sum_{1\le j\le k\le N/2} u_{ij} + \sum_{1\le j'\le k'\le N/2} u_{i'j'} + \sum_{1\le j'\le k\le N/2} u_{j'k} + \sum_{1\le j\le k'\le N/2} u_{jk'}.$$

Here (i, j), $(i'j')$ correspond to electrons and ions, u is Coulombic energy of interaction of particle pairs. After averaging of the sums by (17b) (see [6] and references therein) we obtain the following, by selecting the principal terms with $m_j, m_k = m_e$ ($\hat{k}_1^z$ — component of a unit vector, $N' = N/2$), for different ratios between indices i, j, k: $i \ne j \ne k$; $j = k$; $i = j$, $i = k$; $i = j = k$ (at that, the free index can be located in electron or ion subsystems)

$$(i^4/N)\beta^{-1}\Big\langle \sum_{i,k,j=1}^{N'} e^3 z_i z_j z_k (-)(k_1^z k_2^z)\,\frac{1}{m_j m_k}\,\frac{\partial^2 U}{\partial z_j \partial z_k}$$

$$\times e^{-i(\mathbf{k}\mathbf{r}_i+\mathbf{k}_1\mathbf{r}_j+\mathbf{k}_2\mathbf{r}_k)}\Big\rangle_0 = \frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'(N'-1)}{V^3}\int(\hat{k}_1^z\cdot\nabla_1)$$

$$\times(\hat{k}_2^z\cdot\nabla_2)u_{ee}(|r_1-r_2|)e^{-i(\mathbf{k}\mathbf{r}+\mathbf{k}_1\mathbf{r}_1+\mathbf{k}_2\mathbf{r}_2)}$$

$$\times g_{3e}(r,r_1,r_2)d\mathbf{r}d\mathbf{r}_1d\mathbf{r}_2 + \frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'(N'-1)}{V^3}$$

$$\times\int(\hat{k}_1^z\cdot\nabla_1)^2 u_{ee}(|r_1-r_2|)e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}$$

$$\times g_{3e}(r,r_1,r_2)d\mathbf{r}d\mathbf{r}_1d\mathbf{r}_2 + 2\frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'(N'-1)}{V^3}$$

$$\times\int(\hat{k}_1^z\cdot\nabla_1)^2 u_{ei}(|r_1-r_2|)e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}$$

$$\times g_{eei}(r,r_1,r_2)d\mathbf{r}d\mathbf{r}_1d\mathbf{r}_2 + 2\frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'}{V^2}\int(\hat{k}_1^z\cdot\nabla_1)$$

$$\times(\hat{k}_2^z\cdot\nabla_2)u_{ee}(|r_1-r_2|)e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}g_{ee}(r,r_1)d\mathbf{r}d\mathbf{r}_1$$

$$+\frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'}{V^2}\int(\hat{k}_1^z\cdot\nabla_1)^2 u_{ee}(|r_1-r_2|)e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}$$

$$\times g_{ee}(r,r_1)d\mathbf{r}d\mathbf{r}_1 + 2\frac{e^3}{\beta}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'}{V^2}\int(\hat{k}_1^z\cdot\nabla_1)^2 u_{ei}$$

$$\times(|r_1-r_2|)e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}g_{ei}(r,r_1)d\mathbf{r}d\mathbf{r}_1.$$

$$(i^4/N)\beta^{-2}\Big\langle\sum_{i,k,j=1}^{N} e^3 z_i z_j z_k (k_1^z k_2^z)\,\frac{1}{m_j m_k}\,e^{-i(\mathbf{k}\mathbf{r}_i+\mathbf{k}_1\mathbf{r}_j+\mathbf{k}_2\mathbf{r}_k)}\Big\rangle_0$$

$$=(-)\,\frac{e^3}{\beta^2}\,\frac{k_1^z k_2^z}{m_e^2}\,\frac{N'(N'-1)}{V^3}\int e^{-i(\mathbf{k}\mathbf{r}+\mathbf{k}_1\mathbf{r}_1+\mathbf{k}_2\mathbf{r}_2)}$$

$$\times g_{3e}(r,r_1,r_2)d\mathbf{r}d\mathbf{r}_1d\mathbf{r}_2 + (-)\,\frac{e^3}{\beta^2}\,\frac{(k_1^z k_2^z)^2}{m_e^2}\,\frac{N'}{V^2}$$

$$\times\int e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}g_{ee}(r,r_1)d\mathbf{r}d\mathbf{r}_1 + (-)2\frac{e^3}{\beta^2}\,\frac{(k_1^z k_2^z)^2}{m_e^2}\,\frac{N'}{V^2}$$

$$\times\int e^{-i(\mathbf{k}+\mathbf{k}_1)\mathbf{r}+\mathbf{k}_2\mathbf{r}_1}g_{ee}(r,r_1)d\mathbf{r}d\mathbf{r}_1 + (-)\,\frac{e^3}{\beta^2}\,\frac{(k_1^z k_2^z)^2}{m_e^2}$$

$$\times\frac{N'}{V^2}\,\delta_{k+k_1+k_2}. \tag{19}$$

The second equality in (19) corresponds to the second expression in the right member of 1.1 (see (18b)).

The second, third and fourth summands from item 1 (1.2.−1.4., see (18a)) after averaging have a form similar to (19). Let us consider the main contribution to the last summand from item 1. (1.5.) which includes the magnetic field. This contribution is different from zero at $j=k$.

$$(i^4 N)\Big\langle\sum_{i=1}^{N'} e z_i e^{-ikz_i}\sum_{j=1}^{N'} e z_j e^{-i(k_1^z z_j + k_1^x x_j)}$$

$$\times\sum_{k=1}^{N'} e z_k e^{-i(k_2^z z_k + k_2^x x_k)}\left\{-k_1^x k_2^x\,\frac{e z_j}{c m_j}(\upsilon_j^y H)^x\,\frac{e z_k}{c m_k}(\upsilon_k^y H)^x\right\}\Big\rangle_0$$

$$=\frac{H^2 N'}{V^2}\,k_1^x k_2^x\,\frac{e^5}{c^2}\,\frac{1}{\beta m_e^3}\int e^{-i\left(\mathbf{k}\mathbf{r}+(\mathbf{k}_1+\mathbf{k}_2)\mathbf{r}_1\right)}g_{ee}(r,r_1)d\mathbf{r}d\mathbf{r}_1$$

$$+\frac{H^2 N'}{V^2}\,k_1^x k_2^x\,\frac{e^5}{c^2}\,\frac{1}{\beta m_e^3}\,\delta_{k+k_1+k_2}. \tag{20}$$

The averaged expressions in items 2, 3 represent a sum of four summands whose form coincides with the right member of the second equality in (19) at the values of the products of the vectors before the summands respectively $-(k_1^x k_2^x)^2$, $-(k_1^z k_2^z)^2$, $-(k_1^z k_2^x)^2$, $-(k_1^x k_2^z)^2$.

The averaged expression in item 4 consists of five terms, the form of four of which matches the right member of the second equality in (19) at the values of products of the vectors before the terms: $(k_1^x k_2^x)^2$, $(k_1^z k_2^z)^2$, $(k_1^z k_2^x)^2$, $(k_1^x k_2^z)^2$, the fifth term is equal to the integral in (20) with coefficient $4k_1^x k_2^x k_1^z k_2^z\,\frac{N'}{V^2}\,\frac{e^3}{\beta^2 m_e^2}$.

At $z_i = 1$ (which corresponds to singly charged ions), it is necessary to write out the formulas similar to (19) and (20), which contain correlation functions g_{eei}, g_{eii}, g_{ei} instead of g_{3e}, g_{eei}, g_{ee} and the sign before the expression in the right member changes. Thus, the main contribution to $S_\rho^{2,2}$ ($\sim 1/m_e^2$, $\sim 1/m_e^3$) is equal to the sum of all five terms from items 1−4 determined above. The estimates show a considerable contribution (20) to correlator $S_\rho^{2,2}$ at the intensities of a constant magnetic field achievable in laboratory experiments (plasma parameters: $n_e = 10^{17}-10^{21}\,\mathrm{cm}^{-3}$, $T = 1-10\,\mathrm{eV}$, $B = 10^4-10^5\,\mathrm{Gs}$, at which PGR and SHG should be studied) [5]. Let us compare for estimation the terms (the equal multiplicands are omitted) in sums (19) (2-th sum) and (20) ($\frac{e^3}{\beta^2}\,\frac{(k_1^z k_2^z)^2}{m_e^2}$ and $\frac{H^2}{\beta m_e^3}(k_1^x k_2^x)\frac{e^5}{c^2}$); their ratio is $\sim\frac{k^2}{\beta}/\frac{H^2 e^2}{m_e c^2}$. By expressing the wave vector through the cut-off frequency and by selecting parameter values from the above-mentioned ranges, we can estimate the parameter regions where the contribution of (20) to the frequency moment of correlator $S_\rho^{2,2}$ is insigniciant, considerable or prevailing.

The above-mentioned procedure can be used to calculate other frequency $S_\rho^{r_1,r_2}$ moments of dense charged media.

Conclusion

Frequency moment $S_\rho^{2,2}$ has been analyzed as applied to the conditions in laboratory dense plasma. Evidently, a specific calculation of the main contribution to frequency moment $S_{\rho}^{2,2}$ according to (19), (20) (or other frequency moments according to the corresponding relations found using the suggested procedure in this paper) for two-component fully ionized hydrogen plasma with the classical statistics is a separate problem related to the setting of an interparticle interaction potential (see, for instance, [8–11,16–19]), correlation functions of the 2-nd and 3-rd orders: g_{eei}, g_{eii}, g_{ei} and g_{3e}, g_{eei}, g_{ee} at certain values of thermodynamic parameters: temperature and pressure. The correlations functions from (19), (20) were studied in a large number of papers (see, for instance, [8–11,16–19]).

In their turn, explicit approximations of quadratic response functions should be performed using expressions which have the correct asymptotics in the limit cases, e.g., for rarefied plasma (see [5,14]). The use of values of one correlator (e.g., $S_{\rho}^{2,2}$) at certain (P, T, H) presupposes a one-parameter approximation for the QRF. In order to determine the adjustment parameters, numeric values of correlators must be compared (see (14)) with moments of convolute approximations for tensors $\hat{\chi}^{(2)}_{p,ijk}$ or $\hat{\chi}^{(2)}_{J,ijk}$.

It should be noted that we have discussed the model approach to determination of quadratic response functions under the action of an external field, which is based, in particular, on calculation of their frequency moments. In doing so, we used ratio (10) obtained within the framework of the nonlinear theory of response to external perturbation [12,13]. At the same time, nonlinear phenomena (e.g., PGR and SHG) in plasma are studied using an equation in relation to polarization of the medium and electric field $\mathbf{E}(\mathbf{r}, t)$) (see, for instance, [5,14]), with a QRF under the action of a mean field in the given medium (they will be denoted as $\chi^{(2)}_{p,ijk}$ etc). Therefore, construction of a model for $\chi^{(2)}_{p,ijk}$ according to $\hat{\chi}^{(2)}_{p,ijk}$ requires a clarification related to substitution of the known relation $\mathbf{D} = \hat{\varepsilon}\mathbf{E}$ ($\hat{\varepsilon}$ is permittivity of the medium) into an equation similar to (6).

Thus, in this paper we have justified a model for calculation of quadratic response functions which determine nonlinear phenomena caused by quadratic interaction of electromagnetic waves in a dense charged medium (Coulombic systems, plasma) in a constant magnetic field. This approach uses the rather well-known thermodynamic information about dense charged media and does not require significant computer capacities as compared to implementation of direct numerical modeling of quadratic response functions, which is in principle possible for model systems.

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Conflict of interest

The author declares that he has no conflict of interest.

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