ARTICLES

Magnetic susceptibility of ferrocolloids with frozen texture

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A theoretical analysis of the static and dynamic magnetic susceptibilities of a frozen moderately concentrated ferrocolloid is presented. The freezing of the fluid is assumed to take place in the presence of a constant external magnetic field. The steric and dipole-dipole interactions between ferroparticles are taken into account. We have studied the situations when current magnetic field is parallel and normal to the magnetic field applied during freezing. The influence of interparticle interaction on the real and imaginary parts of initial susceptibility is examined. The analysis of a stochastic resonance in this system is presented and the influence of interparticle interaction and field applied during freezing on signal-noise ratio is estimated.
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I. INTRODUCTION

The interest of investigators in systems of dipole particles embedded in a solid matrix has increased in recent years (see, for example, [1–8]). Ferrocolloid (magnetic liquid) with frozen solvent is an example of such a system. As soon as the solvent is frozen, both the space arrangement of ferromagnetic particles and orientations of their axes of light magnetization become fixed. Thus, the static and dynamic magnetic susceptibilities of such a system may differ essentially from those of ferrocolloid with a solvent in a liquid state.

Theoretical analysis of the static magnetization of very dilute magnetic liquids was performed in [4] and their dynamic response to the external magnetic field was studied in [5,6]. Any interparticle interaction was neglected in these works. However, in many experimental situations (for example, [7,8]) this interaction is very significant.

The aim of this work is the strict theoretical analysis of the static and dynamic magnetic susceptibilities of frozen moderately concentrated magnetic fluids, taking into account magnetic and steric interparticle interactions.

Consider a system of N identical spherical ferromagnetic particles. The particle magnetic moment is constant in its absolute value. The solvent becomes frozen instantaneously, therefore space arrangement of the particles and orientations of their axes of light magnetization after freezing remain as before.

II. EQUILIBRIUM SUSCEPTIBILITY

Let us denote by the indexes I and II all physical parameters corresponding to the situations before and after solvent freezing and use the following designations: $\mathbf{e}_j$ and $\boldsymbol{\nu}_j$ are the unit vectors aligned along the magnetic moment $\mathbf{m}_j$ and light magnetization axis of the jth particle, respectively, $\boldsymbol{\alpha}_J = m\mathbf{H}_J/T_J$ $(J = \text{I, II})$, m is the absolute value of the particle magnetic moment, and $\mathbf{H}_J$ and T_J are the magnetic field and the absolute temperature in energetic units.

The energy of the magnetic anisotropy of a particle may be written as usual,

$$u_a = -T\kappa(\mathbf{e}\cdot\boldsymbol{\nu})^2,$$

where κ stands for the dimensionless parameter of anisotropy. As long as the solvent is in a liquid state, the equilibrium N-particle distribution function has a Gibbs form

$$\psi_\text{I}(\mathbf{r}_1,\ldots,\mathbf{r}_N,\mathbf{e}_1,\ldots,\mathbf{e}_N,\boldsymbol{\nu}_1,\ldots,\boldsymbol{\nu}_N) = \frac{1}{Z_\text{I}}\exp\left[\left(\boldsymbol{\alpha}_\text{I}\cdot\sum_i \mathbf{e}_i\right) + \kappa_\text{I}\sum_i (\mathbf{e}_i\cdot\boldsymbol{\nu}_i)^2 - \frac{1}{2}\sum_{i\neq j} w_{\text{I}ij}\right],$$

$$w_{\text{I}ij} = [w_{d\text{I}ij}(\mathbf{e}_i,\mathbf{e}_j,\mathbf{r}_{ij}) + w_{sij}(\mathbf{r}_{ij})], \quad \mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j, \tag{1}$$

$$Z_I = \int \exp\left[\left(\boldsymbol{\alpha}_I \cdot \sum_i \mathbf{e}_i\right) + \kappa_I \sum_i (\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2 - \frac{1}{2}\sum_{i \neq j} w_{Iij}\right] \prod_k d\mathbf{r}_k d\mathbf{e}_k d\boldsymbol{\nu}_k.$$

Here $w_{dIij}(\mathbf{e}_i, \mathbf{e}_j, \mathbf{r}_{ij})$ and $w_{sij}(\mathbf{r}_{ij})$ are the dimensionless potentials of dipole-dipole and steric interparticle interaction and $\mathbf{r}_k$ is radius vector of the center of the kth particle.

We will assume that the effects of interparticle interaction are not great. This allows us to use the method of virial expansion.

Let us find the equilibrium distribution function on $\mathbf{r}_1, \ldots, \mathbf{r}_N, \boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N$ of the ferrocolloid being in a liquid state. For this purpose, we need to integrate Eq. (1) over all vectors $\mathbf{e}_i$. Using the method of virial expansion in a linear approximation by the Meyer function, we obtain

$$g_I(\mathbf{r_1}, \ldots, \mathbf{r}_N, \boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N) = \int \psi_I \prod_k d\mathbf{e}_k$$

$$\approx \frac{1}{Z_I} \int \exp\left[\left(\boldsymbol{\alpha}_I \cdot \sum_i \mathbf{e}_i\right) + \kappa_I \sum_i (\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2\right]\left(1 + \frac{1}{2}\sum_{i \neq j} f_{Iij}\right)\prod_k d\mathbf{e}_k$$

$$= \frac{1}{Z_I}\left(1 + \frac{1}{2}\sum_{i \neq j} \frac{\Omega_I(ij)}{\Phi(i)\Phi(j)}\right)\prod_k \Phi_I(k),$$

$$\Phi(j) = \Phi(\boldsymbol{\nu}_j) = \int \exp[(\boldsymbol{\alpha}_I \cdot \mathbf{e_i}) + \kappa_I(\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2]d\mathbf{e}_j, \tag{2}$$

$$\Omega_I(ij) = \Omega_I(\boldsymbol{\nu}_i, \boldsymbol{\nu}_j, \mathbf{r}_{ij}) = \int \exp\{[\boldsymbol{\alpha}_I \cdot (\mathbf{e_i} + \mathbf{e_j})] + \kappa_I[(\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2 + (\mathbf{e}_j \cdot \boldsymbol{\nu}_j)^2]\}f_{I12}(\mathbf{e}_i, \mathbf{e}_j, \mathbf{r}_{ij})d\mathbf{e}_i d\mathbf{e}_j,$$

$$f_{I12}(\mathbf{e}_i, \mathbf{e}_j, \mathbf{r}_{ij}) = \exp(-w_{Iij}) - 1.$$

Below we shall consider that in instantaneous freezing of the solvent, vectors $\mathbf{r}_1, \ldots, \mathbf{r}_N$ and $\boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N$ become frozen instantaneously as well. Let us determine an N-particle conditional distribution function φ_{II} over vectors $\mathbf{e}_j$ (all vectors $\mathbf{r}_j$ and $\boldsymbol{\nu}_j$ are fixed). Using again the linear approximation by the Meyer function f_{Iij}, we get

$$\varphi_{II}(\mathbf{e}_1, \ldots, \mathbf{e}_N | \mathbf{r}_1, \ldots, \mathbf{r}_N, \boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N) = \frac{1}{Z_{II}}\exp\left[\left(\boldsymbol{\alpha}_{II} \cdot \sum_i \mathbf{e}_i\right) + \kappa_{II}\sum_i (\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2 - \frac{1}{2}\sum_{i \neq j} w_{IIij}\right],$$

$$\tag{3}$$

$$Z_{II} = \int \exp\left[\left(\boldsymbol{\alpha}_{II} \cdot \sum_i \mathbf{e_i}\right) + \kappa_{II}\sum_i (\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2 - \frac{1}{2}\sum_{i \neq j} w_{IIij}\right]\prod_k d\mathbf{e}_k$$

$$\approx \left(1 + \frac{1}{2}\sum_{i \neq j} \frac{\Omega_{II}(ij)}{\Phi_{II}(i)\Phi_{II}(j)}\prod_k \Phi_{II}(k)\right).$$

Here w_{IIij}, Φ_{II}, and Ω_{II} are determined similar to w_{Iij}, Φ_I, and Ω_I in expressions (1) and (2) with replacement T_I, $\boldsymbol{\alpha}_I$, and κ_I by T_{II}, $\boldsymbol{\alpha}_{II}$, and κ_{II}, respectively.

Taking into account Eqs. (2) and (3), we may write the stationary distribution function for the state after the freezing of the fluid,

$$\psi_{II}(\mathbf{e}_1, \ldots, \mathbf{e}_N, \mathbf{r}_1, \ldots, \mathbf{r}_N, \boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N) = g_I \varphi_{II}$$

$$\approx \frac{1}{Z_I}\prod_k \frac{\Phi_I(k)}{\Phi_{II}(k)}\left(1 + \frac{1}{2}\sum_{i \neq j} W_{ij}\right)\exp\left[\left(\boldsymbol{\alpha}_{II}\sum_i \mathbf{e}_i\right) + \kappa_{II}\sum_i (\mathbf{e}_i \cdot \boldsymbol{\nu}_i)^2 - \frac{1}{2}\sum_{i \neq j} w_{IIij}\right],$$

$$W_{ij} = \frac{\Omega_I(ij)}{\Phi_I(i)\Phi_I(j)} - \frac{\Omega_{II}(ij)}{\Phi_{II}(i)\Phi_{II}(j)}. \tag{4}$$

Using Eq. (4), it is easy to calculate the average value of vector $\mathbf{e}$,

$$\langle \mathbf{e}\rangle = \int \mathbf{e}_1 \psi_{II}\prod_k d\mathbf{e}_k d\mathbf{r}_k d\boldsymbol{\nu}_k.$$

In the linear approximation by the Meyer function (see Appendix A),

$$\langle \mathbf{e} \rangle = \langle \mathbf{e} \rangle_0 + \langle \mathbf{e} \rangle_1,$$

$$\langle \mathbf{e} \rangle_0 = \frac{1}{F_{\mathrm{I}}} \int \mathbf{J}_{\mathrm{II}}(\boldsymbol{\nu}) \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu})}{\Phi_{\mathrm{II}}(\boldsymbol{\nu})} d\boldsymbol{\nu},$$

$$\langle \mathbf{e} \rangle_1 = \frac{n}{F_{\mathrm{I}}^2} (\mathbf{K}_1 + \mathbf{K}_2 + \mathbf{K}_3 - G_{\mathrm{I}} \langle \mathbf{e} \rangle_0),$$

$$F_{\mathrm{I}} = \int \Phi_{\mathrm{I}}(\boldsymbol{\nu}) d\boldsymbol{\nu}, \quad \mathbf{J}_{\mathrm{II}} = \int \mathbf{e} \, \exp[(\boldsymbol{\alpha}_{\mathrm{II}} \cdot \mathbf{e}) + \kappa_{\mathrm{II}} (\mathbf{e} \cdot \boldsymbol{\nu})^2] d\mathbf{e},$$

$$\mathbf{K}_1 = \int \frac{\Phi_{\mathrm{I}}(1)\Phi_{\mathrm{I}}(2)}{\Phi_{\mathrm{II}}(1)\Phi_{\mathrm{II}}(2)} \mathbf{Q}_{\mathrm{II}12}^{\star} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2,$$

$$\mathbf{K}_2 = \int \frac{\Phi_{\mathrm{I}}(1)\Phi_{\mathrm{I}}(2)}{\Phi_{\mathrm{II}}(1)\Phi_{\mathrm{II}}(2)} \frac{\mathbf{J}_{\mathrm{II}}(1)}{\Phi_{\mathrm{II}}(1)} \Omega_{\mathrm{II}12}^{\star} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2,$$

$$\mathbf{K}_3 = \int \frac{\mathbf{J}_{\mathrm{II}}(1)}{\Phi_{\mathrm{II}}(1)} \Omega_{\mathrm{I}12}^{\star} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2,$$ (5)

$$G_{\mathrm{I}} = \int \Omega_{\mathrm{I}12}^{\star} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2,$$

$$\mathbf{Q}_{\mathrm{II}12}^{\star} = \int \mathbf{e} \, \exp\{[\boldsymbol{\alpha}_{\mathrm{II}} \cdot (\mathbf{e}_1 + \mathbf{e}_2)] + \kappa_{\mathrm{II}}[(\mathbf{e}_1 \cdot \boldsymbol{\nu}_1)^2 + (\mathbf{e}_2 \cdot \boldsymbol{\nu}_2)^2]\} \Gamma_{\mathrm{II}}(\mathbf{e}_1, \mathbf{e}_2) d\mathbf{e}_1 d\mathbf{e}_2,$$

$$\Omega_{J12}^{\star} = \int \exp\{[\boldsymbol{\alpha}_J \cdot (\mathbf{e}_1 + \mathbf{e}_2)] + \kappa_J[(\mathbf{e}_1 \cdot \boldsymbol{\nu}_1)^2 + (\mathbf{e}_2 \cdot \boldsymbol{\nu}_2)^2]\} \Gamma_J(\mathbf{e}_1, \mathbf{e}_2) d\mathbf{e}_1 d\mathbf{e}_2 = \int \Omega_J(12) d\mathbf{r}_{12},$$

$$\Gamma_J(\mathbf{e}_1, \mathbf{e}_2) = \int f_{J12}(\mathbf{e}_1, \mathbf{e}_2, \mathbf{r}_{12}) d\mathbf{r}_{12},$$

$$J = \mathrm{I}, \mathrm{II}, \quad n = \frac{N}{V}.$$

Here V is the volume of the system, $\langle \mathbf{e} \rangle_0$ is the value of $\langle \mathbf{e} \rangle$ for the system of noninteracting particles, and n is the numerical concentration of the particles. The expression (5) for $\langle \mathbf{e} \rangle_0$ may be obtained from the theory presented in [3,4], where somewhat different reasonings were used.

The effect of the solvent solidification is especially strong if the particles energy of magnetic anisotropy is high enough, i.e., the strong inequalities $\kappa_{\mathrm{I}}, \kappa_{\mathrm{II}} \ll 1$ hold true. Below we will consider this situation.

If $\kappa_J \gg 1$, $\kappa_J \gg \alpha_{\mathrm{I}}, \alpha_{\mathrm{II}}$ ($J = \mathrm{I}, \mathrm{II}$), then the following asymptotic expressions can be easily obtained:

$$F_J = (4\pi)^2 \frac{\exp(\kappa_J)}{\kappa_J} \frac{\mathrm{sh}\alpha_J}{\alpha_J}, \quad \Phi_J(\boldsymbol{\nu}) = 4\pi \mathrm{ch}(\boldsymbol{\alpha}_J \cdot \boldsymbol{\nu}),$$

$$\mathbf{J}_{\mathrm{II}} = 4\pi \frac{\kappa_{\mathrm{II}}}{\kappa_{\mathrm{II}}} \boldsymbol{\nu} \mathrm{sh}(\boldsymbol{\alpha}_{\mathrm{II}} \cdot \boldsymbol{\nu}),$$ (6)

$$\langle \mathbf{e} \rangle_0 = \frac{\int \boldsymbol{\nu} \mathrm{th}(\boldsymbol{\alpha}_{\mathrm{II}} \cdot \boldsymbol{\nu}) \mathrm{ch}(\boldsymbol{\alpha}_{\mathrm{I}} \cdot \boldsymbol{\nu}) d\boldsymbol{\nu}}{\int \mathrm{ch}(\boldsymbol{\alpha}_{\mathrm{I}} \cdot \boldsymbol{\nu}) d\boldsymbol{\nu}}.$$

The dependencies of $\langle \mathbf{e} \rangle_0$ on $\boldsymbol{\alpha}_{\mathrm{II}}$ are shown in Fig. 1. It is interesting to note that the value of $\langle \mathbf{e} \rangle_0$ for $\boldsymbol{\alpha}_{\mathrm{II}} \to \infty$ is less than unity. This is because the particles axes of light magnetization are fixed. This result is due to the assumption $\kappa_{\mathrm{I}} \gg 1$, $\kappa_{\mathrm{I}} \gg \boldsymbol{\alpha}_{\mathrm{I}}$.
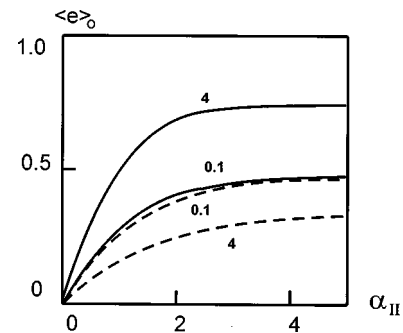

FIG. 1. Equilibrium mean magnetic moment of the particle in the dilute colloid as a function of dimensionless current magnetic field. The solid lines correspond to $\mathbf{H}_{\mathrm{II}} \| \mathbf{H}_{\mathrm{I}}$, dashed lines to $\mathbf{H}_{\mathrm{II}} \perp \mathbf{H}_{\mathrm{I}}$. Numbers of curves are dimensionless fields of solidification $\boldsymbol{\alpha}_{\mathrm{I}}$.

Let us determine the moment $\langle \mathbf{e} \rangle$ in Eq. (5) taking into account the term $\langle \mathbf{e} \rangle_1$ describing the interparticle interaction.

First, we need to calculate Γ_J. Because the particles are spheres of radius a, the integration in the preceding relation is to be carried out over the region $r_{12} > 2a$. In this region $w_{sij} = 0$ and $w_{ij} = w_{dij}$.

As known, the space integral of the Meyer function with dipole-dipole potential depends on the shape of the infinite volume of integration [8]. It is shown in [9] that the correct result may be obtained if this volume is considered as an infinitely long cylinder extended along the current magnetic field. The symmetry axis of this cylinder crosses the center of one of interacting particles (for example, the first one). Namely, in this situation the magnetic field $\mathbf{H}$ figuring in the formula of type (5) is equal to the current magnetic field in the region where two considered particles are located. Calculating $\mathbf{K}_1, \mathbf{K}_2, \mathbf{K}_3$, we may equate this current field to $\mathbf{H}_{\mathrm{II}}$. Calculating the statistical integral Z_{I} and, therefore, G_{I} in Eq. (5) (see Appendix A) we may put current field equal to $\mathbf{H}_{\mathrm{I}}$.

It is impossible to get a short analytical expression for $\mathbf{K}_1, \mathbf{K}_2, \mathbf{K}_3$ for an arbitrary value of the parameter $\boldsymbol{\alpha}_{\mathrm{II}}$. Below, we shall restrict our consideration to the situation $\boldsymbol{\alpha}_{\mathrm{II}} \ll 1$. Assume that the axis Oz is aligned along the field $\mathbf{H}_{\mathrm{I}}$ and the axis Ox lies in the plane $\mathbf{H}_{\mathrm{I}}, \mathbf{H}_{\mathrm{II}}$. Let φ stands for the angle between $\mathbf{H}_{\mathrm{I}}$ and $\mathbf{H}_{\mathrm{II}}$.

In the case κ_{I}, $\kappa_{\mathrm{II}} \gg 1$, $\gamma_{\mathrm{II}} < 1$, and $\alpha_{\mathrm{II}} \ll 1$, parameters $\mathbf{K}_i$ and G_{I} may be derived analytically. After some calculations (see Appendix B) we obtain the following relations:

$$\frac{K_{1z}}{F_{\mathrm{I}}^2} \approx \alpha_{\mathrm{II}} 8v \, \gamma_{\mathrm{II}} J_{\|} \left[\left(\cos^2\varphi - \frac{1}{2}\sin^2\varphi \right) J_{\|} + \frac{3}{2} J_{\perp}\sin^2\varphi \right],$$

$$\frac{K_{1x}}{F_{\mathrm{I}}^2} \approx \alpha_{\mathrm{II}} 8v \, \gamma_{\mathrm{II}} J_{\perp} \left[\left(\sin^2\varphi - \frac{1}{2}\cos^2\varphi \right) J_{\perp} + \frac{3}{2} J_{\|}\cos^2\varphi \right],$$

$$\frac{G_{\mathrm{I}}}{F_{\mathrm{I}}^2} = 8v \, \gamma_{\mathrm{I}} L^2(\boldsymbol{\alpha}_{\mathrm{I}}),$$

$$K_{2i} = 0 \quad (i = z, x),$$

$$\frac{K_{3z}}{F_{\mathrm{I}}^2} = 8\rho \, \gamma_{\mathrm{I}} J_{\|}(\boldsymbol{\alpha}_{\mathrm{I}}) C_{\|}, \quad \frac{K_{3x}}{F_{\mathrm{I}}^2} = 8\rho \, \gamma_{\mathrm{I}} J_{\perp}(\boldsymbol{\alpha}_{\mathrm{I}}) C_{\perp}, \quad (7)$$

$$C_{\|} = \frac{L(\boldsymbol{\alpha}_{\mathrm{I}})}{J_{\|}(\boldsymbol{\alpha}_{\mathrm{I}})} \left[\mathrm{cth}\boldsymbol{\alpha}_{\mathrm{I}} - \frac{3}{\boldsymbol{\alpha}_{\mathrm{I}}} + \frac{6}{\boldsymbol{\alpha}_{\mathrm{I}}^2} L(\boldsymbol{\alpha}_{\mathrm{I}}) - J_{\|}(\boldsymbol{\alpha}_{\mathrm{I}})L(\boldsymbol{\alpha}_{\mathrm{I}}) \right],$$

$$C_{\perp} = \frac{L(\boldsymbol{\alpha}_{\mathrm{I}})}{J_{\|}(\boldsymbol{\alpha}_{\mathrm{I}})} \left[\frac{1}{2} \left(L(\boldsymbol{\alpha}_{\mathrm{I}}) - \mathrm{cth}\boldsymbol{\alpha}_{\mathrm{I}} + \frac{3}{\boldsymbol{\alpha}_{\mathrm{I}}} - 6\frac{L(\boldsymbol{\alpha}_{\mathrm{I}})}{\boldsymbol{\alpha}_{\mathrm{I}}^2} \right) \right.$$

$$\left. - L(\boldsymbol{\alpha}_{\mathrm{I}})J_{\perp}(\boldsymbol{\alpha}_{\mathrm{I}}) \right],$$

$$J_{\|} = 1 - 2\frac{L(\boldsymbol{\alpha}_{\mathrm{I}})}{\boldsymbol{\alpha}_{\mathrm{I}}}, \quad J_{\perp} = \frac{L(\boldsymbol{\alpha}_{\mathrm{I}})}{\boldsymbol{\alpha}_{\mathrm{I}}},$$

$$L(\boldsymbol{\alpha}_{\mathrm{I}}) = \mathrm{cth}(\boldsymbol{\alpha}_{\mathrm{I}}) - \frac{1}{\boldsymbol{\alpha}_{\mathrm{I}}}, \quad v = \frac{4\pi}{3}a^3, \quad \gamma_{\mathrm{II}} = \frac{m^2}{(2a)^3 T_{\mathrm{II}}}.$$

Taking into account Eqs. (5)–(7), we come to the following expressions for the components of $\langle \mathbf{e} \rangle$:

$$\langle e_z \rangle = \boldsymbol{\alpha}_{\mathrm{II}z}\beta_{\|}, \quad \langle e_x \rangle = \boldsymbol{\alpha}_{\mathrm{II}x}\beta_{\perp},$$

$$\beta_{\|} = J_{\|} \left\{ 1 + 8\rho \left[\gamma_{\mathrm{II}} \left(J_{\|} + \frac{3}{2}(J_{\perp} - J_{\|})\sin^2\varphi \right) + \gamma_{\mathrm{I}}C_{\|} \right] \right\},$$

$$(8)$$

$$\beta_{\perp} = J_{\perp} \left\{ 1 + 8\rho \left[\gamma_{\mathrm{II}} \left(J_{\perp} + \frac{3}{2}(J_{\|} - J_{\perp})\cos^2\varphi \right) + \gamma_{\mathrm{I}}C_{\perp} \right] \right\}.$$

Here $\rho = nv$ is the volume concentration of the particles. The components of the tensor of initial susceptibility χ are

$$\chi_{\|,\perp} = \frac{m^2}{v T_{\mathrm{II}}}\rho\beta_{\|,\perp}.$$

We have obtained the analytical expressions (8) in approximation $\boldsymbol{\alpha}_{\mathrm{II}} \to 0$. However, using Eq. (5), it is not difficult to derive numerically the equilibrium susceptibility of the colloid for any value of $\boldsymbol{\alpha}_{\mathrm{II}}$.

III. DYNAMIC SUSCEPTIBILITY

In this section we will assume again that the inequalities $\kappa_{\mathrm{I}}, \kappa_{\mathrm{II}} \gg 1$, and $\gamma_{\mathrm{II}} < 1$ hold true.

Let $f_1(\mathbf{e}|\boldsymbol{\nu})$ be the conditional distribution function over $\mathbf{e}$ at fixed $\boldsymbol{\nu}$ for the frozen system; $g_1(\boldsymbol{\nu})$ is the distribution function over $\boldsymbol{\nu}$. The mean value of the vector $\mathbf{e}$ is

$$\langle \mathbf{e} \rangle = \int \mathbf{e} f_1(\mathbf{e}|\boldsymbol{\nu}) g_1(\boldsymbol{\nu}) d\mathbf{e} \, d\boldsymbol{\nu}. \quad (9)$$

To derive the distribution function f_1 we use the Fokker-Planck equation

$$\frac{\partial f_1}{\partial t} = \zeta \mathbf{K}(f_1 \mathbf{K} u_1) + \zeta \mathbf{K}(f_1 \mathbf{K} u_2) + \zeta T \mathbf{K}^2 f_1,$$

$$\mathbf{K} = \left[\mathbf{e} \times \frac{\partial}{\partial \mathbf{e}} \right],$$

$$(10)$$

$$u_1(\mathbf{e}, \boldsymbol{\nu}) = -T[(\boldsymbol{\alpha}_{\mathrm{II}} \cdot \mathbf{e}) + \kappa_{\mathrm{II}}(\mathbf{e} \cdot \boldsymbol{\nu})^2],$$

$$u_2(\mathbf{e}, \boldsymbol{\nu}) = \sum_j U_j(\mathbf{e}, \boldsymbol{\nu}).$$

Here u_1 is the energy of a single particle taking into account its interaction with the magnetic field $\mathbf{H}_{\mathrm{II}}$ and its energy of anisotropy, $U_j(\mathbf{e}, \boldsymbol{\nu})$ is the mean energy of interaction between this and the jth particles, and ζ is a dissipative coefficient. According to the definition,

$$U_j(\mathbf{e}_1, \boldsymbol{\nu}_1)$$

$$= \int w_{1j}(\mathbf{e}_1, \mathbf{e}_j, \mathbf{r}_{1j}) p_2(\mathbf{e}_j, \boldsymbol{\nu}_j, \mathbf{r}_{1j}|\mathbf{e}_1, \boldsymbol{\nu}_1) d\mathbf{e}_j d\boldsymbol{\nu}_j d\mathbf{r}_{1j}.$$

$$(11)$$

Here p_2 is the conditional binary distribution function of $\mathbf{e}_j, \boldsymbol{\nu}_j, \mathbf{r}_{1j}$ at fixed $\mathbf{e}_1$ and $\boldsymbol{\nu}_1$, and w_{1j} is the potential of interparticle interaction.

Because the particles are hard spheres of radius a, the distance between their centers r_{1j} has to be larger than $2a$. In the region where $r_{1j} > 2a$, we have by the order of value $w_{1j} \sim \gamma_{\mathrm{II}}$. Hence, while determining u_2 in the first approximation by the γ_{II}, one may neglect interparticle interaction in the expression for p_2 and write

$$p_2(\mathbf{e}_j, \boldsymbol{\nu}_j, \mathbf{r}_{1j} | \mathbf{e}_1, \boldsymbol{\nu}_1) = \frac{1}{V} p_1(\mathbf{e}_j, \boldsymbol{\nu}_j), \quad r_{1j} > 2a,$$

$$p_2 = 0, \quad r_{1j} < 2a. \tag{12}$$

Here p_1 is the distribution function of the single particle that does not interact with the other particles.

We assume that as long as the system is in a liquid state, it remains in equilibrium. Then

$$p_1(\mathbf{e}, \boldsymbol{\nu}) = \psi_1(\mathbf{e} | \boldsymbol{\nu}) \varphi_1(\boldsymbol{\nu}), \tag{13}$$

where $\varphi_1(\boldsymbol{\nu})$ is the distribution function for the single particle before freezing, and ψ_1 is the conditional distribution function over $\mathbf{e}$ of this particle after freezing. Repeating the reasonings of Sec. II, we come to

$$\varphi_1(\boldsymbol{\nu}) = \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu})}{F_{\mathrm{I}}}, \tag{14}$$

where Φ_{I} and F_{I} are the same as in Eqs. (2) and (5). If $\kappa_{\mathrm{I}} > 1$, then the asymptotic approximation

$$\varphi_1(\boldsymbol{\nu}) = \frac{1}{4\pi} \frac{\alpha_{\mathrm{I}}}{\mathrm{sh}(\alpha_{\mathrm{I}})} \mathrm{ch}(\alpha_{\mathrm{I}} \cdot \boldsymbol{\nu}) \tag{15}$$

holds true.

It should be noted that the functions f_1 and g_1 are determined for one particle taking into account interparticle interaction, whereas ψ_1 and φ_1 are calculated neglecting this interaction.

The Fokker-Planck equation for the function ψ_1 is

$$\frac{\partial \psi_1}{\partial t} = D\mathbf{K}(\psi_1 \mathbf{K} u_1) + D\mathbf{K}^2 \psi_1,$$

$$D = \zeta T. \tag{16}$$

Equation (16) is too difficult to solve analytically. For its approximate analysis we may consider that for $\kappa_{\mathrm{II}} \gg 1$ and $\alpha_{\mathrm{II}} \ll 1$ the potential u_1 as a function of $\cos\theta = (\mathbf{e} \cdot \boldsymbol{\nu})$ has two minima at $\theta \approx 0, \pi$, a maximum at $\theta \approx \pi/2$, and

$$u_+ = u_1(\theta = 0) = -T(\alpha_{\|} + \kappa_{\mathrm{II}}),$$

$$u_- = u_1(\theta = \pi) = -T(-\alpha_{\|} + \kappa_{\mathrm{II}}), \tag{17}$$

$$u_m = u_1(\theta = \pi/2) = -T\alpha_{\perp},$$

where $\alpha_{\|}$ and $\alpha_{\perp}$ are the components of α_{II} parallel and perpendicular to $\boldsymbol{\nu}$, respectively.

Taking into account that the plot of the function $u_1(\theta)$ has two deep potential pits and a barrier between them, using the method applied in [6], we shall seek the solution of Eq. (16) in the form

$$\psi_1 = n_+(t, \boldsymbol{\nu}) \delta(\theta) + n_-(t, \boldsymbol{\nu}) \delta(\theta - \pi), \tag{18}$$

where $\delta(x)$ is a delta function and n_+ and n_- are the probabilities for the particle to be in states close to $\theta = 0$ and $\theta = \pi_1$, respectively.

Repeating the reasonings of [6], we come to

$$n_{\pm}(t, \boldsymbol{\nu}) = \frac{1}{q(t)} \left[n_{\pm}(0, \boldsymbol{\nu}) q(0) + \int_0^t W_{\mp}(s) q(s) ds \right],$$

$$q(t) = \exp\left(\int_0^t [W_+(s) + W_-(s)] ds \right),$$

$$W_{\pm} = c_{\pm} \exp\left(-\frac{u_m - u_{\pm}}{T} \right),$$

$$c_{\pm} = \zeta k_{\pm} \left(\frac{k_m}{2\pi T} \right)^{1/2}, \tag{19}$$

$$k_{\pm} = \frac{d^2 u_1}{d\theta^2}(\theta = 0, \pi) = (\pm \alpha_{\|} + 2\kappa_{\mathrm{II}})T,$$

$$k_m = -\frac{d^2 u_1}{d\theta^2}\left(\theta = \frac{\pi}{2} \right) = (-\alpha_{\perp} + 2\kappa_{\mathrm{II}})T.$$

Substituting Eqs. (12)–(14) and Eqs. (18) and (19) into Eq. (11) gives

$$U_j(\mathbf{e}, \boldsymbol{\nu}) = -\frac{1}{V} \int \Gamma_{\mathrm{II}}(\mathbf{e}, \mathbf{e}_j) \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu}_j)}{F_{\mathrm{I}}} [n_+(t, \boldsymbol{\nu}_j) \delta(\mathbf{e}_j - \boldsymbol{\nu}_j) + n_-(t, \boldsymbol{\nu}_j) \delta(\mathbf{e}_j + \boldsymbol{\nu}_j)] d\mathbf{e}_j d\boldsymbol{\nu}_j$$

$$= -\frac{1}{V} \int \Gamma_{\mathrm{II}}(\mathbf{e}, \boldsymbol{\nu}_j) \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu}_j)}{F_{\mathrm{I}}} [n_+(t, \boldsymbol{\nu}_j) - n_-(t, \boldsymbol{\nu}_j)] d\mathbf{e}_j d\boldsymbol{\nu}_j$$

$$= -\frac{1}{V} 8\gamma_{\mathrm{II}} v \int \left(e_{1Z} \nu_{1Z} - \frac{1}{2}(e_{1X}\nu_{1X} + e_{1Y}\nu_{1Y}) \right) \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu}_j)}{F_{\mathrm{I}}} [n_+(t, \boldsymbol{\nu}_j) - n_-(t, \boldsymbol{\nu}_j)] d\boldsymbol{\nu}_j. \tag{20}$$

Here we use the Cartesian-coordinate system with the axis OZ aligned along $\mathbf{H}_{\mathrm{II}}$ and the axis OX displaced in the plane $(\mathbf{H}_{\mathrm{I}}, \mathbf{H}_{\mathrm{II}})$. With the help of Eq. (19), we get

$$n_+(t, \boldsymbol{\nu}_j) - n_-(t, \boldsymbol{\nu}_j) = n_+(0, \boldsymbol{\nu}_j) - n_-(0, \boldsymbol{\nu}_j) \frac{q(0)}{q(t)} + \frac{1}{q(t)} \int_0^t [W_-(s) - W_+(s)] q(s) ds. \tag{21}$$

Neglecting the values of the order of α_{II}/κ_{II}, in linear approximation in α_{II},

$$W_-(s)-W_+(s)=\frac{1}{\tau}\alpha_{II}(s)\nu_Z, \quad W_-(s)+W_+(s)=\frac{1}{\tau},$$
(22)
$$\tau=\frac{\pi^{1/2}}{4\zeta T\kappa_{II}^{3/2}}\exp(\kappa_{II}).$$

Substituting Eq. (22) into Eq. (21), after some simple calculations, gives

$$n_+(t,\boldsymbol{\nu}_j)-n_-(t,\boldsymbol{\nu}_j)=[n_+(0,\boldsymbol{\nu}_j)-n_-(0,\boldsymbol{\nu}_j)]\exp\left(-\frac{t}{\tau}\right)$$
$$+\nu_Z\int_0^t\frac{\alpha_{II}(s)}{\tau}\exp\left(\frac{t-s}{\tau}\right)ds. \quad (23)$$

As follows from Eq. (23), τ is the characteristic time of the particle transition over a potential barrier at $\theta=\pi/2$. The initial values of $n_\pm(0,\boldsymbol{\nu})$ are determined according to the conditions preceding solvent freezing. If this state of the system is an equilibrium one, then the single-particle conditional distribution function over $\mathbf{e}$ is

$$\varphi_I(\mathbf{e}|\boldsymbol{\nu})=\frac{\exp[(\alpha_I\cdot\mathbf{e})+\kappa_I(\mathbf{e}\cdot\boldsymbol{\nu})^2]}{\Phi_I(\boldsymbol{\nu})}. \quad (24)$$

Using Eq. (24) at $\kappa_I\gg1$, we get

$$n_\pm(0,\boldsymbol{\nu})=\int_{\mathbf{e}_\pm}\varphi_I d\mathbf{e}\approx\frac{\exp(\pm\alpha_I\cdot\boldsymbol{\nu})}{2\mathrm{ch}(\alpha_I\cdot\boldsymbol{\nu})}, \quad (25)$$

where $\mathbf{e}_\pm$ covers the region of $\mathbf{e}$ close to $\theta=0$ and $\theta=\pi$, respectively.

Substituting Eq. (25) into Eq. (23), we obtain

$$n_+(t,\boldsymbol{\nu}_j)-n_-(t,\boldsymbol{\nu}_j)=\mathrm{th}(\alpha_I\cdot\boldsymbol{\nu})\exp\left(-\frac{t}{\tau}\right)$$
$$+8\gamma\upsilon\,\boldsymbol{\nu}_Z\int_0^t\frac{\alpha_{II}(s)}{\tau}\exp\left(\frac{t-s}{\tau}\right)ds. \quad (26)$$

Let us assume that the dimensionless field α_{II} depends on t as

$$\alpha_{II}(t)=\alpha_0\cos\omega t.$$

Now it follows from Eq. (26) that

$$n_+(t,\boldsymbol{\nu})-n_-(t,\boldsymbol{\nu})$$
$$=\frac{\alpha_0\boldsymbol{\nu}_Z}{1+(\omega\tau)^2}[\cos(\omega\tau)+\omega\tau\sin(\omega\tau)]$$
$$+\left(\mathrm{th}(\alpha_I\cdot\boldsymbol{\nu})-\boldsymbol{\nu}_Z\alpha_0\frac{1+\omega\tau}{1+(\omega\tau)^2}\right)\exp\left(-\frac{t}{\tau}\right). \quad (27)$$

For the times $t\gg\tau$, the second term on the right-hand side of Eq. (27) may be neglected. Using the first term in Eq. (20), taking into account that all particles are identical, in a thermodynamic limit we have

$$u_2(\mathbf{e})=\sum_j\,U_j=-8\rho\gamma_{II}\frac{\alpha_0}{1+(\omega\tau)^2}[\cos(\omega\tau)+\omega\tau\sin(\omega\tau)]$$
$$\times\int\boldsymbol{\nu}_{jZ}\left(e_Z\boldsymbol{\nu}_{jZ}-\frac{1}{2}(e_X\boldsymbol{\nu}_{jX}+e_Y\boldsymbol{\nu}_{jY})\right)\frac{\Phi_I(\boldsymbol{\nu}_1)}{F_I}[n_+(t,\boldsymbol{\nu}_j)-n_-(t,\boldsymbol{\nu}_j)]d\boldsymbol{\nu}_j. \quad (28)$$

To simplify calculations, we shall consider separately the situations when the field $\mathbf{H}_{II}$ is parallel and perpendicular to $\mathbf{H}_I$.

Field $\mathbf{H}_{II}$ is parallel to $\mathbf{H}_I$

The coordinate axes Oz,OZ and Ox,OX coincide, respectively. Using Eq. (28), the expression for the mean energy of the particle interaction with the other particles ($\kappa_I\gg1$) is easily obtained,

$$u_2(\mathbf{e})=-e_z\alpha_0 8\rho\gamma_{II}\frac{1}{1+(\omega\tau)^2}\int\boldsymbol{\nu}_{1z}^2\frac{\Phi_I(\boldsymbol{\nu}_1)}{F_I}d\boldsymbol{\nu}_1$$
$$\approx-(\mathbf{e}\cdot\alpha_0)8\rho\gamma_{II}\frac{\cos(\omega t)+\omega\tau\sin(\omega t)}{1+(\omega\tau)^2}J_{||}(\alpha_I). \quad (29)$$

Substituting u_2 from Eq. (29) into Eq. (10), we come to a problem similar to that for a single particle in the effective dimensionless magnetic field,

$$\alpha_e=\alpha_0\{\cos(\omega t)+B_{||}[\cos(\omega t)+\omega\tau\sin(\omega t)]\}, \quad (30)$$
$$B_{||}=\frac{8\rho\gamma_{II}}{1+(\omega\tau)^2}J_{||}(\alpha_I).$$

Using Eq. (18), the expression for the conditionally mean value of vector $\mathbf{e}$ at fixed $\boldsymbol{\nu}$ is

$$\langle\mathbf{e}\rangle_{\boldsymbol{\nu}}=\boldsymbol{\nu}[n_+(t,\boldsymbol{\nu})-n_-(t,\boldsymbol{\nu})]. \quad (31)$$

Substituting $\alpha_e(t)$ from Eq. (30) to Eq. (26) instead of $\alpha_{II}(t)$ and then the result into Eq. (31), we come to
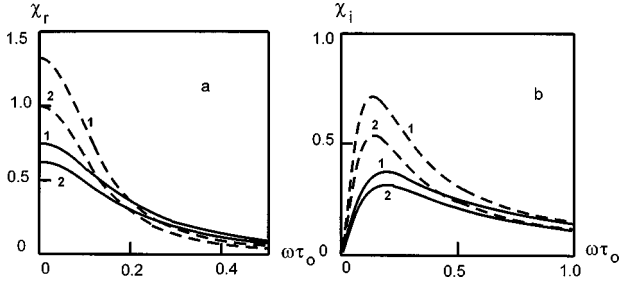
FIG. 2. Real (a) and imaginary (b) parts of initial susceptibility vs dimensionless frequency at $\gamma_I = 0.1$, $\gamma_{II} = 0.5$, $\alpha_I = 1$. Here $\chi_r = g\chi'$, $\chi_i = g\chi''$, $g = K\upsilon/(m^2\rho)$, $\tau_0 = 4\zeta K\pi^{1/2}$. Solid lines, $\rho = 0$; dashed, $\rho = 0.5$. Numbering of curves: 1, $\mathbf{H}_{II} \| \mathbf{H}_I$; 2, $\mathbf{H}_{II} \perp \mathbf{H}_I$.

FIG. 3. Real (a) and imaginary (b) parts of susceptibility as functions of temperature $T = T_{II}$ at $\omega\tau_0 = 1$, $\alpha_I = 1$, $\gamma_I = 0.1$, $\gamma' = m^2/[K(2a)^2] = 1$. Solid lines, $\rho = 0.1$; dashed, $\rho = 0.5$. Parameters χ_r, χ_i, τ_0 and figures at curves are the same as in Fig. 2.

$$\langle \mathbf{e} \rangle_{\boldsymbol{\nu}} = \boldsymbol{\nu}\nu_z \frac{\boldsymbol{\alpha}_0}{1+(\omega\tau)^2}\{[1 + B_{\|} - (\omega\tau)^2 B_{\|}]\cos(\omega t)$$

$$+ \omega\tau(1 + 2B_{\|})\sin(\omega t)\}. \qquad (32)$$

The unconditionally mean value of vector $\mathbf{e}$ is

$$\langle \mathbf{e} \rangle = \int \langle \mathbf{e} \rangle_{\boldsymbol{\nu}} g_2(\boldsymbol{\nu}) d\boldsymbol{\nu}, \qquad (33)$$

where g_2 is a distribution function over orientations of $\boldsymbol{\nu}$. According to the assumptions that before freezing the system is in an equilibrium state and the freezing is instantaneous, this function is to be equal to the distribution function for the colloid being in a liquid state. Using the results of Sec. II, we may obtain approximately (see Appendix C)

$$g_2(\boldsymbol{\nu}) = \frac{\Phi_I(\boldsymbol{\nu})}{F_I} + \frac{n}{F_I}\left(\int \Omega^{\star}(\boldsymbol{\nu},\boldsymbol{\nu}_1) d\boldsymbol{\nu}_1 - \frac{\Phi_I(\boldsymbol{\nu})}{F_I} G_I \right). \qquad (34)$$

When the strong inequality $\kappa_I \gg 1$ is valid, one may obtain the following estimations:

$$\int \Omega_I^{\star}(\boldsymbol{\nu},\boldsymbol{\nu}_1) d\boldsymbol{\nu}_1$$

$$= \frac{1}{2}(4\pi)^2 \left(\frac{\exp(\kappa_I)}{\kappa_I} \right)^2$$

$$\times \Gamma_I\{\mathrm{ch}[\boldsymbol{\alpha}_I \cdot (\boldsymbol{\nu}+\boldsymbol{\nu}_1)] - \mathrm{ch}[\boldsymbol{\alpha}_I \cdot (\boldsymbol{\nu}-\boldsymbol{\nu}_1)]\}.$$

Substituting Eqs. (32) and (34) into Eq. (33), we come to

$$\langle e_z \rangle = \boldsymbol{\alpha}_0[\beta_{\|}'\cos(\omega t) + \beta_{\|}''\sin(\omega t)],$$

$$\beta_{\|}' = \frac{J_{\|}(\boldsymbol{\alpha}_I)}{1+(\omega\tau)^2}\left[1 + 8\rho\left(\gamma_{II}J_{\|}(\boldsymbol{\alpha}_I)\frac{1-(\omega\tau)^2}{1+(\omega\tau)^2} + \gamma_I C_{\|} \right)\right], \qquad (35)$$

$$\beta_{\|}'' = \omega\tau \frac{J_{\|}(\boldsymbol{\alpha}_I)}{1+(\omega\tau)^2}\left[1 + 8\rho\left(\gamma_{II}J_{\|}(\boldsymbol{\alpha}_I)\frac{2}{1+(\omega\tau)^2} + \gamma_I C_{\|} \right)\right].$$

The dependencies of $\beta_{\|}'$ and $\beta_{\|}''$ from ω are given in Fig. 2. For this situation an increase of interparticle interaction leads to an increase of $\beta_{\|}''$ maximum and to a decrease of frequency corresponding to this maximum.
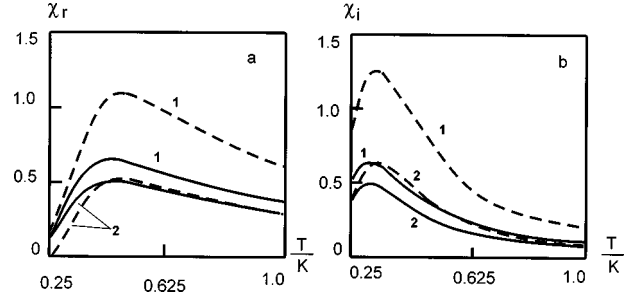
Fields $\mathbf{H}_I$ and $\mathbf{H}_{II}$ are perpendicular to each other

Let us align the axes OX and OZ along the fields $\mathbf{H}_I$ and $\mathbf{H}_{II}$, correspondingly. Using a spherical coordinate system with polar axis OX, we may obtain from Eq. (28) instead of Eq. (29)

$$u_2(\mathbf{e}) = -e_Z \boldsymbol{\alpha}_0 8\rho\gamma_{II}\frac{\cos(\omega t) + (\omega\tau)\sin(\omega t)}{1+(\omega\tau)^2}J_{\perp}(\boldsymbol{\alpha}_I). \qquad (36)$$

Substituting Eq. (36) into Eq. (10), we come to the problem concerning a single particle in the effective dimensionless field,

$$\boldsymbol{\alpha}_e = \boldsymbol{\alpha}_0\{\cos(\omega t) + B_{\perp}[\cos(\omega t) + \omega\tau\sin(\omega t)]\},$$

$$\qquad (37)$$

$$B_{\perp} = \frac{8\rho\gamma_{II}}{1+(\omega\tau)^2}J_{\perp}(\boldsymbol{\alpha}_I).$$

Repeating the foregoing reasonings, we obtain

$$\langle e_x \rangle = \langle e_Z \rangle = \boldsymbol{\alpha}_0[\beta_{\perp}'\cos(\omega t) + \beta_{\perp}''\sin(\omega t)],$$

$$\beta_{\perp}' = \frac{J_{\perp}(\boldsymbol{\alpha}_I)}{1+(\omega\tau)^2}\left[1 + 8\rho\left(\gamma_{II}J_{\perp}(\boldsymbol{\alpha}_I)\frac{1-(\omega\tau)^2}{1+(\omega\tau)^2} + \gamma_I C_{\perp} \right)\right], \qquad (38)$$

$$\beta_{\perp}'' = \omega\tau \frac{J_{\perp}(\boldsymbol{\alpha}_I)}{1+(\omega\tau)^2}\left[1 + 8\rho\left(\gamma_{II}J_{\perp}(\boldsymbol{\alpha}_I)\frac{2}{1+(\omega\tau)^2} + \gamma_I C_{\perp} \right)\right].$$

The plots of $\beta_{\perp}'$ and $\beta_{\perp}''$ versus ω are given in Fig. 2.

The real χ_J' and imaginary χ_J'' parts of initial susceptibility may be written as

$$\chi_J' = \frac{m^2\rho}{\upsilon K}\kappa_{II}\beta_J', \qquad \chi_J'' = \frac{m^2\rho}{\upsilon K}\kappa_{II},\beta_J''$$

$$K = \kappa_J T_J = \mathrm{const}, \qquad J = \|,\perp.$$

The temperature dependencies of $\chi_{\|}$ and $\chi_{\perp}$ susceptibility components are given in Fig. 3. The real χ' and imaginary χ'' parts of the initial susceptibility as functions of T have maxima. These maxima occur due to a competition between two factors. First, the relaxation time τ grows with decreasing temperature. This leads to a decrease of χ'. Second, a
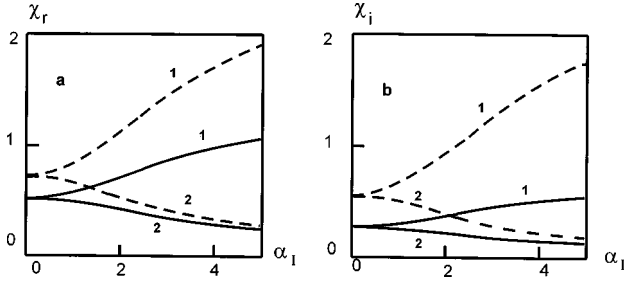
FIG. 4. Real (a) and imaginary (b) parts of susceptibilities as functions of α_I at $\gamma_I = 0.1, \gamma_{II} = 0.5, T_{II}/K = 0.5, \omega\tau_0 = 0.1$. Solid lines, $\rho = 0.1$; dashed, $\rho = 0.5$. Parameters χ_r, χ_i, τ_0 and figures at curves are the same as in Fig. 2.

FIG. 5. Signal-noise ratio vs temperature ($T = T_{II}$) at $\omega\tau_0 = 0.1, \gamma_I = 0.1, \gamma' = 0.25$. (a) $\alpha_I = 1$: solid lines, $\rho = 0$; dashed, $\rho = 0.5$. (b) $\rho = 0.5$: solid lines, $\alpha_I = 0.5$; dashed, $\alpha_I = 1$. Numbering of the curves are the same as in Figs. 2–4.

decrease of temperature results in an increase of coupling between the particle magnetic moment and external field. This leads to an increase of χ'. The interparticle interaction raises both $\chi'_{\|,\perp}$, $\chi''_{\|,\perp}$, and temperatures corresponding to their maxima.

The dependencies of $\chi'_{\|,\perp}$ and $\chi''_{\|,\perp}$ on solidification dimensionless field α_I are shown in Fig. 4. The components of $\chi_\|$ increase and the components of $\chi_\perp$ decrease as α_I increases.

It should be mentioned that the expressions (35) and (38) for $\beta'_\|$ and $\beta''_\|$ at $\omega = 0$ coincide with those for $\beta_\|$ and $\beta_\perp$ in Eq. (8) provided that the angle $\varphi = 0$, ($\mathbf{H}_I\|\mathbf{H}_{II}$) and $\varphi = \pi/2$ ($\mathbf{H}_I \perp \mathbf{H}_{II}$), respectively.

IV. STOCHASTIC RESONANCE

The stochastic resonance (SR) consists in the coherent response of a multistable system to a driven periodic signal. The description of this phenomenon in various physical systems may be found in [9–11]. Theoretical investigations of SR in a very dilute systems of single-domain magnetic particles embedded in a solid matrix have been carried out in [5,6]. Our results for dynamic susceptibility of the frozen ferrocolloid allow us to evaluate the effect of magnetic interparticle interaction on the SR in these systems.

Let e_Z be the component of vector $\mathbf{e}$ along the field $\mathbf{H}_{II}$. Its absolute value depends periodically on t with angular frequency ω_0 and

$$S(\omega) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \langle e_Z(t)e_Z(t+s)\rangle \exp(i\omega s)ds$$

is the spectral density function.

This function may be calculated with the help of expression (26). However, for $\alpha_{II} \ll 1$ it will be easier to use the general theory of linear response and the fluctuation-dissipation theorem. According to this theorem,

$$S(\omega) = \frac{1}{2}\alpha_0^2|\beta_J(\omega)|^2\delta(\omega - \omega_0) + \frac{\beta''_J}{\pi\omega},$$
$$J = \|,\perp, \quad |\beta_J|^2 = (\beta'_J)^2 + (\beta''_J)^2. \tag{39}$$

The first term in this equation for $S(\omega)$ describes the linear response of the system to an external periodic signal; the second one arises due to thermal noise.
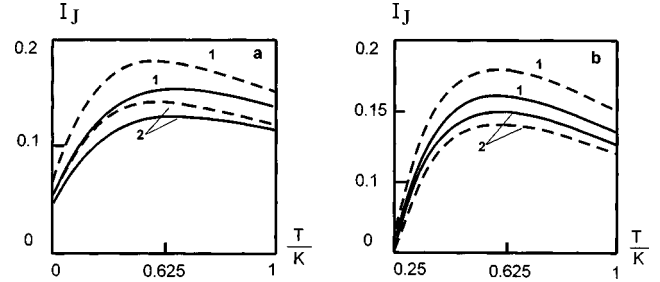
Determine the signal-noise ratio as usual,

$$R_J = \frac{\pi}{2}\frac{m^2 H_{0II}^2}{K^2}\omega_0\tau_0 I_J(\kappa_{II}),$$

$$I_J = \frac{\kappa_{II}^2|\beta_J(\omega_0)|^2}{\beta_J(\omega_0)''\tau_0}, \tag{40}$$

$$\tau_0 = \frac{4\zeta K}{\pi^{1/2}}, \quad J = \|,\perp.$$

Here K is a constant of the particle magnetic anisotropy.
   Let

$$\gamma_{II} = \frac{m^2}{(2a)^3 T_{II}} = \kappa_{II}\gamma',$$

$$\gamma' = \frac{m^2}{(2a)^3 K}. \tag{41}$$

Substituting Eq. (41) into Eq. (40), we may consider I_J as a function of T_{II}/K at other fixed physical parameters of the system and temperature of freezing being equal to T_I.

The results of calculations of I_J are given in Fig. 5. It is easy to see that I_J as a function of T_{II} have maxima. An increase of interparticle interaction leads to an increase of both components of I_J and to a decrease of temperature, corresponding to their maxima. The increase of solidification field α_I causes an increase of $I_\|$ and a decrease of $I_\perp$. All other parameters of the system being equal, $I_\| > I_\perp$.

V. CONCLUSIONS AND DISCUSSIONS

The results obtained evidence that the dipole-dipole interaction increases the initial susceptibility of the frozen ferrocolloid if the current magnetic field $\mathbf{H}_{II}$ is parallel to the field applied during freezing $\mathbf{H}_I$. If $\mathbf{H}_{II}$ is parallel to $\mathbf{H}_I$, then at small values of H_I this interaction leads to an increase, whereas at strong H_I it leads to a decrease of susceptibility.

The maxima χ''_m of the imaginary parts of the dynamic susceptibilities $\chi_\|$ and $\chi_\perp$ as functions of the current field frequency ω increase and the corresponding frequencies ω_m decrease as dipole-dipole interparticle interaction increases; the components of $\chi_\|$ are the increasing functions of the

freezing field H_I; the components of $\chi_\perp$, on the contrary, decrease.

The real parts $\chi'_{||}$ and $\chi'_\perp$ of the susceptibilities as functions of temperature have maxima. The occurrence of these maxima is the result of competition between an increase of the average magnetic moment of the particle and the growth of relaxation time τ as the temperature decreases. The first factor leads to an increase of χ', the second one to a decrease.

If the linear approximation by the current field H_{II} holds well, then the stochastic resonance in a moderately concentrated frozen ferrocolloid has the same principal characteristics as those in dilute systems.

At the same time, the parameters of signal-noise ratio $I_{||}$ and $I_\perp$ corresponding to parallel and perpendicular orientations of the current $\mathbf{H}_{II}$ and applied during freezing $\mathbf{H}_I$ magnetic fields, increase with the parameters of interparticle magnetodipole interaction; $I_{||}$ increases and $I_\perp$ decreases as the field H_I increases.

We have performed our calculations of magnetic susceptibilities in Secs. II and III by using two different methods. Both methods lead to identical results in linear approxima-

tion by multiplying $\rho\gamma_J$ and the dimensionless current field $\boldsymbol{\alpha}_{II}$. However, the method of virial expansion used in Sec. II allows us to receive strict results for any value of $\boldsymbol{\alpha}_{II}$. In this situation one may calculate the parameters $\mathbf{K}_1$, $\mathbf{K}_2$ in Eq. (5) numerically to determine the stationary magnetization. It is not a difficult problem. At the same time, solving kinetic equation (10) in the nonlinear approximation by $\boldsymbol{\alpha}_{II}$ is connected with some principal difficulties. For this reason we decided to present here both methods of calculation of susceptibilities.

ACKNOWLEDGMENTS

APPENDIX A

Derivation of Expressions (5)

According to the definition and taking into account expression (4), we have

$$\langle\mathbf{e}\rangle = \int \mathbf{e}\psi_{II}\prod_k de_k d\mathbf{r}_k d\boldsymbol{\nu}_k$$

$$= \frac{1}{Z_I}\int \prod_k \frac{\Phi_I(k)}{\Phi_{II}(k)}\left(1+\frac{1}{2}\sum_{i\neq j} W_{ij}\right)\mathbf{e}_1\exp\left[\left(\boldsymbol{\alpha}_{II}\cdot\sum_j \mathbf{e}_j\right)+\kappa_{II}\sum_j (\mathbf{e}_j\cdot\boldsymbol{\nu}_j)^2 + \frac{1}{2}\sum_{i\neq j} w_{IIij}\right]\prod_l de_l d\mathbf{r}_l d\boldsymbol{\nu}_l.$$

Consider the integral

$$I = \int \mathbf{e}_1\exp\left[\left(\boldsymbol{\alpha}_{II}\cdot\sum_j e_j\right)+\kappa_{II}\sum_j (\mathbf{e}_j\cdot\boldsymbol{\nu}_j)^2 + \frac{1}{2}\sum_{i\neq j} w_{IIij}\right]\prod_l de_l$$

and let

$$Q_{II1j} = \int \mathbf{e}_1 f_{II1j}\exp\{[\boldsymbol{\alpha}_{II}\cdot(\mathbf{e}_1+\mathbf{e}_j)]+\kappa_{II}[(\mathbf{e}_1\cdot\boldsymbol{\nu}_1)^2+(\mathbf{e}_j\cdot\boldsymbol{\nu}_j)^2]\}de_1 de_j.$$

Then

$$I = \mathbf{J}_{II}(\boldsymbol{\nu}_1)\prod_{k\neq1} \Phi_{II}(\boldsymbol{\nu}_k) + \frac{1}{2}\sum_{i\neq j} f_{II1j}\mathbf{e}_1\exp(ij)de_1 de_j\times\prod_{k\neq i,j} \Phi_{II}(\boldsymbol{\nu}_k)$$

$$= \mathbf{J}_{II}(\boldsymbol{\nu}_1)\prod_{k\neq1} \Phi_{II}(\boldsymbol{\nu}_k) + \sum_{j\neq1} \int f_{II1j}\mathbf{e}_1\exp(1j)de_1 de_j\prod_{k\neq1,j} \Phi_{II}(\boldsymbol{\nu}_k)$$

$$+ \frac{1}{2}\sum_{i\neq j,\ i,j\neq1} \int f_{IIij}\exp(ij)de_i de_j\mathbf{J}_{II}(\boldsymbol{\nu}_1)\prod_{k\neq1,i,j} \Phi_{II}(\boldsymbol{\nu}_k)$$

$$= \mathbf{J}_{II}(\boldsymbol{\nu}_1)\prod_{k\neq1} \Phi_{II}(\boldsymbol{\nu}_k)\left(1+\sum_{j\neq1} \frac{\mathbf{Q}_{II1j}}{\mathbf{J}_{II}(\boldsymbol{\nu}_1)}\Phi_{II}(\boldsymbol{\nu}_j) + \frac{1}{2}\sum_{i\neq j,1\neq i,j} \frac{\Omega_{IIij}}{\Phi_{II}(\boldsymbol{\nu}_i)\Phi_{II}(\boldsymbol{\nu}_j)}\right),$$

$$\exp_{ij} = \exp\{[\boldsymbol{\alpha}_{II}\cdot(\mathbf{e}_1+\mathbf{e}_j)]+\kappa_{II}[(\mathbf{e}_1\cdot\boldsymbol{\nu}_1)^2+(\mathbf{e}_j\cdot\boldsymbol{\nu}_j)^2]\}.$$

Using this expression, we may obtain

$$\langle\mathbf{e}\rangle = \frac{1}{F_I}\int \mathbf{J}_{II}(\boldsymbol{\nu}_1)\prod_{k\neq1} \prod_{\text{all } l} \frac{\Phi_I(\boldsymbol{\nu}_l)}{\Phi_{II}(\boldsymbol{\nu}_l)}\left(1+\sum_{j\neq1} \frac{\mathbf{Q}^\star_{IIij}}{\mathbf{J}_{II}(\boldsymbol{\nu}_1)\Phi_{II}(\boldsymbol{\nu}_j)} - \sum_{j\neq1} \frac{\Omega_{II1j}}{\Phi_{II}(\boldsymbol{\nu}_1)\Phi_{II}(\boldsymbol{\nu}_j)} + \frac{1}{2}\sum_{i\neq j} \frac{\Omega_{IIij}}{\Phi_I(\boldsymbol{\nu}_i)\Phi_I(\boldsymbol{\nu}_j)}\right).$$

In the same approximation

$$Z_{\mathrm{I}} = V^N F_{\mathrm{I}}^N \left(1 + \frac{1}{2} \frac{N(N-1)}{V} \frac{G_{\mathrm{I}}}{F_{\mathrm{I}}^2} \right),$$

$$G_{\mathrm{I}} = \int \Omega_{\mathrm{I}12} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2 d\mathbf{r}_{12}, \quad F_{\mathrm{I}} = \int \Phi_{\mathrm{I}}(\boldsymbol{\nu}) d\boldsymbol{\nu}.$$

Using these expressions and neglecting the potential of interparticle interaction w_{ij}, we come to

$$\langle \mathbf{e} \rangle_0 = \frac{1}{F_{\mathrm{I}}} \int \mathbf{J}_{\mathrm{II}}(\boldsymbol{\nu}) \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu})}{\Phi_{\mathrm{II}}(\boldsymbol{\nu})} d\boldsymbol{\nu}.$$

Then in a linear approximation by the Meyer function, taking into account the identity of all particles in a thermodynamical limit, we get

$$\langle \mathbf{e} \rangle_1 = \langle \mathbf{e} \rangle - \langle \mathbf{e} \rangle_0 \approx \frac{n}{F_{\mathrm{I}}^2} \left(\int \frac{\Phi_{\mathrm{I}}(\nu_1)\Phi_{\mathrm{I}}(\nu_2)}{\Phi_{\mathrm{II}}(\nu_1)\Phi_{\mathrm{II}}(\nu_2)} \mathbf{Q}_{\mathrm{II}12} d\mathbf{r}_{12} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2 \right.$$

$$= \int \frac{\mathbf{J}_{\mathrm{II}}(\boldsymbol{\nu}_1)}{\Phi_{\mathrm{II}}(\boldsymbol{\nu}_1)} \Omega_{\mathrm{I}12} d\mathbf{r_{12}} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2 - \int \frac{\mathbf{J}_{\mathrm{II}}(\boldsymbol{\nu}_1)}{\Phi_{\mathrm{II}}(\boldsymbol{\nu}_1)} \frac{\Phi_{\mathrm{I}}(\boldsymbol{\nu}_1)\Phi_{\mathrm{I}}(\boldsymbol{\nu}_2)}{\Phi_{\mathrm{II}}(\boldsymbol{\nu}_1)\Phi_{\mathrm{II}}(\boldsymbol{\nu}_2)} \Omega_{\mathrm{II}12} d\mathbf{r}_{12} d\boldsymbol{\nu}_1 d\boldsymbol{\nu}_2 - G_{\mathrm{I}} \langle \mathbf{e} \rangle_0 \right).$$

From the last two expressions we may easily obtain the formula (5).

APPENDIX B

Calculation of the Parameters K_1, K_2, K_3, and G_{I}

We shall use designations $\gamma_J = m^2/[(2a)^3 T_J]$, $J = \mathrm{I},\mathrm{II}$.

For $\gamma_J < 1$, one may expand the exponent in the Meyer function f_{J12} [see Eq. (2)] by the powers of potential of dipole-dipole interparticle interaction w_d. In the linear approximation

$$f_{J12} = \frac{m^2}{T_J} \left(3 \frac{(\mathbf{e}_1 \mathbf{r}_{12})(\mathbf{e}_2 \mathbf{r}_{12})}{r_{12}^5} - \frac{(\mathbf{e}_1 \mathbf{e}_2)}{r_{12}^3} \right).$$

Using this approximation, let us calculate Γ_J in Eq. (5). It is convenient to write

$$\Gamma_J = \gamma_J (2a)^3 \Psi_J,$$

$$\Psi_J(\mathbf{e}_1, \mathbf{e}_2) = \int_{\xi > 1} \left(3 \frac{(\mathbf{e}_1 \boldsymbol{\xi})(\mathbf{e}_2 \boldsymbol{\xi})}{\xi^5} - \frac{(\mathbf{e}_1 \mathbf{e}_2)}{\xi^3} \right) d^3 \xi,$$

$$\xi = \frac{\mathbf{r}_{12}}{2a}.$$

For the reasons mentioned under the expression (7), we need to integrate over an infinite cylinder with the axis directed along the vector $\boldsymbol{\alpha}_J$ and crossing the center of one of the interacting particles (for example, the first one).

One may use for ξ a coordinate system where $2aR$ is the distance between a point in space and the cylinder axis, θ is the angle between radius vector of this point and this axis, and φ is the polar angle in plane normal to $\boldsymbol{\alpha}_J$. For vector $\boldsymbol{\nu}$ we will use the Cartesian-coordinate system with the axis OZ aligned with $\boldsymbol{\alpha}_J$.

Using these coordinate systems, we come to

$$\Psi_J(\mathbf{e}_1, \mathbf{e}_2) = -2\pi \int \left(e_{1Z} e_{2Z} - \frac{1}{2}(e_{1X} e_{2X} + e_{1Y} e_{2Y}) \right)$$

$$\times (3\cos^2\theta - 1) \frac{\sin\theta}{R} d\theta \, dR.$$

At first, it is necessary to perform integration over the angle θ at fixed R and then to integrate over R (the integrating volume is the above-mentioned cylinder). Taking into account that the particles are rigid spheres, one may obtain the following expressions for the region of integration over θ:

$$0 \leq \theta \leq \arcsin R, \quad \pi - \arcsin R \leq \theta \leq \pi \quad \text{if } R < 1,$$

$$0 \leq \theta \leq \pi \quad \text{if } R > 1.$$

After simple calculations,

$$\Psi_J = \frac{4\pi}{3} \left(e_{1Z} e_{2Z} - \frac{1}{2}(e_{1X} e_{2X} + e_{1Y} e_{2Y}) \right).$$

Therefore,

$$\Gamma_J(\mathbf{e}_1, \mathbf{e}_2) = 8 \gamma_J v \left(e_{1Z} e_{2Z} - \frac{1}{2}(e_{1X} e_{2X} + e_{1Y} e_{2Y}) \right).$$
(B1)

If $\kappa_J \gg 1$, then

$$\exp(\kappa_J(\mathbf{e} \cdot \boldsymbol{\nu})^2) \approx 2\pi \frac{\exp(\kappa_J)}{\kappa_J} [\delta(\mathbf{e} - \boldsymbol{\nu}) + \delta(\mathbf{e} + \boldsymbol{\nu})]. \quad (B2)$$

Therefore at $\boldsymbol{\alpha}_{\mathrm{II}} \ll 1$,

$$\Phi_I(\boldsymbol{\nu}) \approx 4\pi \frac{\exp(\kappa_I)}{\kappa_I} \mathrm{ch}(\boldsymbol{\alpha}_I \cdot \boldsymbol{\nu}), \qquad (B3)$$

$$\Phi_{II}(\boldsymbol{\nu}) \approx 4\pi \frac{\exp(\kappa_{II})}{\kappa_{II}},$$

$$F_I \approx (4\pi)^2 \frac{\exp(\kappa_I)}{\kappa_I} \frac{\mathrm{sh}(\alpha_I)}{\alpha_I},$$

$$\mathbf{J}_{II}(\boldsymbol{\nu}) \approx 4\pi \frac{\exp(\kappa_{II})}{\kappa_{II}} \boldsymbol{\nu}(\boldsymbol{\alpha}_{II} \cdot \boldsymbol{\nu}).$$

Taking account of

$$\Gamma_J(\mathbf{x}_1, \mathbf{x}_2) = \Gamma_J(-\mathbf{x}_1, -\mathbf{x}_2) = -\Gamma_J(\mathbf{x}_1, -\mathbf{x}_2)$$
$$= -\Gamma_J(-\mathbf{x}_1, \mathbf{x}_2)$$

and expression (B2), one may easily obtain

$$\mathbf{Q}^\star_{II12} \approx (4\pi)^2 \frac{\exp(2\kappa_J)}{\kappa_J^2} \Gamma_{II}(\boldsymbol{\nu}_1, \boldsymbol{\nu}_2) \boldsymbol{\nu}_1(\boldsymbol{\alpha}_{II} \cdot \boldsymbol{\nu}_1),$$

$$\Omega^\star_{I12} \approx (4\pi)^2 \frac{\exp(2\kappa_I)}{\kappa_I^2} \Gamma_I(\boldsymbol{\nu}_1, \boldsymbol{\nu}_2) \frac{\mathrm{ch}[\boldsymbol{\alpha}_I \cdot (\boldsymbol{\nu}_1 + \boldsymbol{\nu}_2)] - \mathrm{ch}[\boldsymbol{\alpha}_I \cdot (\boldsymbol{\nu}_1 - \boldsymbol{\nu}_2)]}{2}, \qquad (B4)$$

$$\Omega_{II12} \approx 0.$$

Now we may derive $\mathbf{K}_1, \mathbf{K}_2, \mathbf{K}_3$. In the process of these calculations it is necessary to suggest that $\boldsymbol{\alpha}_J = \boldsymbol{\alpha}_{II}$. Let us introduce the Cartesian-coordinate system with axis Oz directed along $\boldsymbol{\alpha}_I$ and axis Ox located in the plane $(\boldsymbol{\alpha}_I, \boldsymbol{\alpha}_{II})$.

Let φ be the angle between OZ and Oz (i.e., between $\boldsymbol{\alpha}_I$ and $\boldsymbol{\alpha}_{II}$). Using the ordering correlations between vector components after rotation of coordinate system and Eq. (B1), one obtain

$$\Gamma_{II}(\boldsymbol{\nu}_1, \boldsymbol{\nu}_2) = 8\gamma_{II} v \left[\left(\nu_{1z}\nu_{2z} - \frac{1}{2}\nu_{1x}\nu_{2x} \right) \cos^2\varphi \right.$$

$$\left. + \left(\nu_{1x}\nu_{2x} - \frac{1}{2}\nu_{1z}\nu_{2z} \right) \sin^2\varphi + \frac{3}{2}(\nu_{1x}\nu_{2z} + \nu_{2x}\nu_{1z})\sin\varphi\cos\varphi \right] - \frac{1}{2}\nu_{1y}\nu_{2y}. \qquad (B5)$$

Substituting this expression in Eq. (5), after simple calculations we obtain Eq. (7).

At the calculations of G_I one needs to suggest $\boldsymbol{\alpha}_J = \boldsymbol{\alpha}_I$, i.e., $OZ = Oz$. Substituting Eqs. (B2) and (B4) in Eq. (5), we come to expression (7) for G_I.

APPENDIX C

Using the expression (2), we have

$$g_2(\boldsymbol{\nu}_1) = \int g_I(\mathbf{r}_1, \ldots, \mathbf{r}_N \boldsymbol{\nu}_1, \ldots, \boldsymbol{\nu}_N) \prod_{\mathrm{all}\ k} d\mathbf{r}_k \prod_{k \neq 1} d\boldsymbol{\nu}_k$$

$$\approx \frac{1}{Z_I} \left(1 + \frac{1}{2} \sum_{i \neq j} \frac{\Omega_I(ij)}{\Phi(i)\Phi(j)} \right) \prod_k \Phi_I(k) \prod_{\mathrm{all}\ k} d\mathbf{r}_k \prod_{k \neq 1} d\boldsymbol{\nu}_k$$

$$= \frac{1}{Z_I} \left(\Phi_I(1) V^N F_I^N + \sum_{j \neq 1} \int \Omega_I(1j) d\mathbf{r}_{1j} d\boldsymbol{\nu}_j V^{N-1} F_I^{N-2} + \frac{1}{2} \sum_{i \neq j,\ i,j \neq 1} \Omega_I(ij) d\mathbf{r}_{ij} d\boldsymbol{\nu}_i d\boldsymbol{\nu}_j V^{N-1} F_I^{N-3} \right)$$

$$= \frac{V^N F_I^N}{Z_I} \left(\frac{\Phi_I(1)}{F_I} + \frac{N-1}{V F_I^2} \int \Omega^\star_{I12} d\boldsymbol{\nu}_2 + \frac{(N-1)(N-2)}{2V F_I^2} \int \Omega^\star_{I23} d\boldsymbol{\nu}_2 d\boldsymbol{\nu}_3 \right).$$

Taking into account that

$$G_I = \int \Omega^\star_{Iij} d\boldsymbol{\nu}_i d\boldsymbol{\nu}_j,$$

$$Z_I \approx F_I^N V^N \left(1 + \frac{1}{2} \frac{N(N-1)}{V} \frac{G_I}{F_I^2} \right),$$

$$n = \frac{N}{V}$$

[see Eq. (5) and Appendix B] in the thermodynamical limit we come to the expression (34).

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