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Polarization Oscillations of Dipole Media

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A collisionless kinetic equation with a self-consistent field is obtained for dipole gases. The dispersion properties of the medium in the presence and absence of an external stationary electric field are investigated on the basis of the equation. The dielectric constant and polarizability tensors of such a medium are calculated and the conditions and parameters of the proper longitudinal and transverse oscillations of the polarization vector are found. It is pointed out that such oscillations may build up in a stationary electric field.

INTRODUCTION

THE kinetic theory of polar and paramagnetic gases was developed by a number of workers,[1-6] who paid principal attention to finding the transport coefficients and to their analysis in the presence of external fields. In the present paper we consider the dispersion properties of electric-dipole media and possible oscillation modes that arise in them. Examples of such media are gases and liquids whose molecules have a constant electric dipole moment, and also media with dipoles induced by an external field.

It is well known that the character of the interaction between the particles ensures specific properties of the dispersion dependence of the medium and a set of possible natural oscillations corresponding to this dispersion. Under conditions when the medium consists of neutral particles, electric dipole-dipole interaction is decisive and can make an appreciable contribution to the dielectric tensor.

The analysis is based on the Boltzmann collisionless equation with a self-consistent field. We show below that there exists a region of applicability of the collisionless equation for the description of polar gases. The intermolecular interaction can be taken into account within the framework of the self-consistent-field formalism, which is introduced into the dynamic part of the Boltzmann equation.

1. FUNDAMENTAL EQUATIONS

The kinetic equation for the single-particle distribution function can be obtained from the Liouville equation neglecting the two-particle etc. correlations, by means of the usual procedure of averaging over the dynamic variables of all the particles except one (see, e.g., [7]):

$$\frac{\partial f}{\partial t}+\dot{\mathbf{x}}_i\frac{\partial f}{\partial \mathbf{x}_i}+\frac{\partial f}{\partial \mathbf{p}_i}\frac{\partial}{\partial \mathbf{x}_i}\mathbf{d}_i\int f(\mathbf{x}_j,\mathbf{p}_j,t)\,\nabla_{\mathbf{r}_j}\frac{\mathbf{d}_j\mathbf{r}_{ji}}{r_{ji}^3}\,d\mathbf{x}_j\,d\mathbf{p}_j=0. \quad (1)$$

The interaction is taken into account in Hamiltonian in the dipole-dipole approximation, $\mathbf{x}_i$ and $\mathbf{p}_i$ are the generalized coordinate and the corresponding generalized momentum ($\mathbf{x}_i$ includes the orientation angle coordinates Ω_i of the dipole $\mathbf{d}_i$ and the radius vector of its center of gravity $\mathbf{r}_i$); $\mathbf{r}_{ji} = \mathbf{r}_j - \mathbf{r}_i$. The integral in (1) can be interpreted as the intensity of the self-consistent field of a system of dipoles at the point where the i-th dipole is located.

We shall show that the field $\mathbf{E}(\mathbf{r}_i, t)$ defined in this manner satisfies the Poisson equation

$$\operatorname{div}\mathbf{E}=-4\pi\operatorname{div}\mathbf{P}, \quad (2)$$

where the polarization vector $\mathbf{P}$ is defined by the relation

$$\mathbf{P}=\int \mathbf{d}_i f(\mathbf{x}_i,\mathbf{p}_i,t)\,d\mathbf{p}_i\,d\Omega_i. \quad (3)$$

Indeed, by defining the integral in (1) as the field E and calculating div E, we obtain after simple transformations

$$\operatorname{div}\mathbf{E}=-\int \operatorname{div}_{\mathbf{r}_j}\left[f(\mathbf{x}_j,\mathbf{p}_j,t)\,\mathbf{d}_j\Delta_{\mathbf{r}_j}\frac{1}{r_{ji}}\right]d\mathbf{x}_j\,d\mathbf{p}_j + \int \Delta_{\mathbf{r}_i}\frac{1}{r_{ji}}\operatorname{div}_{\mathbf{r}_j}[\mathbf{d}_j f(\mathbf{x}_j,\mathbf{p}_j,t)]\,d\mathbf{x}_j\,d\mathbf{p}_j. \quad (4)$$

According to the Gauss theorem, the first integral in (4), calculated over the volume $d^3\mathbf{r}_j$, can be transformed into an integral over an infinitely remote surface. It is equal to zero. We use the well known relation

$$\Delta_{\mathbf{r}_i}\frac{1}{r_{ji}}=-4\pi\delta(r_{ji}).$$

We then obtain Eq. (2) from (4) when we take into account the definition (3) of the polarization P.

From the complete system of Maxwell's equations for the system of bound charges (where the current density is $\mathbf{j} = \partial\mathbf{P}/\partial t$), we obtain

$$\Delta\mathbf{E}-\frac{1}{c^2}\ddot{\mathbf{E}}=\frac{4\pi}{c^2}\ddot{\mathbf{P}}-4\pi\operatorname{grad}\operatorname{div}\mathbf{P}. \quad (5)$$

We write the kinetic equation (1) in a more compact form:

$$\frac{\partial f}{\partial t}+\dot{\mathbf{x}}\frac{\partial f}{\partial \mathbf{x}}+\frac{\partial}{\partial \mathbf{x}}(\mathbf{dE})\frac{\partial f}{\partial \mathbf{p}}=0. \quad (6)$$

The collisionless equation (6) has been written out in first order in the small parameter

$$d^2n/T\ll 1, \quad \nu/\omega\ll 1, \quad (7)$$

where n is the particle concentration, T is the absolute temperature in energy units, ν is the collision frequency, and ω are the characteristic frequencies of the problem. The first inequality of (7) follows from the condition for factorization of the distribution function, which is connected with the smallness of the characteristic time of the problem in comparison with the collision-relaxation times.

We shall confine ourselves henceforth to a gas of spherically-symmetrical particles. This simplifies the analysis without loss of generality. The dipole-molecule model will be a hard sphere of mass μ and moment of inertia J, containing a point dipole **d**. The coordinates of the particle are specified by radius vector **r**, which determines the position of the center of gravity of the sphere, and by the Euler angles, which describe the orientation of the vector **d** in space (see the figure). The distribution function is specified in the space of **r**, θ, φ, and of the corresponding momenta. Separating in (6) the angle variables and the corresponding momenta, we represent it in the form

$$\frac{\partial f}{\partial t}+\mathbf{v}\frac{\partial f}{\partial \mathbf{r}}+\omega_\theta\frac{\partial f}{\partial \theta}+\omega_\varphi\frac{\partial f}{\partial \varphi}+\frac{1}{\mu}\nabla_r(\mathbf{dE})\frac{\partial f}{\partial \mathbf{v}}+\frac{M_{\omega_\theta}}{J}\frac{\partial f}{\partial_{\omega_\theta}}+\frac{M_{\omega_\varphi}}{J}\frac{\partial f}{\partial_{\omega_\varphi}}=0. \quad (8)$$

Here $\mathbf{v} = \dot{\mathbf{r}}$, $\omega_\theta = \dot{\theta}$, $\omega_\varphi = \dot{\varphi}$, M_θ and M_φ are the projections of the angular momentum of the dipole $\mathbf{M} = \mathbf{d}\times\mathbf{E}$ on the directions of $\dot{\theta}$ and $\dot{\varphi}$ (see also the figure).

To present Eq. (8) in greater detail, we shall need the components of the dipole moment vector in an immobile coordinate system xyz:[8]

$$d\sin\theta\sin\varphi,\qquad -d\sin\theta\cos\varphi,\qquad d\cos\theta \quad (9a)$$

and the components of the momentum

$$M_{\omega_\theta}=d(E_y\cos\varphi\cos\theta-E_x\sin\varphi\cos\theta+E_z\sin\theta),$$
$$M_{\omega_\varphi}=d(E_x\cos\varphi\sin\theta+E_y\sin\varphi\sin\theta). \quad (9b)$$

The definition (3) of the polarization vector, Maxwell's equation (5) for the field, and the kinetic equation (8) constitute the complete system of equations of the problem under consideration.

2. DISPERSION RELATIONS IN THE ABSENCE OF EXTERNAL FIELDS

To find the natural oscillations of the dipole medium, it is necessary to find the conditions under which the system of equations (3), (5), and (8) has non-trivial solutions in the approximation linear in the perturbation. This procedure is described in the mathematical appendix. We introduce the polarizability tensors α_{ij} and the dielectric tensor, which are connected by the well known relation $\epsilon_{ij} = \delta_{ij} + 4\pi\alpha_{ij}$. Then the dispersion equation for the longitudinal wave ($\mathbf{k} \parallel \mathbf{P}$) in a coordinate system with the z axis directed along the wave vector **k**, take on the usual form

$$\varepsilon_{zz}=0. \quad (10)$$

For a transverse wave ($\mathbf{k}\perp\mathbf{P}$) in the same coordinate system, the dispersion equation is given by the following relation:

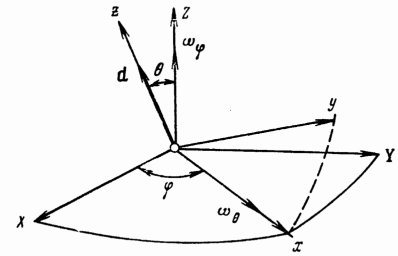
$$N^2=\varepsilon_\perp=\varepsilon_{xx}=\varepsilon_{yy}, \quad (11)$$

where $N = kc/\omega$ is the refractive index of the medium.

The components of the polarizability tensor α_{ij} are calculated in the Appendix. Using the results of these calculations (A.7), we transform the dispersion equation (10):

$$1+\frac{4\pi}{3}\frac{nd^2}{T}\left[1+i\sqrt{\pi}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T}\right)\right]=0, \quad (12)$$

$\omega_T = (2T/J)^{1/2}$, and W is the Kramp function. We shall henceforth neglect the contribution of the translational motion of the particles to the dispersion. This contribution is much smaller than unity for wavelengths ex-



ceeding the dipole dimensions r_0 (of the order of $(kr_0)^2$).

We shall analyze the dispersion equation (10) for two limiting cases of high and low frequencies. We consider first waves with frequencies $\omega \gg \omega_T$. Using the asymptotic representations (A.8) for the Kramp function W, we obtain from (12)

$$\omega^2=\omega_d^2\frac{4\pi}{3}\frac{nd^2}{J}. \quad (13)$$

We now rewrite ϵ_{zz} in the form $\epsilon_{zz} = 1 - (\omega_d/\omega)^2$. From this we can conclude that oscillations with frequencies $\omega \gtrsim \omega_d$ can propagate in the dipole medium. The medium is opaque to frequencies lower than ω_d. We trace here an analogy with the so-called plasma oscillations in a homogeneous non-magnetized plasma. It should be noted that the limits of applicability of the theory, namely the smallness of the parameter $\kappa = nd^2T$, narrow down the region of existence of the natural dipole oscillations of the medium. Such oscillations can be realized only under the condition $4\pi\kappa/3 > 1$. In the other limiting case, $\omega \ll \omega_T$, we have

$$\varepsilon_{zz}=1+{}^4/_3\pi\varkappa(1+i\sqrt{\pi}\omega/\omega_T). \quad (14)$$

It is easy to see that ϵ_{zz} does not vanish, in accordance with (14), for any value of $\omega < \omega_T$, i.e., there are no natural oscillations of the medium in such a frequency region.

We consider now transverse oscillations. The dispersion equation (12) is reduced, with allowance for (A.6), to the form

$$N^2=1+\frac{4\pi}{3}\varkappa\left[1+i\sqrt{\frac{\pi}{2}}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T\sqrt{2}}\right)\right]. \quad (15)$$

As above, we consider two limiting cases: $\omega \gg \omega_T$ and $\omega \ll \omega_T$. In the first case, using the asymptotic form of W at $|z| \gg 1$, we obtain

$$N^2=1-{}^4/_3\pi\varkappa\,\omega_T^2/\omega^2. \quad (16)$$

This relation shows that the refractive index of the medium of dipoles is less than unity for such electromagnetic waves. In the frequency range

$$\omega_T^2<\omega<\omega_T(4\pi\varkappa/3)^{1/2}$$

the refractive index becomes imaginary—these waves do not penetrate into the dipole medium. The length of the skin layer for these waves is of the order of $l < c/\omega_d$.

In the limiting case of low frequencies, $\omega \ll \omega_T$, the dispersion equation (15) takes the form

$$N^2=1+\frac{4\pi}{3}\varkappa\left[1+i\frac{\omega}{\omega_T}\sqrt{\frac{\pi}{2}}\right]. \quad (17)$$

The refractive index of the medium has an imaginary part connected with the Landau damping. Consequently, the wave is absorbed by the medium upon interaction with the resonant dipoles (the frequency of the revolution of the dipoles is of the order of the frequency of the electromagnetic wave).

3. DIPOLE GAS IN A CONSTANT ELECTRIC FIELD

The dipole medium in an external homogeneous constant electric field has a number of distinguishing properties in comparison with such a medium in the absence of a field. These distinguishing properties are due to the change in the character of the unperturbed motion of the dipoles, and to the presence of static polarization connected with the orienting action of the field and with the appearance of a potential energy of the dipole in the external field. All this affects significantly the dispersion properties of the medium of dipoles and in many cases can lead to the appearance of instabilities connected with the conversion of the potential energy of the dipole in the external field into the wave energy. Allowance for the field reduces formally to the need for using for the total electric field in the kinetic equation (8) the sum $\mathbf{E}_0 + \mathbf{E}$, where $\mathbf{E}_0$ describes the intensity of the constant external field and $\mathbf{E}$ is the self-consistent field.

To solve the kinetic equation with allowance for the external field, we shall use the method of integration over the trajectories (this method was used to obtain the results of Sec. 2, see the Appendix). The unperturbed trajectories are obtained in this case by solving the following equations of motion:

$$\ddot{r} = 0, \quad \ddot{\varphi} = 0, \quad \ddot{\theta} + (dE_0/J)\sin\theta = 0 \tag{18}$$

(the field $\mathbf{E}_0$ is directed along the z axis). The last equation describes the motion of the dipole in a plane passing through the vectors $\mathbf{E}_0$ and $\mathbf{d}$. If the total dipole energy ϵ exceeds its energy in the field $\mathbf{E}_0$, then the latter can be regarded as a small perturbation of the free dipole, equal in order of magnitude to $dE_0/\epsilon \approx dE_0/J\omega_\theta^2 \ll 1$. In the first approximation in this small parameter, the trajectory is given by

$$\theta(t') - \theta(t) = \omega_\theta(t - t') + (dE_0/J\omega_\theta^2)\{\sin[\theta(t' - t) + \theta(t)] - \sin\theta(t)\}. \tag{19}$$

In the other limiting case, $\epsilon < dE_0$, the angle of rotation of the dipole with respect to θ is limited, and the solution of the last equation of (18) (see, e.g., [8]) does not have so simple an asymptotic form. The character of the motion of such "captured" dipoles is qualitatively different from the motion of "almost free" particles, a motion not bounded with respect to the angle θ. A consistent allowance for the two mentioned groups of particles calls for integrations over rather complicated trajectories $\theta(t)$, which entails great mathematical difficulties. Qualitative results can be obtained in the limiting case of high frequencies on the basis of the following considerations. Since $\alpha = dE_0/T \ll 1$ in a real situation, we can show that the number of "captured dipoles" is of the order of $\alpha^{1/2}$ in comparison with the total number of the particles. Since the "captured dipoles" are concentrated near small angular velocities $0 < \omega_\theta < (dE_0/J)^{1/2}$, the role of captured particles is insignificant at high frequencies $\omega \gg (dE_0/J)^{1/2}$, namely, at such frequencies this group of particles is practically immobile and does not take part in the high-frequency oscillations of the medium. Estimates show that the total contribution made to the polarization by these particles is of the order of $\alpha^{1/2}E_0 d/J\omega_\theta^2$. We shall henceforth consider the dispersion properties in the region of high frequencies, where the contribution of the "captured dipoles" can be neglected.

The procedure of obtaining dispersion relations for a system of dipoles in a constant electric field is analogous to that described for free dipoles in the Appendix. In the representation (A.4), we obtain for the components $f_{k\omega}^{lm}$ with the aid of trajectories (18), taking (19) into account,

$$\begin{aligned} f^{\pm 10} &= \pm\frac{f_0 d}{2T}\sum_{n,p} J_p(\alpha_1)J_n(\alpha_1)\frac{\omega_\theta E_z}{\omega \pm (p+1)\omega_\theta}e^{\pm i(p-n)\theta}, \\ f^{\pm 11} &= -\frac{f_0 d}{4T}\sum_{n,p} J_p(\alpha_1)J_n(\alpha_1)\frac{\Omega_\pm E_-}{\omega \pm (p+1)\omega_\theta + \omega_\varphi}e^{\pm i(p-n)\theta}, \\ f^{\pm 1-1} &= \frac{f_0 d}{4T}\sum_{n,p} J_p(\alpha_1)J_n(\alpha_1)\frac{\Omega_\mp E_+}{\omega \pm (p+1)\omega_\theta - \omega_\varphi}e^{\pm i(p-n)\theta}, \end{aligned} \tag{20}$$

where

$$f_0 = \frac{nJ}{8\pi^2 T}\left(\frac{\mu}{2\pi T}\right)^{3/2}\exp\left\{\frac{1}{T}\left[E_0 d - \frac{\mu v^2}{2} - \frac{J\omega_\theta^2}{2} - \frac{J\omega_\varphi^2}{2}\right]\right\} \tag{20a}$$

is the stationary distribution function of the dipoles in a constant electric field, $J_n(\alpha_1)$ is a Bessel function, and $\alpha_1 = E_0 d/J\omega_\theta^2$.

Taking the relations (20) into account, and recognizing that $\langle\alpha_1\rangle \ll 1$ (where the angle brackets denote averages over the distribution function), it is easy to obtain the components of the polarizability tensor and of the dielectric constant (see the Appendix). We note that the equation of the trajectory (19) is valid only in the region $\omega_\theta > (E_0 d/J)^{1/2}$. By virtue of the fact that for $\omega > (E_0 d/J)^{1/2}$ the contribution made to the integrals by the region $0 < \omega_\theta < (E_0 d/J)^{1/2}$ is negligibly small, the integration over the trajectories (19) can be extended also into this region.

The dispersion equation for the longitudinal waves has in this case the form

$$\begin{aligned} 1 &+ \frac{4\pi}{3}\varkappa\left[1 + i\sqrt{\pi}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T}\right)\right] \\ &+ i\frac{\pi}{32}\alpha\varkappa\left[1 + i\frac{\sqrt{\pi}}{2}\frac{\omega}{\omega_T}W\left(\frac{\omega}{2\omega_T}\right)\right] = 0. \end{aligned} \tag{21}$$

This equation is valid for frequencies $\omega > (E_0 d/J)^{1/2}$, and reduces in the range $\omega \gg \omega_T$, with the aid of (A.8) to the form

$$\omega^2 - \omega_d^2(1 + i\cdot 3\alpha/32) = 0, \tag{22}$$

whence

$$\omega = \pm\omega_d(1 + {}^3/_{64}\alpha i).$$

The last relation points to a possible instability of the polarization oscillations in a constant electric field, with an increment $\gamma = 3\alpha\omega_d/64$ (in this case $\gamma/\omega_d \ll 1$). Such an instability is connected with the non-equilibrium nature of the dipole distribution in the constant electric field with respect to the angles θ.

In the case $(E_0 d/J)^{1/2} < \omega < \omega_T$, the result is similar to formula (14), with slight shifts of the real and imaginary parts of ω, due both to Landau damping and to the presence of a constant field.

For transverse waves ($\mathbf{k} \parallel \mathbf{E}_0$, $\mathbf{k} \perp \mathbf{P}$), the dispersion equation is given by

$$N^2 = 1 + \frac{\varkappa}{2\pi}\left\{\frac{8}{3}\left[1 + i\sqrt{\frac{\pi}{2}}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T\sqrt{2}}\right)\right] + \alpha\frac{\pi}{8}i\left[1 + \left(\frac{\omega}{\omega_T} - 1\right)i\sqrt{\frac{\pi}{5}}\left(\frac{\omega}{\omega_T\sqrt{5}}\right)\right] + \alpha\frac{\pi}{4}i\left[1 + i\sqrt{\pi}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T}\right)\right]\right\}. \quad (23)$$

In the limit $\omega > \omega_T\sqrt{5}$, we obtain

$$N^2 = 1 - \frac{1}{2}\frac{\omega_d^2}{\omega^2}\left(1 + \frac{21}{128}\alpha\pi i\right), \quad (24)$$

i.e., the transverse wave in the presence of an external field can grow in space. The characteristic growth length is of the order of $l \sim c/\alpha\omega$.

4. DISCUSSION OF RESULTS AND OF THE REGION OF THEIR APPLICABILITY

We have analyzed the dispersion properties of the dipole gas on the basis of the Boltzmann collisionless kinetic equation. The possibility of such an analysis is connected with neglect of the correlations between the dipoles, and also with the larger collision times in comparison with the characteristic times of the processes. These conditions are described by the inequalities (7). Estimates show that the first of these inequalities is, at room temperatures and for typical dipole moments[9] (1–4) D (D = 10^{-18} abs. units), is satisfied up to the concentration 10^{21} cm^{-3}. The second reduces to the relation $0.3r_0n^{1/3}\kappa^{1/6} \ll 1$, where allowance was made for the fact that $\nu \approx \pi\langle\rho\rangle^2 v_T n$; $\langle\rho\rangle$ is defined as the length of splitting of the correlations between two dipoles from the relation[9] $\bar{u}/T = \frac{2}{3}d^4/\langle\rho\rangle^6T^2$, where $\bar{u}$ is the average energy of interaction between the dipoles. At the same parameters, this condition is satisfied at concentrations $n < 10^{24}$ cm^{-3}. Thus, the upper concentration limit is imposed by the applicability of the collisionless approximation, and the lower limit is the threshold of the intensity of the interactions, starting with which the collective oscillations of the dipole medium can take place. This yields the estimate $n \sim 10^{19}$–10^{20} cm^{-3}.

From among the dispersion properties of a dipole medium in an external constant electric field, the most significant is the possibility of buildup of polarization oscillations. The mechanism of this buildup is described in Sec. 3.

We note in conclusion that the dispersion properties of dipole gases, described above, can occur also in ferroelectric crystals with continuously varying orientation of the dipoles,[10,11] and apparently the theory is applicable, without significant changes, to such crystals with cubic symmetry. Nonetheless, the question of collective polarization properties in solids calls for an independent analysis.

We are grateful to V. N. Oraevskiĭ for useful discussions and to O. I. Fisun for a discussion of questions involved in the formulation of the problem.

APPENDIX

Equation (8), when linearized with respect to the perturbation, takes the form

$$\frac{\partial f_1}{\partial t} + \mathbf{v}\frac{\partial f_1}{\partial \mathbf{r}} + \omega_\theta\frac{\partial f_1}{\partial \theta} + \omega_\varphi\frac{\partial f_1}{\partial \varphi} + \frac{1}{\mu}\frac{\partial}{\partial \mathbf{r}}(\mathbf{dE})\frac{\partial f}{\partial \mathbf{v}} + \frac{M_{\omega_\theta}}{J}\frac{\partial f}{\partial \omega_\theta} + \frac{M_{\omega_\varphi}}{J}\frac{\partial f}{\partial \omega_\varphi} = 0,$$

f_1 is the perturbed part of the distribution function and f is a stationary distribution function, which is the solution of the equation

$$\mathbf{v}\frac{\partial f}{\partial \mathbf{r}} + \omega_\theta\frac{\partial f}{\partial \theta} + \omega_\varphi\frac{\partial f}{\partial \varphi} = 0 \quad (A.2)$$

The explicit form of f follows from (20a) at $E_0 = 0$.

The solution of Eq. (A.1) with allowance for (9a) and (9b) and neglecting the translational motion of the dipole, can be written in the form

$$f_1 = -\int_{-\infty}^{t} dt' \frac{fd}{T}[E_x(\omega_\varphi\cos\varphi'\sin\theta' - \omega_\theta\sin\varphi'\cos\theta') + E_y(\omega_\varphi\sin\varphi'\sin\theta' + \omega_\theta\cos\varphi'\cos\theta') + E_z\omega_\theta\sin\theta']. \quad (A.3)$$

The integral in (A.3) is calculated over the unperturbed trajectories of the dipoles

$$\theta(t') - \theta(t) = \omega_\theta(t' - t); \quad \varphi(t') - \varphi(t) = \omega_\varphi(t' - t);$$
$$\theta' \equiv \theta(t'), \quad \varphi' \equiv \varphi(t').$$

Choosing the perturbation in the form $\sim\exp[i(\omega t + \mathbf{k}\cdot\mathbf{r})]$ and putting

$$f_{k\omega}' = \sum_{l,m} f_{k\omega}{}^{lm} e^{i(m\varphi + l\theta)}, \quad (A.4)$$

we obtain after integrating (A.3)

$$f_{k\omega}^{\pm1,0} = \pm\frac{fd}{2T}\frac{\omega_\theta E_z}{\omega \pm \omega_\theta},$$
$$f_{k\omega}^{\pm1,1} = -\frac{fd}{4T}\frac{\Omega_\pm E_-}{\omega \pm \Omega_\pm}, \quad f_{k\omega}^{\pm1,1} = \frac{fd}{4T}\frac{\Omega_\mp E_+}{\omega \pm \Omega_\mp}, \quad (A.5)$$
$$\Omega_\pm = \omega_\theta \pm \omega_\varphi, \quad E_\pm = E_x \pm iE_y.$$

We now substitute the distribution function (A.5) into the equation for the polarization (3) and integrate over the phase volume. For the components of the polarizability tensor we obtain

$$\alpha_{xx} = \alpha_{yy} = \frac{1}{3}\frac{nd^2}{T}\left[1 + i\sqrt{\frac{\pi}{2}}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T\sqrt{2}}\right)\right], \quad (A.6)$$

$$\alpha_{zz} = \frac{1}{3}\frac{nd^2}{T}\left[1 + i\sqrt{\frac{\pi}{2}}\frac{\omega}{\omega_T}W\left(\frac{\omega}{\omega_T}\right)\right], \quad (A.7)$$

where

$$W(z) = \frac{i}{\pi}\int_{-\infty}^{\infty}\frac{e^{-t^2}}{z - t}dt$$

is the Kramp function, which is tabulated in [12].

We present here the asymptotic expressions for W(z):

$$|z| \gg 1, \quad W(z) = \frac{i}{\pi}\sum_{k=0}^{\infty}\frac{\Gamma(k + 1/2)}{z^{2k+1}}, \quad (A.8)$$

$$|z| \ll 1 \quad W(z) = \sum_{k=0}^{\infty}\frac{(iz)^k}{\Gamma(1 + k/2)}, \quad (A.9)$$

$\Gamma(z)$ is the gamma function.

Note added in proof (27 November 1971). It might seem that the Langevin expansion (20a) is stable and should not lead to a buildup of oscillations. Nonetheless, allowance for the strong correlating action of the field in addition to the dipole dipole interaction shows that an infinite spatially-homogeneous medium is not in thermodynamic equilibrium. It is possible that the instability obtained in Sec. 3 is a manifestation of this fact. This question, however, requires a detailed analysis.

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