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FLUCTUATIONS AND TRANSPORT IN AN ELECTRIC FIELD

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UDC 532.5:537

It is shown that thermodynamic fluctuations in a liquid in an applied electric field generate microconvective motion causing mass and heat dispersion. The corresponding dispersion coefficients can be comparable in value or even exceed the coefficients of molecular diffusion and thermal conductivity.

An applied electric field can significantly stimulate heat- and mass-transport processes in liquids with very different electric and magnetic properties [1-3]. The usual interpretation is that this is due to the appearance of specific convective motions in the liquid from Coulomb pondermotive forces and Lorentz forces and also from convective transport of volume charge.

However, these effects do not exhaust all possible effects from the field, since many examples are known where heat and mass exchange are stimulated in situations where convection does not appear. In addition, the necessary condition for the appearance of electro or magnetohydrodynamic convection is that there be nonuniformities in either the properties of the liquid or the external field. Stimulation of heat and mass transport in an applied field is observed in conditions when both the liquid and the field can certainly be considered as uniform. Therefore, besides convection, one concludes that there is another fundamental effect of the electric field on transport processes, in general not involving the violation of mechanical stability of the liquid.

It is shown below that the latter effect comes from the appearance of additional microconvective dispersion in a liquid which is macroscopically at rest. This dispersion is due to random small-scale fluctuations which appear due to the interaction of the external field with random fluctuations of the volume charge. The latter is in turn caused by the usual thermodynamic fluctuations. Since the purpose of the present paper is to demonstrate the existence of this effect, we consider only the simplest examples in a uniform applied electric field and with several simplifying assumptions.

Fluctuations and Dispersion. At small Reynolds numbers the equations of hydrodynamics in the presence of the pondermotive force are given by

$$\gamma\partial\mathbf{v}/\partial t = -\nabla p + \mu\Delta\mathbf{v} + \rho\mathrm{E},\ \mathrm{div}\mathbf{v} = 0. \tag{1}$$

As in [4], the above expression for the pondermotive force is also assumed to be valid for a conducting liquid if the conductivity can be made as small as desired. The unperturbed state is the state of rest where $\mathbf{v} = 0$, $p = \mathrm{const}$, $\rho = 0$, but $\mathbf{E} = \mathbf{E}_0 \neq 0$.

The theory of hydrodynamic fluctuations [5] reduces to (1) and the general equation of heat transport, where the variables are regarded as small fluctuations and local random stresses and heat fluxes are added to the equations. It is then not difficult to obtain directly equations for the correlation functions [6]. Here we will assume that the hydrodynamic fluctuations are generated mainly by fluctuations in the pondermotive force, so that we do not need to introduce additional random terms in (1). The spectral properties of the fluctuations are studied with the help of the correlation theory of stationary random processes [7]: any random function f of the coordinates $\mathbf{r}$ and time t is represented as a Fourier–Stieltjes integral with random measure dZ_f:

Urals State University, Sverdlovsk. Moscow Institute of Chemical Engineering. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 46, No. 3, pp. 398-405, March, 1984. Original article submitted December 7, 1982.

0022-0841/84/4603-0282$08.50

$$f(t,\mathbf{r}) = \int dZ_f e^{i\omega t + i\varkappa r}.$$

The Euler correlation function of two random functions f(t, **r**) and g(t, **r**) can then be represented in the form

$$\langle f(t,\ \mathbf{r})\, g(t+\tau,\ \mathbf{r}+\mathbf{x}) \rangle = \int d\omega \int d\varkappa e^{i\omega\tau + i\varkappa x} \Psi_{fg}(\omega,\ \varkappa),$$

$$\Psi_{f,g}(\omega,\ \varkappa)\, d\omega d\varkappa = \lim \langle\ dZ_f dZ_g^* \ \rangle\ (d\omega,\ d\varkappa \to 0),$$

where $\Psi_{f,g}$ is the appropriate spectral density, and the integration goes over the entire frequency axis ω and over all wave vectors $\varkappa$.

Putting $\rho\mathbf{E} \approx \rho'\mathbf{E}_0$ in (1) and using the above representation for the random variables **v**, p, and ρ, we obtain linear algebraic equations for the random measures. After elimination of the random measure of pressure fluctuations, we find

$$d\mathbf{Z}_v = \left[\mathbf{E}_0 - \frac{(\varkappa \mathbf{E}_0)\varkappa}{\varkappa^2} \right] \frac{dZ_\rho}{\gamma_0 (i\omega + \nu_0 \varkappa^2)}, \tag{2}$$

which couples the random velocity field of the liquid to fluctuations in the volume charge.

The dispersion tensor resulting from velocity fluctuations can be written as an integral with respect to time of the Lagrangian velocity correlation tensor with the help of the well-known Green–Kubo formula. The relation between the Euler and Lagrange correlation functions is one of the central problems in contemporary statistical mechanics, and it still has not been completely solved, although a new approach has recently been developed [8]. For small fluctuations in the first approximation, we can (in general) ignore the difference between the Euler and Lagrange correlation functions and write for the dispersion tensor

$$D_{ij} \approx \int_0^\infty \langle v_i(t,\ \mathbf{r})\, v_j(t+\tau,\ \mathbf{r}) \rangle d\tau = \int_0^\infty d\tau \int d\omega \int d\varkappa e^{i\omega\tau} \Psi_{vi,\ vj}(\omega,\ \varkappa) = \pi \int d\varkappa \Psi_{vi,\ vj}(0,\ \varkappa). \tag{3}$$

where the last equality in (3) was derived by interchanging the order of integration with respect to $d\omega$ and $d\tau$ and using the well-known Fourier integral representation of the delta function.

It follows from (2) that the dispersion induced by fluctuations is anisotropic, with the direction of the unperturbed electric field $\mathbf{E}_0$ being singled out. We choose one of the coordinate axes (the first) along $\mathbf{E}_0$, then D_{ij} is determined by calculating its eigenvalues D_1 and $D_2 = D_3$.

Model Systems. The properties of volume charge fluctuations depend significantly on the type of liquid under consideration. We consider two complementary cases: an ideal weak electrolyte and a weakly conducting liquid. In both cases (for simplicity) we ignore the effect of temperature fluctuations on the properties of the medium. In the case of the electrolyte, we assume that the electrical conductivity is due to a single dissociating impurity, and the valences of positive and negative ions are the same. We also assume that the dissociation is complete and we ignore fluctuations in the dielectric constant. In the case of the weakly conducting liquid, we assume that fluctuations in the dielectric constant and electrical conductivity result only from density fluctuations.

Electrolyte. Let the number concentration of impurity molecules be $n_0 + n'$ where n' gives the concentration fluctuations. The equations of electrodynamics needed in the analysis are (we take into account electroosmotic effects using the Nernst–Planck relation):

$$\mathbf{j} = \sigma\mathbf{E} - r\nabla n + \frac{1}{4\pi} \frac{\partial(\varepsilon\mathbf{E})}{\partial t} + \rho\mathbf{v},\ \operatorname{div}\mathbf{j} = 0,\ \rho = \frac{1}{4\pi}\operatorname{div}(\varepsilon\mathbf{E}), \tag{4}$$

and

$$\sigma = ez(u_1 + u_2)n = sn,\ r = kT(u_1 - u_2) \tag{5}$$

where subscripts 1 and 2 refer to positive and negative ions, respectively.

Using the assumptions discussed above, we linearize (4) and (5), represent the random functions $\mathbf{j}'(\mathbf{r},\ t)$, $\mathbf{E}'(\mathbf{r},\ t)$, $\rho(\mathbf{r},\ t)$ and $n'(\mathbf{r},\ t)$ as Fourier–Stieltjes integrals, and solve the resulting system of linear algebraic equations for the random measures. In particular, we have

$$dZ_\rho = -\frac{r\varkappa^2 + is(\varkappa \mathbf{E}_0)}{i\omega + \omega_*} dZ_n, \quad \omega_* = \frac{4\pi\sigma_0}{\varepsilon_0}, \tag{6}$$

determining dZ_V from (2) in terms of dZ_n.

Weakly Conducting Liquid. In this case (4) will still be correct but $r = 0$, $\sigma = \sigma(\gamma)$, and $\varepsilon = \varepsilon(\gamma)$. The analog to (6) in the linearized theory is

$$dZ_\rho = i\left(\frac{\omega_*}{4\pi}\frac{d\varepsilon}{d\gamma} - \frac{d\sigma}{d\gamma}\right)\frac{\varkappa \mathbf{E}_0}{i\omega + \omega_*} dZ_\gamma. \tag{7}$$

A dielectric medium is of particular interest. We can consider a dielectric to be a weakly conducting medium with vanishingly small conductivity. The results can then be obtained from those for a weakly conducting liquid by putting $d\sigma/d\gamma = 0$ and taking the limit $\omega_* \sim \sigma_0 \to 0$.

Thus, the spectral properties of volume charge fluctuations are determined by those of impurity concentration fluctuations (electrolyte) and density fluctuations (weakly conducting liquid).

Concentration and Density Fluctuations. It is known that fluctuations of the concentration n at different points of space and different times are not correlated in the hydrodynamic limit so that the correlation function is a delta function [6]. Therefore, these fluctuations can be considered as white noise with a spectral density independent of ω and $\varkappa$. However, this is not the case at distances comparable to the linear scale l_m of the internal structure of the medium, or for time intervals of the order of the lifetime τ_m of structural formations; in these cases n' exhibits strong correlations. It is convenient to introduce the "molecular" frequency ω_m and wave number $\varkappa_m$ such that

$$\Psi_{n,\,n}(\omega, \varkappa) = NY(\omega_m - \omega)\,Y(\varkappa_m - |\varkappa|), \tag{8}$$

where Y(x) is the Heaviside unit step function. Equation (8) is a reasonable approximation to the actual dependence of the spectral density on frequency and wave vector when the correlation distance is of order $l_m \sim \varkappa_m^{-1}$ and the correlation time is of order $\tau_m \sim \omega_m^{-1}$. This relation can be justified either with the help of a method related to the well-known Debye method of determining the number of independent harmonics in the Fourier transform of n'(**r**, t) in terms of the number of degrees of freedom of the system (where the integration over wave vector is replaced by a summation over the corresponding Brilluoin zone), or by the device of replacing the delta function in the expression for the correlation function by a function (distribution in the sense of Schwartz) smeared out in space or time. Formally, the situation is completely analogous to that in the spectral theory of concentration fluctuations in dispersive systems [9].

The constant N in (8) can be determined from the requirement that fluctuations in the number of impurity molecules n_V' in volume V satisfy the well-known relation

$$\langle n_V'^2 \rangle = Vn_0. \tag{9}$$

Choosing volume V to be a parallelepiped of lengths L_1, L_2, and L_3 oriented along the coordinate axes and using the Fourier–Stieltjes integral for n'(**r**, t), we obtain

$$n_V' = \int_V d\mathbf{r} \int dZ_n e^{i\omega t + i\varkappa r} = \int dZ_n e^{i\omega t} \prod_{j=1}^{3} \frac{\exp(i\varkappa_j L_j) - 1}{i\varkappa_j}.$$

From this and (8) it follows that

$$\langle n_V'^2 \rangle = 8\int \langle dZ_n dZ_n^* \rangle \prod_{j=1}^{3} \frac{1 - \cos(\varkappa_j L_j)}{\varkappa_j^2} = 8N \int_{-\omega_m}^{\omega_m} d\omega \int_{|\varkappa| < \varkappa_m} d\varkappa$$

$$\times \prod_{j=1}^{3} \frac{1 - \cos(\varkappa_j L_j)}{\varkappa_j^2} \approx 16\, N\omega_m \prod_{j=1}^{3} \lim_{y \to 0} \int_{-\infty}^{\infty} \frac{1 - \cos(\varkappa_j L_j)}{\varkappa_j^2 + y^2} d\varkappa_j = 16\, N\omega_m \prod_{j=1}^{3} \lim_{y \to 0} \frac{\pi}{y}(1 - e^{-yL_j}) = 16\pi^3 N\omega_m V\ (V = L_1 L_2 L_3).$$

Comparing this with (9) we have

$$N = n_0/16\pi^3\omega_m, \tag{10}$$

which then determines the spectral density (8).

Similarly, using in place of (9) the well-known formula for the mean square fluctuation of the number of molecules n_V' in volume V

$$\langle n_V'^2 \rangle = \frac{kT\beta}{v} V n_0, \quad v = \frac{1}{n_0}, \; \beta = -\frac{1}{V}\left(\frac{\partial V}{\partial p}\right)_T, \tag{11}$$

we obtain the following result for the spectral density of $\gamma'(\mathbf{r}, t)$:

$$\Psi_{\gamma,\,\gamma}(\omega, \varkappa) = \Gamma Y(\omega_m - \omega)\, Y(\varkappa_m - |\varkappa|), \quad \Gamma = kT\beta\gamma_0^2/16\,\pi^3\omega_m. \tag{12}$$

Dispersion Coefficients. With the help of the above results, one can easily determine the spectral densities of all random processes and calculate the corresponding correlation functions, including cross-correlations. Here we find only the dispersion coefficients, formally defined by (3).

Electrolyte. In this case from (2), (6) and (8), (10) we have

$$\Psi_{v1,\,v1}(0, \varkappa) = \frac{n_0 E_0^2}{16\,\pi^3 \mu_0^2 \omega_*^2 \omega_m}\left(r^2 + s^2 E_0^2 \frac{\varkappa_1^2}{\varkappa^4}\right)\left(1 - \frac{\varkappa_1^2}{\varkappa^2}\right)^2 Y(\varkappa_m - |\varkappa|),$$

$$\Psi_{v2,\,v2}(0, \varkappa) = \frac{n_0 E_0^2}{16\,\pi^3 \mu_0^2 \omega_*^2 \omega_m}\left(r^2 + s^2 E_0^2 \frac{\varkappa_1^2}{\varkappa^4}\right)\frac{\varkappa_1^2 \varkappa_2^2}{\varkappa^4} Y(\varkappa_m - |\varkappa|).$$

Integrating these expressions as indicated in (3) we get

$$D_1 = \frac{2 n_0 E_0^2 \varkappa_m}{15\,\pi \mu_0^2 \omega_*^2 \omega_m}\left(\frac{1}{7} s^2 E_0^2 + \frac{1}{3} r^2 \varkappa_m^2\right), \tag{13}$$

$$D_2 = \frac{2\, n_0 E_0^2 \varkappa_m}{15\,\pi \mu_0^2 \omega_*^2 \omega_m}\left(\frac{3}{56} s^2 E_0^2 + \frac{1}{24} r^2 \varkappa_m^2\right).$$

Using r, s, and ω_* from (5) and (6) we get for the longitudinal dispersion coefficient

$$D_1 = D_1' + D_1'', \quad D_1' = \frac{1}{840\,\pi^3} \frac{E_0^4}{\nu_0^2}\left(\frac{\varepsilon_0}{\gamma_0}\right)^2 \frac{\varkappa_m}{n_0 \omega_m},$$
$$D_1'' = \frac{1}{360\,\pi^3} \frac{E_0^2}{\nu_0^2}\left(\frac{kT}{ez}\right)^2 \left(\frac{u_1 - u_2}{u_1 + u_2}\right)^2 \left(\frac{\varepsilon_0}{\gamma_0}\right)^2 \frac{\varkappa_m^3}{n_0 \omega_m} \tag{14}$$

and an analogous expression for the transverse dispersion coefficient. The two terms in (14) arise from fluctuations in the conduction current and fluctuations in the diffusion current, respectively.

We estimate the magnitudes of the terms in (14). It is convenient to use Gaussian units. If the formation of associations in the system does not play an important role, then $\varkappa_m \sim n_0^{1/3}$ and $\omega_m \sim D_m \varkappa_m^2 \sim D_m n_0^{2/3}$ where D_m is the molecular diffusion coefficient for the impurity molecules. In Gaussian units, for an aqueous solution we have $\nu_0 \sim 10^{-2}$, $(\varepsilon_0/\gamma_0)^2 \sim 10^3$, $D_m \sim 10^{-6}$–10^{-5}. Putting $E_0 \sim 10^{-2}$ (30 kV/cm) for illustration, assuming that the mobilities of positive and negative ions are different and using the values $kT \sim 10^{-14}$, $e \sim 10^{-10}$, we obtain

$$D_1' \sim (10^{16} \div 10^{17})\, n_0^{-4/3}, \quad D_1'' \sim (10^4 \div 10^5)\, n_0^{-2/3}.$$

When $n_0 \sim 10^{15}$ we have $D_1' \sim 10^{-4}$–10^{-3} and $D_1'' \sim 10^{-6}$–10^{-5}, which is comparable to or larger than the typical value of the molecular diffusion coefficient in a liquid (order 10^{-6}–10^{-5}). For small electrolyte concentrations, fluctuations in the conduction current are most important. With an increase in the concentration, the fluctuations in the diffusion current become more important. The two types of fluctuation are equally important for $n_0 \sim 10^{18}$.

It is obvious that the above relations are not valid for very weak electrolytes since we have ignored the electrical conductivity of the solvent. In addition, errors arise from ignoring fluctuations in the dielectric constant, the effect of temperature fluctuations, and so on.

Weakly Conducting Liquid. From (2), (7), and (12) we have in place of (13):

$$\Psi_{v1,\,v1}(0, \varkappa) = \frac{kT\beta E_0^4}{16\,\pi^3 \nu_0^2 \omega_*^2\, \omega_m}\left(\frac{\omega_*}{4\,\pi}\frac{d\varepsilon}{d\gamma} - \frac{d\sigma}{d\gamma}\right)^2 \frac{\varkappa_1^2}{\varkappa^4}\left(1 - \frac{\varkappa_1^2}{\varkappa^2}\right)^2 Y(\varkappa_m - |\varkappa|),$$

$$\Psi_{v2,\,v2}(0,\,\varkappa) = \frac{kT\beta E_0^4}{16\,\pi^3 v_0^2 \omega_*^2 \omega_m} \left(\frac{\omega_*}{4\,\pi} \frac{d\varepsilon}{d\gamma} - \frac{d\sigma}{d\gamma} \right)^2 \frac{\varkappa_1^4 \varkappa_2^2}{\varkappa^8} Y(\varkappa_m - |\varkappa|),$$

and the formulas for the dispersion coefficients replacing (14) become:

$$D_1 = \frac{2\,kT\beta E_0^4 \varkappa_m}{105\,\pi v_0^2 \omega_*^2 \omega_m} \left(\frac{\omega_*}{4\,\pi} \frac{d\varepsilon}{d\gamma} - \frac{d\sigma}{d\gamma} \right)^2,$$

$$D_2 = \frac{kT\beta E_0^4 \varkappa_m}{140\,\pi v_0^2 \omega_*^2 \omega_m} \left(\frac{\omega_*}{4\,\pi} \frac{d\varepsilon}{d\gamma} - \frac{d\sigma}{d\gamma} \right)^2. \tag{15}$$

A dielectric medium ($\omega_* \sim \sigma_0 \to 0$) is of particular interest. For the longitudinal dispersion coefficient, we have

$$D_1 = \frac{kT\beta E_0^4 \varkappa_m}{840\,\pi^3 v_0^2 \omega_m} \left(\frac{d\varepsilon}{d\gamma} \right)^2 \tag{16}$$

and the formula for the transverse dispersion coefficient is similar.

We put $\beta \sim 10^{-10}$, $E_0^4/v_0^2 \sim 10^{12}$, $(d\varepsilon/d\gamma)^2 \sim 10^2$, where as before we use Gaussian units. Then we have from (16) $D_1 \sim 10^{-14}\varkappa_m/\omega_m$. For fluctuations on the molecular level, as in the case of the electrolyte considered above, the correlation length is of the order of the size of the molecule and the lifetime of the fluctuations is of the order of the characteristic period of molecular vibration. In this case $\varkappa_m \sim l_m^{-1} \sim 10^7$-$10^8$, $\omega_m \sim \tau_m^{-1} \sim 10^8$-$10^{12}$ and D_1 is several orders of magnitude less than the typical value of the molecular diffusion coefficient. But this conclusion is completely changed if there is a supermolecular structure in the liquid (swarms or cybotactic groups) [10]. In both cases $\varkappa_m \sim l_m^{-1}$ and l_m is the characteristic size of the groups; ω_m can then depend on their nature.

For athermal swarms (thermodynamically stable elements of an ordered phase (e.g., crystal) with a purely kinetic origin [10]) we have $\omega_m \sim D_m\varkappa_m^2$, where D_m is the effective coefficient of Brownian diffusion of a single swarm. This can be estimated using the Einstein formula which takes into account the barrier to diffusion in a concentrated swarm: $D_m \sim \zeta kT/6\pi\mu_0 l_m \sim 10^{-14} l_m^{-1}$, where the constraint factor is taken to be $\varsigma \sim 10^{-1}$. Thus, $\varkappa_m/\omega_m \sim 10^{14} l_m^2$, $D_1 \sim l_m^2$, and the dispersion induced by fluctuations becomes comparable to molecular diffusion for $l_m \gtrsim 10^{-3}$ cm; the corresponding inequality on the lifetime has the form $\tau_m \gtrsim 10^{-5}$ sec.

For cybotactic groups (thermal associations which appear as a result of large-scale fluctuations, the orienting effect of external fields on the molecules, etc. [10]), D_m is a molecular diffusion coefficient inside each separate group. It should be approximately equal to the value typical for molecular diffusion in a solid. Using the estimate $D_m \sim 10^{-12}$, we obtain $D_1 \sim 10^{-2} l_m$ and the effect of dispersion is comparable to molecular diffusion for $l_m \gtrsim 10^{-4}$ cm or $\tau_m \gtrsim 10^{-4}$ sec.

The above estimates are very rough and overall too low; this is because we have ignored temperature fluctuations and completely ignored the effect of the applied field on the structure and consequently on the properties of the liquid itself. But even without considering the structuring effect of the field, it is clear that fluctuation effects can be very important in the case of a weakly conducting liquid near a transition into a liquid crystal phase, or a liquid near the melting point.

The general conclusion is that dispersion induced fluctuations can significantly increase the effective diffusion coefficient in widely varying types of liquids. The same conclusion can be made concerning the effective thermal conductivity. Although the origin of the effect is extremely weak thermodynamic fluctuations, it is no less real than other well-known phenomena due to fluctuations: Brownian motion, light scattering by density fluctuations, anomalous behavior of the thermodynamic quantities near a critical point.

The analysis given above shows that dispersion due to fluctuations can help to explain the observed dependences of the effective heat and mass exchange coefficients on the physical properties of the liquid and the regime parameters of heat and mass exchange, including effects which are difficult to understand only on the basis of the classical explanations. A detailed discussion of these effects is outside the scope of this paper. Here we only mention that an important example is various hysteresis phenomena [1, 2] which are obviously related to hysteresis of the internal structure of the liquid.

NOTATION

D, diffusion coefficient; D_{ij}, D_j, dispersion tensor and its eigenvalues, respectively; **E**, electric field strength; e, charge of the electron; k, Boltzmann constant; l_m, linear scale of the internal structure; L_j, linear dimension; n, number concentration of molecules; N, constant defined in (8); p, pressure; r, s, coefficients defined in (5); **r**, radius vector; t, time; T, temperature; u_j, mobility of ions; **v**, velocity; V, volume; z, ion valence; dZ, random measure; β, isothermal compressibility; γ, density; Γ, constant in (12); ε, dielectric constant; ζ, constraint factor; $\boldsymbol{\varkappa}$, wave vector; $\varkappa_m$, maximum wave number; μ, ν, dynamic and kinematic viscosities; ρ, volume charge density; σ, electrical conductivity; τ_m, lifetime of a fluctuation; Ψ, spectral density; ω, frequency; ω_m, maximum frequency; ω_*, parameter defined in (6); subscript 0, state without fluctuations, subscripts 1 and 2, positive and negative ions, and also the longitudinal and transverse directions; a prime denotes a fluctuation, and an asterisk denotes the complex conjugate.

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