Fluctuation-dissipation theory of nonlinear viscosity

G. N. Bochkov and Yu. E. Kuzovlev

N. I. Lobachevskiĭ Gorky State University
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Fluctuation-dissipation relations are considered for thermal perturbations (that is, perturbations of the distribution function) in nonlinear nonequilibrium systems. A general expression for the irreversible fluxes is obtained in terms of the fluctuational characteristics. In the application to hydrodynamics, it is shown that the fluctuational stress tensor is non-Gaussian, and that in consequence of this the dependence of the viscous shearing stress on the velocity gradient is nonlinear and nonanalytic. The change of this nonanalyticity at the critical point is considered, and relations (following from the fluctuation-dissipation relations) are found between the kinetic scale exponents. These examples illustrate the general relation between the form of the dissipative nonlinearity and the character of the statistics of the transfer process. By use of a variational principle, nonlinear interaction of slow viscous modes in two-dimensional hydrodynamics is treated, and the nonanalytic dispersion law is found.

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1. INTRODUCTION

In our previous paper,[1,2] we studied the exact fluctuation-dissipation relations (FDR) that relate to each other the nonlinear dissipative characteristics of a nonequilibrium system and the non-Gaussian statistical characteristics of the fluctuations. The latter, as was shown, uniquely determine the dependence of the macroscopic irreversible fluxes on the forces thermodynamically conjugate to them.[2] The converse problem—recovery of the fluctuations on the basis of phenomenological data—has no unique formal solution. But in practice, there is always concrete information about the system, which makes it possible to make more-or-less justified qualitative assumptions about the type of statistics of the transfer process. In such a case, the FDR enable us to find the quantitative characteristics of the fluctuations. On the other hand, if we know the statistics of the transfer structure, we can predict the form of the dissipative nonlinearity.

The present paper is devoted to an analysis, in a concrete example, of the hydrodynamics of these aspects of the application of FDR. Below, we justify and treat a simple non-Gaussian model of the fluctuational stress tensor in a (d-dimensional) dense gas, and we show that it leads to a nonlinear character of the shear viscosity; more accurately, to a definite nonlinear and nonanalytic dependence of the macroscopic stress tensor on the velocity gradient. This dependence is due to the contribution of long-wavelength viscous (and thermal) hydrodynamic modes to the fluctuational stress tensor.

The result obtained in Sec. 3 agrees qualitatively (and even quantitatively) with the result of the microscopic kinetic theory of Ernst *et al.*[3] (see also Refs. 4 and 5). In Ref. 3, a very complicated analysis is carried out a broken BBGKY (Bogolyubov-Born-Green-Kirkwood-Yvon) chain for a nonequilibrium dense gas of hard spheres (for $d=2,3$). But the results of calculations relating to hydrodynamics cannot depend on the details of the microscopic model and should in principle be obtainable more simply in a stochastic model for macrovariables. And in fact, their derivation (for arbitrary d) from the FDR reduces to the calculation of known integrals. Here it is sufficient to consider equilibrium fluctuations.

In the case $d=3$, the nonlinear corrections to the shear viscosity are relatively small. But the method presented is applicable also to models of continuous media under conditions when the nonlinear effects may be substantial and decisive (for example, in critical states), and to processes of transfer not only of momentum but also of other quantities. As an example, in Sec. 3 we consider the nonanalytic behavior of the shear viscosity at the critical point (here the nonlinearity dominates), and we find relations between the kinetic scale exponents that follow from the FDR.

We note that the FDR[1] relate to dynamic perturbations. But here we have to do with thermal or statistical perturbations; that is, perturbations of the distribution function. These perturbations, of course, are described by thermal forces, which are introduced as parameters of a quasiequilibrium distribution (more accurately, of a nonequilibrium distribution that is the result of the evolution of a quasiequilibrium distribution over a time large on a microscopic and small on a macroscopic scale). In Sec. 2, by the method developed earlier,[6] the nonlinear FDR are obtained for thermal perturbations, and it is shown that they coincide formally with the relations of Ref. 1. A compact canonical form is considered for the transfer equations, which express the fluxes in terms of the thermal forces, and for the equations of evolution of the macrovariables.

The structure that follows from the FDR for the nonlinear evolution equations is such that the latter can be derived by a variational principle, which generalizes the principle of minimum entropy production known in linear nonequilibrium thermodynamics. A statistical version of this principle (for dynamic perturbations) was given in Ref. 2. The variational principle formulated in Sec. 4 (which is formally similar to Hamilton's principle in mechanics) is then applied to the derivation of an approximate dispersion law for interacting viscous modes in two-dimensional hydrodynamics.

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2. THE FDR FOR THERMAL PERTURBATIONS, AND THE NONLINEAR EQUATIONS OF EVOLUTION

1. Let a system with Hamiltonian $\hat{H}_0$ (containing a thermostat at temperature $T\equiv\beta^{-1}$), at a certain instant of time $t=0$, be in a state with the quasiequilibrium density matrix

$$\rho_q(x)=\exp\{\ln\rho_0-x_\alpha\hat{Q}_\alpha+\theta(x)\},\quad \mathrm{Sp}\,\rho_q=1, \tag{1}$$

where $\rho_0\sim\exp(-\beta\hat{H}_0)$ is the equilibrium distribution, and where x_α are the thermal forces conjugate to the macrovariables $\hat{Q}_\alpha$ (the set $\hat{Q}_\alpha$ may also include $\hat{H}_0$). As is well known, the distribution (1) corresponds to the maximum informational entropy for prescribed means $Q_\alpha=\langle\hat{Q}_\alpha\rangle=\mathrm{Sp}(\rho\hat{Q}_\alpha)$. Over a characteristic time τ_μ (the scale of the kinetic stage of relaxation), much smaller than the time τ_m of relaxation of the macrovariables Q_α to equilibrium (the scale of the hydrodynamic stage), there is established instead of (1) a nonequilibrium distribution $\rho(x)$ with the same microscopic entropy and the same controlling parameters x_α as in (1). In the state with this new distribution (the Mori distribution[7]), there are, in contrast to $\rho_q(x)$, nonzero dissipative fluxes

$$I_\alpha\equiv\dot{Q}_\alpha=Y_\alpha(Q)\equiv\mathrm{Sp}\left\{\rho(x)\frac{i}{\hbar}[\hat{H}_0,\hat{Q}_\alpha]\right\}. \tag{2}$$

Because of the inequality $\tau_\mu\ll\tau_m$, the means Q_α in the states $\rho_q(x)$ and $\rho(x)$ coincide and are related to the thermal forces by the formulas of the quasiequilibrium distribution:

$$Q_\alpha=\frac{\partial}{\partial x_\alpha}\theta(x);$$
$$x_\alpha=\frac{\partial}{\partial Q_\alpha}S(Q),\quad S(Q)\equiv x_\alpha Q_\alpha-\theta(x). \tag{3}$$

We shall call the function $S(Q)$ the macroentropy. Its specific meaning may change from problem to problem. It is easy to show that $S(Q)\leqslant 0$; the inequality is possible only in equilibrium with $x_\alpha=0$.

If the set Q_α is sufficient for a closed reduced description, for $t>\tau_\mu$ the density matrix $\rho(x)$ should preserve its form, depending on the time only through the values of the forces at the time. Then (2) and (3) form a closed system of Markov (that is, of first order in the time) equations of evolution.

2. We shall consider the universal properties of the flux functions $Y_\alpha(Q)$, which follow from their relation to the characteristics of the fluctuations (and ultimately from the conservation of the microscopic phase volume). We introduce the characteristic functional of the random fluxes:

$$D(u;x)\equiv\frac{1}{\tau}\ln\mathrm{Sp}\exp\left\{\ln\rho(x)+u_\alpha\int_0^\tau\hat{I}_\alpha(t)\,dt\right\}\equiv u_\alpha Y_\alpha(Q)+\sum_{n=2}^{\infty}\frac{u^n}{n!}D_n(x). \tag{4}$$

Here the interval τ is so chosen that $\tau_\mu\ll\tau\ll\tau_m$. Under this condition, (4) is independent of τ. Furthermore, under this condition $\rho(x)$ in (4) may obviously be replaced by $\rho_q(x)$. By using the method of Ref. 6 (with ρ_q as initial distribution instead of the equilibrium distribution ρ_0), we find the following generating FDR[1)]:

$$D(u-x;\,x)=D(-\varepsilon u;\,\varepsilon x). \tag{5}$$

Here $\varepsilon_\alpha=\pm1$, depending on the temporal evenness or oddness of x_α. This is a generalization of the results of Ref. 1 to thermal perturbations.

If the system is also subject to external dynamic forces $y_\alpha\,(\hat{H}_0\to\hat{H}_0-y_\alpha\hat{Q}_\alpha)$, then instead of (5) one can obtain the formula

$$D(u-x-\beta y;\,x;\,y)=D(-\varepsilon u;\,\varepsilon x;\,\varepsilon y),$$

in which both forms of perturbation figure equally.

Further, we introduce the local nonlinear transfer coefficients $\lambda_n(Q)\equiv\frac{1}{2}(-1)^nD_n$ and the kinetic potential[2]

$$F(x;Q)\equiv x_\alpha Y_\alpha^\circ(Q)+\sum_{n=2}^{\infty}\frac{\lambda_n(Q)}{n!}x^n;$$
$$Y_\alpha^\circ(Q)\equiv{}^1/_2[Y_\alpha(Q)-\varepsilon_\alpha Y_\alpha(\varepsilon Q)],\quad Y_\alpha^{\mathrm{r}}(Q)\equiv Y_\alpha(Q)-Y_\alpha^\circ(Q), \tag{6}$$

where Y_α^r and Y_α^i are the reversible and irreversible components of the fluxes. From (5), with use of (3), we get the general expression for the irreversible fluxes in terms of the fluctuational characteristics, the diffusion coefficients D_n (Ref. 2) or λ_n:

$$\dot{Q}_\alpha=Y_\alpha^\circ(Q)+\lambda_{\alpha\beta}(Q)x_\beta+\frac{1}{2!}\lambda_{\alpha\beta\gamma}(Q)x_\beta x_\gamma+\ldots=\frac{\partial}{\partial x_\alpha}F(x;Q), \tag{7}$$
$$x_\alpha=\frac{\partial}{\partial Q_\alpha}S(Q).$$

These equations give a canonical representation of the nonlinear equations of evolution. Although in the nonlinear range reciprocity relations are not satisfied, the symmetry of the tensors λ_n (Refs. 1 and 2) leads, as is evident from (7), to a definite relation between cross processes of transfer. With respect, however, to "virtual" variations of the forces x_α that are independent of the Q_α, reciprocity relations always hold. One must note such consequences of the FDR (5) as the convexity of the kinetic potential with respect to the x_α and the relations for the entropy production[2]:

$$\mathscr{P}(Q)\equiv\dot{S}(Q)=\frac{\partial S}{\partial Q_\alpha}\dot{Q}_\alpha=2F\left(\frac{\partial S}{\partial Q};Q\right)\geqslant 0. \tag{8}$$

Hence it is evident that the macroentropy serves as the Lyapunov function for equations (7).

3. It is inconvenient to deal with an infinite number of fluctuational parameters λ_n. Furthermore, the expansion (7) may not exist (as will be demonstrated in the next section). Therefore we shall use an integral representation (of the logarithm) of the characteristic functional (4):

$$D(iu;x)=iu_\alpha Y_\alpha(Q)+2\int[\exp(iu_\alpha q_\alpha)-1-iu_\alpha q_\alpha]R(q;Q)\,dq. \tag{9}$$

From (5), (7), and (9) we obtain the FDR

$$Y_\alpha^i(Q)=\int q_\alpha[1-\exp(-q_\gamma x_\gamma)]R(q;Q)\,dq,\quad x=\partial S/\partial Q, \tag{10}$$

$$R(-\varepsilon q;\varepsilon Q)=\exp\left\{-q_\gamma\frac{\partial S(Q)}{\partial Q_\gamma}\right\}R(q;Q). \tag{11}$$

In the Markov approximation under consideration (on a macroscopic time scale), $\exp[\tau D(iu;x)]$ is the characteristic function of the infinitely divisible distribution, so that (9) coincides with the Lévy-Khintchine representation known in the theory of Markov processes.[8] In this representation, the kernel R is nonnegative and determines the frequency of jumps of the amplitude q_α. The probabilistic interpretation of the function R is simple: it is proportional to the power of the Langevin noise sources that occur in the stochastic version of equations (7).

Formula (11) determines the unbalance that occurs, in a departure from equilibrium, between elementary transfer processes of opposite sign. It is natural to introduce, as a general "symmetric" measure of nonequilibrium noise, the function

$$R'(q;\ Q)={}^1/_2(1+e^{-qx})R(q;\ Q).$$

The relations (10) and (11) take the form

$$Y_\alpha{}^i=2\int q_\alpha \operatorname{th}({}^1/_2\, qx)R'(q;Q)\,dq,\quad R'(-\varepsilon q;\ \varepsilon Q)=R'(q;Q). \tag{12}$$

In many cases it may be assumed that the dependence of the intensity R' of the noise sources on the macrostate is slight and smooth, and that the character of the dissipative nonlinearity is determined predominantly by the dependence of R' on q. Then at least over a limited range of values of Q, we may approximately set

$$R'(q;\ Q)=R_0(q)=R|_{x=0}. \tag{13}$$

Formulas (12) and (13) relate the characteristic functional of the equilibrium fluctuations to the irreversible fluxes.

3. A NON-GAUSSIAN MODEL OF THE FLUCTUATIONAL STRESS TENSOR, AND ITS CONSEQUENCES

1. The results of Sec. 2 are easily translated into the language of distributed systems. Thus equations (7) are rewritten in the form

$$\dot{Q}_\alpha=\sigma_\alpha(Q,r)-\nabla_\mu I_{\alpha\mu}(Q,r)=\left\{\frac{\delta}{\delta x_\alpha(r)}-\nabla_\mu\frac{\delta}{\delta\nabla_\mu x_\alpha(r)}\right\}F(\nabla x;x;Q),Q=Q(t,r). \tag{14}$$

Here r is a spatial coordinate. The thermal forces x_α and ∇x_α act as independent variables, since they are related to physically different transfer processes—local and spatial.

Further, let $Q_\alpha(r)$ denote the momentum-density vector. Then

$$\sigma_\alpha=0,\quad I_{\alpha\mu}=I_{\alpha\mu}{}^r+I_{\alpha\mu}{}^i,\quad I_{\alpha\mu}{}^r=\rho^{-1}Q_\alpha Q_\mu+p\delta_{\alpha\mu},$$

where ρ is the density, p the pressure, and $I^i_{\alpha\mu}$ the viscous-stress tensor. The thermal force conjugate to the flux $I^i_{\alpha\mu}$ is

$$z_{\alpha\mu}=\nabla_\mu x_\alpha=-\nabla_\mu\beta\rho^{-1}Q_\alpha.$$

We denote by $\dot{J}_{\alpha\mu}(t,r)$ the fluctuational stress tensor (FST). We also introduce the notation

$$g_{\alpha\mu}=\int_0^\tau dt'\int_V dr'\, J_{\alpha\mu}(t+t',r+r'), \tag{15}$$

where the volume V is much larger than the correlation volume of the FST. On expressing the kinetic potential in a standard manner, on the basis of formulas (5) and (6), in terms of the correlators of the FST, we get from (7), (14), and (15) the spatially local transfer equations[2)]

$$I_{\alpha\mu}{}^i=\frac{1}{2\tau V}\left\{\langle g_{\alpha\mu}, g_{\alpha'\mu'}\rangle z_{\alpha'\mu'}-\frac{1}{2!}\langle g_{\alpha\mu}, g_{\alpha'\mu'}, g_{\alpha''\mu''}\rangle z_{\alpha'\mu'}z_{\alpha''\mu''}+\dots\right\}. \tag{16}$$

2. On assuming that the FST is Gaussian and δ-correlated in space, we get from (16) and (14), with allowance for the isotropy of the medium, the usual linear Navier-Stokes viscous terms. A corresponding theory of hydrodynamic fluctuations was constructed by Landau and Lifshitz.[9] In principle, however, the FST has a finite correlation radius, since it contains a contribution from collective thermal motions of the particles.

The fluctuational stress tensor is also non-Gaussian. In fact, for its "kinetic" part $\dot{J}^k_{\alpha\mu}$ we may write

$$J_{\alpha\mu}{}^k(t,r)=\sum_i\frac{p_{i\alpha}p_{i\mu}}{m_i}\delta(r_i(t)-r),$$

$$g_{\alpha\mu}{}^k=\sum_V\int_0^\tau m_i{}^{-1}p_{i\alpha}(t)p_{i\mu}(t)\,dt,$$

where p_i, r_i, and m_i are the momenta, coordinates, and masses of the particles. The bilinear combination of normal random variables $p_{i\alpha}$ is non-Gaussian. Of course on averaging over a (physically small) volume V this non-Gaussianness diminishes by virtue of the central limit theorem; but that part of it remains that is due to long-range correlations of the particle momenta (collective motions).

The contribution of density fluctuations to $g^k_{\alpha\mu}$ may be neglected, since for the relative fluctuations of the number of particles N within a region of size L we have

$$\langle\delta N^2\rangle/N^2\sim l/LN,$$

where l is the length of the free path. Furthermore, we may neglect the potential part $\dot{J}^p_{\alpha\mu}$ of the FST. It is easy to show that

$$g_{\alpha\mu}{}^p\approx\sum_k\Delta p_{k\alpha}\Delta r_{k\mu},\quad\langle(g_{\alpha\mu}{}^p)^2\rangle\approx mTr_0{}^2n^2\sigma v_T\tau V, \tag{17}$$

where the sum is over all collisions of particles within a four-dimensional volume τV; Δp_k and Δr_k are the changes of momentum and of coordinate during a collision; r_0 is a characteristic radius of interaction, n is the mean number density of the particles, σ is the scattering cross section, and v_T is the thermal velocity. At the same time

$$\langle(g_{\alpha\mu}{}^k)^2\rangle\approx T^2nlv_T{}^{-1}\tau V,\quad l\approx1/n\sigma. \tag{18}$$

The ratio of the two contributions (17) and (18) to the kinetic coefficients in (16) is equal in order of magnitude to $(n\sigma r_0)^2\approx(\pi r_0^3 n)^2$ and is small even in a dense gas.

The indicated considerations lead to this simple model of the equilibrium FST:

$$J_{\alpha\mu}(t,\ r)=\rho v_\alpha(t,\ r)v_\mu(t,\ r), \tag{19}$$

where $v(t,r)$ is a Gaussian random hydrodynamic velocity field.

3. We shall consider the consequences of the model (19).[3)] The characteristic functional of the FST (19) can be calculated exactly if one prescribes the correlator

$$K_{\alpha\mu}(t,\ r)=\langle v_\alpha(t,\ r)v_\mu(0,\ 0)\rangle$$

or the corresponding spectral density $K_{\alpha\mu}(\omega,k)$. But for simplicity we neglect 1) the contribution to $K_{\alpha\mu}$ from the acoustic modes and 2) from the thermal modes and the effects of incompressibility. The first, as can be shown, does not change the final results; the second changes them by a coefficient of order unity. The corresponding simplified correlator is

$$K_{\alpha\mu}=\delta_{\alpha\mu}K, \quad K(\omega;k)=\frac{2T}{\rho}\frac{\nu k^2}{\omega^2+(\nu k^2)^2}. \tag{20}$$

Being interested only in the shear viscosity, i.e., in the relation between I^i_{12} and

$$z_{12}=-\beta\rho^{-1}\nabla_2 Q_1=-\beta\nabla v,$$

we shall calculate the equilibrium characteristic functional (for arbitrary dimensionality d):

$$D(iu;0)=\lim_{\tau V\to\infty}(\tau V)^{-1}\ln\left\langle \exp\left\{iu\int_0^\tau dt\int_V dr\,\rho v_1(t,r)v_2(t,r)\right\}\right\rangle$$
$$=-\frac{1}{2}(2\pi)^{-(1+d)}\int \ln\{1+u^2\rho^2K^2(\omega,k)\}\,d\omega\,dk. \tag{21}$$

The integral with respect to $|k|$ must be cut off from above at some limit $\varkappa_0$. For this purpose we introduce a factor $f(k)=\exp(-k^2/2\varkappa_0^2)$. On reducing (21) to the standard form (9), we get after substitution of (20):

$$R_0(q)=\frac{1}{4}(2\pi)^{-(d+1)}|q|^{-1}\int \exp\left\{-\frac{|q|}{\rho K(\omega,k)}\right\}f(k)\,d\omega\,dk$$
$$=A_d|q|^{-1/2}(|q|+q_0)^{-(d+1)/2},\quad A_d=\frac{T}{\pi^{1/2}\Gamma(d/2)}\left(\frac{T}{2\pi\nu}\right)^{d/2}\Gamma\left(\frac{d+1}{2}\right), \tag{22}$$
$$q_0=T/\nu\varkappa_0^2.$$

We further use the approximation (13). From (12), (13), and (22) we find for the stress tensor the expression

$$I_{12}{}^i=4A_d\int_0^\infty \frac{\operatorname{th}(\frac{1}{2}qz_{12})}{q^{1/2}(q+q_0)^{(d+1)/2}}\,dq. \tag{23}$$

Hence it is evident that the long-wavelength components of the FST [to which correspond the large values of $|q|$ in (22) and (23)] lead to a nonanalytic dependence of the shear stresses on the velocity gradient. For $d=3$, we find from (23) the expansion

$$I_{12}{}^i=4A_3\left\{\frac{\pi}{4\sqrt{q_0}}z_{12}-\left(\frac{z_{12}}{2}\right)^{3/2}C+\ldots\right\},\quad C=\int_0^\infty (y-\operatorname{th}y)\,y^{-5/2}\,dy\approx 1.55, \tag{24}$$

where higher-order terms have been omitted.

The first term must have the form $-\rho\nu\nabla v=\rho T\nu z_{12}$, where ν is the "linear" viscosity that occurs in (20). From this condition, we choose $\varkappa_0$. Then the ratio of the nonlinear and linear terms in (24) will be $\approx 0.1(\nabla v)^{1/2}T\rho^{-1}\nu^{-5/2}$. The calculations in Ref. 3, based on an approximate solution of an abbreviated BBGKY chain, give a ratio about 1.7 times as large. By removing the approximations that were made, one can obtain better numerical agreement.

In the two-dimensional case, the first term of the expansion (23), the largest at small z_{12}, is already singular,

$$I_{12}{}^i=-\frac{T^2}{2\pi\nu}z_{12}\ln z_{12}+\ldots,$$

so that the linear approximation is incorrect. In general, for even dimensionality $d=2m$ the first singular term has the form

$$\sim(-1)^m z_{12}{}^m\ln z_{12},$$

and for odd, $d=2m+1$,

$$\sim(-1)^m z_{12}^{m+1/2}.$$

These terms are independent of the cutoff parameter $\varkappa_0$.

It is easy to understand that the nonanalyticity under consideration is due to the limiting transition $V\to\infty$ in (21) and to the contribution of arbitrarily slow modes. In actuality, the kinetic coefficients are determined by correlators within a finite region, and the relation (23) is smoothed off in some manner to zero.[4)] What is most important in this example is the existence of a very intimate connection between the form of the dissipative nonlinearity and the correlator of the fluctuational flux field.

As a second example, we shall consider kinetic fluctuations, applying to them the same FST model.

4. In the vicinity of the critical point the transport coefficients may diverge-according to the scaling hypothesis, by a power law. Thus it is known[10] that for $d=3$ the shear viscosity diverges weakly:

$$\nu\sim\chi^{-\varphi},$$

where χ is the reciprocal correlation radius, and where the index $\varphi>0$ is close to zero. Consequently, for $\chi\to 0$ the dispersion law $\omega=\nu k^2$ and the linear (for a slight deviation from equilibrium) relation $I_{12}\sim\nu z_{12}=-\beta\nu\nabla v$ become incorrect. At the critical point, again according to scaling, we can write:

$$\omega\sim|k|^y,\quad I_{12}{}^i\sim z_{12}{}^\gamma, \tag{25}$$

We consider the relations between the indices φ, y, and γ that follow from the FDR.

Since the velocity field is not an order parameter, we must suppose that its one-time correlators have no singularities in the critical region and preserve the form

$$\langle v_\alpha(t,r)v_\alpha(t,0)\rangle\sim\delta(r).$$

From this and from (25) it follows that the spectral density has the form

$$K(\omega,k)=\omega^{-1}G(|k|^y/\omega).$$

Substituting this expression in (22) and taking into account that at the critical point the cutoff factor cannot be significant (that is, we may set $f\equiv 1$), we get

$$R_0(q)\sim|q|^{-2-d/y}\quad (d=3). \tag{26}$$

For convergence of the integral in (9), the inequality $d/y<1$ must be satisfied; therefore $y>3$.

Furthermore, we find from (12) and (13)

$$I_{12}{}^i\sim z_{12}^{3/y},\quad \gamma=3/y<1. \tag{27}$$

We note that the function (9) with the kernel (26), of power form, is the logarithm of the characteristic function of a stable distribution,[8] which is invariant with respect to convolution; the latter leads only to a scale transformation of it.

We consider the vicinity of the critical point. Here the spectral density and the kernel R_0 have the form

$$K(\omega,k)=\chi^{-y}G(\omega/\chi^y;\,k/\chi);$$
$$R_0(q)=\tfrac{1}{2}|q|^{-1}\chi^{d+y}B(|q|\chi^y;\,\varkappa_0/\chi). \tag{28}$$

Substituting (28) in (12), we get in the linear approximation

$$I_{12}{}^{l}=z_{12}\chi^{d-y}\int_0^\infty qB(q;\varkappa_0/\chi)\,dq. \qquad (29)$$

The coefficient of z_{12} is by definition proportional to $\chi^{-\varphi}$. From this and from (27) follow the inequalities

$$\varphi\geqslant y-3=\frac{3}{\gamma}(1-\gamma),\quad y\leqslant 3+\varphi,\quad \gamma\geqslant\frac{3}{3+\varphi}. \qquad (30)$$

But the behavior of the integrand in (29) at large q must not depend on the cutoff parameter. Therefore the equalities must evidently hold in (30).

Thus the exponent of the critical dispersion law and the index of divergence of the linear viscosity are connected by the relation

$$y=3+\varphi.$$

This result of our fluctuation-dissipation theory is substantiated by known experimental data. Since the index φ is small (calculations by the theory of interacting Kawasaki modes and by the method of the renormalization group give, respectively, $\varphi\approx 0.054$ and $\varphi\approx 0.084$[11]), the index y must be close to three. In fact, measurements of the width of the Rayleigh scattering line at the critical point give an approximately cubic dispersion law.[10]

The index γ in the critical law of viscous friction is, according to (30), close to unity (from the estimates for φ, we have $\gamma\approx 0.975$). As far as we know, this index has not been considered in the literature.

We note in conclusion that the results of this section have a primarily qualitative relation to the non-Gaussian property of the FST, and that the specific model form (19) is of little importance.

4. VARIATIONAL PRINCIPLE FOR NONLINEAR RELAXATION

1. We shall show that the nonlinear evolution equation (7) in the most general case can be obtained from a simple variational principle (see also the Appendix).

We introduce the "Hamiltonian" $H(x;Q)$ of the system:

$$H(x;Q)=F(x;Q)-F(\partial S/\partial Q;Q).$$

We introduce further the "Lagrangian" (the motivation for these terms will be explained below) $\Lambda(I;Q)$ as a Legendre transformation:

$$\Lambda(I;Q)=x_\alpha I_\alpha-H(x,Q),\qquad I_\alpha=\frac{\partial}{\partial x_\alpha}H(x;Q)=\frac{\partial}{\partial x_\alpha}F(x;Q).$$

We shall prove the inequality

$$\Lambda(I;Q)-I_\alpha\frac{\partial S}{\partial Q_\alpha}\geqslant 0. \qquad (31)$$

We rewrite this expression in the equivalent form

$$\left(x_\alpha-\frac{\partial S}{\partial Q_\alpha}\right)\frac{\partial}{\partial x_\alpha}F(x;Q)-F(x;Q)+F\left(\frac{\partial S}{\partial Q};Q\right) \qquad (32)$$

and consider the condition for an extremum of it with respect to the x_α:

$$\left(x_\beta-\frac{\partial S}{\partial Q_\beta}\right)\frac{\partial^2}{\partial x_\alpha\,\partial x_\beta}F(x;Q)=0.$$

In view of the convexity of the kinetic potential[5]) with respect to the x_α,[2] this condition is satisfied only at the point $x_\alpha=x_\alpha(Q)$; at this point, the matrix of second derivatives is positive definite. Consequently this point is an absolute minimum. Since the function (32) vanishes there, the inequality (31) is proved. The virtual fluxes I_α at the extremum coincide with the right side of (7), and the Lagrangian Λ is equal to the entropy production $\mathscr{P}(Q)$.

From (31) follows the variational principle in the form

$$\int\{\Lambda(\dot Q;Q)-\dot S(Q)\}\,dt=\min(=0) \qquad (33)$$

or in the equivalent form

$$\delta A[Q]=\delta\int\Lambda(\dot Q;Q)\,dt=0, \qquad (34)$$

where Q and $\dot Q$ are varied independently, like the coordinates and velocities in mechanics. The formal resemblance of (34) to Hamilton's principle in mechanics is obvious. Here the role of canonical momenta is played by the virtual thermal forces x_α. The structure of the Lagrangian is such that the "Euler equations" reduce to equations (7) of the first order in the time, while "Hamilton's equations" for $\dot Q_\alpha$ and for $\dot x_\alpha$ duplicate each other.

We consider the trajectory $Q(t)$ on the semi-infinite interval from $t=0$ to $t=\infty$. In view of the stability of the motion expressed by the inequality (8), the value of $Q(\infty)$ is the equilibrium value and is independent of $Q(0)$; therefore for a fixed value of $Q(0)=Q$, the variational principle (33) can be reduced to the form

$$\int_0^\infty\Lambda(\dot Q;Q)\,dt=\min, \qquad (35)$$

which is convenient for concrete applications.

2. We shall apply this variational principle to the problem of nonlinear interaction of modes in two-dimensional hydrodynamics. Small perturbations in a hydrodynamic incompressible liquid decay $\sim\exp(-\nu k^2 t)$; but for $d=2$, this law leads to an infinite diffusion coefficient for the elements of the liquid (under the action of thermal fluctuations). A number of authors have expressed the idea that in the case $d=2$ the linear approximation is incorrect, and it is necessary to take into account the nonlinear interaction of a large number of modes.

In this problem, the role of the Q_α is played by the spatial Fourier components $Q_\alpha(t,k)$ of the velocity field, and the Lagrangian and the variational principle (35) have the form

$$\int_0^\infty dt\int dk\left\{\frac{1}{2\nu k^2}|\dot Q_\alpha(t,k)-Y_\alpha{}^0(k,Q)|^2+\frac{1}{2}\nu k^2|Q_\alpha(t,k)|^2\right\}=\min, \qquad (36)$$

where the reversible components of the fluxes are

$$Y_\alpha{}^0(k,Q)=\left(\delta_{\alpha\beta}-\frac{k_\alpha k_\beta}{k^2}\right)k_\gamma\int\delta(k'+k''-k)\,Q_\beta(t,k')\,Q_\gamma(t,k'')\,dk'\,dk''.$$

We shall seek an approximate solution of the hydrodynamic equations in the form

$$Q_\alpha(t,k)=\exp[-\gamma(k^2)t]\,Q_\alpha(0,k),$$

by minimizing the entropy-production integral (36) with respect to $\gamma(k^2)$. For definiteness we set

$$Q_\alpha(0, k) = v(\delta_{\alpha 1} - k_\alpha k_1/k^2).$$

On substituting these expressions in (36), we get after minimization the following integral equation for $\gamma(k^2)$:

$$\frac{1}{\nu k^2} - \frac{\nu k^2}{\gamma^2(k^2)} = \frac{v^2}{\nu}\int R(\hat{k}, \hat{k}', \hat{k}'')\{\gamma(k^2)+\gamma(k'^2) + \gamma(k''^2)+\gamma((k-k'-k'')^2)\}^{-2}\,dk'\,dk'', \tag{37}$$

where $\hat{k} \equiv k/|k|$; the exact form of the function R plays no role. The integral is cutoff at large $|k'|$ and $|k''|$.

For $d>2$ and for sufficiently small amplitude of the perturbation v, equation (37) has a solution the quadratic dispersion law $\gamma(k^2) = \nu k^2$. But if $d=2$, substitution of this law in the right side of (37) leads to logarithmic divergence of the integral at the point $k=0$ and to the impossibility of making the right side arbitrarily small. This indicates breakdown of the quadratic law in the small-wave-number range.

For an approximate solution of equation (37) at small but finite k^2, we expand the expression in wavy brackets in the vicinity of the points $k'^2 = k^2/3$ and $k''^2 = k^2/3$ and require that the result of the calculation of the integral remain finite for $k^2 \to 0$. Thus we obtain the differential equation ($z \equiv k^2$)

$$\ln\left\{\frac{d\gamma(z)/dz}{\gamma(z)+3\gamma(z/3)}\right\} = \mathrm{const}\left(\frac{d\gamma(z)}{dz}\right)^2,$$

an approximate solution of which has the form

$$\gamma(z) = \nu z + \nu' z\left(\ln\frac{b}{z}\right)^{1/2}, \quad \nu' \neq 0, \tag{38}$$

where ν, ν', and b are certain constants. Consequently the damping constant of slow modes depends nonanalytically on k^2. When k^2 is too small, formula (38) must be replaced by some more accurate equation, but the nonanalyticity obviously remains.

We note the simplicity of the (variational) method based on fluctuation-dissipation theory; it leads economically to the same results as does a detailed theory of interacting modes.[12]

APPENDIX

The variational principle considered above describes the relaxation of a "free" ("closed"[1]) system to equilibrium. But since this principle is a very general consequence of the FDR, it can be extended with certain changes to an open system, in which the external dynamic or thermal forces f_i excite undamped, on the average, fluxes $\Psi_i = \Psi_i(Q)$, and whose stationary state is a nonequilibrium state. Then there is an entropy flux $f_i\Psi_i(Q)$ from the source of the perturbation to the thermostat. The fluxes Ψ_i are determined by the current state of the system Q_α (here the Q_α are "internal" variables).

In order to avoid formal complications, we consider the simple case in which f_i = const and the external forces do not act directly on the thermostat and the local transport coefficients, but make a contribution only to the reversible term of the evolution equation:

$$\dot{Q}_\alpha = B_{\alpha i}(Q) f_i + \frac{\partial}{\partial x_i} F(x; Q), \quad x_\alpha = \frac{\partial S}{\partial Q_\alpha}. \tag{A1}$$

It was shown in Ref. 1 that such a possibility does not contradict the FDR (from the practical point of view, this is the common case). From the FDR it is easy to derive

$$B_{\alpha i}(\varepsilon Q) = -\varepsilon_\alpha \mu_i B_{\alpha i}(Q), \quad \Psi_i(Q) = -B_{\alpha i}(Q)\,\partial S/\partial Q_\alpha, \tag{A2}$$

where $\mu_i = \pm 1$ is the parity of f_i. For the change of macroentropy, we have instead of (8) the expression, no longer of fixed sign,

$$\dot{S}(Q) = \mathscr{P}(Q) - f_i\Psi_i(Q). \tag{A3}$$

We introduce the modified potential

$$\tilde{F}(x; Q) = x_\alpha B_{\alpha i}(Q) f_i + F(x; Q),$$

and then—exactly in accordance with the previous prescription—the Hamiltonian and the Lagrangian $\tilde{\Lambda}(I; Q)$. It is easily seen that

$$\tilde{\Lambda}(\dot{Q}; Q) = \Lambda(\dot{Q} - B(Q)f; Q) - f_i\Psi_i(Q) \tag{A4}$$

and that in consequence of the inequality (31) and the relation (A2), we have

$$\tilde{\Lambda}(\dot{Q}; Q) - \dot{Q}_\alpha \partial S/\partial Q_\alpha \geqslant 0,$$

here the equality is attained only at solutions of the equations (A1). Hence we get a variational principle that generalizes (33) and (34), in the form

$$\int \{\tilde{\Lambda}(\dot{Q}, Q) - \dot{S}(Q)\}\,dt = \min(=0), \qquad \delta\int \tilde{\Lambda}(\dot{Q}; Q)\,dt = 0. \tag{A5}$$

But an analog of the principle (35) in general does not exist, since for given f_i, a stationary stable state may be nonunique or, on the contrary, may not exist at all (and then the system performs, instead of a relaxational motion, a periodic or irregular one). According to (A.3)–(A.5), $\tilde{\Lambda}$ has the meaning of the increment of macroentropy in a virtual process (which is always larger than in a real one).

1) We note that formally (5) is always an exact relation, valid both in the quantum and in the classical case. The assumptions used in the reduced description show up only in the physical interpretation of (5).

2) Of course in the general case, equations (14) lead to spatially nonlocal models of a continuous medium. Furthermore, even in the local case, when (14) contain r- derivatives of no higher than the second order, the FDR permit dependence of the transfer coefficients on the gradients.

3) In (19) one could still add a δ-correlated "molecular" component. But it is unimportant for what follows.

4) It is clear also that for $V\to\infty$, we are actually considering a stationary nonequilibrium state, and the result is not connected with an assumption about separation of the microscope and macroscopic time scales.

5) For a simple proof of this property, it is sufficient to use the nonnegativity of the kernel R in (9) and (10).

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