LIGHT SCATTERING IN A FLUID SUBJECTED TO A TEMPERATURE GRADIENT

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The comparison of the experiments on light scattering in a fluid subjected to a temperature gradient with the theory of nonequilibrium hydrodynamic fluctuations, based on a local equilibrium [18], and with the fluctuating hydrodynamics has been carried out. It is shown that the local equilibrium theory will much better be coordinated to experiment.

A fluid subjected to a temperature gradient is a simple and, consequently, important example of a stationary nonequilibrium state. To the present time, the certain progress in a study of fluctuations in similar sorts of nonequilibrium systems is achieved, in particular due to experiments on light scattering. The initiator of studying the light scattering in nonuniformly heated media was L.I. Mandelstam [1]. The history of the development of researches in this field is presented in [2], therefore we do not stop here on it. We emphasize only the fact that the conventional theory allowing to determine spectra of light scattering in a nonequilibrium fluid in the middle of the 1980s, anyway in West, becomes fluctuating hydrodynamics [3–16]. Fluctuating hydrodynamics is based on the use the Landau–Lifshitz fluctuating forces [17] as the Langevin sources in linear equations describing the dynamics of fluctuations in a nonequilibrium steady state.

The basis for a similar approach to nonequilibrium hydrodynamic fluctuations are the following reasonings. In the nonequilibrium steady state, we do not know the ensemble, over which it is necessary to make averaging. Fluctuations are caused by the molecular movement. As the nonequilibrium macroscopic scale of this state is much more than that of the molecular movement, the nonequilibrium connected, for example, with a temperature gradient should not render any essential influence on molecular sources of fluctuations. Consequently, for the description of nonequilibrium fluctuations, it is possible to use the equilibrium Landau–Lifshitz formulas, having saved the local dependence of thermodynamic parameters in them on coordinates.

The objection of this conception formulated in [18] is that thermal hydrodynamic fluctuations are field fluctuations and have no relation to the molecular level of a description. At the hydrodynamic level, the medium should be examined as continuous. The basic hypothesis used for nonequilibrium continuous media in addition to the Onsager regressive hypothesis is the hypothesis of local equilibrium. Therefore, the ensemble, on which it is necessary to average, is known. On the contrary, fluctuating forces for the examined steady state are unknown. They, however, can easy be determined by the known local-equilibrium ensemble and the regressive equations.

Mathematically, the difference between fluctuating hydrodynamics and the theory developed in [18] is, as will be seen, the Langevin fluctuation-dissipation theorem.

For the fluctuations x_i of hydrodynamic values with time-dependent evolution

$$\dot{x}_i = -\lambda_{ij} x_j, \quad (1)$$

the Langevin fluctuation-dissipation theorem connects the intensities Q_{ij} of fluctuating forces y_i, which should be added in the right part of (1),

$$\langle y_i(t)\, y_j(0)\rangle = Q_{ij}\delta(t), \quad (2)$$

with the matrix λ and the average meanings of the square-law functions of fluctuations x_i at the initial time moment

$$Q_{ij} = \lambda_{ik} \langle x_k x_j \rangle + \lambda_{jk} \langle x_k x_i \rangle . \quad (3)$$

According to fluctuating hydrodynamics, the intensities Q_{ij} in (3) are determined by the Landau–Lifshitz formulas, and λ corresponds to the state under study. Then it is possible to find $\langle x_i x_j \rangle$ from (3), i.e. to determine the ensemble adequate to the examined nonequilibrium state. In particular, for an unstable mode such that $\dot{x} = -\lambda x$ with $\lambda \to 0$, it follows from the fluctuation-dissipation theorem

$$Q = 2\lambda \left\langle x^2 \right\rangle \quad (4)$$

that fluctuations of an unstable mode rise: $\left\langle x^2 \right\rangle \to \infty$.

Actually, the hypothesis of local equilibrium means that $\langle x_i x_j \rangle$ are given; therefore, equality (3) should be read from the right to the left, i.e. it defines a matrix Q for given λ and $\langle x_i x_j \rangle$. For an unstable mode, the conclusion about the growth of fluctuations at a vicinity of the threshold of stability should be changed to the conclusion that the fluctuating force intensity of the unstable mode tends to zero, ($Q \to 0$), together with its dissipative function [19].

Thus, fluctuating hydrodynamics and the locally equilibrium theory of nonequilibrium hydrodynamic fluctuations give different conclusions about the behaviour of fluctuations. Therefore, it is expedient to carry out their comparison with experiment. First of all, this concerns the experiments on Mandelstam–Brillouin light scattering in a fluid subjected to a temperature gradient.

For the dynamic structure factor of a fluid with temperature $T = T_0 + \vec{r}\vec{\nabla}T$, fluctuating hydrodynamics gives [4]

$$S\left(\vec{k},\omega\right) = 2T_0\rho_0 \left\{ \frac{Dk^4}{\left(\omega^2 - c^2k^2\right)^2 + \omega^2 D^2 k^4} - \right.$$

$$\left. - \frac{2\omega^3 D^2 k^4 \vec{k}\vec{\nabla}T}{T_0 \left[\left(\omega^2 - c^2k^2\right)^2 + \omega^2 D^2 k^4 \right]^2} \right\}, \quad (5)$$

where c is the sound propagation velocity, $D = \frac{4}{3}\nu + \xi$, ν and ξ are, respectively, the shear and bulk kinematic viscosities, and ρ_0 is the liquid density. The correction to an equilibrium spectrum is odd in the frequency, which reflects the break of time symmetry. The natural explanation of this breaking is that the number of phonons coming from areas with greater temperature is more than the number of phonons coming from areas with smaller temperature.

If the condition of a small attenuation $ck \gg Dk^2$ is satisfied, it is easy to reduce (5) to

$$S\left(\vec{k},\omega\right) = \frac{T_0\rho_0}{c^2} \left\{ \frac{\Gamma k^2 \left(1 - \epsilon\left(\vec{k},\omega\right)\right)}{\left(\omega - ck\right)^2 + \Gamma^2 k^4} + \right.$$

$$\left. + \frac{\Gamma k^2 \left(1 + \epsilon\left(\vec{k},\omega\right)\right)}{\left(\omega + ck\right)^2 + \Gamma^2 k^4} \right\}, \quad (6)$$

where $\Gamma = \frac{1}{2}D$ such that Γk^2 is the damping coefficient of sound waves, and

$$\epsilon\left(\vec{k},\omega\right) = \frac{c}{Dk^2} \frac{\vec{k^\circ}\vec{\nabla}T}{T_0} \frac{2\omega^2 D^2 k^4}{\left(\omega^2 - c^2k^2\right)^2 + \omega^2 D^2 k^4}. \quad (7)$$

Here, $\vec{k^\circ}$ is a unit vector in the direction of a vector $\vec{k}$. Thus, the Mandelstam–Brillouin doublet is represented by two Lorentzians of different height. The peaks at the frequencies $-ck$ and $+ck$ are named, respectively, the Stokes and anti-Stokes ones.

The modern experimental equipment does not allow one to determine enough precisely the frequency dependence of satellites, however it allows the authentic determination of the integrated intensity of each of them. The basic value measured in experiments is the asymmetry of a spectrum determined by the formula

$$\epsilon = \frac{I_S - I_a}{I_S + I_a}, \quad (8)$$

where I_S and I_a are the integrated intensities of the Stokes and anti-Stokes peaks. After the integration in formula (6) for the asymmetry given by fluctuating hydrodynamics, it was found

$$\epsilon = \frac{c}{2\Gamma T_0} \frac{\vec{k^\circ}\vec{\nabla}T}{k^2}. \quad (9)$$

The first experiments carried out for water [20] have qualitatively confirmed the character of dependence (6) and, at the same time, have shown that the slope of the line $\epsilon_{\exp}\left(\frac{\vec{k^\circ}\vec{\nabla}T}{k^2}\right)$ is equal to 6700 $\mathrm{cm}^{-1}\mathrm{K}^{-1}$, while the calculations by formula (9) give 19000 $\mathrm{cm}^{-1}\mathrm{K}^{-1}$. Thus, the experimental value of the asymmetry is 2.8 times less, though, in the subsequent experiments [21], a fit was achieved for the "best" results, and it also was noticed that the possible discrepancy is connected to the influence of walls and nonlinear effects.

In work [22] devoted to the Mandelstam–Brillouin light scattering by capillary waves on a surface of water subjected to a gradient of temperature along the surface, the same tendency as in [20] was again emphasized: the approximately three-fold reduction of the experimental asymmetry in comparison with the prediction of fluctuating hydrodynamics [23]. We note that, in experiments on a surface, neither the walls, nor nonlinearity are present. About the identical small experimental asymmetry was reported also in work [24], where two different types of the container with a fluid, one of the conventional Rayleigh–Benard type and a tall cylindrical cell, were used. The latter allows one to exclude the effect of boundaries. Authors of [24] have connected the small asymmetry with the nonuniformity of a temperature gradient along the cell.

From the above-stated, it is possible to conclude that the experiments on light scattering and the results following from fluctuating hydrodynamics have an essential quantitative discrepancy.

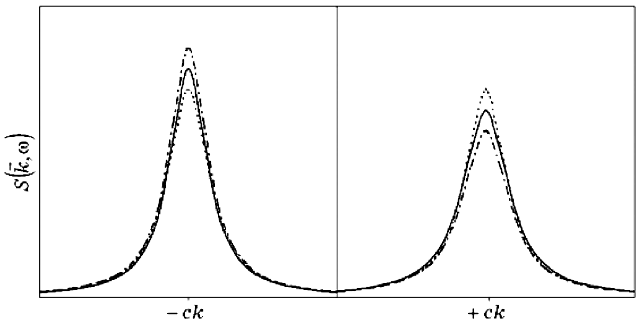
Let us write down now the expression for the structure factor obtained on the basis of the local equilibrium hypothesis in [18]:

$$S\left(\vec{k},\omega\right) = 2T_0\rho_0\left\{\frac{Dk^4}{\left(\omega^2-c^2k^2\right)^2+\omega^2D^2k^4}+\right.$$

$$\left.+\frac{\omega\vec{k}\vec{\nabla}T\left[\left(\omega^2-c^2k^2\right)^2-\omega^2D^2k^4\right]}{T_0\left[\left(\omega^2-c^2k^2\right)^2+\omega^2D^2k^4\right]^2}\right\}. \qquad (10)$$

Under the same condition $ck \gg Dk^2$, this yields formula (6) with the twice smaller $\epsilon\left(\vec{k},\omega\right)$ and, accordingly, the twice smaller asymmetry ϵ, so the slope becomes 9500 $cm^{-1}K^{-1}$. Though this result exceeds the experimental value of 6700 $cm^{-1}K^{-1}$, it is wholly digestible in view of the effect of walls, etc. Below, the plots of the frequency dependences of the equilibrium and nonequilibrium dynamic structure factors (5) and (10) under the condition of small attenuation of sound in the areas of Stokes and anti-Stokes satellites are given in irrespective units.

Thus, we have demonstrated in the present work that the theory of nonequilibrium hydrodynamic fluctuations [18] based on the local equilibrium hypothesis, in addition to the equivalence of various approaches to the solution of the problem, gives also the much better agreement with experiment, than fluctuating hydrodynamics.



Dynamic structure factor $S\left(\vec{k},\omega\right)$ as a function of the frequency ω 1) in an equilibrium liquid (dotted line); 2) in a nonequilibrium liquid according fluctuating hydrodynamics (dot-dashed line); 3) in the same nonequilibrium liquid according the local equilibrium theory [18] (solid line). Owing to the sharpness of peaks, only a narrow frequency box about $\omega = \pm ck$ is plotted

The local equilibrium theory of a light scattering by capillary waves in the case where the temperature gradient is subjected along the liquid surface also give results which have good agreement with experiment and will be present in a separate work.

1. L.I. Mandelstam, Dokl. AN SSSR **2**, 219 (1934).
2. I.L. Fabelinskii, Uspekhi Fiz. Nauk **164**, 897 (1994).
3. V.M. Zaitsev and M.I. Shliomis, Zh. Eksp. Teor. Fiz. **59**, 1583 (1970).
4. A.-M.S. Tremblay, M. Arai, and E.D. Siggia, Phys. Rev. A **23**, 1451 (1981).
5. D. Ronis and S. Patterman, Phys. Rev. A **22**, 773 (1980).
6. G. van der Zwan and P. Mazur, Phys. Lett. **75A**, 370 (1980).
7. K. Kawasaki, J. Phys. A: Math., Nucl., Gen. **6**, L4 (1973).
8. R. Graham, Phys. Rev. A **10**, 1762 (1974).
9. H.N.W. Lekkerkerker, J.-P. Boon, in *Fluctuations, Instabilities, and Phase Transitions*, edited by T. Riste (Plenum Press, New York, 1975).
10. J. Swift, P.C. Hohenberg, Phys. Rev. A **15**, 319 (1977).
11. Y. Tsuhiya, Phys. Soc. Jap. **43**, 1823 (1977).
12. J. Keizer, Phys. Fluids **21**, 198 (1978).
13. D. Ronis, I. Procaccia, and I. Oppenheim, Phys. Rev. A **19**, 307 (1979).
14. T.R. Kirkpatrick, E.G.D. Cohen, and J.R. Dorfman, Phys. Rev. A **26**, 950 (1982).
15. H. van Beijeren and E.G.D. Cohen, J. Stat. Phys. **53**, 285 (1988).
16. M.C. Cross and P.C. Hohenberg, Rev. Mod. Phys. **65**, 851 (1993).
17. L.D. Landau and E.M. Lifshits, Zh. Eksp. Teor. Fiz. **32**, 618 (1957).
18. V.P. Lesnikov, Ukr. J. Phys. **49**, 279 (2004).
19. V.P. Lesnikov, J. Phys. Studies. **1**, 208 (1997).

20. D. Beysens, Y. Garrabos, and G. Zalczer, Phys. Rev. Lett. **45**, 403 (1980).
21. H. Kiefte, M.J. Clouter, and R. Penney, Phys. Rev. B **30**, 4017 (1984).
22. D.S. Chung, K.Y. Lee and E. Mazur, Phys. Lett. A **145**, 348 (1990).
23. M. Grant and R.C. Desai, Phys. Rev. A **27**, 2577 (1983).
24. K. Hattori, K. Sakai, and K. Takagi, J. Phys: Cond. Matter. **10**, 3333 (1998).

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РОЗСІЯННЯ СВІТЛА В РІДИНІ З ГРАДІЄНТОМ ТЕМПЕРАТУРИ

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Р е з ю м е

Проведено порівняння експериментів з розсіяння Мандельштама–Бріллюена в рідині з градієнтом температури з теорією нерівноважних гідродинамічних флуктуацій, основаній на локальній рівновазі [18], та флуктуаційною гідродинамікою. Показано, що локально рівноважна теорія значно краще узгоджується з експериментом.