One More Step to Revision of Kinetics

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A formally exact relation is derived which connects thermodynamically non-equilibrium evolution of gas density distribution after its arbitrary strong spatially non-uniform perturbation and evolution of many-particle correlations between path of some marked particle and its surroundings in equilibrium gas. This relation directly confirms significance of the many-particle correlations even under the Boltzmann-Grad limit and thus validates the earlier suggested revision of kinetics.

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I. INTRODUCTION

The "law of large numbers", first discovered by Bernoulli approximately 300 years ago [1], up to now is world outlook paradigm of physical applications of probabilities. Meanwhile, at the end of his work [1] Bernoulli himself emphasized that if equivalent observations were perpetually continued (and at last probability turned to certainty) then one would conclude that all things obey exact rules or even fate. In the field of probabilistic interpretation of quantum mechanics such conclusion means "nonlocality" not less impressive than the famous EPR paradox. Naturally, one may ask who is counting outcomes of our observations (e.g. castings a die) and makes so that their relative frequencies have definite limits equal to speculative "probabilities" (e.g. all equal to $1/6$) ?

The standard answer is that such godlike role belongs to "independency" of the outcomes: "independency" reduces probability of their series to product of "elemental probabilities". But this is tautology, not the answer, because elemental probability loses meaning in absence of the limit. A good explanation of this logical fact can be found in [2]. Hence, presence (or absence) of the limit is no more than postulate concerning probabilistic measure in a space of observation sequences.

Unfortunately, in statistical physics built on microscopic dynamics the only a priori probabilities at our disposal are probabilities of its initial conditions. In such the theory, as Krylov emphasized almost 60 years ago [3], we should distinguish between actual dynamical (cause-and-effect) dependencies or correlations and statistical ones. Moreover, sometimes they controvert one to another. The determinism of dynamics can produce infinitely long-living actual correlations and thereby arrange definite limits for relative frequencies being perceived as fast decay of statistical correlations and "independency" of observations (similarly, ideality of a die is incarnate actual correlation ensuring "independency" of its castings). Examples are well known from mixing (chaotic) time-reversible dynamics with finite number of degrees of freedom (see e.g. [4, 5]).

And, vice versa, deficiency of actual dynamical dependencies may result in arbitrary long statistical correlations. For example, if earlier collisions of a particle (or charge carrier) with gas of similar particles (or with phonon gas) have no influence on later ones, then such system is indifferent to what rate (relative frequency) of collisions does happen (therefore it would be vain to wait till "probability turns to certainty"). The result is scaleless fluctuations, possessing 1/f-type spectrum, in collision rate of a test particle and thus in its diffusivity and mobility (1/f-noise) [6, 7, 9] (for more details and examples see also [8, 10, 11]).

As it was shown in [9] (see also [10–12]), in such case one has no rights to introduce a priori "probabilities of collisions", all the more, postulate "molecular chaos". Instead, we should honestly investigate dynamical evolution of an initial statistical ensemble, taking in mind Krylov's conjecture that under mixing dynamics, generally, relative frequencies of some phenomenon along particular phase trajectories in no way are connected with some its a priory probability [3].

In case of gas, if wishing to investigate Brownian motion of its particles, we have to solve the BBGKY equations starting from spatially non-uniform statistical ensemble with certainly known (non-random) position of at least one (test) particle. An exact solution is wittingly impossible, but we can presume the gas not too dense and resort to approximative shortened description of collisions in terms of Boltzmannian "collision integrals". However, analysis of this approach in [9] revealed that the Boltzmann's "molecular chaos" hypothesis is incompatible with spatial non-uniformity of the ensemble. As the consequence, in place of the single Boltzmann equation the theory births infinite chain of kinetic equations. They operate with usual one-particle distribution function plus infinite set of specific many-particle distribution functions which represent clusters of (real or virtual) collisions or close encounters of particles and together describe fluctuations in the rate of collisions [9–11].

In view of innovatory character of these specific functions, it seems principal to make sure that they have adequate prototypes beyond approximations. Below we present a proof based on exact relations between such functions and a relaxation law for arbitrary strong perturbations of gas density.

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II. START STATISTICAL ENSEMBLE

First, we will consider statistical ensemble of N particles which are contained in a volume Ω and possess short-range repulsion of one from another, under two successive formal limits: firstly the "thermodynamical" one, $N \to \infty$, $\Omega \to \infty$, $\nu_0 = N/\Omega \to$const, and secondly the "Boltzmann-Grad limit", $\nu_0 \to \infty$, $\sigma \to 0$, $\lambda = (\sigma\nu_0)^{-1} \to$const (λ is mean free path and σ properly defined cross-section of inter-particle collisions).

Let initially, at time moment $t = 0$, our system is described by the N-particle distribution function

$$\rho^{(in)}(\Gamma) \,=\, C^{(in)}\,\rho^{(eq)}(\Gamma)\,\exp\,[\,-\sum_{j=1}^{N} V(\boldsymbol{r}_j)/T\,] \;\;,$$
$$\rho^{(eq)}(\Gamma) \,=\, C^{(eq)}\,\exp\,[\,-H(\Gamma\,)/T\,] \tag{1}$$

Here $\Gamma = \{\Gamma_1, \dots, \Gamma_N\}$ with $\Gamma_j = \{\boldsymbol{r}_j\,, \boldsymbol{p}_j\}$ being variables of j-th particle, H denotes full N-particle Hamiltonian, $\rho^{(eq)}$ is corresponding canonical equilibrium distribution , $V(\boldsymbol{r})$ is a bounded function, and constants $C^{(eq)}$ and $C^{(in)}$ ensure the normalization

$$\int \rho^{(in)}(\Gamma)\,\prod_{j=1}^{N} d\Gamma_j \,=\, \int \rho^{(eq)}(\Gamma)\,\prod_{j=1}^{N} d\Gamma_j \,=\, 1 \tag{2}$$

We will be interested in consequent evolution of the system, that is in distribution function

$$\rho(t,\Gamma) \,=\, \exp\,(\,tL\,)\,\rho^{(in)}(\Gamma) \;\;, \tag{3}$$
$$L \,=\, \sum_{j=1}^{N}\,[\,(\,\partial H/\partial\,\boldsymbol{r}_j)\,\partial/\partial \boldsymbol{p}_j \,-\, (\partial H/\partial \boldsymbol{p}_j)\,\partial/\partial\,\boldsymbol{r}_j\,]\;,$$

where L is the Liouville operator corresponding to Hamiltonian H. Clearly, $\rho^{(in)}$ and $\rho(t)$ represent statistical ensemble which is non-equilibrium because of non-uniform perturbation of particles density

$$\nu(t,\boldsymbol{r}) \,=\, \int\, \rho(t,\Gamma)\,\sum_{j=1}^{N} \delta(\boldsymbol{r}-\boldsymbol{r}_j)\,\prod_{j=1}^{N} d\Gamma_j \tag{4}$$

Alternatively, one can think that $\rho(t)$ comes from equilibrium in presence of an external potential $V(\boldsymbol{r})$ which however is abruptly switched off at $t = 0$.

It is convenient to characterize density perturbations by function

$$\phi(\boldsymbol{r}) \,\equiv\, \exp\,[-V(\boldsymbol{r})/T\,] \,-\, 1$$

Then, introducing functional

$$\mathcal{F}\{\phi\} \,\equiv\, \int\, \rho^{(eq)}(\Gamma)\,\prod_{j=1}^{N}[\,1+\phi(\boldsymbol{r}_j)\,]\,\,d\Gamma_j \;\;, \tag{5}$$

one can easy verify the equalities

$$C^{(in)} \,=\, 1/\mathcal{F}\{\phi\} \;\;, \tag{6}$$
$$\nu(0,\boldsymbol{r}) \,=\, [\,1+\phi(\boldsymbol{r})\,]\,\frac{\delta\ln\mathcal{F}\{\phi\}}{\delta\,\phi(\boldsymbol{r})} \tag{7}$$

In order to perform the thermodynamical limit, we will assume that as a whole the perturbation is finite:

$$\int |\phi(\boldsymbol{r})|\,d\boldsymbol{r} \,<\, \infty \tag{8}$$

Then the limit of the functional (5) looks as

$$\mathcal{F}\{\phi\} \,=\, 1 + \sum_{k=1}^{\infty}\frac{\nu_0^k}{k!}\int F_k(\dots)\,\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j \,=$$
$$=\, \exp\,\sum_{k=1}^{\infty}\frac{\nu_0^k}{k!}\int \Upsilon_k(\dots)\,\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j \;\;, \tag{9}$$

where $F_k(\dots) = F_k(\boldsymbol{r}_1, \dots, \boldsymbol{r}_k)$ are usual non-normalized (or, in other words, "normalized to volume") k-particle spatial distribution functions (or, in other words, "correlation functions") of strictly equilibrium gas. In particular, $F_1(\boldsymbol{r}_1) = 1$.

Functions $\Upsilon_k(\dots) = \Upsilon_k(\boldsymbol{r}_1, \dots, \boldsymbol{r}_k)$ formally relate to F_k like cumulants, or semi-invariants, of a random field relate to its statistical moments: $\Upsilon_2(\boldsymbol{r}_1, \boldsymbol{r}_2) = F_2(\boldsymbol{r}_1, \boldsymbol{r}_2) - F_1(\boldsymbol{r}_1)F_1(\boldsymbol{r}_2) = F_2(\boldsymbol{r}_1, \boldsymbol{r}_2) - 1$, and so on.

In fact, of course, all F_k and Υ_k obey translation invariance, i.e. depend on inter-particle distances $\boldsymbol{r}_i - \boldsymbol{r}_j$ only. At that, any of the cumulants $\Upsilon_k \to 0$ when some of its arguments $\boldsymbol{r}_i - \boldsymbol{r}_j$ goes to infinity as measured in units of characteristic radius r_0 of particles interaction (taking in mind 3-D gas, one can define r_0 e.g. by means of $\sigma = \pi r_0^2$). Therefore, if the function $\phi(\boldsymbol{r})$ is smooth enough, on scales of order of r_0, then

$$\int \Upsilon_k\,\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j \,\to\, \gamma_k\,r_0^{3(k-1)}\int \phi^{\,k}(\boldsymbol{r}_1)\,d\boldsymbol{r}_1 \tag{10}$$

at $k > 1$ with γ_k being "virial coefficients". Under the Boltzmann-Grad limit, $r_0 \to 0$ and the "gas parameter", defined e.g. by $g = (4\pi r_0^3/3)\,\nu_0$, also tends to zero, $g \to 0$. Consequently, gas becomes ideal from the point of view of thermodynamics:

$$\mathcal{F}\{\phi\} \,\to\, \exp\left[\nu_0\int \phi(\boldsymbol{r})\,d\boldsymbol{r}\right] \;\;, \tag{11}$$
$$\frac{\nu(0,\boldsymbol{r})}{\nu_0} \,\to\, 1+\phi(\boldsymbol{r}) \,=\, \exp\,[-V(\boldsymbol{r})/T\,] \tag{12}$$

III. RELAXATION, TIME SYMMETRY AND EQUILIBRIUM-NON-EQUILIBRIUM RELATIONS

However, from the point of view of kinetics our gas remains quite non-ideal. Let us consider relations which substitute (7) and (9) at $t > 0$ and connect, on one hand, non-equilibrium process of relaxation of relative gas density perturbation, $\nu(t,\boldsymbol{r})/\nu_0 - 1$, and, on the other hand, Brownian motion of particles in unperturbed equilibrium gas.

Taking into account that $L\,\rho^{(eq)} f = \rho^{(eq)} L f$ (with f being "arbitrary function" of Γ_j), i.e. the Liouville operator L commutes with any equilibrium distribution, and combining formulas (1), (3) and (4), we can transform the latter to

$$\nu(t,\boldsymbol{r}) \,=\, \frac{1}{\mathcal{F}\{\phi\}} \int \prod_{j=1}^{N} d\Gamma_j \;\, \rho^{(eq)}(\Gamma) \;\times \qquad (13)$$
$$\times\, \sum_{j=1}^{N} \delta(\boldsymbol{r}-\boldsymbol{r}_j)\, \exp{(\,tL)} \prod_{j=1}^{N} [\,1+\phi(\boldsymbol{r}_j)\,]$$

Let us make the change of integration variables here: $\boldsymbol{p}_j \,\Rightarrow\, -\boldsymbol{p}_j$. Since the Hamiltonian is even function (quadratic form) of the momentums while L is odd with respect to them, the only result of this operation is change of sign of time argument before L :

$$\nu(t,\boldsymbol{r}) \,=\, \frac{1}{\mathcal{F}\{\phi\}} \int \prod_{j=1}^{N} d\Gamma_j \;\, \rho^{(eq)}(\Gamma) \;\times \qquad (14)$$
$$\times\, \sum_{j=1}^{N} \delta(\boldsymbol{r}-\boldsymbol{r}_j)\, \exp{(\,-\,tL)} \prod_{j=1}^{N} [\,1+\phi(\boldsymbol{r}_j)\,]$$

which clearly demonstrates time symmetry of gas evolution: $\nu(-t,\boldsymbol{r}) = \nu(t,\boldsymbol{r})$.

Next, recall that operator $\exp{(\,-tL)}$ has twofold sense: if acting onto distribution functions it describes their time-reversed evolution, but if acting onto system's variables (or any function of them) it describes their evolution in real direct time. In particular,

$$\exp{(\,-\,tL)}\;\phi(\boldsymbol{r}_j) \,=\, \phi(\boldsymbol{r}_j(t,\Gamma)) \quad , \qquad (15)$$

where expression $\boldsymbol{r}_j(t,\Gamma)$ means (vector of) coordinates of j-th particle at time t considered as a function of initial state Γ of the whole system taken at $t=0$ (i.e. the set $\boldsymbol{r}_j(t,\Gamma)$ is solution to system's Hamiltonian equations starting from $\boldsymbol{r}_j(0,\Gamma)=\boldsymbol{r}_j$, $\boldsymbol{p}_j(0,\Gamma)=\boldsymbol{p}_j$) .

Hence, obviously, we can rewrite (14) in the form

$$\frac{\nu(t,\boldsymbol{r})}{\nu_0} \,=\, \int \frac{\rho^{(eq)}(t,\Gamma\,|\,\boldsymbol{r})}{\mathcal{F}\{\phi\}} \prod_{j=1}^{N} [\,1+\phi(\boldsymbol{r}_j)\,]\, d\boldsymbol{\Gamma}_j \qquad (16)$$

where $\rho^{(eq)}(t,\Gamma\,|\,\boldsymbol{r})$ is conditional distribution function of system's state Γ at current time t corresponding to condition that initially (at time zero) one of particles was positioned exactly at a given point $\boldsymbol{r}$. Formally,

$$\rho^{(eq)}(t,\Gamma\,|\,\boldsymbol{r}) \,=\, \exp{(\,tL)}\,\rho^{(eq)}(0,\Gamma\,|\,\boldsymbol{r}) \quad ,$$
$$\rho^{(eq)}(0,\Gamma\,|\,\boldsymbol{r}) \,=\Omega\,\rho^{(eq)}(\Gamma\,)\,\frac{1}{N}\sum_{j=1}^{N} \delta(\boldsymbol{r}-\boldsymbol{r}_j) \quad , \qquad (17)$$

where factor Ω/N ensures normalization to unit: $\int \rho^{(eq)}(t,\Gamma\,|\,\boldsymbol{r})\,d\Gamma = 1$, and we already keep in mind the coming thermodynamical limit.

To complete the limit, notice that all of N terms of (17) give equal contributions to integral in (16). In analogy with transition from (5) to (9), the result is

$$\frac{\nu(t,\boldsymbol{r})}{\nu_0} \,=\, \frac{1}{\mathcal{F}\{\phi\}} \int d\boldsymbol{r}' \;[\,1+\phi(\boldsymbol{r}')\,]\,\{\,W_1(t,\boldsymbol{r}'|\boldsymbol{r})\,+$$
$$+\sum_{k=1}^{\infty} \frac{\nu_0^{\,k}}{k!} \int F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r}) \prod_{j=1}^{k} \phi(\boldsymbol{r}_j) d\boldsymbol{r}_j\;\} \quad (18)$$

Here $W_1(t,\boldsymbol{r}'|\boldsymbol{r})$ means probability density of finding at point $\boldsymbol{r}'$ at time t the same test particle what started from the point $\boldsymbol{r}$. This function is normalized to unit:

$$\textstyle\int W_1(t,\boldsymbol{r}'|\boldsymbol{r})\,d\boldsymbol{r}' \,=1 \qquad (19)$$

In other words, this is just the distribution of Brownian displacement of test particle which was under investigation in [11] (and much earlier in [9, 10]).

Functions $F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$ describe joint probabilities of finding the test particle (started from $\boldsymbol{r}$) at point $\boldsymbol{r}'$ and simultaneously some k other particles at points $\boldsymbol{r}_j$. These functions are symmetric with respect to $\boldsymbol{r}_j$ ($j=1...\,k$) and definitely normalized with respect to $\boldsymbol{r}'$ as well as $\boldsymbol{r}$:

$$\int F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\,d\boldsymbol{r}' \,=\, F_k(t,\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r}) \quad (20)$$
$$\int F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\,d\boldsymbol{r} \,=\, F_{k+1}(\boldsymbol{r}_1,\dots,\boldsymbol{r}_k,\boldsymbol{r}') \quad (21)$$

On right-hand side in (20) we see non-normalized (i.e. "normalized to whole volume") distribution functions (correlation functions) characterizing probability of location of k particles at points $\boldsymbol{r}_j$ under condition that one more particle had started from $\boldsymbol{r}$. Functions $F_{k+1}(\boldsymbol{r}_1,\dots,\boldsymbol{r}_k,\boldsymbol{r}')$ on right-hand side of (21) are usual static $(k+1)$-particle distribution functions mentioned in previous Section.

Of course, after the limit all these functions acquire translation invariance: $W_1(t,\boldsymbol{r}'|\boldsymbol{r}) \,=\, W_1(t,\boldsymbol{r}'-\boldsymbol{r})$, etc. With this notion it becomes rather obvious that identities (21) guarantee "conservation of particles": $\int \nu(t,\boldsymbol{r})\,d\boldsymbol{r} \,=\, \int \nu(0,\boldsymbol{r})\,d\boldsymbol{r}$.

Initial conditions for these functions also are clear:

$$W_1(0,\boldsymbol{r}'|\boldsymbol{r}) \,=\, \delta(\boldsymbol{r}'-\boldsymbol{r}) \;\; ,$$
$$F_{k+1}(0,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r}) \,=\, \delta(\boldsymbol{r}\,'-\boldsymbol{r})\,F_{k+1}(\boldsymbol{r}_1,\dots,\boldsymbol{r}_k,\boldsymbol{r}) \;\; ,$$
$$F_k(0,\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r}) \,=\, F_{k+1}(\boldsymbol{r}_1,\dots,\boldsymbol{r}_k,\boldsymbol{r}) \qquad (22)$$

Correspondingly, at $t=0$ (18) reduces to (7) with (9).

In principle, (18) can be qualified as example of "generalized Onsager relations" or "generalized fluctuation-dissipation relations" which follow from time reversibility of microscopic dynamics and establish connections between dissipative nonlinearity and statistics of transport processes, in particular, statistics of equilibrium noise (see e.g. [13, 14] and references therein).

IV. DYNAMICAL MULTI-PARTICLE CORRELATIONS AND THEIR CUMULANTS

The distribution functions what appear in (18) mix up i) average values, ii) static correlations between *positions* of particles due to their potential interaction, and besides iii) specific dynamical correlations between current relative disposition of mutually close (colliding or merely encountering) particles and previous *displacement*, $\boldsymbol{r}' - \boldsymbol{r}$, of one of them (the test particle). Next, we would like to separate those contributions and especially consider the third of them which, as far as I know, never attracted particular attention of classics, although, as we will see, possess quite anomalous spatial scaling as compared with static correlations.

In order to extract joint dynamical correlations in a pure form, we should introduce cumulants, or semi-invariants, similar to cumulant functions $\Upsilon_k(\boldsymbol{r}_1, ..., \boldsymbol{r}_k)$ in (9). With this purpose, first, notice that expression inside braces in (18) has the same formal structure as expression $\langle\, x \exp(\phi\, y)\,\rangle$, where x and y are two random quantities, ϕ is dummy parameter (test parameter), and angle brackets denote averaging procedure. Second, apply the well known general formula (see e.g. Ref. [15])

$$\langle\, x \exp(\phi\, y)\,\rangle \,=\, \langle\, \exp(\phi\, y)\,\rangle \left[\langle x\rangle + \phi\,\langle x, y\rangle + \frac{\phi^2}{2!}\,\langle x, y, y\rangle + \frac{\phi^3}{3!}\,\langle x, y, y, y\rangle + ... \right] \tag{23}$$

with $\langle\, x, y, \dots, y\,\rangle = \langle\, x, y^{(k)}\,\rangle$ being joint cumulants in Malakhov's notations [15].

In our context, similar transformation as performed in (18) yields

$$\begin{aligned} & W_1(t, \boldsymbol{r}'|\boldsymbol{r}) + \sum_{k=1}^{\infty} \frac{\nu_0^k}{k!} \int F_{k+1}(t, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r}) \prod_{j=1}^{k} \phi(\boldsymbol{r}_j)\, d\boldsymbol{r}_j \,= \\ & = \left[1 + \sum_{k=1}^{\infty} \frac{\nu_0^k}{k!} \int F_k(t, \boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r}) \prod_{j=1}^{k} \phi(\boldsymbol{r}_j)\, d\boldsymbol{r}_j \right] \times \\ & \times \left[W_1(t, \boldsymbol{r}'|\boldsymbol{r}) + \sum_{k=1}^{\infty} \frac{\nu_0^k}{k!} \int W_{k+1}(t, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r}) \prod_{j=1}^{k} \phi(\boldsymbol{r}_j)\, d\boldsymbol{r}_j \right] \,\equiv\, \mathcal{F}\{t, \phi\,|\boldsymbol{r}\} \,\times\, \mathcal{W}\{t, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\} \end{aligned} \tag{24}$$

Here new functions $W_{k+1}(t, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r})$, in place of $F_{k+1}(t, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r})$, are just the cumulant functions (analogous to $\langle\, x, y, \dots, y\,\rangle$ in (23)) what correspond to refined dynamical (two-time) inter-particle correlations.

Further, notice that because of the normalization relations (19) and (20) result of integration of the above expression over $\boldsymbol{r}'$ coincides with functional $\mathcal{F}\{t, \phi\,|\boldsymbol{r}\}$ defined by the upper square brackets in (24). Therefore, firstly,

$$\int \mathcal{W}\{t, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\; d\boldsymbol{r}' \,=\, 1 \quad , \quad \text{that is} \quad \int W_{k+1}(t, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r})\; d\boldsymbol{r}' \,=\, 0 \quad , \tag{25}$$

with functional $\mathcal{W}\{t, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\}$ defined by the lower square brackets in (24). Secondly, formula (18) takes the form

$$\frac{\nu(t, \boldsymbol{r})}{\nu_0} \,=\, \frac{\mathcal{F}\{t, \phi\,|\boldsymbol{r}\}}{\mathcal{F}\{\phi\}} \left[1 + \int \phi(\boldsymbol{r}')\, \mathcal{W}\{t, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\; d\boldsymbol{r}' \right] \tag{26}$$

Notice also that, because of the normalization relation (21), integration of (24) over $\boldsymbol{r}$ results in identity

$$\int \mathcal{F}\{t, \phi\,|\boldsymbol{r}\}\; \mathcal{W}\{t, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\; d\boldsymbol{r} \,=\, \frac{1}{\nu_0}\, \frac{\delta \mathcal{F}\{\phi\}}{\delta\, \phi(\boldsymbol{r}')} \tag{27}$$

Besides, combining (24) with initial conditions (22) we obtain one more property of cumulants W_{k+1}:

$$\mathcal{W}\{0, \boldsymbol{r}'|\phi\,|\boldsymbol{r}\} \,=\, \delta(\boldsymbol{r}' - \boldsymbol{r}) \quad , \quad \text{that is} \qquad W_{k+1}(0, \boldsymbol{r}'|\boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r}) \,=\, 0 \quad , \tag{28}$$

thus confirming that indeed they represent correlations of very dynamical nature.

Functions $F_k(t, \boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r})$ also can be replaced by corresponding cumulants if we write

$$\mathcal{F}\{t, \phi\,|\boldsymbol{r}\} \,=\, \exp \sum_{k=1}^{\infty} \frac{\nu_0^k}{k!} \int \Upsilon_k(t, \boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r}) \prod_{j=1}^{k} \phi(\boldsymbol{r}_j)\, d\boldsymbol{r}_j \quad , \tag{29}$$

with $\Upsilon_k(t, \boldsymbol{r}_1, \dots, \boldsymbol{r}_k|\boldsymbol{r})$ being direct analogue of above introduced static cumulant functions $\Upsilon_k(\boldsymbol{r}_1, ..., \boldsymbol{r}_k)$.

Let us discuss and compare two just now introduced sorts of cumulants. Last of them, $\Upsilon_k(t,\boldsymbol{r}_1,...,\boldsymbol{r}_k|\boldsymbol{r})$, at $k>1$ describe conditional joint correlations between k particles under condition that some one more $(k+1)$-th (test) particle earlier, at $t=0$, was localized at point $\boldsymbol{r}$. Certainly, such poor condition can not change short-range character of the correlations, and the latter must decay at same inter-particle distances $|\boldsymbol{r}_i-\boldsymbol{r}_j|\gtrsim r_0$ as unconditioned correlations. Then, quite as in (10),

$$\int \Upsilon_k(t,\boldsymbol{r}_1,...,\boldsymbol{r}_k|\boldsymbol{r})\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j\approx$$
$$\approx r_0^{3(k-1)}\int \gamma_k(t,\boldsymbol{r}_1|\boldsymbol{r})\,\phi^k(\boldsymbol{r}_1)\,d\boldsymbol{r}_1 \qquad (30)$$

All the more, such condition can not influence upon mean gas density. Therefore it is natural to state, in agreement with (22), that difference between the conditional one-particle distribution function, $\Upsilon_1(t,\boldsymbol{r}_1|\boldsymbol{r})=F_1(t,\boldsymbol{r}_1|\boldsymbol{r})$ (expressing conditional gas density in units of ν_0), and the unconditioned one, $\Upsilon_1(\boldsymbol{r}_1)=F_1(\boldsymbol{r}_1)=1$, at $t>0$ is not significantly greater than at $t=0$. We can write this in the form

$$\left|\int[\,\Upsilon_1(t,\boldsymbol{r}_1|\boldsymbol{r})-\Upsilon_1(\boldsymbol{r}_1)\,]\,\phi(\boldsymbol{r}_1)\,d\boldsymbol{r}_1\right|\lesssim \qquad (31)$$
$$\lesssim\left|\int[\,F_2(\boldsymbol{r}_1,\boldsymbol{r})-1\,]\,\phi(\boldsymbol{r}_1)\,d\boldsymbol{r}_1\right|\sim r_0^3\,\overline{\phi}\;,$$

where $\overline{\phi}$ is characteristic magnitude of $\phi(\boldsymbol{r})$.

Formal substantiation of these statements in principle could be obtained from the Bogolyubov-Born-Green-Kirkwood-Yvon (BBGKY) equations

$$\frac{\partial F_n^{(eq)}}{\partial t}=L^{(n)}F_n^{(eq)}+\nu_0\sum_{j=1}^{n}\int L_{j\,n+1}F_{n+1}^{(eq)}\,d\Gamma_{n+1} \qquad (32)$$

for the full phase space n-particle distributions $F_n^{(eq)}=$ $=F_n^{(eq)}(0,\Gamma_1|\Gamma_2,...,\Gamma_{n-1}|\boldsymbol{r})$ connected to our spatial distributions by means of

$$W_1(t,\boldsymbol{r}_1|\boldsymbol{r})=\int F_1^{(eq)}(t,\Gamma_1|\boldsymbol{r})\,d\boldsymbol{p}_1\;,$$
$$F_{k+1}(t,\boldsymbol{r}_1|\boldsymbol{r}_2,...,\boldsymbol{r}_{k+1}|\boldsymbol{r})= \qquad (33)$$
$$=\int F_{k+1}^{(eq)}(t,\Gamma_1|\Gamma_2,...,\Gamma_{k+1}|\boldsymbol{r})\prod_{j=1}^{k+1}d\boldsymbol{p}_j$$

along with (20), and subjected to initial conditions

$$F_n^{(eq)}(0,\Gamma_1|\Gamma_2,...,\Gamma_{n-1}|\boldsymbol{r})=$$
$$=\delta(\boldsymbol{r}_1-\boldsymbol{r})\,F_n(\boldsymbol{r}_1,...,\boldsymbol{r}_n)\prod_{j=1}^{n}G_0(\boldsymbol{p}_j)\;, \qquad (34)$$

with "number one" assigned to the test particle, $L^{(n)}$ standing for n-particle Liouville operator,

$$L_{ij}=\nabla U(\boldsymbol{r}_i-\boldsymbol{r}_j)\cdot(\partial/\partial\boldsymbol{p}_i-\partial/\partial p_j)\;,$$

$U(\boldsymbol{R})$ being interaction potential, and $G_0(\boldsymbol{p})$ equilibrium Maxwellian momentum distribution. Clearly, this is nothing but a task about Brownian motion of test particle in equilibrium gas. But, unfortunately, such equations remain almost uninvestigated beyond rough approximations like the Boltzmannian kinetics or its improvement suggested in [9, 10] (otherwise essays like the present one would be unneeded).

Other conditional cumulants, $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,...,\boldsymbol{r}_k|\boldsymbol{r})$, seem much more interesting objects because describe joint $(k+1)$-order correlations between k "ordinary" particles and besides the test particle itself. The latter is peculiar particle since it is the only we know about wherefrom it came to its current position. Therefore, in fact W_{k+1} represent joint correlations between its preceding displacement $\boldsymbol{r}'-\boldsymbol{r}$ ("Brownian path") and its current surroundings. Moreover, such extra correlations what do not reduce to mere gas density fluctuations already taken into account by Υ_k. Thus we must conclude that cumulants $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,...,\boldsymbol{r}_k|\boldsymbol{r})$ represent fluctuations in the line of collisions of the test particle, and in this sense, in contrary to Υ_k, they are specific dynamical correlations. Correspondingly, coordinates $\boldsymbol{r}_j$ belong to those neighbors of the test particle which currently are or recently were or soon will be involved into collisions with it (or even those which would be in collision if some other neighbor did not interfere with it).

As joint cumulants in general, $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,...,\boldsymbol{r}_k|\boldsymbol{r})$ vanish if one (or more) of its constituents become statistically independent on others. The independency, in its turn, takes place when any of the relative distances $|\boldsymbol{r}_j-\boldsymbol{r}'|$ notably exceeds some characteristic spatial scale l_0. However, the above reasoning prompts that this scale is not the interaction radius r_0, figuring in (10), (30) and (31), but "radius of collision" which is sooner the mean free path λ than r_0.

Indeed, clearly, information about a collision can propagate to as long distance apart it as λ, if not greater. Nevertheless, λ would be overestimate of l_0, since that is propagation along the direction of relative momentum $\boldsymbol{p}_j-\boldsymbol{p}'$ (and velocity) only. At two perpendicular dimensions the collision occupies merely a region $\approx\sigma\sim r_0^2$. Hence, total "volume of collision" (and volume covered by related correlations) is $\approx\sigma\lambda=1/\nu_0$. Correspondingly, it is reasonable to estimate mean (averaged over all directions) "radius of collision" (and mean radius of related correlations) l_0 as $l_0=1/\nu_0^{1/3}$. In other words, l_0 is nothing but typical distance between neighboring gas particles.

For preliminary characterization of cumulants W_{k+1} it is useful to notice also that both the initial conditions (28) and equalities (25) are manifestations of the mentioned general property of cumulants: since at $t=0$ position of the test particle is non-random (certainly equals to $\boldsymbol{r}$), any joint cumulant it enters must disappear (similarly, all $\langle\,x,y,...,y\,\rangle=0$ if x is a constant), while (25) as well reflects the fact that statement "test particle is somewhere" is certainly true.

V. THE BOLTZMANN-GRAD GAS

Next, consider behavior of our cumulant functions $W_{k+1}(t,\boldsymbol{r}'\,|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$ under the Boltzmann-Grad limit when their role becomes especially distinct.

At $g\to 0$, the estimates (10), (11), (30) and, very importantly, (31), lead to the asymptotic

$$\mathcal{F}\{t,\phi\,|\boldsymbol{r}\}\,\to\,\exp\left[\nu_0\int\phi(\boldsymbol{r})\,d\boldsymbol{r}\right]\quad,\quad\frac{\mathcal{F}\{t,\phi\,|\boldsymbol{r}\}}{\mathcal{F}\{\phi\}}\,\to\,1 \tag{35}$$

As the consequence, firstly, identity (27) simplifies to

$$\int\mathcal{W}\{t,\boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\;d\boldsymbol{r}\,=\,1\quad,\qquad\text{that is}\qquad\int W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\;d\boldsymbol{r}\,=\,0 \tag{36}$$

Secondly, our main formula (18), or (26), turns into

$$\frac{\nu(t,\boldsymbol{r})}{\nu_0}-1\,=\,\int\phi(\boldsymbol{r}')\,\mathcal{W}\{t,\boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\;d\boldsymbol{r}' \tag{37}$$

Thirdly, definition (24) of the cumulants $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$ degenerates to

$$\begin{aligned}&W_1(t,\boldsymbol{r}'-\boldsymbol{r})+\sum_{k=1}^{\infty}\frac{1}{k!}\int F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\prod_{j=1}^{k}\varphi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j\,=\\&=\left\{W_1(t,\boldsymbol{r}'-\boldsymbol{r})+\sum_{k=1}^{\infty}\frac{1}{k!}\int W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\prod_{j=1}^{k}\varphi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j\right\}\exp\left[\int\varphi(\boldsymbol{r})\,d\boldsymbol{r}\right]\quad,\end{aligned} \tag{38}$$

where we introduced new dummy function (series expansion variable) $\varphi(\boldsymbol{r})\,\equiv\,\nu_0\,\phi(\boldsymbol{r})$. It is taken in mind, of course, that all functions in the above expressions have not written out argument $g\to 0$ which directly determines $\nu_0\propto\lambda^{-3}g^{-2}$ and $r_0\propto\lambda g$ and indirectly $(\boldsymbol{r}_j-\boldsymbol{r}')$-dependencies of W_{k+1}'s under question.

Fourthly, functional $\mathcal{W}\{t,\boldsymbol{r}'|\phi\,|\boldsymbol{r}\}$ should be reconsidered in the course of the Boltzmann-Grad limit,

$$\mathcal{W}\{t,\boldsymbol{r}'|\phi\,|\boldsymbol{r}\}\,=\,\lim_{g\to 0}\,W_1(t,\boldsymbol{r}'-\boldsymbol{r})\,+\,\lim_{g\to 0}\,\sum_{k=1}^{\infty}\frac{\nu_0^k}{k!}\int W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j\quad, \tag{39}$$

as a result of competition between infinitely growing factor ν_0, on left hand, and infinitely shrinking radius l_0 of decay of $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$'s via $|\boldsymbol{r}_j-\boldsymbol{r}'|$ dependence, on right hand.

Among three qualitatively different variants of the competition, i.e. $\nu_0 l_0^3\,\to\,0$ or $\nu_0 l_0^3\,\to$const or $\nu_0 l_0^3\to\infty$, in fact the middle variant is acceptable only, since, from the point of view of the density relaxation, $\nu(t,\boldsymbol{r})/\nu_0$, the third variant would mean that none initial density perturbation can be considered as a weak one (even if it is infinitesimally small) while the first variant would lead to purely linear relaxation of the perturbation (even if it is arbitrary large). Hence, once again we come to conclusion that $l_0\,\sim\,1/\nu_0^{1/3}\,\propto\,\lambda g^{2/3}$ (and without loss of generality we can put on $l_0\,=\,1/\nu_0^{1/3}$).

In order to properly perform the limits in (39), one should take in the mind identities (25) (and then (36) as well) which are very important since say that results of integrations in (39) must represent full derivatives with respect to $\boldsymbol{r}'$ (and finally also with respect to $\boldsymbol{r}$).

Then, under assumption that $\phi(\boldsymbol{r})$ is not "too arbitrary" but sufficiently smooth field, quite natural expectation for the limit of $(k+1)$-th term of (39) reads

$$\lim_{g\to 0}\,\nu_0^k\int W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})\prod_{j=1}^{k}\phi(\boldsymbol{r}_j)\,d\boldsymbol{r}_j\,=\,-\,\frac{\partial}{\partial\boldsymbol{r}'}\cdot\boldsymbol{I}_{k+1}(t,\boldsymbol{r}'-\boldsymbol{r})\;\phi^k(\boldsymbol{r}') \tag{40}$$

with vector functions $\boldsymbol{I}_{k+1}(t,\boldsymbol{R})$ formally just defined by this expression. Substituting it into (39) and then (39) to (37) and twice applying integration by parts, we obtain

$$\frac{\nu(t,\boldsymbol{r})}{\nu_0} - 1 \;\rightarrow\; \int W_1(t,\boldsymbol{r}'-\boldsymbol{r})\,\phi(\boldsymbol{r}')\,d\boldsymbol{r}' + \sum_{k=1}^{\infty}\frac{1}{(k+1)!}\int w_{k+1}(t,\boldsymbol{r}'-\boldsymbol{r})\;\phi^{k+1}(\boldsymbol{r}')\,d\boldsymbol{r}' \tag{41}$$

with one more set of (scalar) functions defined by

$$w_{k+1}(t,\boldsymbol{R}) \,=\, -\,\partial \boldsymbol{I}_{k+1}(t,\boldsymbol{R})/\partial \boldsymbol{R} \tag{42}$$

According to these definitions and again to identities (25) and (28), with addition of (36), the new functions must obey restrictions

$$\int w_{k+1}(t,\boldsymbol{R})\; d\boldsymbol{R} \,=\, 0 \quad , \qquad w_{k+1}(0,\boldsymbol{R}) \,=\, 0 \quad , \tag{43}$$

$$\int \boldsymbol{I}_{k+1}(t,\boldsymbol{R})\; d\boldsymbol{R} \,=\, 0 \quad , \qquad \boldsymbol{I}_{k+1}(0,\boldsymbol{R}) \,=\, 0 \tag{44}$$

We passed over in silence that in fact transition from left-hand to right-hand side of (40) needs in one more property of cumulants $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$, namely, that they remain bounded under the Boltzmann-Grad limit. In terms of corresponding distribution functions, $F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r}) < \text{const}$ when $l_0 \rightarrow 0$ (at fixed t and $\boldsymbol{R} = \boldsymbol{r}' - \boldsymbol{r}$). From the point of view of the BBGKY equations, hardly this statement might give rise to doubts. What is for a scale of smoothness of $\phi(\boldsymbol{r})$ needed in (40) at $l_0 \rightarrow 0$, possibly, it must stay at the level of mean free path λ.

VI. DISCUSSION AND RESUME

It seems quite obvious that just now considered distribution functions $F_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$, corresponding cumulant functions $W_{k+1}(t,\boldsymbol{r}'|\boldsymbol{r}_1,\dots,\boldsymbol{r}_k|\boldsymbol{r})$ and their children $w_{k+1}(t,\boldsymbol{R})$ are in very close conceptual and mathematical connections with the special full phase space distribution functions $A_n(t,\boldsymbol{R},\boldsymbol{v}_1,\dots,\boldsymbol{v}_n)$, considered in [9] in the framework of the "collisional approximation" to BBGKY equations (in [10] they were renamed to $F_n(t,\boldsymbol{R},\boldsymbol{v}_1,\dots,\boldsymbol{v}_n)$), and their children like $W_n(t,\boldsymbol{R})$ introduced and investigated in [11]. At that, equalities (43) and (44) show that, in opposite to $W_1(t,\boldsymbol{R})$, functions $w_{k+1}(t,\boldsymbol{R})$ can not be treated as spatial probability distributions. The lowercase letter just accentuates this difference between $w_{k+1}(t,\boldsymbol{R})$ and true (non-negative) distribution functions $W_n(t,\boldsymbol{R})$. By implication, however, $w_{k+1}(t,\boldsymbol{R})$ and $W_{k+1}(t,\boldsymbol{R})$ reflect similar properties of statistical ensemble. More careful analysis, with the help of the BBGKY equations, shows that sooner the first is proportional to time derivative of the second.

Detailing of these connections is interesting subject for separate work. At present, the thing what is most important for us is that in essence both the old and new of the enumerated mathematical objects represent the same dynamical multi-particle correlations. This notion, as combined with formally exact relation (26) or its limit form (41) under comprehension of its *non-linearity*, leads to principal conclusion that the specific multi-particle distribution functions like $A_n(t,\boldsymbol{R},\boldsymbol{v}_1,\dots,\boldsymbol{v}_n)$, half-intuitively introduced and argued in [9] as autonomous complementary (in addition to the one-particle distribution) characteristics of spatially non-uniform statistical ensemble, now appear as formally well-grounded concepts of statistical kinetics. After above consideration (and close e-print [16]), we have visual demonstration of primitiveness of classical kinetic theories.

Concretely, to resume, we demonstrated existence and significance of dynamical multi-particle correlations what follow from the above derived chain of relations (18) $\rightarrow$ (26) $\rightarrow$ (37)+(39) $\rightarrow$ (41). These relations connect, on one (left) hand,

coefficient functions in series expansion of space-time-varying density of spatially non-uniform and thus thermodynamically non-equilibrium gas over its initial density perturbation, and, on the other (right) hand,

cumulant correlation functions which describe creation and evolution of joint many-particle correlations in thermodynamically equilibrium gas which is spatially uniform in thermodynamical sense but at the same time non-uniform in statistical sense owing to "informational perturbation" of gas by information about initial position of one of its particles (termed "test particle").

On the left, initial density perturbation can be arbitrary large, therefore generally it causes strongly non-linear relaxation process, may be even producing shock waves. This means that on the right-hand side all many-particle correlations really exist (remaining essentially different from zero even under Boltzmann-Grad limit).

At it was shown earlier, first in [9] (see also [10]) and then in [11, 12], these correlations are not things in themselves. In opposite, they directly govern a true evolution of the one-particle distribution function (as referred to the "test particle") and thus determine statistics of self-diffusion (Brownian motion) of gas particles, at that involving possibility of what can be characterized as "violation of the law of large numbers" and "1/f fluctuations in particles diffusivity and mobility". The salt of such statistical characteristics of gas as $A_n(t,\boldsymbol{R},\boldsymbol{v}_1,\dots,\boldsymbol{v}_n)$ or $w_{k+1}(t,\boldsymbol{R})$ is that they were irretrievably lost by conventional kinetics based on the Boltzmann's "Stosshalansatz" or, in other words, "molecular chaos" hypothesis. Due to it, in the Boltzmannian kinetics (at least under the Boltzmann-Grad limit) the one-particle distribution function ascended the throne as the only and exhaustive characteristics of gas state, since none interparticle correlations have survived.

Immediate cause of the loss of these correlations at very beginning of kinetics was the way of thinking when one identifies actual independency of particles or events, in the sense of dynamics, and their formal independency, in the sense of mathematical probability theory (see Introduction). The "molecular chaos" hypothesis is natural product of such thinking about "collisions" of particles. However, this hypothesis immediately pops up as inadmissible one when concept of "collision" is based on such principal property of dynamics as conservation of phase volume and consequently conservation of probabilities (for more explanations see [9, 10] and references therein). The pay for this dynamical approach to collisions is absence of definite limits for their relative frequencies, i.e. uncertainty of a priory "probability of (one or another sort of) collisions" (if paraphrasing Bernoulli's words [1], already mentioned in the Introduction, probability never turns to certainty). That is why "molecular chaos" hard stays unproved (see O. Lanford's article [18] and related comments in [10]).

Up to now, such revision of kinetics was concerning statistics of self-diffusion in equilibrium gas only [9–11]. But if continued to non-equilibrium situations it must somehow touch also kinetics in its usual sense of the fluid mechanics. Intuition would like to suggest that probable changes in this field must be not revolutionary. If that is the case, then it is interesting, for instance, to clarify what information about the Brownian motion follows from the mentioned relations if one substitutes there results of conventional kinetics, e.g. those coming from the Boltzmann equation (though, to the best of my knowledge [17], none exact spatially non-uniform solution to this equation is known). In any case, we have found new incentives to critical review of kinetics.

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