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Comment on “Lagrangian formulation and symmetrical description of liquid dynamics”

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Trachenko [Trachenko, Phys. Rev. E **96**, 062134 (2017)] has argued for creating a Lagrangian formulation of liquid dynamics. This Comment shows that the proposed two-field Lagrangian does not describe dynamics of liquid in a correct way. Other claims on the symmetric description of liquid dynamics and on the similarity of Maxwell and Frenkel characteristic times are discussed.

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In a recent paper [1] the author claimed that he was able to represent a Lagrangian formulation of liquid dynamics and that “the proposed Lagrangian provides new mechanisms for the interplay between the dissipative and mass terms.” Furthermore, the author claimed he “demonstrated the equivalence of hydrodynamic and solid-state approaches to liquids,” which is represented as “symmetry of liquid description.” In this Comment we discuss statements and results of Ref. [1] that we disagree with.

(1) The essence of liquid dynamics is in the viscoelasticity of liquid; on macroscopic scales the density fluctuations in liquid are constrained by local conservation laws, while on microscopic scales the liquid behaves as an elastic medium. Numerous theories have been applied to the analytical description of the viscoelastic transition in liquids [2–7]. However, in Ref. [1] the author did not start from the whole set of balance equations for conserved quantities, but considered a macroscopic equation for the velocity field only and made use of the well-known empiric *extrapolation* by Frenkel [2,4], which makes sense only for transverse dynamics. Therefore, in making a general claim about the formulation of a general symmetric description of liquid dynamics the author is not correct—the given and well-known Frenkel formulation is solely for transverse components of current fluctuations. The Frenkel extrapolation is equivalent to the treatment of a system of two balance equations: for transverse components of the mass current and of the stress tensor. Furthermore, the relaxation time of longitudial components of the stress tensor coupled to density fluctuations is $\tau_L = D_L/(c_\infty^2 - c_s^2)$ [6–8], with D_L, c_∞, and c_s being longitudinal kinematic viscosity, high-frequency and adiabatic speeds of sound, respectively, in contrast to the case of transverse dynamics. Using the formulation of the Lagrange description of liquid dynamics proposed in Ref. [1], it is impossible to describe longitudinal dynamics with adiabatic propagation of macroscopic sound in liquids and the positive dispersion of acoustic excitations. The general case of liquid dynamics must be formulated via a set of equations containing additionally a continuity equation for density fluctuations and explicit treatment of heat fluctuations.

(2) The author of Ref. [1] admits that in his macroscopic equation “τ formally is Maxwell relaxation time” as it must be according to the well-known theories of transverse excitations in liquids [2,8]. However, after that he makes the claim that “at the microscopic level, Frenkel’s theory approximately identifies this time with the time between consecutive diffusive jumps in the liquid.” Note that in making this claim the author does not show any numeric evidence, although the numerical checks would immediately clarify the possibility to replace the *collective* Maxwell relaxation time τ_M by the *single-particle* Frenkel time τ_F. Moreover, all the “Frenkel line” approach to dynamics of supercritical fluids proposed in Ref. [9] is based on the supposedly fundamental role of the single-particle Frenkel time in collective dynamics. Despite knowing about a large difference between the Maxwell and Frenkel times [10] the author advocates the incorrect identification of τ_M by τ_F in the transverse dynamics. Even a simple application of the Stokes-Einstein ratio provides evidence of an order of magnitude difference between the slow Frenkel time and the fast Maxwell relaxation for dense liquids [11].

Here we show the estimation of the Frenkel time τ_F according to its definition in Ref. [9] as “the average time it takes an atom to move the average interparticle distance” using the mean square displacements $\langle R^2\rangle(t)$ and pair distribution functions $g(r)$, obtained in molecular dynamics (MD) simulations, for the same Ar fluids at $T = 205$ and 450 K and a pressure of 10 kbar as in Ref. [1]. The MD simulations were performed on a system of 2000 particles interacting via a standard Lennard-Jones potential of Ar—first in the NPT ensemble and then, upon equilibration at a given pressure, in the NVE ensemble. It follows from our Fig. 1 [see the locations of straight black lines which correspond to the time needed for the particle to reach $\langle R_{\max}^2\rangle$ with $R_{\max}$ being the position of first maximum of $g(r)$] that the Frenkel time τ_F is equal for Ar fluid to 17.184 ps at $T = 205$ K and to 2.645 ps at $T = 450$ K. For comparison, the Maxwell relaxation times, obtained from shear viscosity η calculated from the Green-Kubo integration of the shear stress autocorrelation function and the high-frequency shear modulus G_∞ (see Ref. [12] for methodology of calculations of G_∞), have much smaller values: $\tau_M = 0.285$ ps at 205 K and $\tau_M = 0.092$ ps at 450 K. These values and their ratios with the Frenkel time τ_F are in agreement with similar estimations in Ref. [10] for supercritical Ar at 280 K and in Ref. [11] for supercritical Ne.

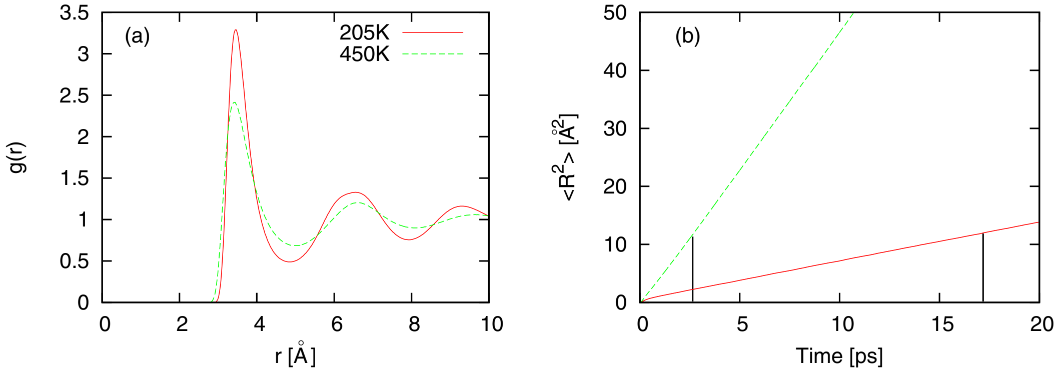

FIG. 1. Frenkel time estimation for supercritical Ar at 205 and 450 K and corresponding pressures as in Ref. [1] via pair distribution functions (a) and mean square displacements (b). The vertical bars in panel (b) show the time needed for particles to reach $\langle R^2_{\max}\rangle$ with $R_{\max}$ being the position of first maximum of $g(r)$, shown in panel (a).

(3) Let us consider the Lagrangian formulation of liquid dynamics proposed by Trachenko [1]. His Eq. (10) (here and hereafter references to the equations of Ref. [1] are implied) for the description of the transverse flow of viscous liquid describes a one-dimensional propagation of damped waves of the transverse velocity field $v(x,t)$ with the the shear wave velocity c. In view of the present dissipative term $\sim\partial v/\partial t$, the equation is not time reversible, and thus the construction of a variational formulation for it meets some problems. One possible way to solve this task is to embed Eq. (10) of Ref. [1] into some self-adjoint set of equations [13]. Such a way is used in some problems of electrodynamics, thermal elasticity, etc. [14], and Ref. [1] follows this way too. For this purpose the author introduces an auxiliary field, $\phi_2(x,t)$ [apart from $\phi_1(x,t)\equiv v(x,t)$], and postulates for this pair of fields the equations (22). Since the set (22) is self-adjoint, the equations can be considered as Euler-Lagrange equations. The corresponding Lagrangian function can be built by means of a known procedure [15]; such a function is presented in Eq. (21).

The proposed Lagrangian description is invariant under time translations $t\mapsto t+\lambda$, $\lambda\in R$. Consequently, there exists a corresponding Noether conserved quantity, called in Ref. [1] the energy. For the solution $\phi_1=\phi_0\exp(-\frac{t}{2\tau})\cos(kx-\omega t)$ to be a damped wave and the dual amplifying wave $\phi_2(x,t)=\phi_1(-x,-t)$, see Eq. (23) in Ref. [1], the energy $E=\phi_0^2\omega^2$ (which actually is a mean energy density) is positive.

Note that the equality (23) implies a partial choice of solution made by hand. Thus the following conclusion is erroneous: "...a Lagrangian formulation of a nonreversible dissipative process necessitates two waves moving in the opposite space-time directions, resulting in this sense in the reversibility of the Lagrangian description."

Actually, the Lagrangian is invariant under a time reversal, i.e., the transformation,

$$t\mapsto -t,\quad \phi_1\mapsto\phi_2,\quad \phi_2\mapsto\phi_1,$$

even without the constraint (23), i.e., when both degrees of freedom ϕ_1 and ϕ_2 are independent of one another. One can select the solution $\phi_2=0$ and arrive at $E=0$ whatever solution for ϕ_1 is chosen. Another choice, $\phi_2(x,t)=-\phi_1(-x,-t)$ instead of Eq. (23), yields a negative value for E. Thus the energy density E (or the Hamiltonian H) is not a useful quantity and has little to do with physical properties of the transverse velocity field $\phi_1(x,t)=v(x,t)$. One should use instead another definition, for example, the density of kinetic energy $\epsilon=\rho v^2/2\equiv\rho\phi_1^2/2$, where ρ is a mass density. The mean value of ϵ reveals dissipation, but then the physical treatment of the field ϕ_2, which reveals the gain of the analogous quantity $\rho\phi_2^2/2$, is questionable.

Nonphysical features of the Lagrangian and Hamiltonian descriptions proposed in Ref. [1] will be more evident from the following discussion. When analyzing wave solutions of his Eq. (10), the author notes the similarity of the dispersion relation (12) to that of the tachyon field [16]. Actually, his Eqs. (10) and (22) and the Lagrangian (21) are likely related to the tachyon theory much closely than to the theory of liquid. Indeed, within the Lagrangian formalism an arbitrary (except nondegenerate, smooth, etc.) transformation of variables is admissible. Let us perform two consequent substitutions of the variables ϕ_1 and ϕ_2. The first one,

$$\phi_1=e^{-t/2\tau}\tilde{\phi}_1,\quad \phi_2=e^{t/2\tau}\tilde{\phi}_2,$$

removes the "dissipative" term (20) from the Lagrangian (21). The second one,

$$\tilde{\phi}_1=\frac{1}{\sqrt{2}}(\psi_1+\psi_2),\quad \tilde{\phi}_2=\frac{1}{\sqrt{2}}(\psi_1-\psi_2),$$

splits the Lagrangian into a difference of two free-field parts:

$$L=\frac{1}{2}\left[\left(\frac{\partial\psi_1}{\partial t}\right)^2-c^2\left(\frac{\partial\psi_1}{\partial x}\right)^2+\frac{1}{4\tau^2}\psi_1^2\right]$$
$$-\frac{1}{2}\left[\left(\frac{\partial\psi_2}{\partial t}\right)^2-c^2\left(\frac{\partial\psi_2}{\partial x}\right)^2+\frac{1}{4\tau^2}\psi_2^2\right].$$

Each line describes the free Klein-Gordon field with imaginary mass $m=i/2\tau$, i.e., a scalar tachyon field [16,17]. The corresponding energy is a difference of partial energies related to the ψ_1 and ψ_2 fields, and thus it can acquire an arbitrary positive or negative value; i.e., it is not positively defined. In other words, such a system has no vacuum, and thus it is nonphysical. The alternative use of a complex field suggested in Ref. [1], say $\phi=\frac{1}{\sqrt{2}}(\phi_1+i\phi_2)$, instead of the real fields ϕ_1 and ϕ_2, retains this nonphysical peculiarity since a choice of variables does not affect the general properties of the Lagrangian.

In summary, the Langrangian formulation of liquid dynamics claimed in Ref. [1] is in fact incorrect, because it is focused solely on the transverse velocity field and not on liquid dynamics in general. Furthermore, as we have shown, the proposed two-field Lagrangian does not correspond to the description of "two waves moving in the opposite space-time directions," but rather corresponds to the two dynamic processes, one of which is losing kinetic energy (in the conventional sense) and the second of which is unphysical with gaining the energy. The proposed Hamiltonian description is ill-posed since it is based on the conserved energy which is neither additive nor positively defined. All that has nothing in common with processes in liquid dynamics. Another proposal by Trachenko to interpret the collective Maxwell relaxation time via the single-particle Frenkel time is incorrect, because of a huge difference between them, which excludes the definitive role of the Frenkel time in the emergence of the gap for transverse excitations [18].

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