Quantum fluids in nanoporous media - effects of the confinement and fractal geometry

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The zero-point energy of atoms of two stable isotopes of helium - $^4$He and $^3$He - is high enough to prevent their solidification even at extremely low temperatures without application of external pressure. So they belong to the class of quantum fluids with strong correlations between atoms but their behavior is quite different at low temperatures. The first one represents a Bose-system and shows superfluid transition at 2.17 K while atoms of $^3$He are governed by Fermi-statistics and superfluid transition can be observed only at much lower temperatures (about 1 mK) when the pairing of two atoms occurs. But even at helium temperatures (1.5-4.2 K) the effects of quantum statistics for $^3$He atoms becomes pronounceable especially in nanoscale confinement (nanoporous media, thin adsorbed layers on solid substrates) and in the presence of nanoscale disorder induced for example by silica aerogel strands. In recent years the problem of correct description of quantum fluids in the confined geometry at nanoscale length has emerged. It has been recognized that the quantum fluids at these circumstances can be considered as a new state of quantum matter due to close values between characteristic lengths for these quantum liquids and the size of geometrical confinement and significant contribution from the surface atoms. So one has to apply new physics to describe such systems with taking into account their complex nature. For example, last two years the attempts to develop the fractionalized two-fluid hydrodynamics for nanoporous media with fractal dimensions have been made. The actuality of such new hydrodynamics becomes very clear for the last development in chemical synthesis of different kind of aerogels with nanopore structure as well as numerous studies of nanoporous substances. One of the interesting obtained results that density waves (the first sound) and temperature waves (the second sound) become strong coupled even in the absence of viscosity, so it is purely geometric effect of fractal space of nanopores. In the present report we will review the procedure, results and discuss the issues for this approach.

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1. Introduction

The zero-point energy of atoms of two stable isotopes of helium - $^4$He and $^3$He - is high enough to prevent their solidification even at extremely low temperatures without application of external pressure. So they belong to the class of quantum fluids with strong correlations between atoms but their behavior is quite different at low temperatures. The first
one represents a Bose-system and shows superfluid transition at 2.17 K while atoms of 3He are governed by Fermi-statistics and superfluid transition can be observed only at much lower temperatures (about 1 mK) when the pairing of two atoms occurs. But even at helium temperatures (1.5-4.2 K) the effects of quantum statistics for $^3$He atoms becomes pronounceable especially in nanoscale confinement (nanoporous media, thin adsorbed layers on solid substrates) and in the presence of nanoscale disorder induced for example by silica aerogel strands. In recent years the problem of correct description of quantum fluids in the confined geometry at nanoscale length has emerge\textsuperscript{1,2,3,4}. It has been recognized that the quantum fluids at these circumstances can be considered as a new state of quantum matter due to close values between characteristic lengths for these quantum liquids and the size of geometrical confinement and significant contribution from the surface atoms. So one has to apply new physics to describe such systems with taking into account their complex nature. For example, last two years the attempts to develop the fractionalized two-fluid hydrodynamics for nanoporous media with fractal dimensions have been made\textsuperscript{5,6}. The actuality of such new hydrodynamics becomes very clear for the last development in chemical synthesis of different kind of aerogels with nanopore structure as well as numerous studies of nanoporous substances. One of the interesting obtained results that density waves (the first sound) and temperature waves (the second sound) become strong coupled even in the absence of viscosity, so it is purely geometric effect of fractal space of nanopores.

In the present report we will review the procedure, results and discuss the issues for this approach. The rest of this paper is organized as follow: In Section 2 we make a little review of existed model of superfluid bulk helium-4. In the next Section 3 properties and peculiarities of aerogel are discussed. In Section 4 possible fractionalization of hydrodynamical model is shown. In Section 5 we speculate about another way of considering complex fractal structure of aerogel for description of superfluid helium behavior. In Section 6 we make a summarization and some novel results are given.

2. Existed model

There are several approaches to describe the behavior of bulk superfluid helium-4. For example, two-fluid model\textsuperscript{7,8}, the microscopic description based on the Gross-Pitaevskii equation\textsuperscript{9,10} and others.

2.1. Two-fluid model

The most famous model is two-fluid model (TFM) proposed by Landau in the past century. In this model superfluid helium with density $\rho$ is considered as a two component system: an uncondensed, normal component with density $\rho_n$ with velocity $v_n$ and a condensed, superfluid component characterized by density $\rho_s = \rho - \rho_n$ with velocity $v_s$. Without dissipative terms one finds the following system of hydrodynamical equations (so-called Landau-Khalatnikov equation\textsuperscript{7,8}).
\[ \frac{d\rho}{dt} + \text{div}(\rho_n \mathbf{v}_n + \rho_s \mathbf{v}_s) = 0, \quad (1) \]
\[ \frac{\partial}{\partial t} (\rho_n \mathbf{v}_n + \rho_s \mathbf{v}_s)_i + \frac{\partial}{\partial x_k} \Pi_{ik} = 0, \quad (2) \]
\[ \frac{dS}{dt} + \text{div} S \mathbf{v}_n = 0, \quad (3) \]
\[ m \frac{\partial \mathbf{v}_s}{\partial t} + m(\mathbf{v}_s \nabla) \mathbf{v}_s + \nabla \mu = 0, \quad (4) \]

The entropy is denoted by \( S \), the mass of an atom by \( m \), the pressure by \( p \) and the chemical potential by \( \mu \). In \( (2) \) the stress tensor \( \Pi_{ik} \) is given by

\[ \Pi_{ik} = \rho_n v_{ni} v_{nk} + \rho_s v_{si} v_{sk} + p \delta_{ik}, \quad (5) \]

and the sum over the index \( k \) is assumed.

One can obtain from these equations that there are two type of collective motion: motion of the fluid where both components move in phase is called first (ordinary) sound, while second sound is associated with out of phase motion of the two components. The above TFM equations describe the flow properties of superfluid \(^4\)He in the bulk of the liquid. They are not valid in the immediate proximity of walls and free surfaces, where the effects of so-called “healing” are encountered. Moreover, Landau-Khalatnikov equations cannot be used in the vicinity of \( \lambda \) point, where large variations of superfluid density in space as well as in time may occur.

For taking into account such “healing” effects, which has quantum nature, one can consider some models derived from nonlinear Schrödinger equations.

### 2.2. Nonlinear Schrödinger equation

The Schrödinger equation itself is applicable only at \( T = 0K \) and small interaction between helium atoms, when all of them are in condensed state. In that case we can introduce macroscopic wave function of condensate. Than velocity field is determined as probability flow of this wave function and it is proportional to phase gradient. He-II is dense fluid of strongly interacted bosons and applicability of NLS is questionable, but it helps us to elucidate basic peculiarities of superfluid dynamics.

If one writes short range pair interaction between particles in form \( U(x-x') = U_0 \delta(x-x') \), \( N_0 = \int |\Psi|^2 \, d^3x \) - number of particles in condensate and \( V_0 \) - total condensate volume, than one can write nonlinear Schrödinger equation in form of

\[ i \frac{\partial \Psi}{\partial t} = -\frac{1}{2} \nabla^2 \Psi + |\Psi|^2 - \Psi. \quad (6) \]

Here average density \( \rho_0 = N_0/V_0 \) is set to unity, unity of length is \( \hbar/\sqrt{\rho_0 U_0} \) and unity of time \( \hbar/(\rho_0 U_0) \).
After substitution $\Psi(x,t) \equiv \sqrt{\rho(x,t)} e^{i\theta(x,t)}$ and disparting real and imaginary parts in one can obtain

$$\frac{\partial \rho}{\partial t} + \nabla \cdot \rho \mathbf{v} = 0 \quad (7)$$

$$\frac{\partial \mathbf{v}}{\partial t} + \nabla v^2 + \frac{1}{\rho} \nabla \frac{\rho^2}{2} = -\nabla \left( \frac{(\nabla \rho)^2}{8\rho^2} - \frac{\nabla^2 \rho}{4\rho} \right), \quad (8)$$

where $\mathbf{v} \equiv \nabla \theta$. Except for high-order derivatives in right hand side of equation (8), which can be omitted in hydrodynamical limit, system of equations (7)-(8) is equivalent to Euler equation for nonrotating ideal fluid with pressure defined as $p(\rho) \equiv \rho^2 / 2$. Pressure depends only on density $\rho$ because fluid is considered at absolute zero.

For considering dynamics of He-II at nonzero temperature, Hills and Roberts have improved TFM by introducing additional terms, that is proportional to the gradient of superfluid component. In that case equation (4) can be rewritten as

$$\frac{\partial \mathbf{v}_s}{\partial t} + \nabla \left( \frac{v^2}{2} + \mu \right) = \nabla \left( \eta(\rho_s) \nabla^2 \rho_s + \frac{1}{2} \frac{d\eta}{d\rho_s} (\nabla \rho_s)^2 \right), \quad (9)$$

where $\eta(\rho_s)$ - some undefined function; stress tensor has to be modified and rewritten as

$$\Pi_{ij} \equiv \rho_n v_i v_j + \rho_s v_i v_j + \eta \nabla_i \rho_s \nabla_j \rho_s + \left( p - \eta \rho_s \nabla^2 \rho_s - \frac{1}{2} \frac{\rho_s \eta}{\rho_n} (\nabla \rho_s)^2 \right) \delta_{ij}. \quad (10)$$

New terms that include spatial derivatives of $\rho_s$ are responsible for macroscopic quantum effects, like healing length.

2.3. Correlated density matrix theory

Another way for describing quantum strongly correlated system (superfluid helium is just one example of such systems) is correlated density matrix theory that provides the method of choice to analyze the microscopic structure of strongly correlated quantum fluids in thermal equilibrium. The main point of this theory is concept of renormalized bosons and fermions. Their mass can depend fully on temperature, density and wave vector. So one may discuss the properties of strongly correlated quantum fluids at finite temperatures with reference to the background gas of renormalized free particles.

2.4. Extended irreversible thermodynamics

Recently the extended irreversible thermodynamics has been applied to describe He-I and He-II. It is shown that the behavior of helium II can be described by means of an extended thermodynamic theory where four fields, namely density, temperature, velocity, and heat flux are involved as independent fields. This model is able to explain the propagation of the two sounds that are typical of helium II, and the attenuation calculated for such sounds is in agreement with the experiment results. After some approximations, this model is reduced to TFM, so it can be considered as some generalization of TFM.
3. Aerogel

Silica aerogels are synthesized via a sol-gel process and hypercritical drying which enable production of tenuous solids with porosity $\phi$ as large as 99.8% and unique acoustic properties. Silica aerogel are known to be good examples of fractal materials. A revealed by small-angle x-ray-scattering (SAXS) experiments or small-angle neutron-scattering (SANS) experiments, they are made of a disorder, but homogeneous, array of connected fractal clusters resulting from the aggregation of primary particles. The analysis of the wave-vector dependence of the scattering intensity $I(q)$ has permitted the determination of two characteristic length scales which are the average size $a \approx 10$ Å of the particles and the average size of the clusters $\xi \approx 100$ Å. At length scales from $a$ to $\xi$ silica aerogels show a fractal behavior.$^{[14]}$

The computational confirmation for cluster structure of aerogel has been obtained by modeling as well as by the geometrical analysis of the diffusion limited cluster-cluster aggregation.$^{[15,16]}$

Also it has been demonstrated$^{[3]}$ that it is long-correlated structure of aerogel that makes an essential influence on liquid $^4$He behavior near $\lambda$-point.

3.1. Behavior of $^4$He inside aerogel

There has been considerable interest in the behavior of superfluid $^4$He in the presence of a random disorder induced by highly open porous media, like aerogel. Understanding the results of acoustic experiments is important when dealing with porous media. Use of liquid $^4$He offers unique advantages due to the existence of the superfluid phase with more than one sound mode. In a porous media where the normal component is clamped by its viscosity and only the superfluid component can move, fourth sound (relative motion of the superfluid and normal fluids) propagates and can be used to determine the superfluid fraction.

The high-porosity aerogels are so soft that the aerogel matrix and the clamped normal fluid moves as the results of pressure and temperature gradients, unlike other porous media. This results in sound modes intermediate between first and fourth sound and a second-sound-like mode. In that case, proposed by Biot theory of acoustic propagation in porous, fluid filled, macroscopically homogeneous and isotropic media, is not applicable.

It is very interesting to study the possible influence of geometrical confinement with fractal dimensionality on the flow properties of superfluid $^4$He in the framework of TFM. In order to shed a light on geometrical factor itself one can neglect here by quantum healing and any dissipative processes.

3.2. Nonextensivity

So hereafter we suppose that effectively aerogel can be considered as a cluster with a fractal mass dimension$^{[17]}$ the nanopores of which are filled in by liquid helium.

Note that thermodynamic limit conditions are violated for helium atoms inside nanopores because of a huge inner pore surface of aerogel (up to 3000 m$^2$/g for an aerogel with density 2 mg/cm$^3$). Namely the ratio of total number of helium atoms $N$ to the
total cluster volume $V$ is not constant at $N, V \to \infty$ and nonextensivity of physical properties takes place for helium atoms in two nanopores. In this case the methods of nonextensive thermodynamics\cite{18} should be applied to construct the two-fluid hydrodynamic model and non-extensive entropy like Tsallis entropy should be introduced. We have to note here that beyond thermodynamic limit even the usual Boltzmann-Gibbs entropy becomes non-additive, but the additivity is restored when thermodynamic limit conditions are taken\cite{19}.

4. Fractionalized and nonextensive hydrodynamical model

In general case a macroscopic quantity $Q(A, B)$ associated with the total system may be expressed in terms of the same quantity associated with the subsystems, $Q(A)$ and $Q(B)$\cite{20}.

$$Q(A, B) = f_{\lambda Q}[Q(A), Q(B)],$$

where $f_{\lambda Q}$ is a symmetric bivariate function depending on a constant $\lambda_Q$. Of course, for given quantity $Q$ there exist many functions which satisfy the composability property (11). However, additional assumptions drastically reduce their number. For example, the thermodynamic equilibrium may be used as a constraint on the form of $f_{\lambda Q}$ in Eq. (11). For energy $E_{\lambda}$\cite{21} and entropy $S_q$\cite{22} of helium inside nanopores it leads to

$$E_{\lambda}(A + B) = E_{\lambda}(A) + E_{\lambda}(B) + \lambda E_{\lambda}(A)E_{\lambda}(B),$$
$$S_q(A + B) = S_q(A) + S_q(B) + qS_q(A)S_q(B),$$

where $\lambda$ and $q$ are parameters of nonextensivity and are determined by the properties of system. Suppose that a local equilibrium between liquid $^4$He in different nanopores takes place. So we can re-define such thermodynamics quantities as temperature and pressure in form of\cite{23}

$$T_{\text{phys}} = \frac{1 + qS_q}{1 + \lambda E_{\lambda}} \left( \frac{\partial E_{\lambda}}{\partial S_q} \right)_V,$$
$$p_{\text{phys}} = \frac{T_{\text{phys}}}{1 + qS_q} \left( \frac{\partial S_q}{\partial E} \right)_V.$$

Further, it is possible to introduce the spatial pressure and density distributions in fractal cluster as it has been made in our previous work\cite{5}

$$p_{\text{phys}}(r) = p_f(r)\chi_p(r),$$
$$\rho_{\text{phys}}(r) = \rho_f(r)\chi_p(r),$$

where $r$ is distance from center of fractal cluster, $p_f(r)$ is distribution of pressure in pore, $\chi_p(r)$ is a fractal factor-function. In the case of Euclidean space with $D = 3$ this factor-function should be equal to unit: $\chi_p(r) \equiv 1$. Such kind of fractionalization procedure can be applied to any thermodynamic quantity $A$ i.e.

$$A = A_f(X, Y, Z, \ldots)\chi_A(r),$$
$$\lim_{D \to 3} \chi_A = 1.$$
From the defined thermodynamic quantities (14,15) and eqn. (16) the fractal factor-functions for energy, entropy and temperature are derived

$$\chi_E = \chi_S = \frac{\chi_p}{1 + \lambda E_f(1 - \chi_p)}, \quad (20)$$

$$\chi_T = \frac{1 + qS_f\chi_S}{1 + qS_f} \frac{1 + \lambda E_f}{1 + \lambda E_f \chi_E}. \quad (21)$$

Because all thermodynamic quantities should be expressed in terms of physical (observable) variables, one can propose the following definition of the generalized free energy

$$F = E - T_{phys}S_{phys} = E - \left( \frac{\partial E}{\partial S_{phys}} \right)_{S_{phys}}. \quad (22)$$

which represents no more than the Legendre transformation. So the entropy differential equals to

$$dS_{phys} = \frac{1 + \lambda E_f}{1 + qS_q} dS_q. \quad (23)$$

In the first order with respect to $\lambda E_f$ and $qS_q$ the entropy can be written as:

$$S_{phys} = \frac{1}{q} \ln(1 + qS_q) + \lambda \int E_f(\chi_q)dS_q \approx S_q - qS_q S_q^2 + \lambda \int E_f dS_q. \quad (24)$$

Finally the fractal factor-function for $S_{phys}$ is

$$\chi_{Sp} = \chi_S + \chi_p(\chi_p - 1) \left( \frac{\lambda E_f - qS_f}{2} + \lambda H \right), \quad (25)$$

where $H = \frac{1}{S_f} \int E_f dS_f$. After substitution the fractionalized thermodynamical quantities into TFM one can derive the main system of equations for the fractionalized twofluid hydrodynamic model

$$\frac{\partial \rho_f}{\partial t} \chi_p + \text{div}(\rho_{sf} \chi_p v_s + \rho_{nf} \chi_p v_n) = 0, \quad (26)$$

$$\frac{\partial \rho_f \sigma_f}{\partial t} \chi_{Sp} + \text{div}(\rho_f \sigma_f \chi_{Sp} v_n) = 0, \quad (27)$$

$$\rho_{sf} \frac{\partial v_s}{\partial t} = -\frac{\rho_{sf}}{\rho_f} \nabla(p_f \chi_p) + \rho_{sf} \sigma_f \chi_p \chi_{Sp} \nabla(T_f \chi_T), \quad (28)$$

$$\rho_{nf} \frac{\partial v_n}{\partial t} = -\frac{\rho_{nf}}{\rho_f} \nabla(p_f \chi_p) - \rho_{nf} \sigma_f \chi_p \chi_{Sp} \nabla(T_f \chi_T), \quad (29)$$

where $\chi_{Sp} = 1 + (\chi_p - 1) \left( \frac{\lambda E_f - qS_f}{2} + \lambda H \right)$. From eqns. (26,27,28,29) two equations for
waves of pressure and temperature follow

\[ \frac{1}{u_1^2} \frac{\partial^2 p_f}{\partial t^2} = \nabla^2 p_f + 2 \frac{\nabla \chi p \nabla p_f}{\chi p} + \frac{\nabla^2 \chi p p_f}{\chi p}, \]  
(30)

\[ \frac{1}{u_2^2} \frac{\partial^2 T_f}{\partial t^2} = \nabla^2 T_f (1 + (\chi_p - 1)M) + \nabla T_f \left( M \nabla \chi_p - (1 - M) \frac{\nabla \chi_p}{\chi p} \right) + 
+ T_f (qS_f - \lambda E_f) \left( \nabla^2 \chi - \frac{(\nabla \chi p)^2}{\chi p} \right) + 
+ \frac{1}{u_2^2} \rho_f \left( \frac{\partial T_f}{\partial \sigma_f} \right)_{\rho_f} \left( \nabla \chi_p \nabla p_f + \frac{(\nabla \chi p)^2}{\chi p} p_f \right), \]  
(31)

where \( u_1^2 = (\partial p_f/\partial \rho_f)_S \), \( u_2^2 = \rho_f \sigma_f^2 / \rho_f (\partial T_f/\partial \sigma_f)_{\rho_f} \) are the squared first and second sound velocities respectively and \( M = (\lambda E_f + qS_f)/2 + \lambda H, N = (3\lambda E_f - qS_f)/2 + \lambda H \).

On figure 1 the profiles for pressure wave and for the temperature wave induced by it are shown. It is seen from eqn. (31) that the coupling between pressure and temperature waves appears even in the absence of \(^4\)He viscosity and aerogel skeleton inertia, which is undoubtedly the effect of not only fractional dimensionality of nanopore space, but also of the nonextensive nature of thermodynamical quantities for He-II inside nanopores.

5. Fractional nonlinear Schrödinger equation

Let us assume that confinement (aerogel strands) forbid realization of some Feynman’s paths for particular helium atom in contrast to helium atom in the free space. For taking into account such peculiarities one can propose to make use of fractional Schrödinger equation\(^{24}\).

Fractional Schrödinger equation (FSE) is derived from path integration over Levy’s
paths and is given by

\[ i\hbar \frac{\partial \psi(r,t)}{\partial t} = K_\alpha (-\hbar^2 \nabla^2)^{\alpha/2} \psi(r,t) + V(r,t) \psi(r,t). \] (32)

Here Riesz fractional derivative is introduced as

\[ (-\hbar^2 \nabla^2)^{\alpha/2} \psi(r,t) = \frac{1}{(2\pi\hbar)^{3/2}} \int d^3p e^{i\mathbf{p} \cdot \mathbf{r} / \hbar} |\mathbf{p}|^\alpha \varphi(p,t), \] (33)

\[ \varphi(p,t) = \int d\mathbf{r} e^{-i\mathbf{p} \cdot \mathbf{r} / \hbar} \psi(r,t). \] (34)

5.1. Galilean noninvariance of fractional Schrödinger equation

Let us consider transformation of equation (32) when inertial reference frame is changed. With such transformation spatial coordinates and time are changed as

\[ t' = t, \quad r' = r - vt, \]

where \( v \) is some constant relative velocity. Then time and fractional spatial derivative is rewritten as

\[ \partial_t f = \left( \partial_{t'} - v \partial_{r'} \right) f, \] (35)

\[ \left( \nabla_r^2 \right)^{\alpha/2} = \left( \nabla_{r'}^2 \right)^{\alpha/2}. \] (36)

Let us assume that in old reference frame K FSE has form

\[ i\hbar \frac{\partial \Psi(r,t)}{\partial t} = K_\alpha (-\hbar^2 \nabla^2)^{\alpha/2} \Psi(r,t). \] (37)

While changing reference frame to \( K' \), old wave function is written as

\[ \Psi(r,t) = \varphi(r',t') e^{if(r',t')}, \] (38)

where \( \varphi \) denote wave function in new reference frame. Fractional derivative of product is revealed by generalized Leibniz rule:

\[ D_x^\alpha (\varphi e^{if}) = \sum_{n=0}^{\infty} \frac{\Gamma(\alpha + 1)}{\Gamma(\alpha - n + 1)\Gamma(n + 1)} D_x^\alpha \varphi D_x^{-n} e^{if}. \] (39)

After substitution eqn. (38) in (37) with taking into account eqn. (39) one can derive

\[ (i\hbar \partial_{t'} \varphi - \hbar \varphi \partial_{r'} f - i\hbar v \nabla_r \varphi + \hbar v \varphi \nabla_r f) e^{if} = K_\alpha (-\hbar^2)^{\alpha/2} \sum_{n=0}^{\infty} \frac{\Gamma(\alpha + 1)}{\Gamma(\alpha - n + 1)\Gamma(n + 1)} D_{r'}^\alpha \varphi D_{r'}^{-n} e^{if}. \] (40)

For Galilean invariance of eqn. (40) one has to keep only time derivative and spatial fractional derivative of order \( \alpha \). Let us equate to zero coefficient near the rest derivatives of wave function \( \varphi(r,t) \). Then we can obtain that \( v = 0, \partial_{t'} f = 0, \nabla_{r'} f = 0 \), that is \( f(r',t') = \text{const} \). It leads to

\[ r' = r, \]
\[ t' = t, \]
\[ \Psi' = \Psi e^{i\lambda}, \]
where $\lambda$ is some constant. Thus, fractional Schrödinger equation is **Galilean noninvariant**, that is it changes its form with changing inertial reference frame. As a consequence one needs to choose the especial reference frame where, for example, nanoporous media is in a rest as well as to introduce additional potential into two-fluid hydrodynamic model.

6. Conclusion

It was shown that fractionalized set of hydrodynamical equations with taking into account nonextensivity of He-II inside nanopores leads to coupling between the first and the second sounds that appears even in the absence of viscous friction, which is undoubtedly the effect of fractional dimensionality of nanopore space and nonextensive nature of helium droplets. It was proposed that for better description of behavior of superfluid in nanoporous media with complex fractal structure one can use fractional Schrödinger equation. It leads to Galilean noninvariance and as a consequence one needs to choose the especial reference frame where, for example, nanoporous media is in a rest as well as to improve two-fluid hydrodynamic model by introducing there additional potential.

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