Model of fluid flow in nanotube: classical and quantum features

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Abstract. Different applications of nanostructures are related with fluid flows through these systems. In its turn, experiments show that flows in nanostructures have many specific features, which can’t be explained in classical terms only. Particularly, flow through nanotube is extremely fast in comparison with the analogous classical one. We suggest a model based on the possibility of existence of molecular clusters (Frenkel crystallites) in the fluid. There are some experimental evidences of such phenomenon. Under this assumption one needs to take into account quantum effects. Particularly, the boundary condition, which plays the crucial role for the flow in nanostructures, takes the form of sliding condition instead of the adhesion condition for the classical flow. The parameters of the boundary conditions are determined by solving of quantum scattering problem for the particle of the fluid by the wall potential. Main features of the flow are described in the framework of the model. For very narrow nanotubes another phenomena have an influence on the flow—possibility of existence of solitons in nanotube walls. These soliton solutions are similar to Davidov’s solitons in molecular chains. This model of flow is also described.

1. Introduction
Last years, fluid and gas flows in nano-sized domains are intensively studied [1]. At present, there are no general equations of nano-hydrodynamics. Usually, the molecular dynamics is used for computations [2]. As for analytical approaches, the simplest one involves introducing the slip condition at the boundary [3]. There is also a hybrid method, incorporating the continuous approach and the molecular dynamics (which is used to analyze the fluid structure and determine the rheological law) [4]. In [5], fluctuations are taken into account when deriving the hydrodynamic equations. In [6] viscous-elastic behavior of water in nano-sized gaps was experimentally studied. Great increase of effective viscosity of water in a channel of nano-width is observed. For nano-channel flow the molecular structure of the medium plays a crucial role. It is similar to Brownian motion [7] and body motion through rarefied gas [8]. More precisely, flows in nano-channels are influenced by local heterogeneity of molecular structure of the liquid if its size is compared with the channel width. A hypothesis about the existence of locally-ordered structures in liquid is in [9]. During several decades...
scientists discuss whether there are local domains in liquid (crystallites) in which the molecular structure is similar to that for crystal [10]. Investigations of fluid flows in nano-sized domains show that it is strongly influenced by local ordering of nano-sized scale. Experiments ([6], [11]) show that the effective viscosity of water in nano-channel with hydrophilic walls is essentially greater than the corresponding macroscopic value. Calculations in the framework of molecular dynamics [11] show that there are ordered structures (like periodic) of sizes less than nanometer. Computing experiments in [12] lead to appearance of ice-like state in nanotubes of small diameters under normal conditions. Experimental and theoretical investigations of water state in carbon nanotube [13 – 14] show that there is ice-like envelope with liquid water inside in the nanotube. Increasing of effective fluid viscosity via channel diameter was marked in [15] for channels of few micrometers diameters. Influence of walls isn’t evident. In some experiments there are increasing of effective viscosity, but in others the opposite effect takes place. Particularly, the experiments in [16] show that the fluid flow speed through carbon nanotube (of few nanometers diameter) is essentially greater than one calculated in the framework of classical fluid theory. Possible model of such phenomenon – superfluidity in nano-channel – is suggested in [17]. As it was mentioned earlier, the effective viscosity can be both extremely small in some cases and extremely great in other situations (in comparison with its classical value). Such non-trivial correlation between the nanotube diameter and the viscosity, possibly, is related with local ordering in the liquid. The first variant of such approach is in [18], where the model of nanotube flow close to crystallite model is suggested. In the present paper the relation of the effective viscosity of a fluid in nanotube with the dynamics of locally ordered nano-sized clusters is described. The number of such clusters increases if the temperature tends to the crystallization point. The sizes of such clusters are close to the diameters of hydro-silicate nanotubes. For this reason, it is of importance to study the existence of crystal clusters of this kind in a liquid inside nanotubes and the influence of clusters on the mass transport through the tube. Computations based on molecular dynamics (with the Lennard-Jones potential) show that solid nano-sized clusters can exist both in the nanotube and outside it. The clusters (crystallites) can have a size of order 1 nm, which is close to the internal diameter of silicate nanotube. Note that the computed size of crystallite depends on parameters of the interaction potential, i.e., on the chemical composition of the liquid. Moreover, in unbounded domains, the clusters are also nano-sized. In the case of narrow nanotube with elastic walls (e.g., carbon nanotube) there are elastic waves in walls [19]. It has great influence on the flow. We suggest a solvable model of such flow. Particularly, it is shown that wall soliton induces a flow.

2. Flow of liquid with crystallites

We suggest models taking into account the existence of crystallites in the nanotube. The first model takes into account energy transformations. Namely, we determine the power of pressure drop and compare it with the energy loss due to viscosity and melting.

We use quantum mechanical treatment to explain the type of the boundary condition on the inner surface of nanotube

$$v_s|_s - v_s = L_s \frac{\partial v_s}{\partial n}|_s,$$

where \(v_s\) and \(L_s\) are the characteristic velocity and length (so-called sliding velocity and sliding length).

The character of the flow in the nanotube depends on the correlation between the equilibrium size of the crystallite and the nanotube diameter. We try to find the effective viscosity \(\mu_{eff}\) of the fluid in the nanotube which is defined by the following way. Consider the correlation between our nanotube (possibly, containing crystallites) and classical tube (with the Poiseille flow) of the same size and with the same pressure drop and flow rate. The viscosity of the Poiseille flow having the same parameters is called the effective viscosity of the flow in the nanotube. Note that there is some space between the crystallite and the nanotube wall. It is occupied by so-called non-autonomous phase (of width \(h\))
having the properties of a fluid with another viscosity $\mu_0$ [20]. Size correlations lead to a few particular cases. Introduce some notations. Let $D$ be the nanotube diameter, $L$ be its length, $H_e$ be the equilibrium size of the crystallite and $h_e$ be the equilibrium width of the non-autonomous phase.

The character of the flow depends on the existence of the crystallite inside the tube. For small nanotube diameter $(D < 2h_e)$ there is no crystallite. If $2h_e \leq D < H_e + 2h_e$ then there exists a crystallite inside the nanotube but its diameter is less than the equilibrium one. If $H_e + 2h_e \leq D < 2H_e + 3h_e$ then there are not greater than one crystallite in a cross-section of the nanotube. Correspondingly, one obtains three different expressions for the effective viscosity:

$$
\mu_D = \begin{cases} 
\frac{gD^2}{32vL} + \mu_0 \left( \frac{0.5D}{0.5D + L} \left( 1 - \frac{v}{v} \right)^2 \right), & D < 2h_e \\
\frac{gD^2}{32vL} \left( 1 - \left( \frac{D - h_e}{H_e} \right)^2 \right) + \mu_0 \frac{h_e (D - h_e)}{(h_e + L_e)^2} \left( 1 - \frac{v}{v} \right)^2, & 2h_e \leq D < H_e + 2h_e \\
\mu_0 \frac{h (D - h)}{(h + L_e)^2} \left( 1 - \frac{v}{v} \right)^2, & H_e + 2h_e \leq D < 2H_e + 3h_e
\end{cases}
$$

Here

$$
h = \begin{cases} 
h_e, & H_e + 2h_e < D < \alpha H_e + 2h_e, \\
0.5(D - \alpha H_e), & \alpha H_e + 2h_e \leq D < 2H_e + 3h_e.
\end{cases}
$$

We assume that for $H_e + 2h_e < D < \alpha H_e + 2h_e$ the width of the non-autonomous phase preserves ($h_e$) and the cross-section of the crystallite increases from $H_e$ to $\alpha H_e$ ($\alpha$ is some dimensionless parameter). For larger values of $D$ ($\alpha H_e + 2h_e \leq D < 2H_e + 3h_e$), the width of the crystallite preserves ($H = \alpha H_e$) and the width of the non-autonomous phase $h$ increases: $h = 0.5(D - \alpha H_e)$.

The character of obtained viscosity via diameter dependence is in good correlation with the experimental data [15], [16].

3. Sliding boundary condition and surface waves

Boundary conditions play crucial role in nanotube flow. The character of this condition is determined by the quantum interaction between liquid and walls. Consider the corresponding quantum problem. Let one have a nanotube having a periodic set of atoms (centers) at the wall (a cylinder of radius $R$ with $OZ$ axis. These centers are at $N$ vertices of polygons. Polygons are parallel, and the distance between neighbor polygons is $h$. To describe the system we use zero range potential method [24]. Using of $\delta$-interaction allowing one to simplify considerably the procedure of spectral analysis [25, 26]. Correct mathematical description of point-like interaction is given in the framework of the theory of self-adjoint extensions of symmetric operators. Formally, the Hamiltonian $H$ of the system is a perturbation of free Hamiltonian (i.e. the Laplace operator $-\Delta$ by periodic system of zero range potentials which are in the vertices of the above mentioned polygons (nodes):

$$
H = -\Delta + \sum_{m \in Z_n} \sum_{n \in Z_s} \alpha \delta(r - r_{mn}),
$$

where $\alpha$ is the intensity of the perturbation, $r_{mn}$ is the radius-vector of a node having the following cylindrical coordinates: $r_{mn} = (R \cos(\theta n), R \sin(\theta n), s h), \quad s \in Z_n, n \in Z_s, \quad \theta = 2\pi / N$.

The construction of our model is as follows. First, we restrict the Laplace operator onto the set of functions vanishing at the nodes. The restricted operator is symmetric and non-self-adjoint. It has self-
adjoint extensions which gives us the model operator [25]. To choose the particular extension it is necessary to satisfy the condition at the nodes. The continuous spectrum of the model operator contains values corresponding to modes of waveguide type concentrated near the wall of the nanotube. The corresponding wave function has the following form:

\[ \psi(r) = \sum_{n,s} c_{ns} G(r_{ns}, r; E), \]

where the coefficients \( c_{ns} \in \mathbb{C} \), are determined by the “boundary” conditions at the nodes, \( E \) is the energy of the mode, \( G(r_{ns}, r; E) \) is the Green function of the Laplace operator:

\[ G(r, r'; E) = \frac{\exp(-\sqrt{-E} |r - r'|)}{4\pi |r - r'|}. \]

Wave function has the following asymptotics in a neighborhood of each point \( r_{ns} \):

\[ \psi(r) = \frac{a_{-1}}{|r - r_{ns}|} + a_0 + \alpha (|r - r_{ns}|), \quad r \to r_{ns}. \]

The “boundary” condition at the node \( r_{ns} \) gives us the following correlation: \( a_0 = \alpha a_{-1} \Rightarrow \alpha = 0 \). This condition means that for each centre \( r_{ns} \):

\[ \lim_{|r - r_{ns}| \to 0} \frac{1}{|r - r_{ns}|} \frac{\partial}{\partial |r - r_{ns}|} (|r - r_{ns}| \psi) = \alpha. \]

Using well-known asymptotic expansion for \( G(r_{ns}, r; E) \) in a neighborhood of each point \( r_{ns} \), one obtains the following system:

\[ (-\alpha - \sqrt{-E}) c_{ns} + 4\pi \sum_{n',s'(n',s') \neq (n,s)} c_{n's'} G(r_{ns} - r_{n's'}, E) = 0, \quad s = 0, \pm 1, ..., n \in \mathbb{Z}_N. \]

Then, the dispersion equation takes the form

\[ \alpha = -\sqrt{-E} - \tilde{G}(j, q; E), \]

\[ \tilde{G}(j, q; E) = \sum_{n,s \neq (0,0) \in (0,0)} \frac{\exp(-iqs - i\theta jn) \exp(-\sqrt{-E} \sqrt{2R^2(1 - \cos(\theta n)) + h^2 s^2})}{\sqrt{2R^2(1 - \cos(\theta n)) + h^2 s^2}} \]

Now, it is simple to describe the spectrum. Particularly, there are bands corresponding to solutions having the type of surface waves concentrated near the nanotube wall and moving along it. To explain fast flow in nanochannels, which is observed in experiments, some authors (see, e.g., [27]) formally replace the no-slip condition by the sliding one. The parameter, speed of sliding, is chosen empirically. We have shown above that there exist waveguide modes concentrated near the channel wall and have found its parameters. The existence of these surface waves leads to the sliding condition. One can determine the sliding speed by the following way. Consider the scattering of a particle by the wall potential to determine the momentum transmitted to the wall. We use a way analogous to that in [37].

Let \( H_0 \) be the unperturbed Hamiltonian, \( H_1 \) be the perturbation (wall potential \( V \)), \( \psi_a \) be the solution of the scattering problem by the wall potential \( V \), corresponding to the particle with fixed momentum \( p_a \). The momentum operator \( \hat{p} \) commutes with \( H_0 \), \( \Phi_a \) is the eigenfunction of \( \hat{p} \) \((\hat{p}\Phi_a = p_a \Phi_a)\). Solution \( \psi_a \) corresponds to \( \Phi_a \), i.e. satisfies the equation:
\[ \psi_a = \Phi_a + (E - H_0 + i0)^{-1} H \psi_a, \quad (E - H_0) \psi_a = H \psi_a, \]

One can determine the mean value of the operator \((ih)^{-1}[\hat{p}, H]\) = \((ih)^{-1}[\hat{p}, H_i]\):

\[
(\psi_a, (ih)^{-1}[\hat{p}, H_i] \psi_a) = 2\hbar^{-1} p_a \text{Im}(\Phi_a, H \psi_a) + 2\pi \hbar^{-1} \sum_b \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b). \tag{1}
\]

If one takes the identical operator instead of \(\hat{p}\), then (1) transforms to the form

\[
0 = 2\hbar^{-1} \text{Im}(\Phi_a, H \psi_a) + 2\pi \hbar^{-1} \sum_b \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b). \tag{2}
\]

One gets the expression for \(\text{Im}(\Phi_a, H \psi_a)\) from (2) and inserts it into (1). Then,

\[
(\psi_a, (ih)^{-1}[\hat{p}, H_i] \psi_a) = 2\hbar^{-1} \sum_b (p_b - p_a) \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b). \tag{3}
\]

Note that the right hand side of (3) is the mean momentum transmitted during the scattering of \(\psi_a\). Summation over all states \(\psi_a\) gives us the full mean transmitted momentum. We are interested in the longitudinal transmitted momentum \(\delta p\):

\[ \delta p = \gamma(p, -p_a), \quad \gamma = 2\pi \hbar^{-1} \sum_b \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b), \]

\[ p_a = \frac{\sum_b \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b)}{\sum_b \left| \frac{\Phi_b}{\psi_a} \right|^2 \delta(E_a - E_b)}. \]

Using the value of transmitted momentum one can obtain the boundary condition which takes the form

\[ \frac{\partial v}{\partial n} = \frac{1}{L_s} (v - v_s), \quad L_s = \frac{a}{\tau'}, \]

where \(\tau\) is the mean time of molecule transition between two equilibrium states (“Frenkel transition”) [28], \(a\) is the distance between these equilibrium positions, \(m\) is the mass of liquid particle.

### 4. Wall soliton and flow in nanostructure

For the case of narrow channel another effect can play an important role. It should be stressed that boundary condition, wall structure and profile play crucial role. Namely, it is necessary to take into account vibration and waves in molecular chains forming the nanotube wall. There are experimental evidences of such wall vibration [19]. The most interesting is solitary wave. There are different models describing such solutions (see, e.g., [29]). We shall consider so-called “Davidov soliton”, stable solution of this type in long molecular chain. The analogous waves are also in nanotubes [30].

Consider the soliton influence on the flow. The soliton moves in the nanotube wall and looks like a moving local extension (or constriction) of the tube. Let the area of the cross-section of the tube be \(S = S(x - Vt)\), where \(x\) is the longitudinal coordinate, \(V\) is the soliton velocity. Using the continuity equation

\[ \frac{\partial S}{\partial t} + \frac{\partial (Su)}{\partial x} = 0, \]

where \(u\) is velocity of the flow \((u = V + (v - V) S_0/S)\), \(v\) and \(S_0\) are the flow velocity and the area of the cross-section outside the domain occupied by the soliton, one can obtain the following rough estimation.
\[ v = \bar{v} + 2 \left( 1 - \frac{<S>}{S_0} \right) \frac{V - \bar{v}}{1 + v_s/v}, \]

where \( <S> \) is the average area of the nanotube cross-section, \( \bar{v} \) is the flow in the same nanotube without soliton. To obtain better result it is necessary to describe the flow in more details (see below).

The description of the wall soliton is quantum mechanical problem. Nanotube wall can be treated as a system of molecular chains (or molecular net) [29-31]. Let us consider an exciton of effective mass \( m \) interacting with displacement \( u(x,t) \) of molecules having masses \( M \) from the equilibrium positions at points \( x = na \). The corresponding Hamiltonian is [31]

\[ H = \frac{1}{a} \int \left( \frac{\hbar^2}{2m} |\psi|^2 + \frac{1}{2} M u^2 + MV_0^2 U(\rho) - \frac{\delta \rho |\psi|^4}{M} \right) dx, \]

where \( \rho \) is the relative decreasing of the equilibrium distance \( a \) between neighbor molecules, \( \rho = -u_x \), \( V_0 \) is the longitudinal sound speed in the linear approximation of the chain, \( \delta \) is the energy which characterizes the interaction of the exciton and the displacement, \( U(\rho) \) is the dimensionless potential of intermolecular interaction having minimum at \( \rho = 0 \), \( \psi \) - normalized exciton wave function. The Hamiltonian leads to the following system of equations:

\[ i\hbar \psi_t + \frac{\hbar^2}{2m} \psi_{xx} + \delta \rho \psi = 0, \quad u_x - V_0^2 U(\rho) u_{xx} = \frac{\delta}{M} |\psi|^2. \]

The system has a stable solution having the form of solitary wave of the profile \( \Phi \) depending on \( U \). Particularly, for cubic nonlinearity, \( U(\rho) = \frac{1}{2} \rho^2 + \frac{\gamma}{3} \rho^3 \):

\[ \Phi(\xi) = \frac{1}{2} \left( \frac{3\sigma}{4} \right)^{1/3} \left( \frac{\tau}{\gamma} \right)^{1/3} \text{sech}^2 \left( \frac{1}{2} \left( \frac{\tau \sigma^2}{6\gamma} \right)^{1/3} \xi \right). \]

The speed \( V \) of this soliton is as follows:

\[ V = V_0 \frac{1 + \frac{\tau M}{\gamma m} \left( 1 - \frac{3}{5} \frac{(\gamma \sigma)^{1/3} (\tau / 6)^{2/3}}{\sigma} \right)}{1 + \frac{\tau M}{\gamma m}}, \quad \sigma = \frac{2 \delta m a^2}{\hbar^2}, \quad \tau = \frac{\delta}{MV_0^2}. \]

Geometrically, this solution seems as a local extension (or constriction) of the tube, which moves with velocity \( V \). This motion leads to the motion of liquid in the neighborhood of this extension (restriction). The estimation of the Reynolds number shows that the Stokes approximation is appropriate. To describe the flow we use a model in which the local perturbation of the boundary is replaced by the point-like one (stokeslet approximation) [32, 33]. The mathematical background of the model is the theory of self-adjoint extensions of symmetric operators [34]. Consider two-dimensional straight channel (strip). The result for cylindrical channel is absolutely analogous. The Stokes flow for this case is described by the stream function \( \psi \) satisfying the biharmonic equation (for axisymmetric flow in a cylindrical domain the Laplace operator is replaced by the Stokes operator). As for the boundary conditions, it is more convenient to study moving boundary and fixed singularity, i.e. we assume that the normal derivative of the stream function isn’t zero (it is equal to the wall velocity).

Let us describe briefly the model of point-like perturbation for the Stokes flow. The starting point is the operator \( \Delta_0^2 \) which is the closure of the restriction of \( \Delta^2 \) onto the set of smooth functions vanishing in a neighborhood of zero. The domain of the operator is
A model operator \( \Delta_{0}^{*} \) is obtained as a self-adjoint extension of symmetric operator \( \Delta_{0}^{2} \). Due to the correlation \( \Delta_{0}^{2} \subset \Delta_{0}^{2} \subset \Delta_{0}^{2} \) one can search the extension as a restriction of the adjoint operator. The domain of the operator \( \Delta_{0}^{2} \) consists from the elements of the following form:

\[
u(x) = \sum_{i,j=1}^{2} c_{ij} g_{x_{ij}}(x) + \sum_{i=1}^{2} c_{i} g_{x_{i}}(x) + \xi(x)(a_{0} + \sum_{i=1}^{2} a_{i} x_{i} + \sum_{i,j=1}^{2} a_{ij} \beta_{ij} x_{i} x_{j}) + u_{0}(x).
\]

Here \( u_{0} \in D(\Delta_{0}) \), \( \beta_{ij} = 1, i \neq j, \beta_{ii} = 2^{-1} \), \( \xi(x) \) – smooth cutting function: \( \xi(x) = 1, |x| \leq 1, \xi(x) = 0, |x| \geq 2 \). To construct the model operator one should establish some correlation between these coefficients in the asymptotic expansion, for example, \( U_{0} = A U_{1} \), \( A = A^{*} \), \( U_{1} = (c_{1}, c_{2}, c_{11}, c_{12}, c_{22}) \), \( U_{0} = (a_{0}, a_{1}, a_{2}, a_{11}, a_{12}, a_{22}) \). As for detailed description of all classes of extensions, see [35]. The model allows one to obtain the streamfunction (and, consequently, the picture of the flow) in an explicit form. Calculations show that there is an eddy which follows the soliton, i.e. the soliton produces a flow in nanotube (see Fig. 1)

![Fig. 1.](image-url)

The work was supported by the Program “Development of the Scientific Potential of High School of Russia” (project 2.1.1/4215) and the Program “Scientific Staff of Innovative Russia 2009-2013” (project NK-526P_24).

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