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Liquid in a tube oscillating along its axis

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HIGHLIGHTS

- The oscillations under consideration have been analyzed numerically.
- Two different regimes of oscillations have been identified.
- The results are of interest in the context of the QCM-D sensors.

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ABSTRACT

The Quartz Crystal Microbalance with Dissipation (QCM-D) sensing technique has become widely used to study various supported thin films and adsorption of biological macromolecules, nanoparticles, aggregates, and cells. Such sensing, based on tracking shear oscillations of a piezoelectric crystal, can be employed in situations which are far beyond conventional ones. For example, one can deposit tubes on the surface of a sensor, orient them along the direction of the sensor surface oscillations, and study liquid oscillations inside the oscillating tubes. Herein, we illustrate and classify theoretically the regimes of liquid oscillations in this case. In particular, we identify and scrutinize the transition from the regime with appreciable gradients along the radial coordinate, which are qualitatively similar to those near the oscillating flat interface, to the regime where the liquid oscillates nearly coherently in the whole tube. The results are not only of relevance for the specific case of nanotubes but also for studies of certain mesoporous samples.

Oscillations at a liquid–solid interface induce oscillations in the liquid. An interesting special case here is shear oscillations when the interface oscillates in its plane. In this case, due to viscosity, the oscillations in the liquid are successively more and more damped with increasing distance from the interface. If (i) the interface is flat and oscillates harmonically, (ii) the liquid compression is negligible, and (iii) the thickness of the liquid layer is much larger than the penetration depth of the perturbation, the established oscillations of liquid are well known to be described analytically [1]. In particular, the liquid velocity, v, is oriented along the interface, depends only on the coordinate, $x \ge 0$, perpendicular to the interface ($x=0$ corresponds to the interface), and satisfies the following equation:

$$\frac{\partial v}{\partial t} = \nu \frac{\partial^2 v}{\partial x^2}, \tag{1}$$

where ν is the coefficient of kinematic viscosity. The solution of this equation (Fig. 1) is

$$v = v_0 \exp(-x/\delta)\cos(x/\delta - \omega t), \tag{2}$$

where ω is the frequency of oscillations, $\delta = (2\nu/\omega)^{1/2}$ is the penetration depth, and v_0 is the maximal interface velocity. The force acting per unit interface area is accordingly given by

$$\mathcal{F} = \eta \left.\frac{\partial v}{\partial x}\right|_{x=0} = -\rho(\omega\nu)^{1/2} v_0 \cos(\omega t + \pi/4), \tag{3}$$

where ρ is the liquid density, and $\eta = \rho\nu$ is the coefficient of dynamic viscosity. These equations show that due to the interplay of liquid viscosity and inertia there is an increasing "phase-lag" of the oscillations in the liquid with increasing distance from the surface (note, e.g., that the $x/\delta = \omega t$ condition moves from left to right as δ increases).

In reality, the liquid oscillations described above may occur in various situations. In particular, such oscillations represent one of the ingredients of the function of the QCM-D sensing systems [2–5] which have become widely used to study various thin films and

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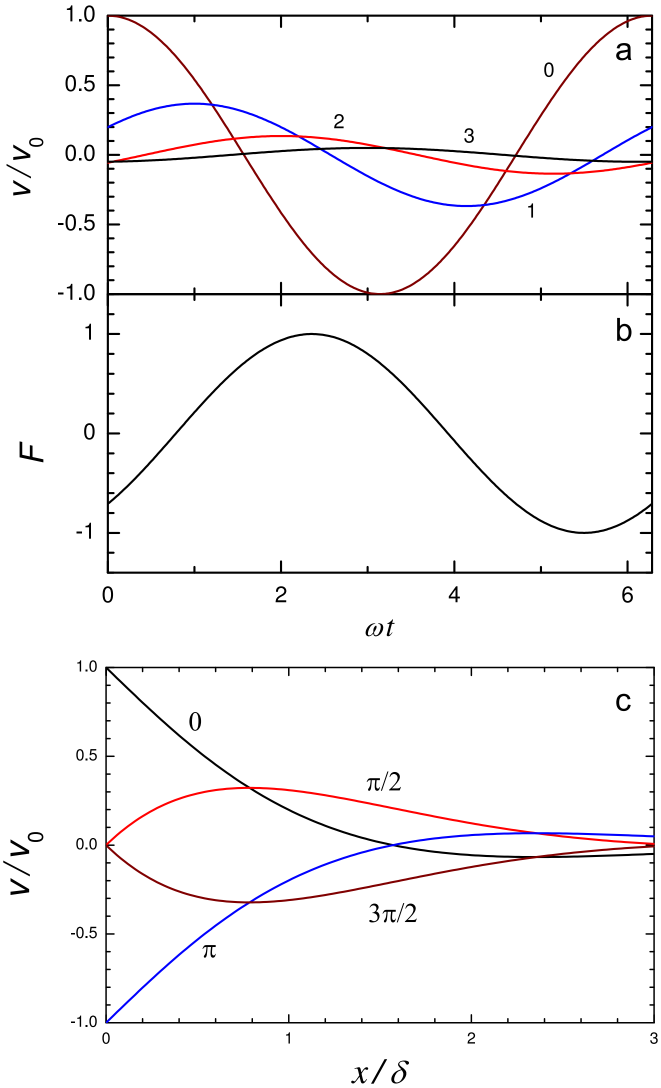

Fig. 1. Shear liquid oscillations near the flat wall harmonically oscillating along its plane: (a) the normalized liquid velocity, v/v_0, as a function of time during one oscillation period ($0 \le \omega t \le 2\pi$) for $x/\delta = 0$, 1, 2, and 3 (Eq. (2)); (b) as (a) for the normalized force, $F = \mathcal{F}/[\rho(\omega\nu)^{1/2}]$, acting per unit area of the wall (Eq. (3)); (c) the normalized liquid velocity as a function of the coordinate perpendicular to the wall for $\omega t = 0$, $\pi/2$, π, and $3\pi/2$ (Eq. (2)).

adsorption of biological macromolecules (e.g., proteins, DNA, polymers, and polyelectrolytes), nanoparticles, aggregates (e.g., vesicles or virions), and cells. In this setup, the typical frequency and penetration depth are 5 MHz and (in water) 250 nm, respectively (for other liquids, δ may be smaller or larger). In principle, the QCM-D sensors can be employed in the situations which are far beyond the conventional ones. For example, this technique was employed in situ to quantify, in real time, adsorption of dye and coadsorbates on flat and mesoporous TiO_2 films [6]. Following this line, one can in principle deposit or fabricate tubes on the surface of a sensor and orient them along the direction of the surface oscillations. In this context (and perhaps in some other contexts as well), it is thus of interest to clarify how liquid will oscillate in a tube oscillating along its axis. This is a main goal of our communication.

Concerning the subject under consideration, we may note that oscillations of liquid in a tube were earlier analyzed in different contexts (see, e.g., articles [7–16] and references therein). Usually, a tube was considered to be fixed. In particular, Womersley [7] described the liquid oscillations induced by oscillations of the pressure gradient. His results have been used directly or with extensions in many subsequent experimental and theoretical studies (see, e.g., [8,9] and references therein). Some other related situations were described as well [10–16].

If a tube oscillates along its axis, the liquid located inside oscillates along this axis as well. Assuming that the tube cross section is circular, we can use the polar coordinate, $0 \le r \le R$ (R is the tube radius), in order to describe liquid oscillations. In particular, Eq. (1) can be replaced by

$$\frac{\partial v}{\partial t} = \frac{\nu}{r}\frac{\partial}{\partial r}r\frac{\partial v}{\partial r}. \tag{4}$$

Near the tube wall, the liquid velocity is equal to the wall velocity, i.e.,

$$v(R) = v_0\cos(\omega t). \tag{5}$$

The second boundary condition can be obtained taking into account that due to the symmetry there is no force in the center, and accordingly

$$\left.\frac{\partial v}{\partial r}\right|_{r=0} = 0. \tag{6}$$

Eq. (4) can formally be solved by using the zeroth-order Bessel functions. With the boundary conditions (5) and (6), the corresponding solution is, however, cumbersome, and to illustrate the final results one should perform numerical calculations. Under such circumstances, direct numerical integration of Eq. (4) appears to be more convenient. Following the latter way, we used dimensionless variables, $\varrho = r/R$ and $\tau = \omega t$. With these variables, Eqs. (4)–(6) are read as

$$\frac{\partial v}{\partial \tau} = \frac{A}{\varrho}\frac{\partial}{\partial \varrho}\varrho\frac{\partial v}{\partial \varrho}, \tag{7}$$

$$v(R) = v_0\cos(\tau), \quad \text{and} \quad \left.\frac{\partial v}{\partial \varrho}\right|_{\varrho=0} = 0, \tag{8}$$

where $A = \nu/\omega R^2$ is the dimensionless parameter determining qualitative features of the liquid oscillations. The integration was performed by employing the conventional discrete scheme at $0.05 \le A \le 5$ with $\Delta\tau = 10^{-6}$ and $\Delta\varrho = 0.01$. With these steps, the integration was proved to be accurate.

The results of our calculations (Figs. 2–4) show that with increasing A there is transition from the regime with appreciable gradients along r which are qualitatively similar to those described by Eqs. (1) and (2) (cf. Fig. 1 with Fig. 2 for $A=0.05$; note that $r/R = 1$ is at the tube–liquid interface and should be compared with $x/\delta = 0$ in Fig. 1) to the regime where the liquid oscillates nearly coherently in the whole tube (see, e.g., Fig. 4 for $A=5$).

To characterize the regimes of liquid oscillations, it is instructive to compare the maximal liquid-induced shear force, $\rho\nu(\partial v/\partial r)$, acting per unit area of the wall in a tube with that, $\rho(\omega\nu)^{1/2}v_0$, acting on the flat wall (Eq. (3)). The corresponding dimensionless parameter defined as the ratio of these forces is

$$p_1 = \left(\frac{\nu}{\omega}\right)^{1/2}\frac{1}{v_0}\left.\frac{\partial v}{\partial r}\right|_{r=R} \equiv \frac{A^{1/2}}{v_0}\left.\frac{\partial v}{\partial \varrho}\right|_{\varrho=1} \equiv A^{1/2}D, \tag{9}$$

where $D \equiv (\partial v/\partial \rho)/v_0$ is the normalized velocity gradient near the wall (at $\rho = 1$). Another relevant dimensionless parameter can be obtained by dividing the maximal force, $\rho\nu(\partial v/\partial r)$, acting per unit area of the wall by that, $0.5\rho\omega R v_0$, needed to induce coherent liquid oscillations:

$$p_2 = \frac{2\nu}{\omega R v_0}\left.\frac{\partial v}{\partial r}\right|_{r=R} \equiv \frac{2A}{v_0}\left.\frac{\partial v}{\partial \varrho}\right|_{\varrho=1} \equiv 2AD. \tag{10}$$