



Soviet-era science, translated into English

PHYSICS

U. B. BAZARON, Corresponding Member of the Academy of
Sciences of the USSR B. V. DERYAGIN,

1965

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Abstract

Full Text

PHYSICS

U. B. BAZARON, Corresponding Member of the Academy of Sciences of the USSR
 B. V. DERYAGIN,
 A. V. BULGADAEV

ON THE SHEAR ELASTICITY OF BOUNDARY LAYERS OF LIQUIDS

Investigations by one of the authors with collaborators (¹⁻³) have shown that thin films of liquids at the boundary with a solid body can possess special properties distinct from bulk properties. The great scientific and practical importance of studying these properties is generally known. Among the little-studied questions in the physicochemistry of thin films of liquids are their mechanical properties. The study of the elastic properties of boundary layers is of great significance, in particular, for such a practically important problem as boundary friction.

The present communication is devoted to the investigation of the shear elasticity of films of certain liquids. One of the authors of the present communication, in a paper (⁴) published in 1932, showed the presence of shear elasticity in films of water in the gap between glass surfaces and its dependence on the thickness of the layer. In the present work the investigation of shear elasticity was carried out by an ultrasonic method on an X-cut piezoquartz resonator in the form of a bar with a rectangular cross section. By this method, in the work (⁵), one of the authors detected the presence of shear elasticity in certain liquids.

Fig. 1. Mounting of a quartz crystal between two needles:
 1 –piezoquartz, 2 –facing plate, 3 –liquid film

The idea of the method, proposed by L. I. Mandel' shtam and S. E. Khaikin for determining the nature of the interaction forces between two contacting surfaces, is as follows. A piezoquartz crystal, fixed between two steel needles at points on the nodal line, is excited at its fundamental resonant frequency. The lateral face of the oscillating quartz, undergoing tangential displacements, is brought into contact with the surface of another solid body. When small quasi-elastic forces arise at the contact, the resonant frequency of the oscillatory system, according

to a well-known theorem of the theory of oscillations, must increase. In our experiments, between the quartz and the second solid body there was a thin film of the liquid under investigation of specified thickness (Fig. 1), obtained by the draining method ⁽⁶⁾.

By simple reasoning it can be shown ^(7,8) that the shift of the resonant frequency $\Delta\omega_0$ is expressed through the coefficient of bond stiffness k in the case of contact of solid surfaces, in first approximation, by the relation

$$\Delta\omega_0 = \frac{k}{2M\omega_0}, \quad (1)$$

where ω_0 is the resonant frequency of the quartz, and M is the mass of the quartz.

In the presence between the quartz and the solid facing plate of a thin film of liquid of thickness d , formula (1) will have the form

$$\Delta\omega_0 = \frac{GS}{2M\omega_0 d}, \quad (2)$$

where G is the shear modulus of the film, and S is the contact area.

In view of the non-obviousness of formula (1), we shall briefly dwell on its derivation. When a solid body is placed on the oscillatory system, an additional ...

positive coupling. The entire system, in the presence of coupling, is regarded as conservative, and the change in the natural frequency is calculated. The kinetic and potential energies of the piezoquartz rod in free oscillations with the fundamental frequency are written as follows:

$$T_0 = \frac{S\rho l}{4} A^2 \omega_0^2; \quad U_0 = \frac{SE}{4l} A^2,$$

where S is the cross-sectional area of the piezoquartz, l is its length, ρ is the density, E is Young's modulus, ω_0 is the frequency, and A is the amplitude of displacement at the free ends. The equation of motion of the mass m (the plate) has the form $m\ddot{\xi} + k\xi = kx$, where x is the displacement of the attachment point, ξ is the displacement of the mass m , and k is the stiffness of the coupling. If $x = A \cos \omega t$, then

$$\xi = \frac{A \cos \omega t}{1 - \omega^2/\omega_1^2},$$

where $\omega_1 = \sqrt{k/m}$ is the natural frequency of the mass m , and ω is the frequency of the system in the presence of coupling. The kinetic and potential energies in the presence of coupling are, respectively,

$$T = \frac{S\rho l}{4} A^2 \omega^2 + \frac{m}{2} \frac{A^2 \omega^2}{(1 - \omega^2/\omega_1^2)^2},$$

$$U = \frac{SE}{4l} A^2 + \frac{k}{2} \left(1 - \frac{1}{1 - \omega^2/\omega_1^2}\right)^2 A^2.$$

Fig. 2. Dependence of $\Delta\omega/2\pi$ on the reciprocal film thickness for vaseline oil

By Rayleigh' s theorem, the frequency of the system in the presence of coupling is found by equating the kinetic and potential energies. Assuming that the frequency shift $\Delta\omega = \omega - \omega_0$ is sufficiently small, after simplifications one can obtain the approximate relation

$$\frac{\Delta\omega_0}{\omega_0} = \frac{1}{2(M + m)} \frac{k\omega_0^2/\omega_1^4 - m}{(1 - \omega_0^2/\omega_1^2)^2},$$

where $M = S\rho l$.

Under the condition that $m \ll M$ and $\omega_1^2 \ll \omega_0^2$ (i.e., k is small—a weak coupling), formula (1) is obtained.

In applying this formula to our case, where there is a thin liquid film between the plate and the piezoquartz, we assumed in advance that, for sufficiently small tangential displacements, the liquid film behaves as a perfectly elastic body. We further assumed that the thickness of our film is much less than the wavelength of the shear oscillations that are possible in the given liquid. The validity of this assumption will be shown below. Thus, under tangential displacements of the piezoquartz, the film will undergo a deformation of pure shear, and since the shear modulus of the liquid is certainly small, the additional elastic coupling effected by the film is also small. Consequently, in the present case the entire line of reasoning described above is applicable, except that instead of k in formula (1) there will stand the expression $k = GS/d$, as a result of which we obtain formula (2). For the shear modulus we obtain the expression

$$G = 2M\omega_0 d \Delta\omega_0 / S. \quad (3)$$

In the event that the film thickness were comparable with the wavelength of the shear oscillations, or much greater than it, the relation between the frequency shift, the shear modulus of the film, and its thickness would be more complicated. In this case one could make use of Smoluchowski' s theory of oscillations of a two-layer system ⁽¹⁰⁾, and formula (2) would turn out to be the limiting case of this theory.

We used X-cut quartz, cut at 5°, for which Poisson' s ratio is zero ⁽⁹⁾. But this quartz, for the given dimensions, at resonance interacts strongly with low-frequency flexural vibrations. Thus there was a weak normal component, which

Fig. 3

Figure 2: Fig. 3

Fig. 4

Figure 3: Fig. 4

in our case caused a negative frequency shift that was practically independent of the film thickness and therefore was easily excluded from consideration.

The experiments were carried out with polished surfaces. The second surface was a small prism of fused quartz with dimensions $7.5 \times 5 \times 4.5$ mm³. The working surfaces were washed in organic solvents and then cleaned in a glow discharge. The liquids under study were repeatedly filtered. The resonant frequency of the quartz was 74.4 kHz. The experiments were carried out at room temperature (22°).

Fig. 3. Dependence of $\Delta\omega/2\pi$ on the reciprocal film thickness for castor oil

From formula (2) it is clear that the frequency shift $\Delta\omega_0$ is inversely proportional to the thickness of the liquid interlayer d . This means that, if the film has a constant shear modulus independent of thickness, $\Delta\omega_0$ must vary directly proportionally to $1/d$. The studies were carried out at minimal amplitudes of the quartz vibrations in order to eliminate the influence of the force of viscous friction and of destruction of the film, which reduce the resonant frequency of the quartz as a result of energy dissipation. For this purpose, curves were plotted for the dependence of the frequency shift on the amplitude of the vibrations, and these curves were extrapolated to zero amplitude. The experimental results for three liquids are presented in Figs. 2, 3, and 4. The curves were plotted for different vibration amplitudes: curve 1 corresponds to the maximum vibration amplitude, curve 2 to half the amplitude, and curve 3 to the data extrapolated to zero amplitude.

Fig. 4. Dependence of $\Delta\omega/2\pi$ on the reciprocal film thickness for oleic acid

The dependences constructed from the extrapolated data correspond almost exactly to straight lines passing through the origin. This shows that the liquids tested possess shear elasticity and that, within the investigated thickness range, no dependence of the shear elasticity on thickness is observed. From formula (3) the shear modulus can be calculated. For vaseline oil, castor oil, and oleic acid, respectively, the following values are obtained: $G_1 = 3.8 \cdot 10^5$ dyn/cm², $G_2 = 2.4 \cdot 10^6$ dyn/cm², $G_3 = 4.2 \cdot 10^5$ dyn/cm². It is important to note that these results give the values of the coefficients of shear elasticity of the liquids in the bulk.

Formula (2), as stated above, is valid if the thickness of the liquid interlayer is much smaller than the wavelength of shear vibrations in the liquid. The practically exact fulfillment of the relation $\Delta\omega_0 d = \text{const}$ confirms the validity of this

conclusion and the legitimacy of calculating the shear modulus by formula (3). However, this also follows from the calculation of the wavelength of vibrations in the liquid, which was of the order of 100μ .

Some side phenomena are observed for castor oil and oleic acid at small thicknesses and large amplitudes of the quartz vibrations. On curve 1 of Fig. 3, a maximum of the frequency shift is expressed. This indicates that under these conditions the resonant frequency of the quartz is strongly affected by viscous friction in the film. In the case of oleic

for the acid (curve 1 in Fig. 4), abrupt decreases and increases in the frequency shift are observed, and these changes are not reproducible. Such behavior of oleic acid can be explained by the appearance, in the boundary film, of slip planes, which are absent at small oscillation amplitudes. This conclusion is consistent with the study of the boundary viscosity of oleic acid by the blowing method (³).

Buryat
Comprehensive Scientific Research Institute,
Siberian Branch of the Academy of Sciences of the USSR

Institute of Physical Chemistry,
Academy of Sciences of the USSR

Received
17 VIII 1964

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