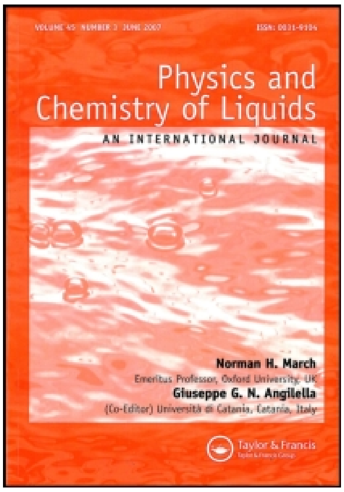

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LETTER

Forces Acting on Solid Spheres and Bubbles Moving Through Dense Liquid

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It is proposed that the force on a solid sphere moving in a dense liquid can be written, when reduced suitably to dimensionless units, as a function of three parameters: (i) The Reynolds number (ii) the ratio of two forces characteristic of the liquid and (iii) the ratio of two lengths, namely an interface thickness divided by the sphere radius. The relation of this to the Stokes regime is exhibited. The additional parameters entering when the sphere is replaced by a bubble are finally noted.

KEY WORDS: Reynolds number, liquid structure factor, melting temperature.

For a macroscopic solid sphere of radius R moving through a dense liquid with speed v, the magnitude F of the force acting on the sphere is first written as a product $F_0 f$. Here F_0 is constructed solely from thermodynamic properties of the unperturbed liquid. It is then argued that the dimensionless function f depend on three quantities, R_e, D_l and a. Each of these is again dimensionless, R_e being the Reynolds number, D_l being solely dependent on the liquid-state properties, while a is the ratio of two lengths: an interface thickness divided by R. In the Stokes limit

$$F_{\text{Stokes}} = 6\pi\eta v R, \tag{1}$$

where η is the shear viscosity, a non-equilibrium property of the liquid. This result (1) is known to be correct for small Reynolds number R_e, defined by

$$R_e = \frac{vR}{\eta} d, \tag{2}$$

where d is the mass density of the liquid. While the present letter is largely based on the limit of small Reynolds number R_e, we shall explore below the basic way the force F depends on the three dimensionless parameters just referred to, one of these being, of course R_e in Eq. (2).

As above, we first separate the magnitude F of the force acting on the sphere into a product of two factors, F_0 and a dimensionless function f:

$$F = F_0 f. \tag{3}$$

If the number density of the liquid is ρ and the thermal energy is $k_B T$, then we shall define F_0 in terms of the corresponding 'perfect gas pressure' $\rho k_B T$ times an 'area' $\rho^{-2/3}$. If the liquid structure factor is $S(q)$, then a well known result of fluctuation theory is that, in the long wavelength limit $q \to 0$:

$$S(0) = \rho k_B T \kappa_T \tag{4}$$

where κ_T is the isothermal compressibility of the liquid. Hence we can write for F_0:

$$F_0 = \frac{S(0)}{\kappa_T} \rho^{-2/3}. \tag{5}$$

Next, we note from Eq. (1) that

$$F_{\text{Stokes}} = F_0 \left[\frac{6\pi\eta v R \rho^{2/3} \kappa_T}{S(0)} \right] \tag{6}$$

which we shall proceed to write in the form

$$F_{\text{Stokes}} = F_0 R_e D_l \tag{7}$$

where D_l is defined by

$$D_l = \left(\frac{6\pi\eta^2}{d} \frac{\rho^{2/3} \kappa_T}{S(0)} \right), \tag{8}$$

which is the ratio of two forces. Evidently, as with F_0 in Eq. (5), D_l depends only on unperturbed liquid state properties. In contrast, however, to F_0, D_l involves the non-equilibrium quantity η, through a force $\alpha\eta^2/d$.

Thus, the first obvious result for the function f in Eq. (3) is that, in the Stokes limit, it takes the form

$$f_{\text{Stokes}} = R_e D_l. \tag{9}$$

This form (9) is evidently a very special limit of

$$f \equiv f(R_e, D_l, a). \tag{10}$$

Here, in addition to the two dimensionless parameters R_e and D_l defined in Eqs. (2) and (8) respectively, we have also introduced the ratio a of two lengths: an interface thickness $l_{\text{interface}}$, measuring the distance over which the homogeneous liquid density ρ is perturbed by the presence of the macroscopic sphere, and the sphere radius R;

$$a = l_{\text{interface}}/R. \tag{11}$$

It seems clear that a is a very small number and can be put equal to zero in the Stokes limit, where also the Reynolds number R_e is a small parameter. We turn immediately below therefore to discuss D_l in dense liquids at a characteristic temperature, which we take to be the melting temperature.

Andrade[1] has given a semiempirical formula for the shear viscosity η of dense liquids at the melting temperature T_m. While his kinetic theory arguments would not command ready acceptance nowadays[2], Brown and March[3] have proposed a route to pass from a Green–Kubo formula for viscosity to the form of Andrade's result (see also the later work of Zwanzig[4] and March[5]). The result is

$$\eta_{T_m} = C(k_B T_m)^{1/2} M^{1/2} \rho^{2/3}. \tag{12}$$

with C a pure number and M the atomic mass. Substituting in Eq. (8) and utilizing Eq. (4) yields for D_l the approximate form at the melting temperature:

$$D_l|_{T_m} = 6\pi C^2. \tag{13}$$

It is of interest to stress that the above, admittedly approximate, argument suggests that if we compare a variety of dense liquids at a characteristic temperature T_m, then D_l is almost constant. Of course, it must also be noted that η appearing in Eq. (8) is a strong function of temperature. Evidently, since the viscosity of liquids decreases with increasing temperature, $D_l|_{T_m}$ in Eq. (13) is an upper bound to D_l at arbitrary T.

Though the main emphasis of this letter has to do with the sphere radius R of macroscopic dimensions, it is of interest to note that there is also a microscopic limit for which the force on the 'solid sphere' can be usefully discussed in terms of the dynamical structure factor $S(q, \omega)$ of the dense liquid. This generalizes the (static) liquid structure factor $S(q)$ introduced above, and for the classical liquids under discussion in the present work:

$$\int_{-\infty}^{\infty} S(q, \omega)\, d\omega = S(q). \tag{14}$$

$S(q, \omega)$, in fact, measures the probability that a neutron incident on the liquid will transfer momentum $\hbar q$ and energy $\hbar\omega$ to the liquid. The rate of change of momentum can then be related to the average of $\hbar q$, say $\overline{\hbar q}$, with respect to the probability $S(q, \omega)$. Since such an expression, from Newton's Law, gives the force F, we interpret this as meaning that F/F_0 tends to a finite value in this microscopic limit. Though F_0, through Eq. (5), is determined by thermodynamic quantities, $\overline{\hbar q}$ is to be expected to depend on microscopic properties of the liquid and even at a characteristic temperature such as T_m will vary from one liquid to another. Although the role of the

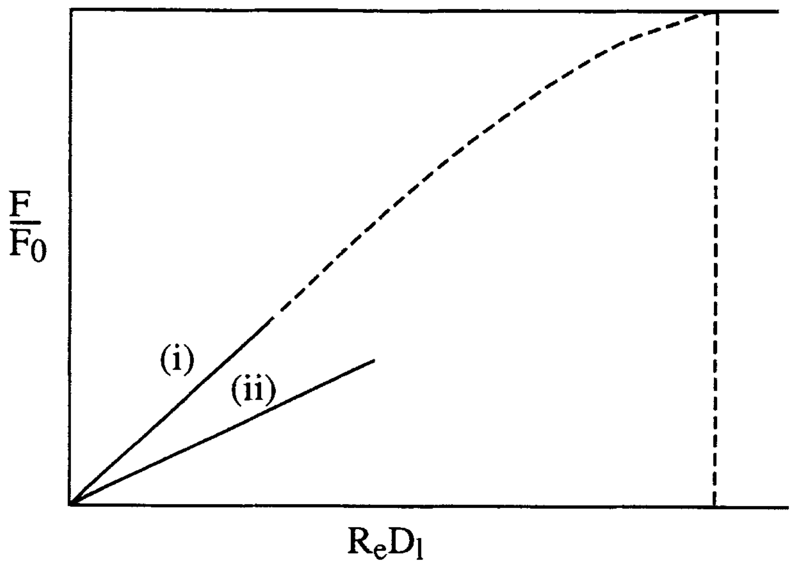

Figure 1. Dimensionless measure of force F on solid sphere moving through liquid vs $R_e D_l$ at characteristic temperature T_m. Top horizontal line is determined solely by $S(q, \omega)$ of the unperturbed dense liquid, provided $f(R_e, D_l, a = 0)$ is assumed to depend only on the single variable $R_e D_l$. Slope of line (i) at origin is unity. Lower line (ii) shows schematically reduction of slope for the case of bubble according to Eq. (16).

parameter a is quite different in this microscopic limit, we propose in Figure 1 a schematic form of F/F_0 for dense liquids at temperature T_m. In the form shown, since $D_l|_{T_m}$ has been demonstrated to be approximately constant, we have plotted F/F_0 versus $R_e D_l$, when the Stokes result (7) has evidently slope unity. The dashed curve in Figure 1 indicates how the constant value proposed above is reached continuously from the Stokes limit, at a value of $R_e D_l$ indicated by the vertical dashed line.

We wish to add a brief comment on the case when the rigid sphere discussed above is replaced by a bubble. The new features to be incorporated into Eqs. (3) and (10) are then the additional dimensionless parameters

$$K = \hat{\eta}/\eta \tag{15}$$

and also the ratio of mass densities $\hat{d}/d$, $\hat{\eta}$ and $\hat{d}$ referring to bubble properties. In fact, in the macroscopic limit, the result (1) of Stokes is then to be generalized into the Hadamard–Rybezynski result[6]

$$F = 4\pi\eta v R \frac{2 + 3K}{2[1 + K]}, \tag{16}$$

which reduces to Eq. (1) in the limit when the viscosity $\hat{\eta}$, and hence K in Eq. (15), tends to infinity. In other words, in the macroscopic limit, the density ratio does not enter. Of course, a discussion of the role of the dimensionless parameter a may well be necessary when one examines in detail the boundary conditions to be imposed in solving the Navier–Stokes equation. Such questions are raised, and discussed, in the work of Paranjape[7] and Paranjape and Robson[8] and will not therefore be elaborated further here.

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