

## SLOW MOTION OF TWO DROPLETS AND A DROPLET TOWARDS A FLUID OR SOLID INTERFACE

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**Abstract**—Starting from the general formula of Haber *et al.* (1973) for the drag force arising when two drops are approaching each other along their line of centers in an unbounded liquid, asymptotic expressions for great and small separations are derived. In the former case a generalization of the well-known formulae of Rybczynski (1911), Hadamard (1911) and Lorentz (1907) is obtained. In the latter, new approximate formulae, valid for great and small drop viscosity are derived. The approach of a liquid drop toward a solid plane boundary is considered as well.

### 1. INTRODUCTION

From the fluid dynamic point of view interest in emulsions is mainly due to the coupling of the flow in both phases—the drops and the continuous phase. This is, however, one of the reasons for the complicated character of the processes in these systems. There are also other effects which interfere with the flow coupling and make the theoretical treatment of emulsion hydrodynamics rather involved. For example, the experimental studies of Sonntag & Strenge (1970), Hartland (1967, 1969), Scheele & Leng (1971), McKay & Mason (1963) reveal, that due to viscous forces, emulsion droplets are deformed when approaching another interface. The initial deformation of the droplets surfaces, usually called a “dimple”, evolves with closer approach and finally a film with nearly uniform thickness forms between the droplets.

Because of the complexity of these process no consistent theory incorporating all these effects has been developed; only particular cases have been considered. For example, in the papers of Murdoch & Leng (1971), Reed *et al.* (1974), Ivanov & Traykov (1976) the flow pattern in thin emulsion films with plane-parallel interfaces has been analyzed. On the other hand, Radoev & Ivanov (1972) and Dimitrov & Ivanov (1975) have shown, that the deformation (due to viscous forces) of a gas bubble moving toward an interface can be considered as a small perturbation of the primary spherical shape. That is why it is interesting to study the mutual approach of two underformable spherical drops when the distance between them is small. This is a necessary initial step to the solution of the more general problem of drops with deformable interfaces.

We shall consider two important particular systems: (1) two drops approaching each other along their line of centers, (2) a single drop moving normally to an infinite solid plane surface. The comparison between these systems is of considerable physical interest because in the former case both interfaces are freely moving, whilst in the latter case the interface solid/fluid is tangentially immobile. It will be shown in section 4 that the change of the boundary condition results in a striking difference in the behaviour of these systems. We shall assume that the system considered is free of surface active impurities and the continuous phase is an unbounded immobile fluid. A steady state process at low Reynolds numbers is considered. Bart (1968) has solved this problem for a droplet approaching a flat interface and Wacholder & Weihs (1972) for a slow motion of a fluid sphere in the vicinity of another sphere or a plane boundary. Their solutions were generalized by Haber *et al.* (1973) for two droplets of different radii and viscosities, suspended in a third unbounded fluid. The purpose of our treatment is to

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obtain asymptotic expressions valid at large and small separations. These asymptotic formulae lead to some interesting conclusions, which are difficult to deduce from the complicated general solution.

The complete solution required derivation of expressions for for the drag force, the stream function and the pressure field. We confine ourselves to the drag force  $F$  only, since it is one of the most relevant characteristics of the process. The respective equations for the two systems under consideration are obtained from the general solution of Haber *et al.* (1973). When two equal drops with radii  $R_c$  and viscosities  $\mu$  are moving toward each other with a velocity  $V$ , we have:

$$F = 4\pi\mu^* Va \sum_{n=1}^{\infty} K_n \{ 2[(2n+1) \sinh 2\alpha + 2 \cosh 2\alpha - 2e^{-(2n+1)\alpha}] + \bar{\mu}[(2n+1)^2 \cosh 2\alpha + 2(2n+1) \sinh 2\alpha - (2n+3)(2n-1) + 4e^{-(2n+1)\alpha}] \} / \left\{ 4 \sinh \left( \frac{2n+3}{2} \alpha \right) \sinh \left( \frac{2n-1}{2} \alpha \right) + \bar{\mu} [2 \sinh (2n+1)\alpha - (2n-1) \sinh 2\alpha] \right\}, \quad [1]$$

Here  $\mu^*$  is the viscosity of the dispersion medium.

For an emulsion drop moving with velocity  $V$  normally towards a solid plane the drag force has the form:

$$F = 8\pi\mu^* Va \sum_{n=1}^{\infty} K_n \{ (2n+1) \sinh 2\alpha + 2 \cosh 2\alpha + 2e^{-(2n+1)\alpha} + \bar{\mu} [(2n+1)^2 \sinh^2 \alpha + (2n+1) \sinh \alpha + 2 - 2e^{-(2n+1)\alpha}] \} / \left\{ 2 \sinh (2n+1)\alpha - (2n+1) \sinh 2\alpha + \bar{\mu} \left[ 4 \sinh^2 \left( \frac{2n+1}{2} \alpha \right) - (2n+1)^2 \sinh^2 \alpha \right] \right\}. \quad [2]$$

The following notations are introduced above:

$$K_n = \frac{(n+1)}{(2n+2)(2n-1)}; \quad \bar{\mu} = \mu/\mu^*; \quad a = R_c \sinh \alpha; \quad R_c + h = a \operatorname{cptgh} \alpha \quad [3]$$

and  $(2R_c + h)$  is the distance between the centres of the spheres.

## 2. MUTUAL APPROACH OF TWO DROPS

Unlike the case of a single drop moving in an infinite liquid medium, considered by Rybczinski (1911) and Hadamard (1911), the drag force in [1] depends on the dimensionless parameter  $\alpha$  (i.e. on the ratio  $h/R_c$ ). Thus, the expression for the drag force could be written in the form:

$$F = 6\pi\mu^* VR_c f(\alpha, \bar{\mu}).$$

Two limiting cases are possible with respect to the values of  $\alpha$ :  $\alpha \gg 1$  and  $\alpha \ll 1$  corresponding to  $h/R_c \gg 1$  and  $h/R_c \ll 1$  respectively. From [3] it follows, that  $\exp(-\alpha) \approx R_c/2h$  at  $\alpha \gg 1$ . We can simplify [1] by introducing this approximation and expanding the terms of the sum in series with respect to the ratio  $R_c/h$ . Since only the term with  $n = 1$  gives contribution to the linear approximation, we thus obtain:

$$F = 6\pi\mu^* VR_c \frac{2/3 + \bar{\mu}}{1 + \bar{\mu}} \left( 1 + \frac{2/3 + \bar{\mu}}{1 + \bar{\mu}} \frac{R_c}{h} \right). \quad [4]$$

The factor before the brackets coincides with Rybczinski-Hadanard's equation. Since [4] is valid for arbitrary values of the viscosity ratio  $\bar{\mu}$  it gives as a limiting case (with  $\bar{\mu} \rightarrow \infty$ ) the drag force for the case of two solid spheres at great separation. The resulting relation is analogous to that derived by Lorentz (1907), see his [14]. For the behaviour of emulsions, the other asymptotic case,  $\alpha \ll 1$ , i.e.  $h/R_c \ll 1$  is of greater importance. From [3] we have the following approximate relations:

$$a \approx \sqrt{2R_c h}; \quad \alpha \approx \sqrt{2h/R_c}. \quad [5]$$

Analysis of [1] in this case is more complicated. The hyperbolic functions in the sum depend on the product  $n\alpha$  and since  $n \rightarrow \infty$  their expansion in power series with respect to  $n$  will not give the correct asymptotic result. Indeed, if one uses the approximation  $\sinh(n\alpha) \approx n\alpha$  the series in [1] will diverge. For the case of a solid sphere approaching a plane solid surface Cox & Brenner (1967) have developed a systematic method for deriving small gap width asymptotic expressions. Their method is well suited for our problem, but its application leads to very complicated calculations because of the presence of the terms with  $\bar{\mu}$  in [1] and [2]. On the other hand we are interested only in the leading terms (at  $\alpha \rightarrow 0$ ) of the asymptotic series. [These terms correspond to the term with  $1/\epsilon$  in [5.3] of Cox & Brenner (1967).] Since in this limit the outer solution is immaterial, it seems justified to carry out the summation in [1] and [2] by replacing there the hyperbolic functions by the approximate expressions:

$$\sinh(n\alpha) \approx n\alpha e^{n\alpha}; \quad \cosh(n\alpha) \approx e^{n\alpha}. \quad [6]$$

With  $n\alpha \ll 1$  (inner solution), [6] gives the correct results and with  $n\alpha \gg 1$  (outer solution) it qualitatively describes the asymptotic behaviour of  $\sinh(n\alpha)$  and  $\cosh(n\alpha)$ . In section 4 we shall compare some results, derived by this semi-intuitive asymptotic solution and by the systematic approach of Cox & Brenner (1967).

By means of [6], from [1] we have

$$F = 96\pi\mu^* V \frac{R_c}{\alpha} \sum_{n=1}^{\infty} \frac{n(n+1)}{(2n+3)^2(2n-1)^2} \frac{(2n+1)\alpha + \bar{\mu}}{3 + (2n+1)\alpha\bar{\mu}} e^{-(2n+1)\alpha}. \quad [7]$$

Carrying out the summation in [7] (see appendix I) we obtain an equation for the drag force at  $\alpha \ll 1$ , which is valid at arbitrary values of  $\bar{\mu}$ :

$$F = \frac{3}{2} \pi\mu^* V \frac{R_c}{\alpha^2} \left\{ \frac{9\beta}{9-4\beta^2} \left( \frac{\pi^2}{4} - \alpha \ln \frac{2}{\alpha} \right) + 4 \frac{4\alpha^2 - \beta^2}{(3-2\beta)^2} \left[ 2 \frac{9-\beta^2}{(3+2\beta)^2} \left( \ln \frac{2}{\alpha} - 2\alpha e^{3(\alpha/\beta)I} \right) - 1 \right] \right\} \quad [8]$$

where

$$\beta = \alpha\bar{\mu} \approx (\mu/\mu^*)\sqrt{2h/R_c}; \quad I = \int_1^{\infty} \frac{e^{-3[1+(1/\beta)]\alpha x}}{1-e^{-2\alpha x}} dx. \quad [9]$$

When the viscosity of the drops is great ( $\bar{\mu} \gg 1$ ),  $\beta$  can reach high values, even if  $h/R_c \ll 1$ . Since  $\alpha/\beta \ll 1$ , [A.8] can be substituted for  $I$  (see appendix II) and the remaining terms in curly brackets in [8] can be expanded in a power series with respect to  $\beta^{-1}$ . Keeping only the linear term in the series we obtain:

$$F = \frac{3}{2} \pi\mu^* V \frac{R_c^2}{h} \left( 1 - \frac{3\pi^2}{32} \frac{\mu^*}{\mu} \sqrt{\frac{R_c}{2h}} \right). \quad [10]$$

The factor before the brackets in [10] is equal to the drag force  $F_s'' = 3\pi\mu^*VR_c^2/2h$  for the case of two solid spheres at small distance ( $h/R_c \ll 1$ ). It can be obtained by direct integration of the lubrication theory equations. As expected, the mobility of the liquid interface decreases the drag force ( $F < F_s''$ ). This effect becomes more appreciable with decreasing thickness  $h$ .

The case  $\beta \ll 1$  can be realised either at small  $\bar{\mu}$  (i.e. when the drop viscosity is low) or at high  $\bar{\mu}$  but very small distances ( $h/R_c \ll 1$ ). The integral  $I$  in [8] depends both on  $\alpha$  and on the ratio  $(\beta/\alpha) = \bar{\mu}$ . Therefore, two different asymptotic expansions are possible (recall that  $\alpha \ll 1$ ): first, with  $\beta \ll \alpha$  the asymptotic form is derived by substituting [A.9] for  $I$  (see appendix II) in [8] and expanding the remaining functions in curly brackets in power series with respect to  $\beta$  and  $\beta/\alpha$ ; second, with  $\beta \gg \alpha$  we substitute [A.8] for  $I$  (see appendix II) in [8] and expand the remaining functions in curly brackets in [8] in power series with respect to  $\beta$  and  $\alpha/\beta$ . Thus we find the following asymptotic equations for the drag force:

$$F = 2\pi\mu^*VR_c \ln(R_c/h) \quad \text{with } \bar{\mu} \ll 1, \quad [11]$$

$$F = (3\pi^3/8)\mu VR_c \sqrt{(R_c/2h)} \quad \text{with } \bar{\mu} \gg 1. \quad [12]$$

Equation [11] corresponds to the case of two nondeformable gas bubbles with freely moving surfaces. It does not follow from the lubrication approximation, because the drag force with freely moving interfaces depends considerably on the energy dissipation in regions rather remote from the axis of symmetry, where this approximation is no longer valid. However, Dimitrov & Radoev (1976) were able to obtain a more general form of the lubrication approximation which allowed them to derive directly [11].

### 3. MOTION OF A DROP TOWARDS A SOLID PLANE

The analysis of [2], giving the drag force for the case of an emulsion droplet approaching a solid plane, is similar to that presented above. At  $\alpha \gg 1$  [2] yields:

$$F = 6\pi\mu^*VR_c \frac{2/3 + \bar{\mu}}{1 + \bar{\mu}} \left( 1 + \frac{9}{8} \frac{2/3 + \bar{\mu}}{1 + \bar{\mu}} \frac{R_c}{h} \right). \quad [13]$$

The derivation of [13] entirely coincides with that of [4]. At  $h/R_c \rightarrow \infty$  [13] gives Rybczinski & Hadamard's formula, while at  $\bar{\mu} \rightarrow \infty$  it gives Lorentz' (1907) relation

$$F = 6\pi\mu^*VR_c \left( 1 + \frac{9}{8} \frac{R_c}{h} \right). \quad [14]$$

At  $\alpha \ll 1$ , using [6], from [2] we obtain:

$$F = 384\pi\mu^*V \frac{R_c}{\alpha} \sum_{n=1}^{\infty} \frac{K_n}{(2n+3)(2n+1)^2} \frac{1 + (2n+1)\beta}{4 + (2n+1)\beta} e^{-(2n+1)\alpha} \quad [15]$$

Since the further treatment of [15] is similar to that of [7], for the sake of brevity we quote the final results only.

From [15] at  $\beta \gg 1$  we find:

$$F = 6\pi\mu^*V \frac{R_c^2}{h} \left( 1 - \frac{9\pi^2}{32} \sqrt{\left(\frac{R_c}{2h}\right) \frac{\mu^*}{\mu}} \right). \quad [16]$$

At  $\bar{\mu} \rightarrow \infty$  [16] yields the well known Taylor's formula for the drag force  $F_s' = 6\pi\mu^*VR_c^2/h$  at the approach of a solid sphere towards a solid plane, see Brenner (1962).

At  $\beta \ll 1$  [15] yields.

$$F = \frac{3}{2} \pi \mu^* V \frac{R_c^2}{h} \left( 1 + \frac{9\pi^2}{32} \sqrt{\left(\frac{2h}{R_c}\right) \frac{\mu}{\mu^*}} \right) = \frac{F'_s}{4} \left( 1 + \frac{9\pi^2}{32} \sqrt{\left(\frac{2h}{R_c}\right) \frac{\mu}{\mu^*}} \right). \quad [17]$$

The factor  $F'_s/4$  before the brackets in [17] is the drag force for the case of an underformable gas bubble, approaching a solid plane. This result also can be deduced from the lubrication theory.

#### 4. DISCUSSION

It is worth noting that  $h \ll R_c$ , a significant difference exists between the drag forces for the processes of mutual approaching of two emulsion droplets and of a droplet approaching a solid plane. For relatively great separations and highly viscous droplets, i.e. at  $\beta \gg 1$ , this difference is mainly due to geometrical factors. Then the droplets behave like solid spheres and the leading terms in the respective equations [10] and [16] (the factors before brackets) are  $F''_s$  and  $F'_s = 4F''_s$ . This is a natural consequence of the similar mobilities of all interfaces.

The situation is however quite different at  $\beta \ll 1$ . The leading term in the drag force for the system droplet/solid plane (see [17]) then increases four times (the droplet behaves like a bubble) but has the same functional dependence on the parameters of the system as in the case  $\beta \gg 1$  (cf. [16] and [17]). This is due to the tangential immobility of the solid plane which does not allow the liquid flow in the dispersion medium and hence in the droplet, to grow very strongly even when  $\bar{\mu} \rightarrow 0$ . For this reason the energy dissipation in the droplet is always negligible.

In the case of two droplets at very small distance  $h$ ,  $\alpha$  can become sufficiently small to ensure the validity of the condition  $\beta = \alpha \bar{\mu} \ll 1$  even when  $\bar{\mu} \gg 1$ . The respective equation [12] for the drag force is entirely different both from [16] and [17]. A peculiar feature of this equation is the independence of the drag force on the viscosity of the dispersion medium. A similar result was obtained by Ivanov & Traykov (1975) for a plane-parallel film, formed between two droplets. This effect is related to the mobility of both interfaces of this system which leads to a predominant dissipation of energy in the droplets. When the droplet viscosity is relatively low ( $\bar{\mu} \ll 1$ ) it is possible for the liquid motion within the droplets to be intensive, but the energy dissipated there is sufficiently small to be neglected. The droplets behave then like bubbles and the drag force is given by [11]. The mobility of the interfaces in this system strongly facilitates the liquid flow and the drag force is of several orders of magnitude smaller than the drag force  $F'_s/4$  for the system bubble/solid interface (more exactly, their ratio equals  $(4h/3R_c) \ln(R_c/h)$ ).

The exact expression for the drag force for a bubble, approaching a gas-liquid interface (see [30] with  $\mu_2 = \mu^*$  and  $\mu_1 = \mu_3 = 0$  in Bart (1968)) differs from the respective equation for two bubbles. The latter follows from [1] with  $\bar{\mu} = 0$  only by a factor 4 in the RHS, so that the drag force will be four times greater for the former system. Despite the great difference in interface mobility, the ratio of the drag forces for these systems is the same as for the systems solid sphere/solid plane and two solid spheres:  $F'_s/F''_s = 4$ . This confirms the conclusion that the drastic difference in the behaviour between the systems two droplets and droplet-solid plane at  $\beta \ll 1$  is due to the tangential immobility of the solid plane.

In order to check the validity of our asymptotic procedure, we give now a brief derivation of [11] by means of the method of Cox & Brenner (1967). We have chosen for this verification the case of two bubbles, because the correctness of our procedure for solid spheres ( $\bar{\mu} \rightarrow \infty$ ) in the limit  $\alpha \rightarrow 0$  is confirmed by the fact that [10] and [16] give correctly the respective limiting expressions. If in the other limiting case of two bubbles ( $\bar{\mu} = 0$ ) the results are also correct, it can be supposed that they will not be wrong for intermediate values of  $\bar{\mu}$ .

With  $\bar{\mu} = 0$  from [1] in the limit  $\alpha \rightarrow 0$  we have

$$F = 2\pi\mu^* a \sum_{n=1}^{\infty} K_n \frac{(2n+1) \sinh 2\alpha + 2 \cosh 2\alpha - 2e^{-(2n+1)\alpha}}{\sinh\left(\frac{2n+3}{2}\alpha\right) \sinh\left(\frac{2n-1}{2}\alpha\right)} \quad [18]$$

$$\approx 4\pi\mu^* VR_c \alpha \sum_{n=1}^{\infty} K_n \frac{(2n+1)\alpha + 1 - e^{-(2n+1)\alpha}}{\sinh\left(\frac{2n+3}{2}\alpha\right) \sinh\left(\frac{2n-1}{2}\alpha\right)}$$

Let us introduce the integers  $N_i$  and  $N_0 = N_i + 1$ ,  $N_i$  being in the region of validity of both the inner and the outer expansions. In the inner expansion we use the approximations  $\sinh x \approx x$  and  $e^{-x} \approx 1 - x$  and in the outer expansion we can neglect with respect to  $h$  the terms of the order of unity. Thus, from [18] we get (see also [3])

$$F/4\pi\mu^* VR_c = \alpha \left[ \frac{8}{\alpha} \sum_{n=1}^{n=N_i} \frac{n(n+1)(2n+1)}{(2n+3)^2(2n-1)^2} + \frac{1}{4} \int_{N_0}^{\infty} \frac{2n+1 - e^{-2n\alpha}}{\sinh^2 \alpha n} dn \right]$$

$$= -\frac{1}{2} + 2 \sum_{n=1}^{n=N_i} \frac{1}{2n-1} + \frac{1}{2} \int_{N_0\alpha}^{\infty} \frac{x - \cosh x \sinh x - \sinh^2 x}{\sinh^2 x} dx. \quad [19]$$

Since  $N_i$  increases strongly as  $\alpha$  decreases, we can replace in [19] both  $N_i$  and  $N_0$  by the same limiting value  $N_m$ . Then the most important terms in the sum and in the integral in [19] will be  $(1/2) \ln N_m$  and  $-2 \ln(N_m\alpha)$  respectively. Hence from [19] (see also [5]) we obtain:

$$F/4\pi\mu^* VR_c = \ln N_m - \ln(N_m\alpha) + \dots = -\ln \alpha + \dots \rightarrow \frac{1}{2} \ln \frac{R_c}{h}. \quad [20]$$

The last limiting form of [20] coincides with [11]. This confirms the correctness of our procedure when the leading term, corresponding to  $\alpha \rightarrow 0$ , is calculated. Although the first correction to the leading term is immaterial for the present work, we have calculated it for the system solid sphere–solid plane, using our procedure. We have obtained the same functionality,  $\ln \alpha$ , as in [2.47] of Cox & Brenner (1967), but the numerical coefficient before this term is  $2/3$  instead of the correct value  $2/5$ . This result suggests, that with higher values of  $\alpha$ , when the outer expansion becomes important, our procedure will lead to expressions which are correct only qualitatively (cf. the comment [6]).

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## APPENDIX I

*Derivation of [8]*

The algebraic factor in the general term of the sum in [7] is represented as a sum of common fractions:

$$\frac{n(n+1)}{(2n+3)^2(2n+1)^2} \frac{(2n+1)\alpha + \bar{\mu}}{3 + (2n+1)\beta} = \frac{A_1}{(2n+3)^2} + \frac{A_2}{2n+3} + \frac{B_2}{(2n-1)^2} + \frac{B_1}{2n-1} + \frac{C}{3 + (2n+1)\beta}; \quad [\text{A.1}]$$

and the 'constants'  $A_1$ ,  $A_2$ ,  $B_1$ ,  $B_2$  and  $C$ , which are functions of  $\alpha$  and  $\beta$ , are determined using the standard procedure. Putting [A.1] in [7] and suitably rearranging the terms in the sum, we obtain:

$$F = 96\pi\mu^* V \frac{R_c}{\alpha} \left\{ \sum_{n=1}^{\infty} \left[ \frac{A_2 e^{4\alpha} + B_2}{(2n-1)^2} + \frac{A_1 e^{4\alpha} + B_1}{2n-1} + \frac{C}{3 + (2n+1)\beta} \right] e^{-(2n+1)\alpha} - A_2 \left( e^\alpha + \frac{e^{-\alpha}}{2} \right) - A_1 \left( e^\alpha + \frac{e^{-\alpha}}{3} \right) \right\}. \quad [\text{A.2}]$$

The summands in [A.2] are transformed, making use of the properties of the geometric progression, as follows:

$$\begin{aligned} \sum_{n=1}^{\infty} \frac{e^{-(2n+1)\alpha}}{2n-1} &= e^{-2\alpha} \sum_{n=1}^{\infty} \left[ \int_{\alpha}^{\infty} e^{-(2n-1)x} dx \right] = e^{-2\alpha} \int_{\alpha}^{\infty} \left[ \sum_{n=1}^{\infty} e^{-(2n-1)x} \right] dx \\ &= e^{-2\alpha} \ln \left[ \frac{(1 + e^{-\alpha})}{(1 - e^{-\alpha})} \right] / 2 \rightarrow \ln(2/\alpha) / 2; \end{aligned} \quad [\text{A.3}]$$

$$\sum_{n=1}^{\infty} \frac{e^{-(2n+1)\alpha}}{(2n-1)^2} = e^{-2\alpha} \sum_{n=1}^{\infty} \int_{\alpha}^{\infty} dx \int_x^{\infty} e^{(2n-1)y} dy \rightarrow \frac{1}{2} \left( \frac{\pi^2}{4} - \alpha \ln \frac{2}{\alpha} \right); \quad [\text{A.4}]$$

$$\sum_{n=1}^{\infty} \frac{e^{-(2n+1)\alpha}}{3 + (2n+1)\beta} = \frac{\alpha}{\beta} e^{3\alpha/\beta} \int_1^{\infty} \frac{e^{-3(1+1/\beta)\alpha x}}{1 - e^{-2\alpha x}} dx \quad [\text{A.5}]$$

The final limiting expressions in [A.3] and [A.4] are valid for  $\alpha \ll 1$ . Substituting in [A.2] the values of  $A_1, A_2, B_1, B_2$  and  $C$  and [A.3]–[A.5], we get [8].

#### APPENDIX II

*Asymptotic representation of the integral I in [8]*

With  $\alpha \ll 1$ , the integral

$$I = \int_1^{\infty} \frac{e^{-3(1+1/\beta)\alpha x}}{1 - e^{-2\alpha x}} dx \quad [\text{A.6}]$$

has two asymptotic representations depending on the ratio  $\alpha/\beta = 1/\bar{\mu}$ .

When  $\alpha/\beta \ll 1$  we can expand  $I$  in power series with respect to  $\alpha/\beta$ :

$$I = \int_1^{\infty} \frac{e^{-3\alpha x}}{1 - e^{-2\alpha x}} dx - 3 \frac{\alpha}{\beta} \int_1^{\infty} \frac{x e^{-3\alpha x}}{1 - e^{-2\alpha x}} dx + \dots, \quad [\text{A.7}]$$

where the integrals can be taken in closed form. In the limit  $\alpha \ll 1$  the result reads:

$$I \rightarrow \frac{1}{2} \left[ -1 + \frac{1}{2} \ln \frac{2}{\alpha} + \frac{3}{\beta} \left( 1 - \frac{\pi^2}{8} \right) \right] \quad [\text{A.8}]$$

When  $\alpha/\beta \gg 1$ , we have (note that  $\alpha \gg 1$ ):

$$I \approx \frac{1}{2\alpha} \int_1^{\infty} \frac{e^{-3(1+1/\beta)\alpha x}}{x} dx = -\frac{1}{2\alpha} E_i[-3(1+1/\beta)\alpha] \rightarrow \frac{\beta}{6\alpha^2} e^{-3\alpha/\beta} \left( 1 - \frac{\beta}{3\alpha} \right); \quad [\text{A.9}]$$

where

$$E_i(-x) = \int_x^{\infty} \frac{e^{-t}}{t} dt$$

is the exponential integral.