

# Thermophoresis in liquids

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The force on an immobile particle in a liquid in the presence of a constant temperature gradient is calculated. The force is proportional to the temperature gradient and to the particle size raised to the 3/2 power. The possibility of observing the effect in experiment is discussed.

The motion of solid particles under the influence of heat flow (called thermophoresis) has been thoroughly investigated in gases, where it is the result of Maxwellian thermal slip (see, e.g., Ref. 1). The force acting on a particle immersed in a gas in which a constant temperature gradient is maintained is proportional to the temperature gradient and to the first power of the particle size.

The nature of thermophoresis in liquids is different. It is one of the fluctuational phenomena in hydrodynamics.<sup>2-10</sup> The force acting on the particle is the result of its interaction with thermal acoustic fluctuations. It will be calculated on the basis of general liquid-dynamics equations derived by the author earlier,<sup>2</sup> in which account is taken of thermal fluctuations. The result reported below shows that the force acting on a solid particle in a non-uniformly heated liquid is proportional to the particle size raised to the 3/2 power, i.e., it is the fluctuational mechanism which governs for macroscopic particles.

Consider an immobile liquid in which a small constant temperature gradient is maintained and in which a spherical solid particle of radius  $R$  is immersed. The usual hydrodynamics equations show that at constant pressure the particle remains immobile if no external forces are applied to it (there is no thermophoresis), and an inhomogeneous temperature distribution is produced around it<sup>11</sup>:

$$T(\mathbf{r}) = G r (1 + \gamma R^3 / r^3), \quad (1)$$

where  $G$  is the temperature gradient at infinity,  $\gamma = (\kappa_l - \kappa_s) / (2\kappa_l + \kappa_s)$ ,  $\kappa_l$ ,  $\kappa_s$  are the thermal-conductivity coefficients of the liquid and solid, respectively, and the origin is at the center of the sphere.

The phenomenon of interest to us comes into play when fluctuational corrections to the hydrodynamic equations are taken into account.<sup>2-10</sup> In this case, just as in Refs. 2 and 9, only acoustic fluctuations play any role, since they are the only ones having a "propagating" character, in contrast to the purely dissipative nature of all the other types of hydrodynamic perturbations (shear and thermal waves).

When acoustic fluctuations are taken into account, the dynamics equations for the liquid constitute (see Ref. 2) a closed system for the hydrodynamic variables (velocity  $\mathbf{v}$ , pressure  $P$ , ...) and the nonequilibrium distribution function  $f$  of the acoustic fluctuations. In ordinary classical liquids it is convenient to so normalize  $f$  that the quantity

$$\int \frac{d^3k}{(2\pi)^3} f(\mathbf{k}, \mathbf{r}),$$

( $\mathbf{k}$  is the wave vector of the fluctuations) is the average energy density of the fluctuations. In fact,  $f$  is equal to the non-

equilibrium part of the usual phonon distribution function, multiplied by the energy of the phonons. Under stationary conditions the function  $f$  satisfies the kinetic equation

$$\mathbf{s} \partial f / \partial \mathbf{r} + f / l = -\mathbf{s} \nabla T, \quad (2)$$

where  $\mathbf{s} = \mathbf{k} / |\mathbf{k}|$ ,  $l = (\lambda k^2)^{-1}$  is the mean free path of the acoustic perturbations with wave vector  $\mathbf{k}$ ,

$$\lambda = \frac{1}{\rho c} \left[ \left( \frac{4}{3} \eta + \eta' \right) + \kappa_l \left( \frac{1}{c_v} - \frac{1}{c_p} \right) \right],$$

$\rho$  is the density of the liquid,  $c$  is the speed of sound,  $\eta$  and  $\eta'$  are respectively the first- and second-viscosity coefficients, and  $c_p$  and  $c_v$  are the specific heats. It is necessary to substitute in the right-hand side of (2) the temperature gradient (1) calculated without allowance for the fluctuations.

The distribution function  $f$  must satisfy Eq. (2), the usual condition that it be finite everywhere including infinity, and also some boundary conditions on the surface of the solid particles. These conditions are determined by the interaction of the acoustic oscillations of the liquid with the surface of the solid. The character of this interaction was investigated in detail (see Ref. 12) in connection with the question of the Kapitza jump. The results of the investigations show that at not too low temperatures the most realistic condition is that of total accommodation of the phonons with the solid surface. This is equivalent to the condition  $f = 0$  for the reflected acoustic oscillations, i.e.,  $|\mathbf{r}| = R$  at  $\mathbf{s} \cdot \mathbf{r} > 0$ .

The additional terms in the equations for the hydrodynamic variables are expressed in terms of the distribution function. In the approximation in which the liquid is incompressible we have under stationary conditions

$$\text{div } \mathbf{v} = 0, \quad \nabla P = \eta \Delta \mathbf{v} + \mathbf{p}. \quad (3)$$

Here

$$\mathbf{p} = - \int \frac{d^3k}{(2\pi)^3} \mathbf{s} \left( \mathbf{s} \frac{\partial f}{\partial \mathbf{r}} \right)$$

and  $P$  stands in fact for the sum

$$P_0 + \frac{\rho}{c} \left( \frac{\partial c}{\partial \rho} \right)_\sigma \int \frac{d^3k}{(2\pi)^3} f,$$

where  $P_0$  is the local equilibrium pressure of the liquid and  $\sigma$  is the entropy per unit mass.

Introducing, for a specified  $\mathbf{s}$ , the coordinates  $\zeta = \mathbf{r} \cdot \mathbf{s}$  and  $\rho = \mathbf{r} - \mathbf{s}(\mathbf{s} \cdot \mathbf{r})$ , we rewrite the kinetic equation (2) in the form

$$\partial f / \partial \zeta + f / l = -s \nabla T = -s \mathbf{G} + (s \nabla) (\mathbf{g} \nabla) \gamma R^3 G / r, \quad (4)$$

where  $\mathbf{g} = \mathbf{G} / G$ .

The subsequent calculations can be substantially simplified by using the following artifice. We seek the solution of Eq. (4) in the form of a sum  $f_1 + f_2$ , where  $f_1$  and  $f_2$  are solutions of Eq. (4) in the case when the right-hand side contains only the first or respectively the second term. We assume here that the boundary conditions for  $f_1$  and  $f_2$  are the same as for  $f$ , i.e., they are finite everywhere (including infinity), and the condition  $f_{1,2} = 0$  at  $r = R$  if  $\mathbf{s} \cdot \mathbf{r} > 0$ .

We obtain the function  $f_1$  by simple calculations:

$$f_1 = -l(s \mathbf{G}) + \varphi_1, \quad (5)$$

where

$$\varphi_1^{in} = l(s \mathbf{G}) \exp \left[ -\frac{\zeta - (R^2 - \rho^2)^{1/2}}{l} \right], \quad \varphi_1^{out} = 0,$$

the superscript *in* denotes that the corresponding equation holds at  $\zeta > (R^2 - \rho^2)^{1/2}$  and  $\rho < R$ , while *out* is used for  $\rho > R$  or for  $\rho < R$  and  $\zeta < - (R^2 - \rho^2)^{1/2}$ .

We introduce an auxiliary function  $\psi(\mathbf{k}, \mathbf{r})$  that satisfies the same boundary conditions as  $f, f_1$ , and  $f_2$ , as well as the equation

$$\partial \psi / \partial \zeta + \psi / l = - (s \nabla) (-\gamma R^3 G / r). \quad (6)$$

The function  $\psi$  is thus the distribution function of the acoustic fluctuations in the case of a spherically symmetric distribution of the temperature  $T(r) = -\gamma R^3 G / r$ . Comparison of the first parts of Eqs. (4) and (6) shows that the function  $(\mathbf{g} \cdot \nabla) \psi$  satisfies the same equation as  $f_2$ . These two functions differ therefore by a certain solution of the homogeneous equation. To find the latter, we note that the function  $\psi$  is discontinuous at  $\rho = R$  and  $\zeta > 0$ . The operator  $(\mathbf{g} \cdot \nabla)$  transforms these discontinuities into  $\delta$ -like singularities that should not be possessed by the function  $f_2$ .

From Eq. (6) we have

$$\begin{aligned} \psi^{in} &= \gamma R^3 G e^{-\zeta/l} \int_{(R^2 - \rho^2)^{1/2}}^{\zeta} d\zeta' e^{\zeta'/l} \frac{\partial}{\partial \zeta'} (\rho^2 + \zeta'^2)^{-1/2}, \\ \psi^{out} &= \gamma R^3 G e^{-\zeta/l} \int_{-\infty}^{\zeta} d\zeta' e^{\zeta'/l} \frac{\partial}{\partial \zeta'} (\rho^2 + \zeta'^2)^{-1/2}. \end{aligned}$$

The discontinuity  $\Delta \psi$  of the function  $\psi$  is defined at  $\rho = R$  and  $\zeta > 0$  as

$$\Delta \psi = (\psi^{out} - \psi^{in}) |_{\rho=R} = -G \gamma R^3 e^{-\zeta/l} \int_0^{\infty} d\zeta' e^{-\zeta'/l} \zeta' (R^2 + \zeta'^2)^{-1/2},$$

from which we find that the singular part of the function

$$\begin{aligned} Q(r) &= h_1' - h_2' + \frac{3}{r} h_1 - \frac{1}{r} h_2 + h_3 - \chi' - \frac{3}{r} \chi, \quad \chi(r) = \frac{3\gamma}{64} \left( \frac{R}{r} \right)^5 (\pi \lambda r)^{-1/2} \int_0^{\infty} w dw \left[ w + \left( 1 + \frac{R^2}{r^2} \right)^{1/2} \right]^{-1/2} \left( w^2 + \frac{R^2}{r^2} \right)^{-1/2}, \\ h_1(r) &= \frac{(\pi \lambda r)^{-3/2}}{32} \int_{(1-R^2/r^2)^{1/2}}^1 d \cos \theta \cos^2 \theta \left[ \cos \theta - \left( \cos^2 \theta - 1 + \frac{R^2}{r^2} \right)^{1/2} \right]^{-3/2} \\ &= \frac{(\pi \lambda r)^{-3/2}}{32} \left[ \frac{4}{45} \left( 1 - \frac{R^2}{r^2} \right)^{3/4} + \frac{1}{12} \left( 1 - \frac{R}{r} \right)^{3/2} + \frac{1}{20} \left( 1 + \frac{R}{r} \right)^2 \left( 1 - \frac{R}{r} \right)^{-1/2} - \frac{1}{4} \left( 1 + \frac{R}{r} \right) \left( 1 - \frac{R}{r} \right)^{1/2} \right. \\ &\quad \left. + \frac{1}{36} \left( 1 + \frac{R}{r} \right)^3 \left( 1 - \frac{R}{r} \right)^{-3/2} \right], \end{aligned}$$

$(\mathbf{g} \cdot \nabla) \psi$  is equal to

$$\Delta \psi \frac{\mathbf{g} \rho}{\rho} \delta(\rho - R) = \Delta \psi [(\mathbf{g} \mathbf{n}) - (\mathbf{g} \mathbf{s})(\mathbf{n} \mathbf{s})] \frac{r}{R} \delta(\rho - R),$$

where  $\mathbf{n} = \mathbf{r} / r$ .

The sum  $(\mathbf{g} \cdot \nabla) \psi + \psi_1$ , where

$$\begin{aligned} \psi_1 &= -\frac{\mathbf{g} \rho}{\rho} \delta(\rho - R) \Delta \psi = \gamma R^3 e^{-\zeta/l} \int_0^{\infty} d\zeta' e^{-\zeta'/l} \zeta' (R^2 + \zeta'^2)^{-1/2} \\ &\quad \times \frac{r}{R} \delta(\rho - R) [(\mathbf{G} \mathbf{n}) - (\mathbf{G} \mathbf{s})(\mathbf{n} \mathbf{s})], \quad (7) \end{aligned}$$

has no  $\delta$ -function singularities and satisfies the condition for  $f_2$ , but it does not vanish at  $r = R$  if  $\mathbf{s} \cdot \mathbf{n} > 0$ . By virtue of Eq. (6) and the condition  $\psi = 0$  we have for  $r = R$  and  $\mathbf{s} \cdot \mathbf{n} > 0$

$$\mathbf{g} \nabla \psi = (\mathbf{g} \mathbf{n}) \partial \psi / \partial r = -(\mathbf{g} \mathbf{n}) \gamma R G = -\gamma R (\mathbf{n} \mathbf{G}).$$

As a result,

$$f_2 = (\mathbf{g} \nabla) \psi + \psi_1 + \psi_2,$$

where the function  $\psi_2$ , which satisfies the homogeneous equation (4) with the condition  $\psi_1 = \gamma R (\mathbf{G} \cdot \mathbf{n})$  at  $r = R$  and  $\mathbf{s} \cdot \mathbf{n} > 0$ , is determined by the equations

$$\psi_2^{in} = \gamma R (\mathbf{G} \mathbf{n}) \exp \left[ -\frac{\zeta - (R^2 - \rho^2)^{1/2}}{l} \right], \quad \psi_2^{out} = 0. \quad (8)$$

The artifice used is effective for the following reasons. To solve Eqs. (3) it is in fact necessary to know only the non-potential part of the vector  $\mathbf{p}$ . The potential part can be excluded by renormalizing the pressure  $P$ , and therefore does not enter in the solution. If  $f$  is replaced in the equation for  $\mathbf{p}$  by  $\psi$ , a potential expression is obtained, for in this case, by virtue of the symmetry of the problem, the vector  $\mathbf{p}$  is directed everywhere along  $\mathbf{n}$  and the coefficient is a certain function of the distance  $r$ . Nor is the potential character disturbed by applying the operator  $(\mathbf{g} \cdot \nabla)$  to the vector  $\mathbf{p}$ . Recognizing also that by virtue of Eq. (2) we have

$$\int \frac{d^3 \mathbf{k}}{(2\pi)^3} \mathbf{s} \left( \mathbf{s} \frac{\partial f}{\partial \mathbf{r}} \right) = - \int \frac{d^3 \mathbf{k}}{(2\pi)^3} \frac{\mathbf{s}}{l} f + \text{grad},$$

where the grad symbol means that the corresponding equality is accurate up to a certain potential term, we have

$$\mathbf{p} = \int \frac{d^3 \mathbf{k}}{(2\pi)^3} \frac{\mathbf{s}}{l} (\varphi_1 + \psi_1 + \psi_2) + \text{grad}.$$

This yields, after simple transformations

$$\text{rot } \mathbf{p} = [\mathbf{G} \mathbf{n}] Q(r), \quad (9)$$

where

$$\begin{aligned}
h_2(r) &= \frac{(\pi\lambda r)^{-3/2}}{32} \int_{(1-R^2/r^2)^{1/2}}^1 d\cos\theta \left[ \cos\theta - \left( \cos^2\theta - 1 + \frac{R^2}{r^2} \right)^{1/2} \right]^{-3/2} \\
&= \frac{(\pi\lambda r)^{-3/2}}{32} \left[ \frac{4}{5} \left( 1 - \frac{R^2}{r^2} \right)^{-1/2} - \left( 1 - \frac{R}{r} \right)^{-1/2} + \frac{1}{5} \left( 1 + \frac{R}{r} \right) \left( 1 - \frac{R}{r} \right)^{-1/2} \right], \\
h_3(r) &= \frac{3\pi}{32} \frac{R}{r} \frac{\gamma\lambda}{(\pi\lambda r)^{3/2}} \int_{(1-R^2/r^2)^{1/2}}^1 \cos\theta d\cos\theta \left[ \cos\theta \right. \\
&\quad \left. - \left( \cos^2\theta - 1 + \frac{R^2}{r^2} \right)^{1/2} \right]^{-3/2} = \frac{3\pi}{32} \frac{R}{r} \frac{\gamma\lambda}{(\pi\lambda r)^{3/2}} \left[ -\frac{1}{2} \left( 1 - \frac{R}{r} \right)^{-1/2} \right. \\
&\quad \left. + \frac{4}{9} \left( 1 - \frac{R^2}{r^2} \right)^{-1/4} + \frac{1}{18} \left( 1 + \frac{R}{r} \right)^2 \left( 1 - \frac{R}{r} \right)^{-1/2} \right], \quad \cos\theta = \text{sn}.
\end{aligned}$$

We proceed to solve the system of hydrodynamic equations (3). The first of these equations shows that the velocity of the liquid is given by  $\mathbf{v} = \text{curl } \mathbf{A}$ , where the pseudovector  $\mathbf{A}$  is defined, by virtue of the symmetry of the problem, as

$$\mathbf{A} = [\mathbf{G}\nabla a] = a' [\mathbf{G}\mathbf{n}].$$

Here  $a$  is a certain scalar function of the distance  $r$ , so that  $\text{div } \mathbf{A} = 0$ . The prime denotes differentiation with respect to  $r$ . Eliminating the pressure by taking the curl of the second equation of (3), we obtain, taking (9) into account,

$$\eta(\Delta^2 a)' = Q(r).$$

Integration of the last equation yields

$$\begin{aligned}
a'(r) &= c_1 r^3 + c_2 r + c_3 + \frac{c_4}{r^2} \\
&\quad - \frac{1}{30\eta r^2} \int_r^\infty dr_1 Q(r_1) (r^5 - 5r^3 r_1^2 + 5r_1^3 r^2 - r_1^5), \quad (10)
\end{aligned}$$

where  $c_1, c_2, c_3, c_4$  are arbitrary constants. The velocity of the liquid is

$$\mathbf{v} = \text{rot } \mathbf{A} = (a'' + a'/r)\mathbf{G} - (a'' - a'/r)\mathbf{n}(\mathbf{G}\mathbf{n}). \quad (11)$$

Assume that the solid in question is maintained at rest by an applied external force ( $-\mathbf{F}$ ). The velocity  $\mathbf{v}(\mathbf{r})$  of the liquid is then zero for all  $\mathbf{n}$  not only at  $r = R$ , but also as  $r \rightarrow \infty$ . In this formulation of the problem, the sought quantity is the force  $\mathbf{F}$  exerted on the body by the liquid. From (11) we find that  $c_1 = c_2 = 0$ ,

$$c_3 = \frac{R^3}{12\eta} \int_R^\infty dr Q(r) \left( 1 - 3\frac{r^2}{R^2} + 2\frac{r^3}{R^3} \right), \quad (12)$$

$$c_4 = -\frac{R^5}{2\eta} \int_R^\infty dr Q(r) \left( \frac{1}{10} - \frac{r^2}{6R^2} + \frac{r^5}{15R^5} \right). \quad (13)$$

The constant  $c_3$ , as seen from (11)–(13), determines the asymptotic behavior of the velocity at sufficiently large  $r$ :

$$\mathbf{v}(\mathbf{r}) = c_3 [(\mathbf{G}\mathbf{n})\mathbf{n} + \mathbf{G}]/r. \quad (14)$$

From the second equation of the system (3) we obtain the pressure  $P$  (reckoned from its value at  $r = \infty$ ):

$$P(r) = 2\eta c_3 \mathbf{G}\mathbf{n}/r^2. \quad (15)$$

The force  $\mathbf{F}$  is determined by the integral

$$\mathbf{F}_i = -\oint \Pi_{ik} n_k dS$$

of the momentum-flux tensor  $\Pi_{ik}$  over a sphere of radius  $r \rightarrow \infty$ . In view of the rapid decrease of the function  $Q(r)$ , this tensor is determined at large distances  $r$  by the usual equation

$$\Pi_{ik} = P\delta_{ik} - \eta(\partial v_i/\partial x_k + \partial v_k/\partial x_i).$$

Substituting here (14) and (15), we get

$$\mathbf{F} = -\frac{2\pi}{3} \mathbf{G} R^3 \int_R^\infty dr Q(r) \left( 1 - 3\frac{r^2}{R^2} + 2\frac{r^3}{R^3} \right). \quad (16)$$

The use of the explicit expression given above for the function  $Q(r)$  yields ultimately

$$\mathbf{F} = -\mathbf{G} (R/\lambda)^{3/2} (I_1 + \gamma I_2), \quad (17)$$

where  $I_1 \approx 0.049$  and  $I_2 \approx 0.071$  are numerical constants given by

$$\begin{aligned}
I_1 &= \frac{1}{16\pi^{1/2}} \int_0^1 dx x^{-3/2} (1+x) \left[ \frac{1}{4} (1+x)(1-x)^{3/2} \right. \\
&\quad \left. - \frac{4}{45} (1-x)(1-x^2)^{3/2} \right. \\
&\quad \left. - \frac{1}{12} (1-x)^{3/2} - \frac{1}{20} (1+x)^2 (1-x)^{1/2} - \frac{1}{36} (1+x)^3 (1-x)^{-1/2} \right] \\
&\quad - \frac{1}{48\pi^{1/2}} \int_0^1 dx x^{-3/2} (x^2 + x + 4) \left[ (1-x)^{1/2} - \frac{1}{5} (1+x)(1-x)^{-1/2} \right. \\
&\quad \left. - \frac{4}{5} (1-x)(1-x^2)^{-1/2} \right], \\
I_2 &= \frac{3}{32\pi^{1/2}} \int_0^1 x^{1/2} dx (1-x^2) \int_0^\infty w dw (w^2 + x^2)^{-3/2} [w + (1+x^2)^{1/2}]^{-1/2} \\
&\quad + \frac{1}{16\pi^{1/2}} \int_0^1 x^{-1/2} dx (2+x) \left[ -\frac{1}{2} (1-x)^{1/2} \right. \\
&\quad \left. + \frac{4}{9} (1-x)^2 (1-x^2)^{-1/2} \right. \\
&\quad \left. + \frac{1}{18} (1+x)^2 (1-x)^{-1/2} \right].
\end{aligned}$$

It is interesting that the sign of the force  $F$  acting on the splid depends on the ratio of the thermal conductivities of the solid and of the liquid. The force is directed along the heat flow (counter to the temperature gradient) at  $\kappa_s/\kappa_l < 8.0$  and in the opposite direction for the inverse inequality.

A convenient method that might be used to observe thermophoresis in a liquid was used in the experiments of Seidel *et al.*<sup>13</sup> on the equilibrium of small hydrogen particles in a volume filled with helium in which a temperature gradient was present. In the absence of thermophoresis, gravity places the hydrogen particles on a level where the densities of the hydrogen particles and of the helium are equal. The thermophoresis force  $F$  displaces the equilibrium position by an amount  $h$  given by ( $g$  is the free-fall acceleration)

$$F = \frac{4}{3} \pi R^3 g \frac{\partial \rho}{\partial T} G h.$$

The displacement  $h$  does not depend on the temperature gradient, and is inversely proportional to  $R^{3/2}$  for the case of solid impurity particles. The function  $h(R)$  determines therefore the difference between the equilibrium positions of particles of radius  $R$  and relatively large particles.

Seidel *et al.*<sup>13</sup> investigated the equilibrium of liquid and solid hydrogen particles in helium. They reported observation of the discussed effect for the liquid but not solid hydrogen particles. The liquid particles are acted upon by a specific thermophoresis mechanism due to the temperature dependence of the surface tension on the interface between the two liquids<sup>13</sup>; this tension leads to a force  $F$  proportional to  $R^2$ , meaning larger than  $R^{3/2}$ . Thermophoresis for liquid particles is thus considerably stronger than for solid ones.

The calculations for solid particles can be easily carried out using the equations above and the known<sup>14</sup> data on the properties helium, and putting by way of estimate  $\kappa_s = \infty$ ,  $\lambda = (4/3)(\eta/\rho c)$ . Under conditions close to those

used by Seidel *et al.*,<sup>13</sup> i.e., at 15 atm and 11 K, we obtain  $h \approx 2 \cdot 10^2$  cm for  $R \approx 1 \mu\text{m}$ . The displacement  $h$  is independent of the temperature gradient if the latter is small, and reaches quite reasonable values. The temperature gradient, however, influences substantially the time to establish equilibrium. The velocity of a solid particle acted upon by a force  $F$  at  $G \approx \text{K/cm}$ , determined from the known Stokes equation, is under these conditions  $F/6\pi\eta R \approx 10^{-5}$  cm/s. The relaxation time is thus of the order of  $2 \cdot 10^3$  s, i.e., quite appreciable but fully acceptable. It can therefore be assumed that a quantitative investigation of the discussed effect for solid particles is experimentally perfectly realistic.

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