

Lift force on spherical nanoparticles in shear flows of rarefied binary gas mixtures

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In this work, we study the lift force on spherical nanoparticles suspended in a shear flow of rarefied binary gas mixtures. Analytical formulae are developed using the gas kinetic theory by considering non-rigid-body intermolecular interactions between the particle and gas molecules. It has been shown that the lift force formulae can be reduced to those in pure gases. It is also found that the direction of the lift force on nanoparticles in binary gas mixtures can be changed by varying the temperature, gas–particle interaction and/or gas concentrations.

Key words: kinetic theory, particle/fluid flow, rarefied gas flow

1. Introduction

A small particle in a shear flow usually experiences a force perpendicular to the flow direction due to the flow velocity gradient (Stone 2000). This phenomenon was first observed in capillaries where blood corpuscles tended to keep away from the walls (Poiseuille 1836). The force acting on the particle is termed ‘lift force’, which directly affects the particle migration and plays a crucial role in a variety of areas such as aerosol mechanics, chemical engineering, environmental science and biology (Herron, Davis & Bretherton 1975; Gavze & Shapiro 1998; Loth 2008; Zheng & Silber-Li 2009). Therefore, it is of great importance to develop a quantitative description for the lift force in shear flows.

In a gaseous medium, the Knudsen number Kn ($Kn = \lambda/R$, where λ is the mean free path of the gas and R is the particle radius) plays a key role in the study of the lift force. In the continuum regime ($Kn \ll 1$), the linear analysis based on the Stokes solution predicts no lift force (Bretherton 1962; Dandy & Dwyer 1990). Thus, the lift force can be attributed to the small inertia effects, and perturbation methods have been used to study the inertia terms in the Navier–Stokes equations (McLaughlin 1991; Asmolov 1999). Saffman (1965) gave an asymptotic solution for the lift force on a rigid sphere at low Reynolds numbers. The Saffman lift force in this regime is in the same direction as the flow velocity gradient, which is consistent with many

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experimental results (Merzkirch & Bracht 1978; Busnaina, Taylor & Kashkoush 1993). The particle in a shear flow may rotate in response to the torque induced by the shear stress (Bagchi & Balachandar 2002). Nevertheless, there is very weak coupling between the shear effect and the rotation effect at low Reynolds numbers (Saffman 1965; Kurose & Komori 1999; Bagchi & Balachandar 2002). In the free-molecule regime ($Kn \gg 1$), the velocity distribution of the gas molecules is not affected by the presence of the nanoparticles and the collisions between gas molecules and the particle dominate the gas–particle momentum transfer (Li 2009). The magnitude and direction of the lift force in this case can be determined by evaluating the momentum transfer upon collisions between gas molecules and the particle. Kröger & Hütter (2006) studied the lift force on a translationally moving particle in a shear flow at high Knudsen numbers. Their investigation suggests that the direction of the lift force is opposite to the flow velocity gradient, which is different from Saffman’s result. Several years ago, Liu & Bogy (2008, 2009) derived an expression for the lift force on spherical particles in a gas shear flow based on the rigid-body collision model between gas molecules and the particle, which is given by

$$F_L = -\frac{1}{3}\pi\rho GR^2\lambda V, \quad (1.1)$$

where ρ is the gas density, \mathbf{G} is the velocity gradient in the shear flow and V is the velocity of the particle relative to the gas. The lift force in (1.1) is also in the opposite direction to the velocity gradient, which confirms the prediction by Kröger & Hütter (2006). It should be noted that the rigid-body collision assumption for (1.1) is reasonable for relatively large particles. For nanoparticles, the molecular interactions between gas molecules and the particle may be significant and play an essential role (Li & Wang 2003a,b, 2004, 2005; Wang & Li 2011, 2012). Hence, (1.1) may not work well for nanoparticles. Recently, Luo *et al.* (2016) considered the influence of gas–particle intermolecular interactions on the lift force in pure gases and obtained a lift force formula, which is given by

$$F_L = \frac{1}{3}\pi\rho GR^2\lambda V \left(5\frac{\Omega_{gp}^{(1,1)*}}{\Omega_g^{(2,2)*}} - 6\frac{\Omega_{gp}^{(1,2)*}}{\Omega_g^{(2,2)*}} \right), \quad (1.2)$$

where $\Omega_{gp}^{(1,1)*}$ and $\Omega_{gp}^{(1,2)*}$ are reduced collision integrals, which refer to gas–particle interactions, and $\Omega_g^{(2,2)*}$ is the reduced collision integral for gas–gas interactions. Equation (1.2) includes (1.1) as a special case as the reduced collision integrals are equal to 1 for rigid-body collisions. Moreover, it is found that the lift force on nanoparticles could be in the same direction as or in the opposite direction to the velocity gradient, depending on the gas–particle interaction potential and temperature.

In practical applications, the motion of particles in shear flows of gas mixtures is often encountered (Zou *et al.* 2007; Kleinstreuer, Zhang & Donohue 2008; Salmanzadeh *et al.* 2012). For binary gas mixtures, where the velocity distribution of gas molecules depends on the concentration distribution and the interactions between the two gas species, a nonlinear dependence of the lift force on the gas component concentration is expected according to the Chapman–Enskog theory (Chapman & Cowling 1970; Wang & Li 2011). Unfortunately, the lift force on nanoparticles in shear flows of gas mixtures is poorly understood. This is primarily due to the complexity of mathematical analysis of the velocity distribution function in gas mixtures.

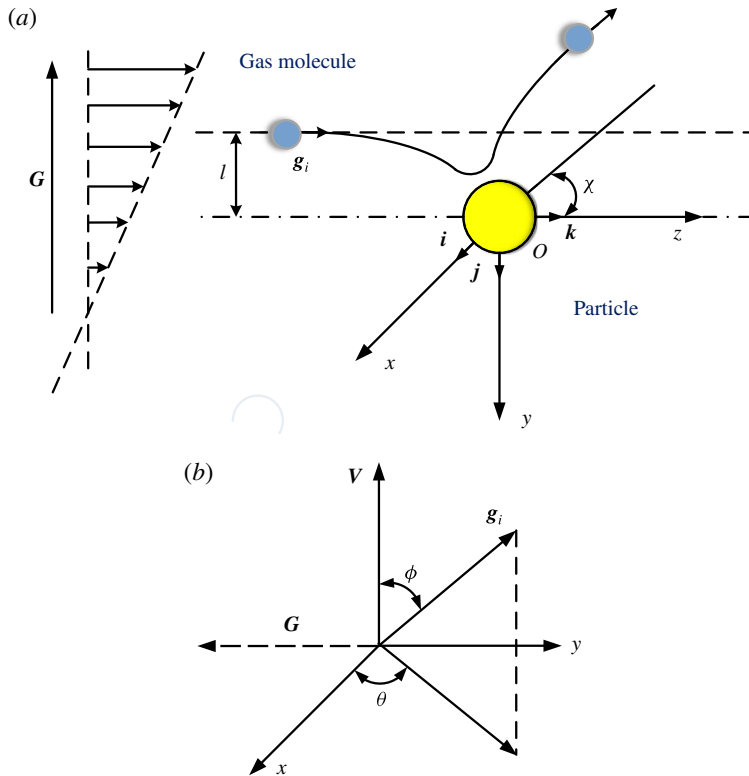


FIGURE 1. (Colour online) (a) Collision model between a gas molecule and a small particle in a linear shear flow. (b) Relationship among various vectors.

This work is devoted to generalize our previous work in pure gases to binary gas mixtures. The rest of the paper is organized as follows. Section 2 gives the assumptions and the velocity distribution function of the gas molecules in a binary gas mixture under a velocity gradient. In §3, the formulae for the lift forces on nanoparticles in a binary gas mixture are derived on the basis of gas kinetic theory. It is shown that the lift force formulae can be simplified to those in pure gases. As an example, the lift forces on a silver nanoparticle in gas mixtures are illustrated in §4. Finally, we conclude the paper in §5.

2. Assumptions and velocity distribution function

Consider a spherical particle moving at velocity \mathbf{V}_p in a linear shear flow, where \mathbf{V}_p is in the same direction as the velocity of the shear flow. The local mass velocity of the gas mixture is \mathbf{V}_0 , and \mathbf{v}_i is the velocity of the gas molecule relative to \mathbf{V}_0 , i.e. \mathbf{v}_i is the peculiar velocity and the subscript i denotes the gas species i . The particle velocity relative to the local mass velocity of the gas is \mathbf{V} ($\mathbf{V} = \mathbf{V}_p - \mathbf{V}_0$), and its direction is perpendicular to the velocity gradient of the shear flow \mathbf{G} . A reference frame is introduced with origin located at the mass centre of the particle, as shown in figure 1. The relative velocity \mathbf{V} is in the positive z -direction and the velocity gradient \mathbf{G} is in the negative y -direction.

Based on the gas kinetic theory (Chapman & Cowling 1970; Bird 1994; Li & Wang 2003a,b), the force acting on the particle can be obtained by finding the

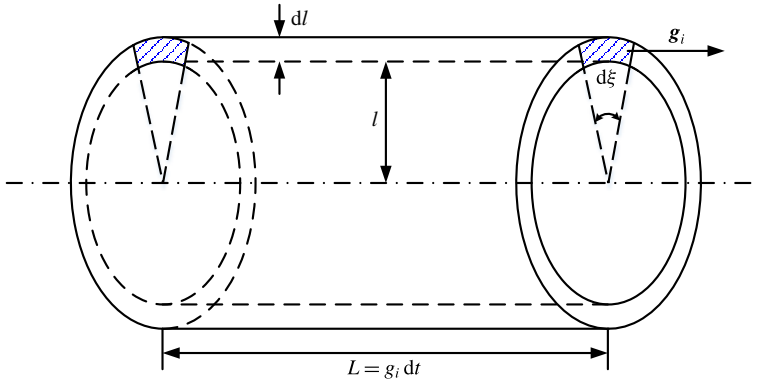


FIGURE 2. (Colour online) Gas molecules travel in a cylindrical region.

total momentum transfer from the gas molecules to the particle. The gas molecules travelling in a cylindrical shell with the same impact parameter l and the same relative velocity g_i ($g_i = v_i - V$) will undergo collisions with the particle during a short time dt . Then, the length of this cylinder is $L = g_i dt$ (see figure 2), and the number of gas molecules in the small sector of this cylindrical shell is $f_i g_i l dl d\xi dt$, where f_i is the velocity distribution of the gas molecules. The total momentum transfer of the gas molecules in the whole real space and velocity space can be obtained by integrating the momentum transfer of the gas molecules over l , ξ and v_i . As a result, the force experienced by the particle upon collisions with gas molecules is given by

$$F_i = m_{ip} \int_{v_i} g_i g_i f_i Q(g_i) dv_i, \quad i = 1, 2, \tag{2.1}$$

where $m_{ip} = m_i m_p / (m_i + m_p)$ is the reduced mass, with m_p and m_i being the particle mass and gas molecular mass of species i , respectively. In (2.1), $Q(g_i)$ represents the collision cross-section, which depends on the scattering scenario of the gas molecules upon collisions with the particle. Specular and diffuse scattering are the two limiting models of collision (Hirschfelder *et al.* 1954; Chapman & Cowling 1970). For specular scattering,

$$Q_s(g_i) = 2\pi \int_0^\infty (1 - \cos \chi) l dl, \tag{2.2}$$

with χ being the scattering angle given by

$$\chi = \pi - 2l \int_{r_m}^\infty \frac{dr}{r^2 \sqrt{1 - \frac{l^2}{r^2} - \frac{U(r)}{m_{ip} g_i^2 / 2}}}, \tag{2.3}$$

where r is the separation between the particle and gas molecule, r_m is the distance of closest encounter and $U(r)$ is the gas–particle interaction potential. The collision cross-section for diffuse scattering is defined as

$$Q_d(g_i) = 2\pi \left[\int_0^{l_0} \left(1 + \frac{1}{g_i} \sqrt{\frac{\pi k_B T}{2m_{ip}}} \sin \frac{\chi}{2} \right) + \int_{l_0}^\infty (1 - \cos \chi) \right] l dl, \tag{2.4}$$

where k_B is the Boltzmann constant, T is the temperature l_0 is the critical impact parameter. Diffuse scattering can only occur if the impact parameter is less than l_0 and the gas molecule collides with the particle physically; if the impact parameter is greater than l_0 , then the gas molecule just grazes the particle and the scattering is considered to be specular.

As mentioned above, the velocity distribution of the gas molecules is required to calculate the force acting on the particle. In the free-molecule regime, whereby the mean free path of the gas is much larger than the radius of the spherical particle, the interactions between the incident gas molecules and those reflected by the particle can be ignored. Hence, the velocity distribution of gas molecules can be obtained by neglecting the presence of the particle (Liu & Bogoy 2008). For a binary gas mixture, the velocity distribution function of the two gas species is more complicated than that of pure gases. According to the Chapman–Enskog theory (Chapman & Cowling 1970), in the presence of a velocity gradient, the second-order approximation to the velocity distribution function is $f_i = f_i^{(0)} + f_i^{(1)}$, where the first term $f_i^{(0)}$ is the molecular velocity distribution function in uniform state, and the second term $f_i^{(1)}$ accounts for the flow velocity gradient. Thus, the integral in (2.1) consists of two terms, the first of which is the drag force and the second of which is the lift force. The present paper focuses on the lift force, which involves $f_i^{(1)}$ (Chapman & Cowling 1970; Bird 1994)

$$f_i^{(1)} = -2f_i^{(0)} \mathbf{B}_i : \nabla \mathbf{V}_0, \quad i = 1, 2, \tag{2.5}$$

where $f_i^{(0)}$ is the velocity distribution function in uniform state given by

$$f_i^{(0)} = n_i \left(\frac{m_i}{2\pi k_B T} \right)^{3/2} \exp\left(-\frac{m_i v_i^2}{2k_B T} \right), \quad i = 1, 2, \tag{2.6}$$

where n_i is the number density of gas i , and \mathbf{B}_i is a symmetrical and non-divergent tensor, which is obtained as a series of Sonine polynomials. Under the first-order approximation,

$$\mathbf{B}_i = b_i \frac{m_i}{2k_B T} \mathbf{v}_i \overset{\circ}{\mathbf{v}}_i, \quad i = 1, 2, \tag{2.7}$$

where b_i is a function of gas composition, temperature, gas molecular mass and the intermolecular potential among gas molecules. The superscript ‘ \circ ’ above $\mathbf{v}_i \mathbf{v}_i$ denotes that the tensor is non-divergent. For the flow in figure 1, $f_i^{(1)}$ can be written as

$$f_i^{(1)} = -f_i^{(0)} b_i \frac{m_i}{k_B T} v_{y,i} v_{z,i} G, \quad i = 1, 2, \tag{2.8}$$

where $v_{y,i}$ and $v_{z,i}$ are the peculiar velocity components in y - and z -directions, respectively.

3. Lift force in a binary gas mixture

3.1. Lift force formula on a spherical nanoparticle by gas i

By substituting (2.8) into (2.1), the lift force $F_{L,i}$ can be written as

$$\mathbf{F}_{L,i} = -\frac{m_{ip} m_i b_i G}{k_B T} \int_{\mathbf{v}_i} g_i \mathbf{g}_i f_i^{(0)} v_{y,i} v_{z,i} Q(g_i) d\mathbf{v}_i. \tag{3.1}$$

As shown in figure 1(b), ϕ and θ are the colatitude and azimuthal angles of \mathbf{g}_i in the frame of reference, respectively. The relative velocity vector \mathbf{g}_i is therefore given by

$$\mathbf{g}_i = g_i \sin \phi \cos \theta \mathbf{i} + g_i \sin \phi \sin \theta \mathbf{j} + g_i \cos \phi \mathbf{k}. \tag{3.2}$$

Since the lift force is collinear with the velocity gradient, only the y -component of \mathbf{g}_i needs to be considered, and it is easy to check that the first and third terms in (3.2) vanish upon integration over the angles in spherical coordinates. This simplifies (3.1) to

$$\mathbf{F}_{L,i} = \frac{m_{ip}m_i b_i G}{k_B T} \int_{v_i} g_i^3 \sin^2 \phi \sin^2 \theta (g_i \cos \phi + V) f_i^{(0)} Q(g_i) d\mathbf{v}_i. \tag{3.3}$$

Substituting (2.6) into (3.3), we obtain the lift force (a detailed derivation is given in appendix A)

$$\mathbf{F}_{L,i} = \frac{8}{15} m_{ip} \sqrt{2\pi k_B T / m_i} G V R^2 n_i b_i [5\Omega_{ip}^{(1,1)*} - 6\Omega_{ip}^{(1,2)*}], \tag{3.4}$$

where $\Omega_{ip}^{(k,q)*}$ is the reduced collision integral given by

$$\Omega_{ip}^{(k,q)*} = \int_0^\infty \frac{e^{-\gamma_i^2} \gamma_i^{2q+3} Q^k(g_i) d\gamma_i}{[(q+k)!/2] \{1 - [1 + (-1)^k]/(2+2k)\} \pi R^2}, \tag{3.5}$$

and $\gamma_i = g_i \sqrt{m_i / (2k_B T)}$. More details can be found in appendix A.

3.2. Total lift force on a spherical nanoparticle in binary gas mixtures

Since the mass of the particle is usually much larger than that of a gas molecule, i.e. $m_i/m_{ip} \approx 1$, then the total lift force can be expressed as

$$\begin{aligned} \mathbf{F}_L = \mathbf{F}_{L1} + \mathbf{F}_{L2} = & \frac{8GVR^2 \sqrt{2\pi k_B T}}{15} [n_1 b_1 \sqrt{m_1} (5\Omega_{1p}^{(1,1)*} - 6\Omega_{1p}^{(1,2)*}) \\ & + n_2 b_2 \sqrt{m_2} (5\Omega_{2p}^{(1,1)*} - 6\Omega_{2p}^{(1,2)*})]. \end{aligned} \tag{3.6}$$

In (3.6), the coefficients b_1 and b_2 are given by (Chapman & Cowling 1970)

$$b_1 = \frac{\beta_1 b_{22} - \beta_2 b_{12}}{\mathcal{B}}, \quad b_2 = \frac{\beta_2 b_{11} - \beta_1 b_{12}}{\mathcal{B}}, \tag{3.7a,b}$$

where $\beta_1 = 5n_1 / (2n^2)$, $\beta_2 = 5n_2 / (2n^2)$ and \mathcal{B} is the symmetric determinant,

$$\mathcal{B} = \begin{vmatrix} b_{11} & b_{12} \\ b_{12} & b_{22} \end{vmatrix}. \tag{3.8}$$

For binary gas mixtures, the expressions for the elements of determinant \mathcal{B} are

$$b_{11} = \sqrt{\frac{k_B T}{2\pi m_0}} \left\{ \frac{4n_1^2}{n^2} \sqrt{\frac{2}{M_1}} \Omega_{11}^{(2,2)} + \frac{16n_1 n_2}{3n^2} M_2 \sqrt{\frac{1}{M_1 M_2}} \left[5M_1 \Omega_{12}^{(1,1)} + \frac{3}{2} M_2 \Omega_{12}^{(2,2)} \right] \right\}, \tag{3.9}$$

$$b_{22} = \sqrt{\frac{k_B T}{2\pi m_0}} \left\{ \frac{4n_2^2}{n^2} \sqrt{\frac{2}{M_2}} \Omega_{22}^{(2,2)} + \frac{16n_1 n_2}{3n^2} M_1 \sqrt{\frac{1}{M_1 M_2}} \left[5M_2 \Omega_{12}^{(1,1)} + \frac{3}{2} M_1 \Omega_{12}^{(2,2)} \right] \right\}, \tag{3.10}$$

$$b_{12} = \sqrt{\frac{k_B T}{2\pi m_0}} \left\{ \sqrt{\frac{1}{M_1 M_2}} \left(-\frac{16n_1 n_2}{3n^2} \right) M_1 M_2 \left[5\Omega_{12}^{(1,1)} - \frac{3}{2} \Omega_{12}^{(2,2)} \right] \right\}, \tag{3.11}$$

where $M_1 = m_1/m_0$, $M_2 = m_2/m_0$ and $m_0 = m_1 + m_2$. Here, $\Omega_{ij}^{(k,q)}$ is defined as (Hirschfelder *et al.* 1954)

$$\Omega_{ij}^{(k,q)} = \int_0^\infty e^{-\gamma_{ij}^2} \gamma_{ij}^{2q+3} Q^k(g_{ij}) d\gamma_{ij}, \tag{3.12}$$

which depends on the intermolecular interactions between gas molecules. It should be noted that the mass in the dimensionless quantity γ_{ij} is the reduced mass between two gas molecules.

For simplicity, hereafter, (3.9)–(3.11) will be represented by

$$b_{11} = \sqrt{\frac{k_B T}{2\pi m_0}} \tilde{b}_{11}, \quad b_{22} = \sqrt{\frac{k_B T}{2\pi m_0}} \tilde{b}_{22}, \quad b_{12} = \sqrt{\frac{k_B T}{2\pi m_0}} \tilde{b}_{12}, \tag{3.13a–c}$$

where \tilde{b}_{11} , \tilde{b}_{22} and \tilde{b}_{12} denote the terms in the curly braces in (3.9), (3.10) and (3.11), respectively. Then (3.7) can be rewritten as

$$b_1 = \frac{\beta_1 b_{22} - \beta_2 b_{12}}{\mathcal{B}} = \frac{\beta_1 \tilde{b}_{22} - \beta_2 \tilde{b}_{12}}{\sqrt{\frac{k_B T}{2\pi m_0}} (\tilde{b}_{11} \tilde{b}_{22} - \tilde{b}_{12}^2)} = \sqrt{\frac{2\pi m_0}{k_B T}} \tilde{b}_1, \tag{3.14a}$$

$$b_2 = \frac{\beta_2 b_{11} - \beta_1 b_{12}}{\mathcal{B}} = \frac{\beta_2 \tilde{b}_{11} - \beta_1 \tilde{b}_{12}}{\sqrt{\frac{k_B T}{2\pi m_0}} (\tilde{b}_{11} \tilde{b}_{22} - \tilde{b}_{12}^2)} = \sqrt{\frac{2\pi m_0}{k_B T}} \tilde{b}_2. \tag{3.14b}$$

Putting (3.14) into (3.6), the total lift force reads

$$\mathbf{F}_L = \frac{16\pi m_0 \mathbf{G} V R^2}{15} [n_1 \tilde{b}_1 \sqrt{M_1} (5\Omega_{1p}^{(1,1)*} - 6\Omega_{1p}^{(1,2)*}) + n_2 \tilde{b}_2 \sqrt{M_2} (5\Omega_{2p}^{(1,1)*} - 6\Omega_{2p}^{(1,2)*})]. \tag{3.15}$$

It should be noted that M_i and $\Omega_{ip}^{(k,q)*}$ in (3.15) are dimensionless, whereas $n_i \tilde{b}_i$ has the same unit as $1/R^2$. Letting $\Pi_i^* = R^2 n_i \tilde{b}_i \sqrt{M_i} (5\Omega_{ip}^{(1,1)*} - 6\Omega_{ip}^{(1,2)*})$, the lift force can be expressed as

$$\mathbf{F}_L = \frac{16\pi m_0 \mathbf{G} V}{15} (\Pi_1^* + \Pi_2^*), \tag{3.16}$$

where Π_1^* and Π_2^* are dimensionless quantities, which contain the interactions between gas species and the particle. It is worth mentioning that \mathbf{F}_{L1} and \mathbf{F}_{L2} in (3.6) are coupled with each other via the coefficients b_1 and b_2 , which depend on the interactions between the two gas species. Thus, the total lift force \mathbf{F}_L is not simply the linear summation of the lift force exerted by each gas component.

3.3. Lift force in a pure gas

It is expected that (3.6) can be simplified to (1.2) for pure gases if the two species are identical. Letting $\Omega_{1p}^{(1,1)*} = \Omega_{2p}^{(1,1)*} = \Omega_{gp}^{(1,1)*}$, $\Omega_{1p}^{(1,2)*} = \Omega_{2p}^{(1,2)*} = \Omega_{gp}^{(1,2)*}$ and $m_1 = m_2 = m_g$, equation (3.6) becomes

$$F_L = \frac{8GVR^2 \sqrt{2\pi k_B T m_g}}{15} (5\Omega_{gp}^{(1,1)*} - 6\Omega_{gp}^{(1,2)*})(n_1 b_1 + n_2 b_2), \tag{3.17}$$

and (3.9)–(3.11) can be rewritten as

$$b_{11} = \sqrt{\frac{k_B T}{\pi m_g}} \left\{ \frac{4n_1^2}{n^2} \Omega_g^{(2,2)} + \frac{8n_1 n_2}{3n^2} \left[\frac{5}{2} \Omega_g^{(1,1)} + \frac{3}{4} \Omega_g^{(2,2)} \right] \right\}, \tag{3.18}$$

$$b_{22} = \sqrt{\frac{k_B T}{\pi m_g}} \left\{ \frac{4n_2^2}{n^2} \Omega_g^{(2,2)} + \frac{8n_1 n_2}{3n^2} \left[\frac{5}{2} \Omega_g^{(1,1)} + \frac{3}{4} \Omega_g^{(2,2)} \right] \right\}, \tag{3.19}$$

$$b_{12} = \sqrt{\frac{k_B T}{\pi m_g}} \left(\frac{8n_1 n_2}{3n^2} \right) \left[\frac{3}{4} \Omega_g^{(2,2)} - \frac{5}{2} \Omega_g^{(1,1)} \right]. \tag{3.20}$$

By using (3.7) and (3.8),

$$n_1 b_1 + n_2 b_2 = \frac{5}{2} \left[\frac{n_1^2 b_{22} - 2n_1 n_2 b_{12} + n_2^2 b_{11}}{n^2 (b_{11} b_{22} - b_{12}^2)} \right]. \tag{3.21}$$

Then, by substituting (3.18)–(3.20) into (3.21), we obtain

$$n_1 b_1 + n_2 b_2 = \frac{5}{8 \sqrt{\frac{k_B T}{\pi m_g} \Omega_g^{(2,2)}}}. \tag{3.22}$$

As a result, (3.17) is simplified to

$$F_L = \frac{\sqrt{2\pi m_g}}{3\Omega_g^{(2,2)}} GR^2 V [5\Omega_{1p}^{(1,1)*} - 6\Omega_{1p}^{(1,2)*}], \tag{3.23}$$

which is the same as (1.2) by using $\rho = m_g n$, $\Omega_g^{(2,2)*} = \Omega_g^{(2,2)} / (2\pi\sigma_g^2)$ and $\lambda = 1/(\sqrt{2}\pi\sigma_g^2 n)$, where σ_g is the collision diameter of the gas molecules.

As another test of consistency, we consider the case in which the concentration of one gas component tends to zero. For instance, in the limit of $n_2 \rightarrow 0$ and $n_1 \rightarrow n$, $b_1 n_1 = 5\sqrt{\pi m_1} / (8\Omega_{11}^{(2,2)} \sqrt{k_B T})$, $b_2 n_2 = 0$ and (3.6) converges to that in a pure gas.

For large particles, the gas–particle interactions can be neglected and the rigid-body collision model is reasonable. In this case, (3.23) can be easily reduced to (1.1) by assuming rigid-body collisions. However, as the particle size decreases to the nanoscale, the influence of the van der Waals interactions between the gas molecules and particle on lift force becomes notable. This influence is manifested in the term $\Omega_{ip}^{(k,q)*}$.

The lift force given by (1.2) could be positive ($5\Omega_{gp}^{(1,1)*} > 6\Omega_{gp}^{(1,2)*}$) or negative ($5\Omega_{gp}^{(1,1)*} < 6\Omega_{gp}^{(1,2)*}$), depending on the gas–particle interactions. In most cases, the

direction of the lift force on nanoparticles in pure gases (1.2) is negative, i.e. opposite to the velocity gradient, which is consistent with (1.1). However, the positive lift force takes place in a certain temperature range for a given gas–particle interaction. The physical reason for the direction change is that the gas molecules on the low-velocity side transfer more momentum to the nanoparticle due to its larger scattering angles upon non-rigid-body collisions. As for binary gas mixtures, the total lift force is a nonlinear combination of individual lift forces contributed from species 1 and 2. A direction change of the lift force is expected if the lift forces of gas species 1 and 2 are in opposite directions, as will be demonstrated in the following section.

4. Lift force on a silver spherical nanoparticle in gas mixtures

As an example, a suspended silver nanoparticle in a Ne–Ar binary gas mixture is considered. A widely used model to describe the van der Waals interactions is the Lennard-Jones (LJ) 12–6 potential,

$$U_{12-6}(r) = 4\varepsilon \left[\left(\frac{\sigma}{r}\right)^{12} - \left(\frac{\sigma}{r}\right)^6 \right], \tag{4.1}$$

where ε and σ represent the binding energy and collision diameter, respectively. The LJ potential parameters for Ne ($\sigma_1 = 2.82 \text{ \AA}$ and $\varepsilon_1/k_B = 32.8 \text{ K}$), Ar ($\sigma_2 = 3.47 \text{ \AA}$ and $\varepsilon_2/k_B = 114.0 \text{ K}$) and Ag ($\sigma_p = 2.57 \text{ \AA}$ and $\varepsilon_p/k_B = 4075.0 \text{ K}$) are adopted from the literature (Svehla 1962; Hippler, Troe & Wendelken 1983; Agrawal, Rice & Thompson 2002). The potential parameters for the interactions between different species 1 and 2 are obtained using the conventional Lorentz–Berthelot rules, i.e. $\sigma_{12} = (\sigma_1 + \sigma_2)/2$ and $\varepsilon_{12} = \sqrt{\varepsilon_1 \varepsilon_2}$.

Figure 3 shows the dimensionless quantity Π_i^* of a silver nanoparticle in a rarefied Ne–Ar gas mixture as a function of reduced Ne concentration n_1/n . It is seen that F_L converges to $F_{L,1}$ (open circle) or $F_{L,2}$ (open triangle) as $n_1 \rightarrow n$ or $n_2 \rightarrow n$, respectively. It is also found that Π_i^* varies nonlinearly with Ne concentration. In (3.6), it seems that the total lift force is simply a linear superposition of the lift forces given by the gas components. However, both b_1 and b_2 depend on the composition ratio and the interactions between the two gas species, by which the dependence of the total lift force on the concentration is nonlinear.

As discussed by Luo *et al.* (2016), if the LJ 12–6 potential function is employed for the gas–particle interaction, a positive lift force that directs towards the high-velocity side exists in a certain reduced temperature range, $0.417 < T^* = k_B T / \varepsilon < 0.951$ (Wang & Li 2012; Khrapak 2014; Luo *et al.* 2016). Therefore, the lift forces acting on the nanoparticle in pure neon gas (F_{L1}) and pure argon gas (F_{L2}) become positive when the temperature is roughly in the ranges of 154–347 K and 286–648 K, respectively. If the temperature is extremely high or low, as shown in figure 3(a) for $T = 100 \text{ K}$ and figure 3(d) for $T = 700 \text{ K}$, since the lift forces in both gas species are negative, the total lift forces are definitely negative. At moderate temperatures, positive F_{L1} (Ne) and/or F_{L2} (Ar) can take place, as shown in figures 3(b) and 3(c). As previously mentioned, the total lift force consists of two contributions from species 1 and 2. If the lift forces of gas species 1 and 2 are in opposite directions, it is expected that the direction of the total lift force changes as the gas concentration varies. For example, in the case of $T = 500 \text{ K}$ (see figure 3c), F_{L1} (Ne) is negative and F_{L2} (Ar) is positive; as a result, the direction of F_L changes from positive to negative as the Ne concentration varies. This means that in a certain temperature range, there exists a critical value of

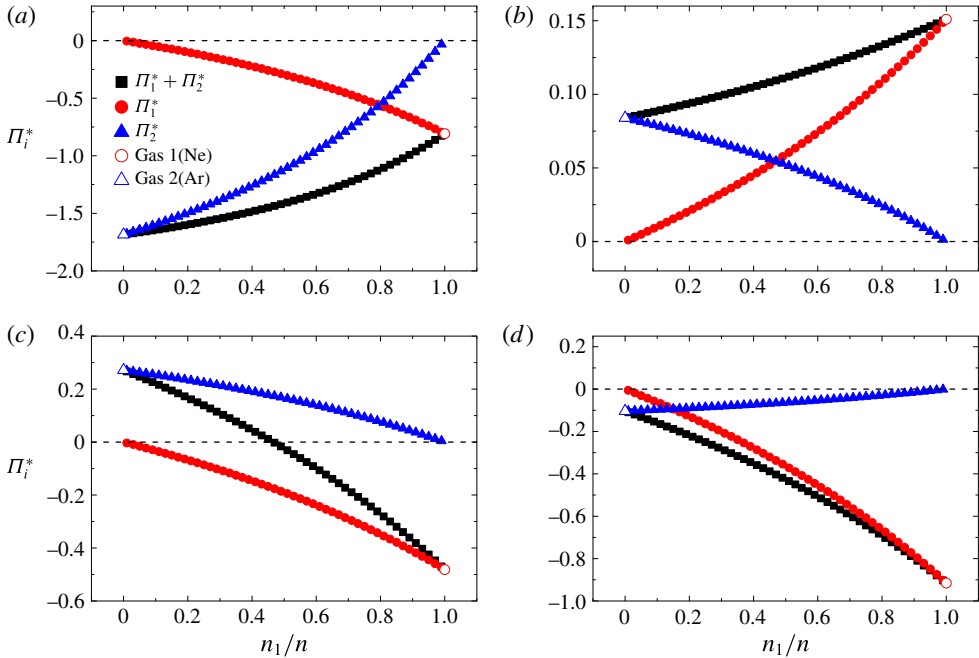


FIGURE 3. (Colour online) Variation of Π_i^* as a function of reduced Ne concentration n_1/n at $T=100$ K (a), $T=300$ K (b), $T=500$ K (c) and $T=700$ K (d). Open symbols corresponding to the results in pure gases.

n_1/n , where the total lift force changes its direction. However, the complex nonlinear dependence of the lift force on the gas concentrations makes it difficult to predict this critical value.

Figure 4 shows the dimensionless quantity Π_i^* of a silver nanoparticle in other gas mixtures (He–Ar and He–Kr gas mixtures). The collision diameter σ and binding energy ε/k_B for He and Kr are 2.55 Å, 10.22 K and 3.66 Å, 178.9 K, respectively (Svehla 1962). As can be seen from (3.16), the total molecular mass of the two species m_0 appears in the coefficient of $(\Pi_1^* + \Pi_2^*)$, and Π_1^* depends on $1/m_0$ as n_1/n converges to 1. Since the total molecular mass of He and Ar is different from that of He and Kr, the values of Π_1^* in figures 4(a) and 4(b) are not equal to each other as n_1/n converges to 1 (pure He gas). It has been checked that the values of Π_1^* in He–Ar and He–Kr gas mixtures are equal to each other if Π_1^* is multiplied by m_0 . By comparing figures 3(b) with 4, it is seen that the dependence of the total lift force on the concentration is still nonlinear and the degree of the nonlinearity between Π_i^* and n_1/n is different for different gas mixtures. The nonlinearity in the case of figure 4(b) is the most distinct one because the differences between the potential parameters and the molecular masses of He and Kr are the largest. Similarly, for the case in figure 3(b), the nonlinearity is weak because the potential parameters and the molecular mass of Ne are close to those of Ar. This is consistent with the discussion in §3.3: if the potential parameters and molecular mass of species 1 are identical to those of species 2, the formulae reduce to those in pure gases and the nonlinearity disappears.

It should be noted that the LJ 12–6 potential might not be an adequate model to describe the interactions between gas molecules and a particle. A rigorous description

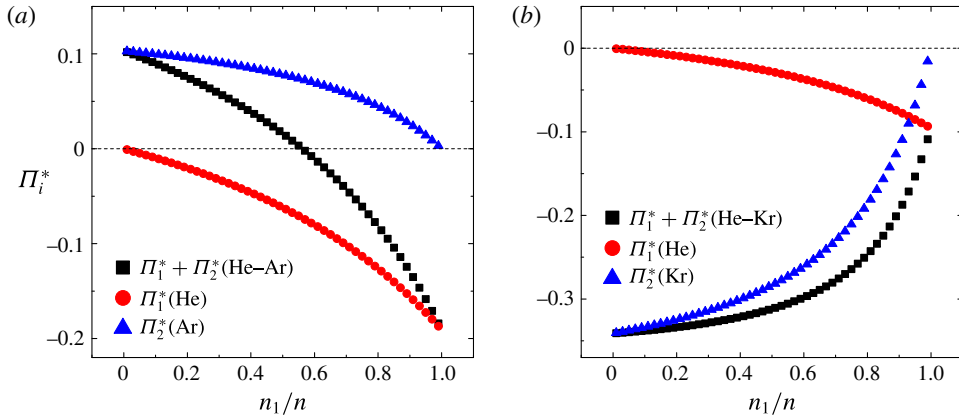


FIGURE 4. (Colour online) Variation of Π_i^* as a function of reduced He concentration n_1/n in He–Ar gas mixtures at $T = 300$ K (a) and in He–Kr gas mixtures at $T = 300$ K (b).

of the gas–particle interaction is unavailable. As a coarse approach, the potential for gas–particle interactions can be determined as a sum of the LJ interactions of gas molecules with all atoms in the particle. If the distribution of atoms in the particle is assumed to be continuous, integration can be carried out in place of summation (Rouquerol *et al.* 2013). In this way, the interaction between gas molecules and a single solid lattice plane is given by (Everett & Powl 1976)

$$U_{10-4}(r) = \frac{10}{3} \varepsilon \left[\frac{1}{5} \left(\frac{\sigma}{r} \right)^{10} - \frac{1}{2} \left(\frac{\sigma}{r} \right)^4 \right], \tag{4.2}$$

and the interaction between gas molecules and a semi-infinite slab of solid is given by (Everett & Powl 1976)

$$U_{9-3}(r) = \frac{3}{\sqrt{10}} \varepsilon \left[\frac{2}{15} \left(\frac{\sigma}{r} \right)^9 - \left(\frac{\sigma}{r} \right)^3 \right]. \tag{4.3}$$

Although there is no evidence that these two potential functions are suitable for gas–particle interactions, in the present paper, we consider them as a possible choice for illustrative purposes. It is expected that the r dependence for the gas–particle interaction potential is covered by these three cases (12–6, 10–4 and 9–3) or their combinations (Luo *et al.* 2016).

In figure 5, $\Pi_1^* + \Pi_2^*$ is depicted as a function of reduced Ne concentration n_1/n in a Ne–Ar gas mixture at various temperatures, wherein (4.1)–(4.3) are employed to model the interactions between the particle and gas molecules. Being similar to the results in figure 3, for interaction potentials given by (4.1)–(4.3), the dependence of $\Pi_1^* + \Pi_2^*$ on the single-species concentration ratio is also nonlinear. A positive lift force can be observed at certain temperature associated with the direction change from positive to negative with increasing Ne concentration. Furthermore, at the same concentration of Ne, the temperature value corresponding to the lift force direction change (if there is a change in lift force direction) increases from 12–6, to 10–4, to 9–3 potentials. This trend may be due to the increasing attractive interactions between gas molecules and the particle. Since we can observe the direction change of the lift

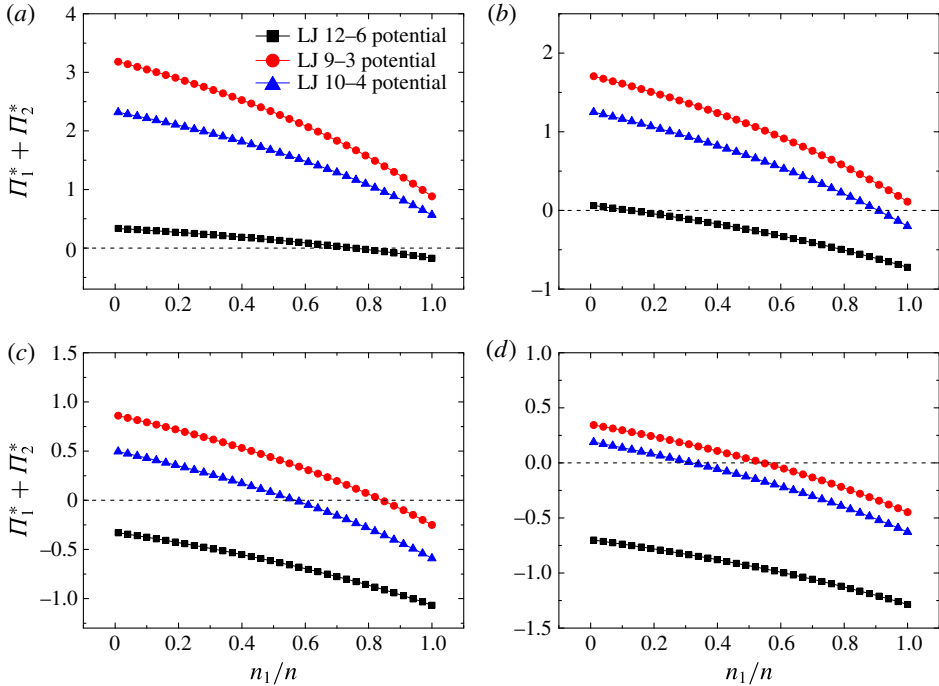


FIGURE 5. (Colour online) Variation of $\Pi_1^* + \Pi_2^*$ (Ne–Ar) as a function of reduced Ne concentration n_1/n for the different gas–particle interaction potential functions at $T = 400$ K (a), $T = 600$ K (b), $T = 800$ K (c) and $T = 1000$ K (d).

force with all three types of interaction potentials, it is expected that the direction change can be observed experimentally. Owing to the computational complexity of the reduced collision integrals, it is not easy to offer a rigorous treatment of the gas–particle interactions. Further intensive experimental studies and theoretical analyses are needed on this issue.

5. Conclusions

To summarize, the lift force on spherical nanoparticles in shear flows of rarefied binary gas mixtures is investigated. In the light of the importance of the molecular interactions between the gas molecules and a nanoparticle, we take into account the non-rigid-body collision effect in our model. On the basis of gas kinetic theory, we derive an analytical lift force formula by calculating the momentum transfer upon collisions between the nanoparticle and surrounding gas molecules. The expression involves terms of the collision integrals, which relies on the intermolecular interactions. The present formula is consistent with its counterpart in pure gases and can be simplified to that in the case of rigid-body collisions. For a particle suspended in a binary gas mixture, the total lift force acting on the particle is considered as the combination of the individual lift forces contributed by the two species, and the dependence of the total lift force on the gas concentration is nonlinear, which can contribute to the molecular interactions between two gas species.

Furthermore, it is illustrated that the positive lift force, which is in the opposite direction of the lift force in the case of rigid-body collisions and drives nanoparticles

towards high-velocity regions, may occur in binary gas mixtures. The emergence of a positive lift force is a result of the higher gas–particle momentum transfer in the low-velocity region compared with that in the high-velocity region. Three types of gas–particle interaction potential models are employed to evaluate the lift force. It is found that the direction of the lift force acting on nanoparticles depends on temperature, gas concentrations and potential parameters and the direction change of the lift force can be observed with varying gas concentration.

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Appendix A. Derivation of lift force on a spherical nanoparticle by gas i

By substituting (2.6) into (3.3), one obtains

$$\mathbf{F}_{L,i} = \frac{\sqrt{2\pi}m_{ip}n_i b_i \mathbf{G}}{4\pi^2} \left(\frac{m_i}{k_B T}\right)^{5/2} \int_{v_i} g_i^3 \sin^2 \phi \sin^2 \theta (g_i \cos \phi + V) \exp\left(-\frac{m_i v_i^2}{2k_B T}\right) Q(g_i) dv_i. \quad (\text{A } 1)$$

Considering the fact that the particle relative velocity V is much smaller than the peculiar velocity of the gas molecules v_i , the exponential term in (A 1) can be simplified to

$$\exp\left(-\frac{m_i v_i^2}{2k_B T}\right) = \exp\left(-\frac{g_i^2 + 2g_i V \cos \phi + V^2}{2k_B T/m_i}\right) \approx \exp\left(-\frac{m_i g_i^2}{2k_B T}\right) \exp\left(-\frac{m_i g_i V \cos \phi}{k_B T}\right), \quad (\text{A } 2)$$

where $\exp[-m_i g_i V \cos \phi / (k_B T)]$ can be expanded to

$$\exp\left(-\frac{m_i g_i V \cos \phi}{k_B T}\right) = 1 - \frac{m_i g_i V \cos \phi}{k_B T} + \frac{1}{2} \left(\frac{m_i g_i V \cos \phi}{k_B T}\right)^2 - \frac{1}{6} \left(\frac{m_i g_i V \cos \phi}{k_B T}\right)^3 + \dots \quad (\text{A } 3)$$

Since $k_B T \sim m_i g_i^2 \gg m_i g_i V$ and $m_i g_i V / (k_B T) \ll 1$, the second- and higher-order Taylor expansion terms in (A 3) can be neglected. Therefore, (A 2) can be simplified to

$$\exp\left(-\frac{m_i v_i^2}{2k_B T}\right) = \exp\left(-\frac{m_i g_i^2}{2k_B T}\right) \left(1 - \frac{m_i g_i V \cos \phi}{k_B T}\right). \quad (\text{A } 4)$$

Moreover, in the short time dt , \mathbf{V} can be treated as constant, thus

$$dv_i = dg_i = g_i^2 \sin \phi d\phi d\theta dg_i. \quad (\text{A } 5)$$

Substituting (A 4) and (A 5) into (A 1), we obtain

$$\begin{aligned} \mathbf{F}_{L,i} &= \frac{\sqrt{2\pi}m_{ip}n_i b_i \mathbf{G}}{4\pi^2} \left(\frac{m_i}{k_B T}\right)^{5/2} \int_{v_i} g_i^3 \sin^2 \phi \sin^2 \theta (g_i \cos \phi + V) \\ &\times \exp\left(-\frac{m_i g_i^2}{2k_B T}\right) \left(1 - \frac{m_i g_i V \cos \phi}{k_B T}\right) Q(g_i) dv_i \end{aligned}$$

$$\begin{aligned}
 &= \frac{\sqrt{2\pi}m_{ip}n_i b_i \mathbf{G}}{4\pi} \left(\frac{m_i}{k_B T}\right)^{5/2} \int_0^\infty \int_0^\pi g_i^5 \sin^3 \phi (g_i \cos \phi + V) \\
 &\quad \times \exp\left(-\frac{m_i g_i^2}{2k_B T}\right) \left(1 - \frac{m_i g_i V \cos \phi}{k_B T}\right) Q(g_i) d\phi dg_i. \quad (\text{A } 6)
 \end{aligned}$$

Integrating over ϕ , we have

$$F_{L,i} = \frac{\sqrt{2\pi}m_{ip}n_i b_i \mathbf{G} V}{\pi} \left(\frac{m_i}{k_B T}\right)^{5/2} \int_0^\infty \left(\frac{1}{3}g_i^5 - \frac{1}{15} \frac{m_i g_i^7}{k_B T}\right) \exp\left(-\frac{m_i g_i^2}{2k_B T}\right) Q(g_i) dg_i. \quad (\text{A } 7)$$

By using the definition of the reduced collision integrals (3.5) and $\gamma_i = g_i \sqrt{m_i/(2k_B T)}$, equation (A 7) can be rewritten as

$$\begin{aligned}
 F_{L,i} &= \frac{8m_{ip}n_i b_i \mathbf{G} V}{15\pi} \sqrt{2\pi k_B T/m_i} \int_0^\infty (5\gamma_i^5 - 2\gamma_i^7) \exp(-\gamma_i^2) Q(g_i) d\gamma_i \\
 &= \frac{8}{15} m_{ip} \sqrt{2\pi k_B T/m_i} \mathbf{G} V R^2 n_i b_i [5\Omega_{ip}^{(1,1)*} - 6\Omega_{ip}^{(1,2)*}], \quad (\text{A } 8)
 \end{aligned}$$

which is (3.4).

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