Marangoni instabilities of drops of different viscosities in stratified liquids

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(Received xx; revised xx; accepted xx)

1. Velocity magnitudes around a bouncing drop

To show that the velocity around a bouncing drop is indeed oscillatory, we measure the velocity around a bouncing drop at instances when the drop is close to its highest and lowest positions. The velocities are measured by PIV and shown in figure 1(*a*) and (*b*), respectively. The two frames correspond to 52 s and 67 s of the Supplementary Movie 3, respectively. The drop radius is $R = 0.59\,\mathrm{mm}$, and the concentration gradient of the mixture is $\mathrm{d}w_\mathrm{e}/\mathrm{d}y \approx 40\,\mathrm{wt}\%$. As can be seen from the legends, the maximum velocity magnitudes decreases from $\approx 0.2\,\mathrm{mm/s}$ at 52 s to $\approx 0.07\,\mathrm{mm/s}$ at 67 s.

For the PIV measurements, PSP (Polyamid Seeding Particles, Dantec Dynamics, Denmark) particles of diameter 20 µm are added in both ethanol and water at 0.4 mg/mL before making the linearly stratified mixture. The images are recorded with the same light-lens-camera system, and they are later analyzed by PIVlab toolbox in MATLAB (Thielicke & Stamhuis 2014; Thielicke 2014). Square interrogation windows of 32×32 pixels corresponding to grid cells of $115\,\mu\mathrm{m} \times 115\,\mu\mathrm{m}$ with an overlap of 50 % are used to obtain the velocity vectors. The relaxation time of the particle is estimated by

$$t_0 = \left(1 + \frac{\rho}{2\rho_\mathrm{p}}\right)\frac{\rho_\mathrm{p} d_\mathrm{p}^2}{18\mu} \tag{1.1}$$

where $\rho_\mathrm{p} = 1.05\,\mathrm{g/cm^3}$ and $d_\mathrm{p} = 20\,\mu\mathrm{m}$ are the density and diameter of the particle, $\rho \approx 911\,\mathrm{kg/m^3}$ and $\mu \approx 2\,\mathrm{mPa\cdot s}$ are the density and dynamic viscosity of the ethanol-water mixture. Then the Stokes number of the particles are calculated to be $St = t_0 u_\mathrm{max}/d_\mathrm{p} = 3 \times 10^{-7} \ll 1$, so that the tracking particles are considered to follow the flow field faithfully.

2. Measurement errors

Let $s(q)$ be the standard deviation of the quantity q, and $s_\mathrm{r} = s(q)/q$ be the relative deviation of quantity q. Quantity q can be a directly measured quantity like R, w_e, h, or can be any other calculated quantity, like $\mathrm{d}w_\mathrm{e}/\mathrm{d}y$, $\mathrm{d}\rho/\mathrm{d}y$, μ, D, Ma, Ra, etc.

Given the limited height of the container (30 mm in height), strictly linearly stratified mixtures are only made in the middle of the container, within a range of, say $\Delta h \approx 20\,\mathrm{mm}$. Take the case where $\mathrm{d}w_\mathrm{e}/\mathrm{d}y \approx 40\,\mathrm{m^{-1}}$ as an example (see figure 2). Above and

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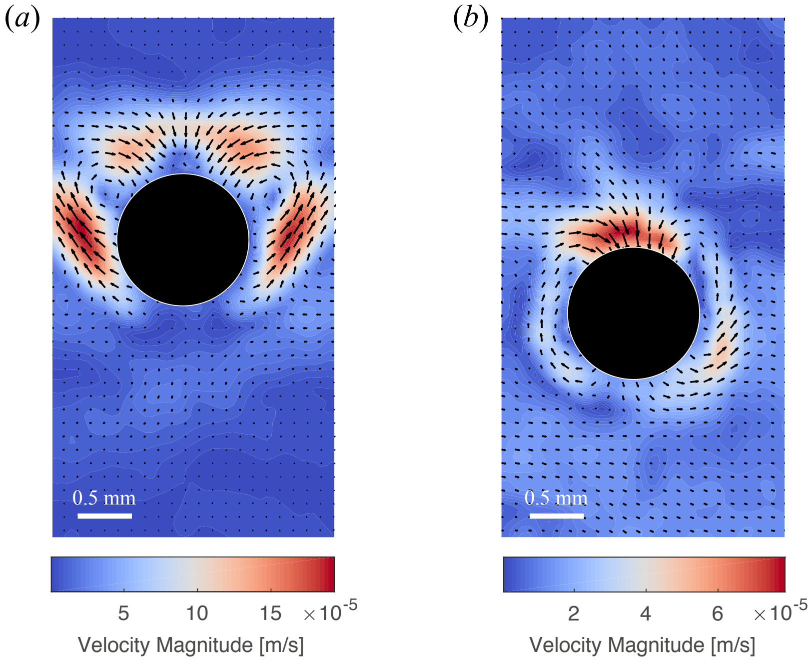

FIGURE 1. Velocities around a bouncing drop measured by PIV, at instances when the drop is close to its highest position (*a*) and lowest position (*b*). The drop radius is $R = 0.59\,\mathrm{mm}$, and the concentration gradient of the mixture is $\mathrm{d}w_\mathrm{e}/\mathrm{d}y \approx 40\,\mathrm{wt\%}$.

below the strictly linearly stratified region, we have $\approx 5\,\mathrm{mm}$ thick layers of ethanol-water mixtures with roughly uniform ethanol concentrations of $w_\mathrm{t} = 80.0\,\mathrm{wt\%}$ and $w_\mathrm{b} = 17.7\,\mathrm{wt\%}$, respectively. Notice that the measured ethanol weight fraction in the top layer is decreasing as the height h increases, this is due to the preferential evaporation of ethanol from the top layer. The maximum ethanol weight fraction in the top layer is $w_\mathrm{t} = 79.8\,\mathrm{wt\%}$ at $h = 12.74\,\mathrm{mm}$, meaning the measurement error for the top layer is $s_\mathrm{r}(w_\mathrm{t}) < 1\,\%$. These two solutions with concentrations w_t and w_b are in fact used to set up the linearly stratified region in between, so that w_t changes to w_b smoothly. The purpose of these two uniform layers is to increase the accuracy of w_e measured by laser deflection: They are made by mixing certain mass ratios of ethanol and water. The mass of ethanol and water are measured on a scale with precision down to 0.1 mg. Each time about 40 g of the mixture is made. Consequently, the relative measurement deviations (or uncertainties) of w_t and w_b are less than $1\,\%$. Consequently, the relative uncertainty of $\Delta w = w_\mathrm{t} - w_\mathrm{b}$ is also smaller than $1\,\%$, i.e., $s_\mathrm{r}(\Delta w) < 1\,\%$.

The uncertainties in h measurement is $s(h) = 0.03\,\mathrm{mm}$, so that $s(\Delta h) = 0.06\,\mathrm{mm}$ leads to $s_\mathrm{r}(h) < 1\,\%$. The gradient is calculated by $\mathrm{d}w_\mathrm{e}/\mathrm{d}y = \Delta w/\Delta h$. Then the relative uncertainty of $\mathrm{d}w_\mathrm{e}/\mathrm{d}y$ is:

$$s_\mathrm{r}(\mathrm{d}w_\mathrm{e}/\mathrm{d}y) = \sqrt{s_\mathrm{r}^2(\Delta w) + s_\mathrm{r}^2(\Delta h)} < 1\,\% \tag{2.1}$$

Since $s_\mathrm{r}(w_\mathrm{e}) < 1\,\%$, we have $s_\mathrm{r}(\mu + \mu') = s_\mathrm{r}(\mu) < 1\,\%$, $s_\mathrm{r}(D) < 1\,\%$, and $s_\mathrm{r}(\rho) < 1\,\%$. Thus $s_\mathrm{r}(\mathrm{d}\rho/\mathrm{d}y) < 1\,\%$.

The largest uncertainty originates from the measurement of the drop radius R. For example, for the drop $R_4 \approx 66\,\mu\mathrm{m}$ shown in figure 2(*d*), it is levitating but also close to the transition point. Its diameter is about 38 pixels wide in our camera, and the uncertainty in measuring it is 1 pixel. This gives us an estimation of the relative uncertainty of R:

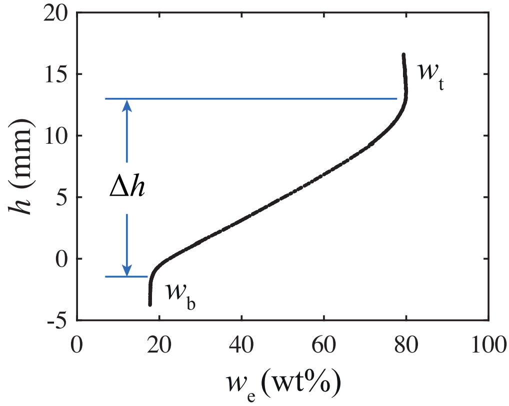

FIGURE 2. The ethanol weight fraction w_e in the whole container at $dw_e/dy \approx 40\,m^{-1}$. Strictly linearly stratified mixture is only made in the middle of the container within a range of $\Delta h \leqslant 20\,mm$. Above and below this region, we have $\approx 5\,mm$ thick layers of ethanol-water mixtures with roughly uniform ethanol concentrations of $w_t = 80.0\,wt\%$ and $w_b = 17.7\,wt\%$, respectively. The measured ethanol weight fraction in the top layer is decreasing as the height h increases because of the preferential evaporation of ethanol from the top layer. The maximum ethanol weight fraction in the top layer is $w_t = 79.8\,wt\%$ at $h = 12.74\,mm$, giving a measurement error $s(w_t) < 1\,wt\%$ for the top layer.

$s_r(R) \approx 2.6\,\%$. Then:

$$\begin{aligned} s_r(Ra) &= \sqrt{16 s_r^2(R) + s_r^2(d\rho/dy) + s_r^2(\mu+\mu') + s_r^2(D)} \\ &\approx 4 s_r(R) \\ &\approx 10.4\,\%. \end{aligned} \tag{2.2}$$

This is the error in calculating the Rayleigh number of this drop.

In addition, the error of the Marangoni number of this drop is:

$$\begin{aligned} s_r(Ma) &= \sqrt{4 s_r^2(R) + s_r^2(d\sigma/dy) + s_r^2(\mu+\mu') + s_r^2(D)} \\ &\approx 2 s_r(R) \\ &\approx 5.2\,\%. \end{aligned} \tag{2.3}$$

Since

$$\frac{Ra}{Ma} = \frac{\mu+\mu'}{\mu}\frac{d\rho}{d\sigma} g R^2, \tag{2.4}$$

we have

$$\begin{aligned} s_r\left(\frac{Ra}{Ma}\right) &= \sqrt{s_r^2(\mu+\mu') + s_r^2(\mu) + 4 s_r^2(R) + s_r^2(\frac{d\rho}{dy}) + s_r^2(\frac{d\sigma}{dy})} \\ &\approx 2 s_r(R) \\ &\approx 5.2\,\%. \end{aligned} \tag{2.5}$$

This is the error in calculating Ra/Ma of this drop.

Notice that for larger drops, $s_r(R)$ is smaller because the drop occupies more pixels in the recorded image. Thus, $s_r(Ra)$, $s_r(Ma)$ and $s_r(Ra/Ma)$ are all smaller for larger drops.

In the meantime, the experimental determination of levitating or bouncing of the drop could also induce some error. Since in the experiments we cannot vary the drop radius R and the gradient dw_e/dy continuously, it is impossible to find exactly the starting

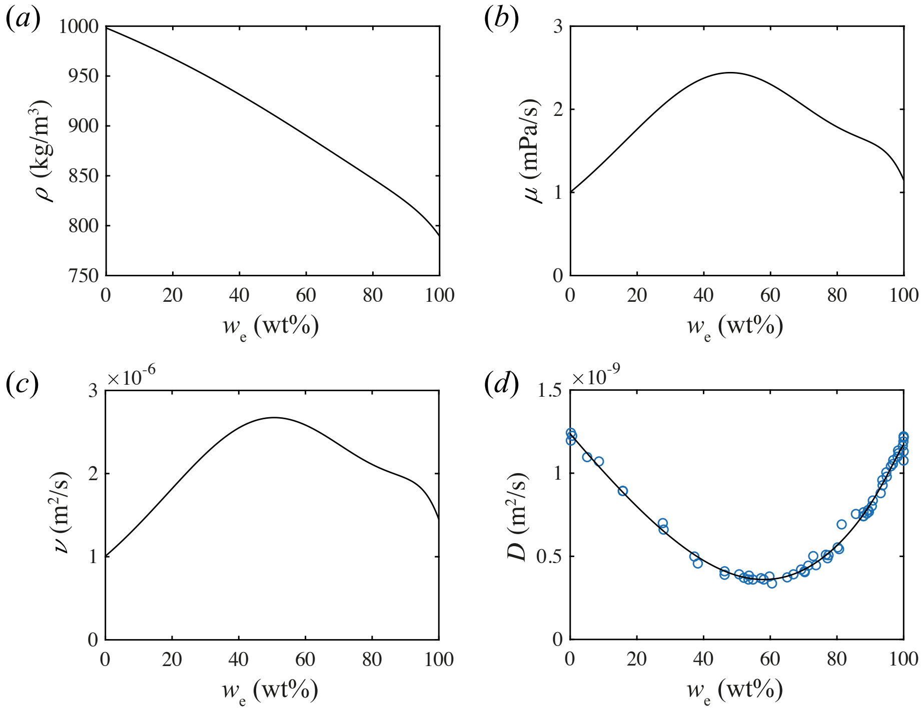

FIGURE 3. Physical properties of the ethanol-water mixture at different ethanol weight fractions $w_{\rm e}$. Density ρ, dynamic viscosity μ and kinematic viscosity ν at 20 °C are shown in (a), (b) and (c), respectively. Data is taken from Khattab *et al.* (2012). Diffusivity of ethanol D is shown in (d). Blue data points are taken from Pařez *et al.* (2013), and the black solid line is a polynomial fit from the data points.

point of the bifurcation. Thus, in the experiments we have used an easily distinguishable criterion to determine if the drop is bouncing: If the bouncing amplitude $h_{\rm A}$ of the drop is larger than its radius R, then the drop is considered to be bouncing.

3. Physical properties of the ethanol-water mixture

Physical properties of the ethanol-water mixture changes with the ethanol weight fraction $w_{\rm e}$. Density ρ and dynamic viscosity μ of the mixture are taken from Khattab *et al.* (2012), and the ethanol diffusivity in the mixture is taken from Pařez *et al.* (2013). ρ, μ, ν and D as functions of ethanol weight fraction $w_{\rm e}$ are shown in figure. 3.

4. Movie captions

Supplementary Movie 1: Two 100 cSt silicon oil drops of different sizes levitating & bouncing in a linearly stratified ethanol-water mixture. The sizes of the drops are $R_1 = 54 \pm 2\,\mu$m and $R_2 = 155 \pm 2\,\mu$m. The gradient of the ethanol weight fraction is $\mathrm{d}w_{\rm e}/\mathrm{d}y = 40\,\mathrm{m}^{-1}$.

Supplementary Movie 2: The flow field around a levitating drop shown by tracer particles. The gradient is $\mathrm{d}w_{\rm e}/\mathrm{d}y = 10\,\mathrm{m}^{-1}$. PSP (Polyamid seeding particles, Dantec Dynamics, Denmark) particles of diameter 20 μm are added in both ethanol and water at 0.4 mg/mL before making the linearly stratified mixture.

Supplementary Movie 3: The flow field around a large bouncing drop shown by tracer particles. The gradient is $\mathrm{d}w_\mathrm{e}/\mathrm{d}y \approx 40\,\mathrm{m}^{-1}$. PSP particles of diameter $20\,\mu\mathrm{m}$ are added in both ethanol and water at $0.4\,\mathrm{mg/mL}$ before making the linearly stratified mixture.

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