Interaction between two drops ascending in a linearly stratified fluid

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Abstract

Three-dimensional numerical simulations of the interaction between two drops rising in a thermally stable stratified fluid are presented. The governing equations are solved by a finite-volume/front-tracking method. The influence of stratification on the dynamics of two drops moving in line and side by side is studied. For the case of nearly spherical drops ascending side by side, drops remain in the horizontal alignment during the interaction; but the rate of separation, after their initial collision, decreases as the stratification becomes stronger. We demonstrate that, in contrast to the case of homogeneous fluid, two drops moving in tandem in a linearly stratified fluid retain their in-line configuration. In addition, the vortical structure behind the drops ascending in a linearly stratified fluid is shown to be the reason for the smaller magnitude of rise velocity of drops compared to that in a homogeneous fluid.

Keywords: Stratified fluid, Drop, Finite volume, Front-tracking method.

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1) Introduction

Even though the motion of drops and bubbles in stratified fluids has several natural and industrial applications such as bubbles rising across pycnoclines in marine and aquatic environments, oil spills, ocean sequestration of CO₂, and bubble mixers used for lake/reservoir destratification and aeration, most studies of stratified flows have been carried out for solid particles. In a step stratification, where a sharp pycnocline separates two layers of homogeneous-density fluid, Srdic-Mitrovic *et al.* [1] observed up to an order of magnitude increase in the drag force of a rigid sphere in the range of $1.5 < Re < 15$, $3 < Fr < 10$, and $Pr = 700$ compared to the homogeneous case, where Re , Fr , and Pr are Reynolds, Froude, and Prandtl numbers, respectively. Torres *et al.* [2] studied the motion of a rigid sphere in a linearly stratified fluid and showed that as stratification becomes weaker, the behavior of the fluid is more similar to the homogenous fluid. The author revealed that an enhanced drag occurs due to the buoyancy of a shell of lighter fluid dragged down by the sphere as it sediments in a heavier fluid. The drag increase at higher Reynolds numbers ($25 \leq Re \leq 100$ and $0.2 \leq Fr \leq 200$, $Pr = 700$) occurs due to the formation of a jet downstream of the particle as isopycnals return to their neutrally buoyant density level [3]. The drag force acting on a horizontal grid of bars moving vertically in a linearly stratified fluid has been considered in experimental investigations by Higginson *et al.* [4]. Their results showed that due to the displacement of fluid elements in a stratified fluid, the drag on the grid is larger than the one in a homogeneous case. Yick *et al.* [5] demonstrated that the Richardson number is more appropriate

parameter rather than the Froude number to describe the motion of a rigid particle at small Reynolds numbers.

Doostmohammadi *et al.* [6] have investigated the motion of a squirmer propelling in a linearly stratified fluid at low Reynolds numbers and showed that life of small organisms can be affected by stratification due to modification in their energy expenditure, detectability, and nutrient uptake.

To the best of our knowledge, little is known about the motion of bubbles and drops in stratified fluids. Young *et al.* [7] experimentally investigated the motion of a bubble in the presence of temperature gradient. They demonstrated that it is possible to keep the bubble stationary by exerting a sufficient negative temperature gradient. Zhang *et al.* [8] obtained the migration velocity of a drop in a vertical temperature gradient by considering the inertial effects at Prandtl numbers of $O(1)$. The motion of a droplet in a sharply stratified fluid with the drop viscosity equal to that of surrounding liquid has been reported by Blanchette and Shapiro [9]. They assumed the density of the drop to be larger than the upper and lower fluids, the Weber number to be small, and the Reynolds number to be of $O(10)$. They performed two cases of simulations; first, they assumed similar surface tension between the drop and two fluid layers, and then they considered increase or decrease in the surface tension during the motion of the drop. They observed that the amount of entrained fluid for the drops is smaller than that of solid particles. Also, they found that the drop accelerates when it falls into a layer of lesser surface tension, and decelerates when settling into a layer of greater surface tension. Bayareh et al. [10] quantified the drag enhancement for a rising drop in a

64 linearly stratified fluid and showed that it scales with $Fr^{2.28}$ in the range of $4 \leq Fr \leq 16$.
 65 They investigated the role of drop deformability on the vortical structure at the lee of
 66 the drop and on the oscillatory motion of the drop around its neutrally buoyant position.
 67 They also demonstrated that the flow structure behind a drop and a rigid sphere is
 68 similar, but the drop is less affected compared to a rigid particle due to the slip condition
 69 on the surface of the drop. Dabiri et al. [11] studied the role of density stratification on
 70 the rising motion of a swarm of crude oil drops in stratified water. They demonstrated
 71 that the probability of cluster formation increases in a stratified fluid compared to a
 72 homogenous one. Here in this paper, we consider hydrodynamic interaction between
 73 two rising drops in a linearly stratified fluid. This is an important problem since it is the
 74 building block of cluster formation in swarms of rising drops in stratified fluids.
 75 Even though the interaction between two bubbles or drops in a stratified fluid has not
 76 been studied, their motion in a homogenous fluid has been explored for a long time.
 77 These investigations have been performed for two bubbles in a vertical alignment ([12],
 78 [13], and [14]), two side-by-side bubbles [15], [16] and for two bubbles with different
 79 initial orientation [17].
 80 The interaction between two spheres rising in tandem in a homogenous fluid is described
 81 by drafting-kissing-tumbling (DKT) process that is commonly considered for the
 82 sedimentation of rigid particles [16]. If two particles are aligned horizontally, the trailing
 83 particle is sucked into the wake of the leading one (drafting). After that two particles
 84 collide with each other (kissing) and then rotate around each other (tumbling). They
 85 eventually settle in a horizontal alignment which is stable configuration. The occurrence

86 of DKT phenomenon for the case of two in-line bubbles depends on the Reynolds
87 number, the deformability, initial distance between the bubbles and properties of
88 ambient fluid. Velez-Cordero *et al.* [14] experimentally and numerically investigated
89 the drafting-kissing-tumbling (DKT) process in shear thinning and Newtonian fluids.
90 They found that two bubbles rising in a Newtonian fluid experience DKT process at low
91 Reynolds number ($Re = 1.3$) but the tumbling phase does not occur in shear-thinning
92 fluids. On the other hand, while two deformable bubbles show DKT phenomenon at
93 Reynolds number around $Re = 23$, two spherical bubbles tend to approach and rotate
94 around each other before they have a collision [15]. Katz and Meneveau [19]
95 experimentally investigated the binary interaction of nearly spherical air bubbles in pure
96 water and observed that two bubbles tend to collide and coalesce at $0.2 \leq Re \leq 35$. Two
97 non-deformable drops reach an equilibrium distance due to the balance between wake
98 suction and pressure gradient. In the case of bubbles, they demonstrated that the
99 coalescence of bubbles is due to the stronger effect of wake suction compared to the
100 pressure gradient in the gap between the bubbles.

101 Zhang and Fan [20] simulated the motion of in-line rising bubbles in a Newtonian fluid
102 at Reynolds number of $O(100)$ and found that the rise velocity of spherical bubbles is a
103 function of the distance between the bubbles. In contrast to Zang and Fan who assumed
104 laminar wake during their simulations, Yuan and Prospreti [21] considered the vortex
105 dynamics behind the in-tandem bubbles in a viscous fluid and showed that there is an
106 equilibrium position for the distance between the bubbles depending on their Reynolds
107 number. The same results have been reported by Legendre *et al.* [16] for the interaction

108 between two rising bubbles for the side by side configuration in a viscous fluid. They
109 showed that depending on the bubbles initial distance, there may exist an equilibrium
110 distance or they may move away from each other. Hallez and Legendre [17] extended
111 the obtained results of Yuan and Prosperetti [21] and Legendre *et al.* [16] by studying
112 several initial distances and initial orientation angles θ ($\theta = 0$ for side-by-side bubbles
113 and $\theta = 90$ for in-line bubbles). It is known that the bubbles are attracted for $0 < \theta < \theta_a$
114 and repelled for $\theta_a < \theta < 90$ (θ_a is a critical angle [20]). Hallez and Legendre [17]
115 characterized the interaction between two bubbles for three critical angles that are
116 affected by the Reynolds number and the distance. They reported that the force acting
117 on the drops rising in tandem leads to the side-by-side configuration, regardless of the
118 distance and the Reynolds number.

119 Several investigators studied the unsteady motion of two spherical bubbles in potential
120 flow ([22, 23, and 24]). They demonstrated that two bubbles attract each other when
121 $55 < \theta < 128$ and they repel each other when $\theta < 55$ or $\theta > 128$. We can conclude that side-
122 by-side configuration is a stable configuration.

123 The dynamic behavior of more than two rising bubbles for the in-line configuration
124 (known as the bubble chain) has been investigated by several researchers ([19, 25, 26,
125 and 27]). For example, Ruzicka [26] studied the transition dynamics of two, three, and
126 more ascending bubbles. His results demonstrated that the rise velocity of a pair of
127 bubbles rising in line is larger than that of a single bubble. Also, he showed that the
128 fixed-end chain (with two boundary bubbles) has greater tendency to form large bubble
129 clusters compared to the free-end chain (no boundary bubbles).

130 The purpose of the present article is to study the interaction between two non-neutrally
 131 buoyant rising drops in a linearly stratified fluid. We study the role of stratification on
 132 the dynamics of motion of two rising drops for in-line and side-by-side configurations.
 133 We consider the formation of the vortical structure behind the drops and explain how
 134 the rear vortex alters the motion of drops. We additionally discuss the drafting, kissing,
 135 and tumbling phenomenon for two drops ascending in tandem in a stratified fluid.

136

137 **2) Formulation and Numerical Implementation**

138 The fully resolved three-dimensional motion of a pair of drops in a linearly stratified
 139 fluid is simulated. For drops smaller than the vertical extent of the stratified region, a
 140 linearly stratified fluid is a more appropriate model than a sharp stratification. Thus, the
 141 density of background fluid can be described as $\rho_b = \rho_0 - \gamma z$, where ρ_0 is the reference
 142 density of the fluid, z is the vertical position and γ the background density gradient.
 143 Since the fluid inside and outside the drop is incompressible and Newtonian, the
 144 governing equations under the Boussinesq approximation can be written as

145

$$\nabla \cdot \mathbf{u} = 0 \quad (1)$$

146

$$\rho_0 \frac{D\mathbf{u}}{Dt} = -\nabla p + (\rho - \check{\rho})\mathbf{g} + \nabla \cdot [\mu(\nabla\mathbf{u} + \mathbf{u}^T)] + \int \sigma k' \mathbf{n}' \delta^{\beta'}(\mathbf{X} - \mathbf{X}') dA' \quad (2)$$

147

$$\frac{DT}{Dt} = \kappa \nabla^2 T \quad (3)$$

$$\rho = \rho_0(1 - \beta T_0) \quad (4)$$

where $\mathbf{u}$ is the velocity vector, p the pressure, T the temperature, $\mathbf{g}$ the acceleration of gravity, σ the surface tension coefficient, and β is the thermal expansion coefficient. ρ , μ , and κ are the local density, viscosity, and thermal diffusivity of the fluid. $\bar{\rho} = \frac{1}{V} \int_V \rho dV$ is the mean density over the entire computational domain V . k' is twice the mean curvature of the interface of the drop, $\mathbf{n}'$ is the unit vector normal to the interface, dA' is the surface element at the interface of the drop, $\delta^{\beta'}$ is the three-dimensional delta Dirac function which is discontinuous at $\mathbf{X}'$, a point at the interface. A uniformly distributed force of $-\bar{\rho}\mathbf{g}$ is added to the entire computational domain to avoid the downward acceleration of the entire domain by gravity. Therefore, the net momentum flux of the computational domain across outer boundaries $\partial\Omega$ is zero ($\int_{\partial\Omega} \rho \mathbf{u} dA = 0$). This term compensates for the hydrostatic head. The last term in equation (2) represents interfacial forces between the continuous and dispersed phases and it is evaluated at point $\mathbf{X}$. The Stokes theorem is used to calculate the force on a surface element [28]:

$$\mathbf{F}_\sigma = \sigma \int_{\delta A} k' \mathbf{n}' dA = \sigma \int_{\delta A} (\mathbf{n}' \times \nabla) \times \mathbf{n}' dA = \sigma \oint_s \mathbf{t} \times \mathbf{n}' ds \quad (5)$$

164

165 where $\mathbf{t}$ is a tangential vector to the edge of the element. Equations (3) and (4) in the
 166 continuous phase can be combined as

167

$$\frac{\partial \rho}{\partial t} + \mathbf{u} \cdot \nabla \rho = \kappa \nabla^2 \rho \quad (6)$$

168

169 The density difference across the computational domain along z direction is γL , where
 170 L is the length of computational domain in vertical direction. The equation governing a
 171 salt-stratified fluid is identical to equation (5). In that case, κ describes diffusivity of salt
 172 rather than diffusivity of temperature. A color function, α , is defined to be zero inside
 173 and unity outside the drop and is calculated using the Heaviside function [28]:

174

$$\nabla \alpha = - \int_A \delta(x - x')(y - y')(z - z') \mathbf{n} dA' \quad (7)$$

175

176 Therefore, fluid properties within the entire domain, Ω , can be written as $\rho = \alpha \rho_f +$
 177 $(1 - \alpha) \rho_d$ and $\mu = \alpha \mu_f + (1 - \alpha) \mu_d$. Subscript f refers to continuous phase and d to
 178 the dispersed phase.

179 Various methods have been used to simulate multiphase flows, such as Volume-of-Fluid
180 (VOF), level-set, and front-tracking methods. Here, a front-tracking technique [13] is
181 implemented to track the interface of different phases. A computational code is
182 developed based on the finite-volume method and projection scheme to solve
183 incompressible Navier-Stokes equations. The details of the numerical method have been
184 presented and fully verified in [29].

185 Equations (1), (2), and (3) are solved over the entire domain utilizing a finite volume
186 method that employs a fixed nonuniform staggered grid by means of a conventional
187 operator splitting method to enforce the continuity equation. The time discretization is
188 obtained using a second-order TVD (total variation diminishing) Runge-Kutta method
189 [30]. The spatial derivatives in the convection term are evaluated by means of the
190 QUICK (Quadratic Upstream Interpolation for Convective Kinetics) scheme, leading to
191 third order accuracy [31]. The code is validated against the published numerical and
192 experimental results in our previous publications ([10], [11]). We also performed a grid
193 resolution study to verify that the grid resolution used in our simulations is sufficient to
194 capture the dynamics of the motion of drops in linear stratification. We determine the
195 accuracy of the code by using different number of points across the drop diameter. It is
196 found that $256 \times 256 \times 480$ grid that provides 20 points per diameter is sufficient to
197 resolve the flow. Increasing the grid resolution to 40 points across the drop diameter
198 will result in 1.7% change in the rise velocity and 2.8% change in the deformation of
199 the drop. The grid spacing for the two cases of 20 points and 40 points across the drop

200 diameter is $0.05d$ and $0.025d$, respectively, where d is the drop diameter. The thickness
 201 of density and momentum boundary layers can be estimated as

202

$$\delta_m \sim O\left(\frac{d}{\sqrt{Re}}\right) \quad (8)$$

203

204 and

205

$$\delta_d \sim O\left(\frac{d}{\sqrt{Re \, Pr}}\right) \quad (9)$$

206

207 Respectively [32]. These values for the drop with Reynolds number $Re_d \approx 200$ based on
 208 the drop rising velocity correspond to $\delta_m \sim 0.0707d$ and $\delta_d \sim 0.0267d$. As shown in
 209 detail in our earlier publication [33], this grid resolution can capture the flow field near
 210 the drop as well as its trajectory [33]. The motion of a drop in a stratified fluid can be
 211 characterized by the following dimensionless parameters. The Reynolds number, $Re =$
 212 $\rho_f w d / \mu_f$, the Froude number, $Fr = w / (N d)$, where w is drop characteristic velocity,
 213 $N = \sqrt{-(g/\rho_f) d \rho / dz}$ indicates the buoyancy frequency, the Eötvös number,
 214 $Eo = (\rho_f - \rho_d) g d^2 / \sigma$, the Prandtl number, $Pr = \nu_f / \kappa$, and the density and the
 215 viscosity ratios, $\eta = \rho_d / \rho_f$ and $\lambda = \mu_d / \mu_f$. The Prandtl number expresses the ratio of
 216 the diffusivity of momentum, ν_f , and the diffusivity of the stratifying agent, κ . For
 217 example, $Pr = 7$ for temperature-stratified water and $Pr = 700$ for salt-stratified water.

218 Here, the Prantdtl number and the Reynolds number are set to be 7 and 790, respectively.
 219 Since the terminal velocity of the drop is not known a priori, $w_s = \sqrt{gd}$ is used to scale
 220 the velocity. We also use $l_s = d$ and $t_s = \sqrt{d/g}$ to normalize length and time,
 221 respectively. Also, the ratio of thermal diffusivity of drop to the thermal diffusivity of
 222 ambient fluid is set to be unity in this paper. The realistic properties of nearly spherical
 223 crude oil drops in the ocean are used as our base data: $\rho = 890\text{kg/s}$, $\mu = 5 \times 10^{-3}\text{Pa.s}$
 224 and $\sigma = 27\text{mN/m}$. Also, we have used the properties of water as an ambient fluid: $\rho =$
 225 1000kg/s , $\mu = 10^{-3}\text{Pa.s}$. These values lead to the following dimensionless parameters:
 226 $Eo = 0.639$, $\lambda = 5$, $\eta = 0.89$, and $Re = 790$. The range of Froude number used in this
 227 work is $10 \leq Fr \leq 22$, which corresponded to $0.51 \leq N \leq 1.1$.

228

229 **3) Results and discussion**

230 First, we explain the interaction between two drops rising in a side-by-side configuration
 231 and discuss the role of stratification on the deformation, rise velocity, and distance. After
 232 that, we consider the DKT mechanism for the in-line initial configuration of two rising
 233 drops.

234

235 **3.1. Side-by-side configuration**

236 A pair of spherical bubbles rising side by side in a potential flow of a homogeneous
 237 fluid is attracted toward each other due to the flow acceleration in the gap between the
 238 bubbles. Legendre et al. [16] studied the motion of a pair of spherical bubbles at a finite

Reynolds number regime and found that at high Reynolds numbers ($Re = 300$) the
 irrotational mechanism is still responsible for the attraction force between the bubbles.
 In the low Reynolds number regime, however, the diffusion of vorticity in the gap
 between the bubbles is blocked by the other bubble, resulting in a lower velocity in the
 gap and repulsive force acting on the bubbles. Here, we investigate the role of
 stratification on the interaction between two deformable drops rising side by side over
 a range of Froude numbers, and for the dimensionless distance of $d^* = 1.25$, where d^*
 $= l/l_s$ and l is the dimensional value of initial distance between the drops. We have
 performed three-dimensional simulation of hydrodynamic interaction of a pair of drops
 rising in a linearly stratified fluid. The computational domain is a rectangular channel
 with periodic boundary conditions in all dimensions. The drops release from rest in a
 stagnant flow and the density of the background fluid is initially linearly changing with
 depth. The schematic of the problem is shown in Fig. 1. The role of drop deformability,
 Eötvös number, on the interaction of drops in a linearly stratified fluid is also discussed.

253

254 **3.1.1. Nearly spherical drops**

The details of the rising dynamics of two side-by-side drops as a function of
 dimensionless time, $t^* = t/t_s$ are plotted in Fig. 2 for homogenous and stratified fluids.
 As illustrated in Fig. 2a, the drops initially accelerate and deform due to the non-uniform
 pressure field. The drops are initially attracted towards each other since the Reynolds
 number based on the drops' rising velocity is on the order of $Re_d \sim 200$ (see Fig. 2b).

260 After the drops collide, they rebound with a superficial super-elasticity. As illustrated
 261 in Fig. 2c, the lateral velocity of the drops after collision is much higher than the pre-
 262 collision velocity. This superficial super-elasticity was also observed experimentally for
 263 bubbles rising in a homogenous fluid by Sanda et al. [34]. Their experimental results
 264 show a reduction in the rising velocity of the bubbles during the collision. However, in
 265 the present numerical results, the drops are released from the rest and are still
 266 accelerating when the collision occurs. Even though, no deceleration is observed here,
 267 the acceleration rate reduces during the collision. The acceleration of the drops in a
 268 stratified fluid is very close to a homogenous fluid up to $t^* = 6$. At this time the
 269 stratification effect reduces the acceleration of the drops and, for the case of the stronger
 270 stratification ($Fr = 11$), the drops start to decelerate. The bouncing motion of the drops
 271 in a stratified fluid is also suppressed (see Fig. 2c).

272 The comparison of the motion of the drops in homogeneous and stratified fluids
 273 demonstrates that the deformation of drops is smaller in a stratified fluid. To quantify
 274 the drop deformation, we calculate a scalar measure of the deformation that is defined
 275 as $\chi = \sqrt{I_1/I_2}$, where I_1 and I_2 are the largest and smallest eigenvalues of the second
 276 moment of inertia tensor, respectively, and $\chi = 1$ corresponds to a spherical drop. The
 277 second moment of inertia tensor is given by [13]:

$$I_{ij} = \int (x_i - x_{i_o})(x_j - x_{j_o})dV \quad (10)$$

279

280 where x_{i_o} and x_{j_o} are the coordinates of the drop center and V is the volume of the drop.
 281 The deformation of drops versus dimensionless time is plotted in Fig. 2d. In a stratified
 282 fluid, buoyancy effects tend to restore the deflected constant density contours
 283 (isopycnals) dragged by the drops, which leads to a reduction in the rise velocity of the
 284 drops. As a result, the deformation of drops is smaller due to a lower instantaneous
 285 Weber number. Fig. 2d shows that the deformation increases as the drops approach each
 286 other and reaches a maximum when the drops arrive at their minimum distance. At this
 287 position, the blocking effect of the vorticity hinders the lateral motion of the drops, and
 288 a reduction in the deformation of the drops is observed. As drops move away from each
 289 other, their deformation increases again and reaches a quasi-steady-state. The variation
 290 of normalized lateral velocity of the left drop, shown in Fig. 2c, confirms that the sign
 291 of the lateral velocity changes when the first deformation peak occurs. It should be noted
 292 that these simulations are performed at a constant Eötvös number, $Eu = 0.639$.
 293 As the Froude number decreases ($Fr \simeq 11$), the buoyancy force more effectively restores
 294 the perturbed isopycnals to their neutrally buoyant levels and hampers the upward
 295 motion of the drops. In addition to buoyancy force, the generation of rear vortical
 296 structure induces a significant decrease in the rise velocity of drops. The mechanism of
 297 the formation of the vorticity behind the drop can be explained as follows: when a drop
 298 rises in a linearly stratified fluid, it perturbs the density layers. The deflection of
 299 isopycnals leads to the formation of a secondary vortex behind the drop. This can be
 300 explained by examining the vorticity equation in the outer fluid under the Boussinesq
 301 approximation:

302

$$\frac{D\boldsymbol{\omega}}{Dt} = (\boldsymbol{\omega} \cdot \nabla)\mathbf{u} + \nu \nabla^2 \boldsymbol{\omega} - \frac{g}{\rho_o} \nabla \rho_f \times \hat{\mathbf{k}}, \quad (11)$$

303

304 where $\boldsymbol{\omega}$ is the vorticity vector and $\hat{\mathbf{k}}$ is the unit vector along the z axis. The term $\nabla \rho_f \times$
 305 $\hat{\mathbf{k}}$ is nonzero behind the drop and is responsible for the formation of the secondary
 306 vortex. Fig. 3 shows the deflection of isopycnals along with corresponding
 307 dimensionless vorticity contours, $\omega^* = \omega t_s$. The asymmetry of the vorticity distribution
 308 behind each individual drop observed in Fig. 3 is due to the small distance between the
 309 drops. A buoyant jet is generated behind each drop and its structure is best illustrated
 310 by the velocity field generated by the rising motion of the drops (see Fig. 4). The two
 311 rear buoyant jets merge into a single strong jet behind the drops. The contribution of
 312 two buoyant jets provides a higher resistance to the ascent of a pair of drops compared
 313 to a single drop. As drops further separate from each other after the collision, the single
 314 jet branches into two jets behind each drop.

315 The spatial distribution of nearly spherical drops shows that the side-by-side
 316 configuration is a stable configuration similar to the homogeneous case; however, the
 317 rate of lateral separation decreases in the presence of stratification due to a weaker
 318 repulsion force. We can conclude that, as the Froude number decreases, the drop
 319 agitation becomes smaller and thus, the probability of the formation of horizontal drop
 320 clusters enhances.

321

3.1.2. Deformable drops

To examine the effect of the drop deformability on the behavior of two drops rising side by side, we have performed two simulations at Eötvös numbers, $Eo = 0.639$ and 3. The lateral distance and the deformation as a function of dimensionless time are plotted in Fig. 5a-b. In the presence of stratification, the deformability significantly alters the motion of drops. As shown in Fig. 5a, the minimum magnitude of distance between deformable drops is larger than that between two spherical ones. This is due to the fact that deformable drops are oblate and thus have a larger center to center distance at a time of contact. Fig. 5a shows that the drop velocity after collision is larger for a spherical drop compared to a deformable one. Fig. 5b shows drop deformation for two different Eötvös numbers. The drop with a larger Eötvös number has significantly larger deformation.

3.2. In-line configuration

We now examine the effect of stratification on the interaction between a pair of drops rising in tandem. The distance between the center of drops is initially set to $d^* = 1.25$ (Fig. 6). Bunner and Tryggvason [13] studied the role of bubble deformation on their motion in a homogeneous fluid and reported that a spherical bubble tends to move away from the wake, but a deformable bubble moves into the wake. They showed that two deformable in-line bubbles accelerate, approach, collide, and then rotate around each other and DKT phenomenon occurs, while two spherical bubbles tend to tumble without a collision ($Eo = 1$ and $Re \simeq 25$). Even though the second stage of DKT process does

not occur at this Reynolds number, it has been experimentally observed for two small spherical bubbles at low Reynolds numbers, $Eo = 0.9$ and $Re = 1.3$ ([14]).

3.2.1. Nearly spherical drops

Similar to the case of two nearly spherical drops ascending side-by-side, the stratification plays a crucial role in determining the dynamics of the interaction between two drops rising in tandem. The horizontal distance, r^* , and vertical distance, z^* , between the drops are plotted in Fig. 7a-b as a function of dimensionless time for different values of Froude number. A pair of drops rising in a stratified fluid at $Fr = 22$ exhibits the same dynamics as those in a homogeneous fluid, but the stratification considerably suppresses the lateral movement of drops and hinders their vertical motion. Drops tend to tumble and rotate around each other in both cases; however, the rate of lateral separation decreases in the presence of stratification.

As shown in Fig. 8, two spherical drops will retain an in-line configuration when ascending in a stronger stratified fluid. This Figure illustrates a snapshot of the motion of drops for different Froude numbers at dimensionless time, $t^* = 30$. Even though in a homogeneous fluid and a weakly stratified fluid, the local velocity gradient in the wake of the leading drop pushes the trailing drop away from the wake, this does not occur in a more strongly stratified fluid. In other words, the in-line configuration is more stable as the Froude number decreases. The pair of drops drags heavy fluid layers, producing a strong buoyancy force. As a consequence, two spherical drops tend to keep their vertical alignment before collision. The strength of stratification determines how drops

collide. The vertical distance decreases abruptly at very low Froude numbers due to the high rate of restoration of isopycnals to their neutrally buoyant levels. When the leading drop reaches its neutrally buoyant density level, the trailing drop approaches and collides with the leading one. The instantaneous average rise velocity of a pair of drops, ω_{av} , is plotted in Fig. 9 for two different initial alignments of drops at $Fr = 22$. Illustrated here is the tendency for two in-line drops to rise faster than two side-by-side drops in the presence of stratification similar to a homogenous case.

373

3.2.2. *Deformable drops*

Here, we consider the effect of deformation on the motion of two inline drops rising in a homogeneous fluid. Our simulations confirm that the DKT phenomenon is affected by the deformability. In contrast to nearly spherical drops, three stages of DKT process are observed for the case of deformable drops. Two nearly spherical drops tend to rotate around each other (tumble) without a collision (Fig. 10). The distance of the drops in a stratified fluid is illustrated in Fig. 11 for $Eo = 0.639$ and 3. Initially two drops approach each other while they keep their in-line configuration ($t^* < 20$). After this, two deformable drops collide with each other and rebound due to a superficial elasticity. During this interaction, the spherical shape of drops is approximately recovered (see Fig. 12) and the drops tend to increase their vertical separation ($t^* \simeq 40$). When the leading drop reaches the neutrally buoyant level, it starts to oscillate around it due to the competition between buoyancy and inertial forces. The trailing drop reaches, kisses, separates, and eventually rotates around the leading drop. The motion of two spherical

388 drops is the same as two deformable drops for $t^* > 40$. Fig. 13 shows the snapshots of
389 isopycnals and normalized velocity contours for deformable drops.

390 Overall, we expect that a dilute suspension of drops rising in tandem does not produce
391 horizontal clusters at low-to-moderate Froude numbers. A horizontal cluster of nearly
392 spherical or deformable drops can be formed where the drops approach their neutrally
393 buoyant density levels.

394

395 **4. Conclusions**

396 The dynamics of the interaction between two drops rising in a linearly stratified fluid is
397 investigated using a finite-volume/front-tracking method. In the flow regime studied in
398 this paper, the dynamical behavior of two in-line and side-by-side drops is significantly
399 affected by stratification. Stratification reduces drop deformability as well as the vertical
400 and lateral motion of two side-by-side drops. In contrast to a homogeneous fluid, two
401 in-line drops retain their in-line configuration at low-to-moderate Froude numbers.
402 After the drops approach their neutrally buoyant density level, they oscillate, and
403 eventually the trailing drop rotates around the leading one.

404

405 **Acknowledgment**

406 This work is supported by NSF grants CBET-1066545 and CBET-1414581.

407

408

409

410 **Nomenclature**

d	Drop diameter
d^*	Dimensionless distance between the drops
dA'	Surface element at the surface
Eo	Eötvös number
Fr	Froude number
F_σ	Force on a surface element
g	Acceleration due to gravity
I_1	Largest eigenvalues of the second moment of inertia tensor
I_2	Smallest eigenvalues of the second moment of inertia tensor
k'	Twice the mean curvature
$\hat{\mathbf{k}}$	Unit vector along the z axis
l	Dimensional value of initial distance between the drops
l_s	Length scale
L	Length of computational domain
n'	Unit vector normal to the interface
N	Buoyancy frequency
p	Pressure
Pr	Prandtl number
Re	Reynolds number
Re_d	Reynolds number based on drop rising velocity

S	Edge of element
t	Time
$\mathbf{t}$	Tangential vector to the edge of element
t^*	Dimensionless time
t_s	Time scale
$\mathbf{u}$	Velocity vector
V	Computational domain
w	Drop characteristic velocity
w_s	Velocity scale
x_{i_o}	Coordinate of the drop center
x_{j_o}	Coordinate of the drop center
$\mathbf{X}$	Eulerian coordinate vector
$\mathbf{X}'$	Lagrangian coordinate vector
z	Vertical position

411

412 Greek letters

α	Color function
β	Thermal expansion coefficient
γ	The background density gradient
$\delta^{\beta'}$	Three-dimensional delta Dirac function
η	Density ratio
θ	Orientation angle
θ_s	Critical angle

κ	Diffusivity of the fluid
λ	Viscosity ratio
μ	Kinematic viscosity
μ_d	Viscosity of dispersed phase
μ_f	Viscosity of continues phase
ν_f	Diffusivity of momentum
ρ	Density
ρ_o	Reference density
ρ_b	Density of background fluid
$\hat{\rho}$	Mean density
σ	Surface tension coefficient
χ	Drop deformation
ω	Vorticity
$\omega *$	Dimensionless vorticity

413

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Figures

Figure 1: Schematic of two drops rising side by side in a linearly stratified fluid. The density profile is not to scale.

Figure 2: (a) The rise velocity, $w^* = w/W_S$, (b) the dimensionless distance between the center of drops, (c) the lateral velocity, $v^* = v/W_S$, and (d) the drop deformation for two drops released side by side as a function of dimensionless time for different values of Froude numbers at $Eu = 0.639$ and $Re = 790$.

Figure 3: The snapshots of the deflection of isopycnals and dimensionless vorticity contours are shown for $Fr = 11$, $Re = 790$ and $Eo = 0.639$.

Figure 4: The snapshots of the deformation of isopycnals and normalized velocity contours are shown for $Fr = 11$, $Re = 790$ and $Eo = 0.639$.

Figure 5: (a) The lateral distance and (b) the drop deformation versus dimensionless time for two drops rising side by side at $Fr = 11$ and $Re = 790$.

Figure 6: Schematic of two drops rising in tandem in a linearly stratified fluid.

Figure 7: (a) The horizontal and (b) vertical distances between two drops rising in tandem for different values of Fr at $Eo = 0.639$ and $Re = 790$.

Figure 8: The snapshots of the deflection of isopycnals and dimensionless velocity contours of drops rising in tandem are shown for different values of Froude number at $t^* = 30$, $Re = 790$ and $Eo = 0.639$. (a) Homogeneous, (b) $Fr = 22$, (c) $Fr = 11$.

Figure 9: Instantaneous average rise velocity of drops as a function of dimensionless time at $Fr = 22$, $Re = 790$ and $EO = 0.639$. Solid line: stratified fluid, dashed line: homogeneous fluid.

Figure 10: (a) The horizontal and (b) vertical distances between two drops rising in a homogeneous fluid versus dimensionless time for two Eötvös numbers at $Fr = 11$ and $Re = 790$.

Figure 11: (a) The horizontal and (b) vertical distances between two drops rising in tandem for different values of Eo at $Fr = 11$ and $Re = 790$.

Figure 12: Drop deformation as a function of dimensionless time for two Eötvös numbers at $Fr = 11$ and $Re = 790$. Solid line: leading drop, dashed line: trailing drop.

Figure 13: The snapshots of the deflection of isopycnals and dimensionless vorticity contours are shown for $Fr = 11$, $Re = 790$ and $Eo = 3$.