

# Dynamics of droplet coalescence on a vibrating vertical surface

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| 1 | Dynamics of droplet coalescence on a vibrating vertical surface   |
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| 9 |   |

#### 10 Abstract

A mass-spring-damper theoretical model with a phenomenological equation is 11 12 established to clarify the underlying physics of the dynamics of droplets on a vertical 13 surface driven by vibration. It is found experimentally and theoretically that the phase 14 shift between droplet and plate appears and peaks at a lower frequency for a larger droplet. At a certain frequency, two droplets could move in the opposite direction. 15 16 Based on the phase shift mechanism, we propose a strategy aimed at promoting droplet coalescence. Compared to the necessity of precise control of frequency for resonance-17 18 induced events, the strategy accepts a higher tolerance for frequency, at which oppositemotion-induced droplet coalescence could occur. The optimal frequency where there is 19 20 a maximum phase shift between two droplets is derived and a large-bandwidth 21 frequency range, which allows at least 90% maximum phase shift, is defined. The good 22 agreement between the experimental and theoretical results collectively shows that the motion of the larger droplet is in the opposite direction to that of the smaller one only 23 24 at large-bandwidth frequency range and the two droplets coalesce with high enough amplitude. Our findings are helpful for the utilization of vibrating surfaces for droplet 25 26 removal.

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#### 28 I. Introduction

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Vapor condensation is critical and common in urban environmental control,<sup>1</sup> power generation,<sup>2</sup> food processing,<sup>3</sup> as well as water recovery<sup>4-6</sup> and treatment.<sup>7</sup> Rising energy costs, environmental impact and carbon emissions have aroused persistent concerns over energy conservation in condensation processes. Considering the important role that vapor condensing plays in industrial processes, a higherperformance design of vapor condensation would be positive from both an economic and environmental perspective.

Dropwise condensation (DWC) and filmwise condensation (FWC) are two common 36 37 modes of vapor condensation. In DWC, discrete droplets nucleate, grow, and then coalesce on a cool surface. The extra surface that is not covered by droplets continues 38 to achieve condensation. In FWC, the whole surface is covered by a continuous 39 condensate film, which causes a rapid increase in thermal resistance. They could suffer 40 41 from ten-fold lower heat transfer coefficients (HTC) than in DWC.<sup>8</sup> Moreover, reducing 42 the area of the drop-covered surface, as soon as nucleation begins, is the key to 43 achieving higher overall condensation HTC. For DWC on a vertical surface, when the diameter of a droplet exceeds the capillary length, it can slide off by gravity. This 44 45 entrains other droplets during the sliding and refreshes extra condensation surface exposed directly to vapor. Consequently, the key to enhancing DWC is to accelerate 46 47 drop removal, which is harder on randomly oriented surfaces.9

Superhydrophobic surfaces have been developed to promote sliding, detachment and 48 combination of small droplets.<sup>10, 11</sup> When two neighboring drops coalesce, a 49 spontaneous jumping effect due to surface energy release is stimulated.<sup>12</sup> This is 50 reported to enhance condensation by an order of magnitude in HTC as compared to 51 DWC on an ordinary surface.<sup>13</sup> Various micro/nanostructured superhydrophobic 52 53 surfaces can be obtained by chemical (etching, oxidation) or mechanical techniques (additive manufacturing).<sup>14</sup> However, the durability of these surfaces remains an issue, 54 55 as the structures and coatings tend to degrade rapidly under high temperature and 56 humidity conditions as well as scale deposits. Moreover, from the point of view of 57 condensate nucleation, hydrophobic surfaces require a higher degree of supersaturation 58 than ordinary surfaces.<sup>15</sup> Increase in the overall performance is thus compromised, as a

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lower condensation source temperature is required. Therefore, active removal ofdroplets from ordinary surfaces is highly desirable.

Vibration of the condensation surface is an effective active technique which could 61 overcome the above challenges. Droplet deformation and displacement driven by 62 vibration are key mechanisms for promoting surface detachment. Depending on the 63 direction, frequency and amplitude of vibration, droplets behave differently. On a 64 horizontal surface vibrating along its normal direction, a backward motion of droplet is 65 observed when vibration frequency is smaller than a quarter of the eigen frequency or 66 67 within a narrow domain around 1.5 times eigen frequency.<sup>16</sup> On a vertical surface vibrating along its longitudinal axis, if the vibration frequency is equal to the resonance 68 frequency of the droplet, a maximal deformation can be obtained. In addition, if the 69 amplitude of vibration is high, the deformation can overcome the contact angle 70 hysteresis and the contact line starts to move.<sup>17, 18</sup> The higher the contact angle, the 71 72 greater this effect. Vibration in the transversal direction of the condensation surface also 73 enhances detachment from superhydrophobic surfaces. As shown in Fig. 1(b) and (c), vertically and horizontally oriented substrates with superhydrophobic treatment are 74 investigated respectively by Moradi et al.<sup>19</sup> and Sun et al.<sup>20</sup> They achieved droplet 75 detachment by high-amplitude vibration at the resonance frequency. Statistical results 76 77 show a reduction in the average size of the departing droplet and a significant increase (by 8.6) in droplet sliding speed compared to non-vibrated ones.<sup>21</sup> Consequences on the 78 79 condensation heat transfer enhancement are noteworthy: heat transfer rate improves by more than 70% over the stationary case when longitudinal vibrations at 100 and 200 Hz 80 are applied to a vertical hydrophobic surface with contact angle  $\theta = 114 \pm 4^{\circ}$  (Fig. 81 1(a)).<sup>22</sup> A further strategy for promoting the detachment of droplets of various sizes can 82 be adopted with cyclic variable frequency vibration. Shedding events on a 83 superhydrophobic surface increase by 120% with this strategy.<sup>19</sup> The positive effect of 84 vibration on condensation has also been reported elsewhere.23,24 A common feature of 85 the above reports is they are limited to a specific frequency strongly related to the drop 86 87 size. This is due to the low damping effect from reduced adhesion force between a 88 hydrophobic surface and droplets. Careful adjustment of frequency is thus necessary,

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which also requires a complex power supply and signal processing system. Moreover,
to the best of our knowledge, no literature studies the merging effect of two neighboring

91 droplets driven by vibration.

Fig. 1 summarizes how our study is distinguished from the previous ones. The present 92 study aims to experimentally and theoretically investigate the dynamics of droplet 93 vibration on a vertical surface, with particular focus on the phase shift between two 94 neighboring droplets at large-bandwidth frequency. Based on the theoretical model, we 95 96 propose an operating strategy to enhance the coalescence of droplets of different sizes 97 to form larger droplets which are more likely to fall. This approach is experimentally validated with a device designed to explore a large frequency spectrum. Advantages of 98 our study include: ordinary surface without hydrophobic treatment, large bandwidth 99 vibration instead of only the resonance frequency. 100



102 103

#### 104 II. Materials and methods

105 A. Experimental set-up

106 The experimental set-up is shown schematically in FIG. 2(a). A sinusoidal signal is

107 generated by a signal generator (CENTRAD GF 266) and then amplified by TIRA

108 Analog Amplifier BAA 120. A vibrator (TIRA TV-51110) is fed by the amplified signal

109 to vibrate a plate along its vertical axis. The copper plate studied has a contact angle of

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about 96° (FIG. 2(b)), as measured by a contact angle system (KRÜSS DSA25B). For
visualization, a high-speed camera (FASTCAM SA3) with a macro lens (Sigma
MACRO 105mm F2.8 EX DG OS HSM) records the vibration of droplets on the plate
at 10 000 fps (frames per second). The whole system (plate, vibrator, high-speed camera)
is placed into a humidity-regulated hermetic chamber. A highly humid environment (98%
RH) helps avoid the evaporation of droplets. The droplets on the plate are prepared by
a sprayer and their sizes depend roughly on the number of spraying.



FIG. 2. (a) Schematic representation of the experimental setup. (b) Water-plate contact angle.

119 B. Image processing procedure

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Image processing is performed using ImageJ<sup>25</sup> and MATLAB<sup>26</sup>. Fig. 2(a) shows the 120 121 main procedure from raw image to drop morphology analysis. First, sequential images 122 from the high-speed camera are converted to binary ones after sharpening, contrast 123 conditioning, and threshold adjustment. This allows to capture the contours of the 124 droplets. Then two successive images, where the target droplets are in their highest and 125 lowest positions, are stacked together. After, we get a time-averaged elliptical contour of the target droplet. Finally, the stacked image containing only the target shape is 126 127 processed with MATLAB to get the dimensions of the major  $(l_{d1})$  and minor axes  $(l_{d2})$ . Similarly, the stacking method is used to obtain the length of a fixed point  $(l_p)$  on the 128 129 plate, which gives the vibration amplitude. We define a dimensionless parameter AR130 (Amplification Ratio) to evaluate the displacement of the center of gravity of the droplet 131 with respect to the plate:

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132 
$$AR_e = \frac{l_{d1} - l_{d2}}{l_p}$$
 (1)

133 Depending on the frequency, there is a phase shift  $\varphi_e \in [0, \pi]$  between the plate and a 134 droplet. It is calculated from the difference between the two frames where the plate and 135 the target droplet respectively start to move downward or upward.

136 
$$\varphi_e = 2\pi \frac{\tau_p^i - \tau_d^i}{\tau_d^{i+1} - \tau_d^i}$$
 (2)

137 where  $\tau$  represents the sequence of frame and superscript *i* represents the *i*<sup>th</sup> cycle. For 138 example, the frames where the droplet and plate reach the crest are chosen as standard 139 frames. Then the sequence of the standard frames for the *i*<sup>th</sup> and (*i*+1)<sup>th</sup> cycle are used 140 to calculate  $\varphi_e$  in Eq. (2).



#### 142 III. Physical model of a droplet subjected to forced vibration

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144

141

FIG. 3. (a) Schematic diagram of the force on a droplet without/with displacement. (b)

145 Corresponding visualization images, left: deformation only, right: deformation + displacement.

146 A generally used model to describe the drop-surface movement is the mass-spring-

147 damper system. Sarkar and Schowalter proposed a second-order ordinary differential

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148 equation (ODE) model to qualitatively describe the droplet motion.<sup>27, 28</sup> We adapt this mass-spring-damper model to our situation to clarify the underlying mechanism of 149 150 vibration-induced droplets motion (in particular, the phase shift between two droplets and their displacement). The vibrating droplet is illustrated in Fig. 3(a). We take the 151 centroid of the droplet, without considering gravity, as the coordinate origin. Hence, in 152 the steady state, droplets of different diameters have different displacements due to the 153 influence of gravity. With limited plate displacement and at non-resonance frequency, 154 the motion is the result of a balance between the driving force,  $F_d$ , the gravitational 155 force,  $F_{g}$ , the restoring force,  $F_{r}$ , and the shear force when displacement occurs,  $F_{dis}$ . 156 Accordingly, Newton's 2<sup>nd</sup> law of drop movement writes: 157

158 
$$m_d \frac{\partial x_d^2}{\partial^2 t} = F_d + F_g + F_r + F_{dis}$$
 (3)

159 The driving force between the wall and the droplet due to viscosity is a function of the 160 velocity gradient and is expressed as:

161 
$$F_{d} = \mu S_{\cup} \frac{\partial u}{\partial y} = \frac{\mu S_{\cup}}{\delta} \left( u_{p} - u_{d} \right)$$
(4)

162 where  $\delta = \sqrt{\frac{2\nu}{\omega_p}}$  is the Stokes-layer thickness with  $\nu$  the kinematic viscosity,

163 
$$S_{\cup} = \frac{\pi d^2}{4}$$
 is the contact area between the bottom of the droplet and the plate,  $\mu$  is the

164 dynamic viscosity, 
$$u_d = \frac{\partial x_d}{\partial t}$$
 is the velocity of the droplet and  $u_p = \frac{\partial x_p}{\partial t}$  is the velocity

165 of the plate where  $x_p = X_p \sin(2\pi f_p t)$  is the equation of motion for the plate, with  $X_p$ 

- 166 the amplitude of vibration, m.
- 167 The gravitational force is expressed as:

$$168 F_g = m_d g = \rho V_d g (5)$$

169 With a given contact angle  $\theta$ , the volume of the droplet  $V_d$  can be determined as<sup>29</sup>:

170 
$$V_d = \frac{\pi d^3}{24} \left( \frac{2 - 3\cos\theta + \cos^3\theta}{\sin^3\theta} \right)$$
(6)

7

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### 171 where d is the diameter of droplet measured according to the image from the high-speed

172 camera.

174 
$$F_r = -\frac{4\gamma S_{\cap} h}{d^2} \left( x_d - x_p \right)$$
(7)

175 where  $S_{\cap}$  is the droplet surface area in contact with air at equilibrium state. It is 176 determined as below:

177 
$$S_{\cap} = \frac{\pi d^2 \left(1 - \cos\theta\right)}{2\sin^2\theta}$$
(8)

178 In Eq. (7), *h* is a non-dimensional function depending on the wetting property of the 179 surface. Celestini et al.<sup>30</sup> proposed a universal curve of *h* according to  $\theta$  (°), expressed 180 by Eq. (9):

181 
$$h = -8e^{-7}\theta^3 + 8e^{-5}\theta^2 - 0.026\theta + 2.6589$$
 (9)

182 With driving frequency closer to the resonance frequency of the droplet and amplitude being high enough, droplet displacement could occur. Fig. 3(b) shows images of a 183 184 vibrating droplet without/with displacement. In case of displacement, two issues are to be corrected in the model. First, as the amplitude increases, the hypothesis of a 185 sufficiently weak external force does not hold.<sup>30</sup> Secondly, a shear force is applied to 186 the droplet due to the relative motion at the droplet-wall interface. Sellier proposed a 187 188 formula for the shear force by integrating the wall shear stress over the whole footprint of the droplet.<sup>31</sup> However, the no-slip condition and the constant droplet profile cannot 189 190 apply when displacement occurs. Here we propose a phenomenological equation expressed like a Gaussian distribution around the resonance frequency. The shear force 191 192 due to displacement  $F_{dis}$  is described as:

193 
$$F_{dis} = -\frac{\beta}{\varepsilon\sqrt{2\pi}} e^{\frac{-(f_p - f_r)^2}{2\varepsilon^2}} \left( u_d - u_p \right)$$
(10)

194 where  $f_r$  is the resonance frequency of the droplet.

195 Dimensional analysis implies the units of  $\beta$  and  $\varepsilon$  are kg/s<sup>2</sup> and 1/s respectively.

196 Therefore, we define  $\beta$  as the additional tension at resonant state, which is the product

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197 of the mass of the droplet and the square of its resonance frequency:

$$198 \qquad \beta = m_d f_r^2 \tag{11}$$

199 The parameter  $\varepsilon$  is the standard deviation representing the influence of the additional 200 tension at resonance. Hence we define  $\varepsilon$  as the efficient frequency. After fitting 201 experimental results, we found that  $\varepsilon$  can be expressed as below:

$$202 \qquad \varepsilon = \frac{f_r}{2} \tag{12}$$

203 Substituting Eqs. (4), (5), (7) and (10) into Eq. (3) yields:

$$m_{d} \frac{\partial x_{d}^{2}}{\partial^{2} t} = \frac{\mu S_{\cup}}{\delta} \left( 2\pi f_{p} X_{p} \cos\left(2\pi f_{p} t\right) - \frac{\partial x_{d}}{\partial t} \right) + m_{d} g$$
204
$$-\frac{4\gamma S_{\cap} h}{d^{2}} x_{d} - \frac{\beta}{\varepsilon \sqrt{2\pi}} e^{\frac{-(f_{p} - f_{r})^{2}}{2\varepsilon^{2}}} \left( \frac{\partial x_{d}}{\partial t} - 2\pi f_{p} X_{p} \cos\left(2\pi f_{p} t\right) \right)$$
(13)

Eq. (13) can be rewritten as:

206

$$m_{d} \frac{\partial x_{d}^{2}}{\partial^{2} t} + \left(\frac{\mu S_{\cup}}{\delta} + \frac{\beta}{\varepsilon \sqrt{2\pi}} e^{\frac{-(f_{p} - f_{p})^{2}}{2\varepsilon^{2}}}\right) \frac{\partial x_{d}}{\partial t} + \frac{4\gamma S_{\cap} h}{d^{2}} x_{d} =$$

$$m_{d} g + \left(\frac{\beta}{\varepsilon \sqrt{2\pi}} e^{\frac{-(f_{p} - f_{p})^{2}}{2\varepsilon^{2}}} + \frac{\mu S_{\cup}}{\delta}\right) 2\pi f_{p} X_{p} \cos\left(2\pi f_{p} t\right)$$
(14)

207 Simplifying the equation, we get:

208 
$$\frac{\partial x_d^2}{\partial^2 t} + 2p \frac{\partial x_d}{\partial t} + \omega_r^2 x_d = g + \frac{X'}{m_d} \cos(\omega_p t)$$
(15)

209 where damping ratio p and resonance frequency  $\omega_r$  are determined as:

210 
$$p = \frac{\mu S_{\cup} + \frac{\beta \delta}{\varepsilon \sqrt{2\pi}} e^{\frac{-(f_p - f_r)^2}{2\varepsilon^2}}}{2\delta m_d}$$
(16)

211 
$$\omega_r = \sqrt{\frac{4\gamma S_{\cap} h}{m_d d^2}}$$
(17)

212  $f_r = \frac{\omega_r}{2\pi}$  is the resonance frequency in Hz.

213 A new time-independent parameter, X', is used to simplify the above expression, as:

214 
$$X' = \left(\frac{\beta}{\varepsilon\sqrt{2\pi}}e^{\frac{-(f_p-f_r)^2}{2\varepsilon^2}} + \frac{\mu S_{\cup}}{\delta}\right)2\pi f_p X_p$$
(18)

By solving Eq. (15) with initial conditions  $u_d(0) = 0$  and  $F_r(0) = m_d g$ , the analytical solution can be derived as:

217 
$$x(t) = x_H(t) + x_P(t)$$
 (19)

In the above solution,  $x_H(t) = e^{-pt} \left( C_1 \cos(\omega_r \cdot t) + C_2 \sin(\omega_r \cdot t) \right)$  is the homogenous solution with  $\omega_r \cdot = \sqrt{\omega_r^2 - p^2}$ . It approaches zero as steady state achieves and this term could be neglected.

221 
$$x_{p}(t) = \frac{X}{m_{d}\sqrt{(2\omega_{p}p)^{2} + m_{d}(\omega_{r}^{2} - \omega_{p}^{2})^{2}}} \sin(\omega_{p}t + \varphi) + \frac{g}{\omega_{r}^{2}}$$
 is the particular solution

222 representing the steady-state vibration, where  $X_d = \frac{X'}{m_d \sqrt{\left(2\omega_p p\right)^2 + m_d \left(\omega_p^2 - \omega_p^2\right)^2}}$  is

- 223 the amplitude of the droplet's center of gravity.
- 224 Theoretical phase shift  $\varphi_t$  is determined by Eq. (20):

225 
$$\tan \varphi_r = \frac{-2\omega_p p}{\omega_r^2 - \omega_p^2}$$
(20)

226 Theoretical amplification ratio (the ratio of two amplitudes) is calculated by:

227 
$$AR_{t} = \frac{X_{d}}{X_{p}} = \frac{l_{d1} - l_{d2}}{l_{p}} = \frac{2\sqrt{\pi} f_{p} \left(\beta e^{\frac{-(f_{p} - f_{r})^{2}}{2e^{2}}} + \mu S_{\cup}\right)}{\delta \varepsilon m_{d} \sqrt{8(\omega_{p} p)^{2} + 2m_{d} (\omega_{r}^{2} - \omega_{p}^{2})^{2}}}$$
(21)

Finally, another critical parameter useful for droplets merging is the distance between the two borders (bottom of the upper drop and top of the lower one). The border positions are roughly calculated by  $X_{d}$ -d/2 with a circular shape hypothesis.

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#### 232 IV. Results and discussion

233 A. Experimental validation using the dynamic of single droplets

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FIG. 4. Comparisons between theoretical (Theo.) and experimental (Exp.) results. (a) Phase shift.
(b) Amplification ratio. Good agreement can be found for large droplets. For the smallest droplet at *d*=1.00 mm, the experimental AR curve shifts to the right. This is due to the influence of evaporation during the experiment.

239 We begin by investigating the dynamics of individual droplets of three different sizes 240 (1.00, 1.31 and 1.55 mm). Different exciting frequencies from 50 to 250 Hz are applied. 241 The natural frequency of the three droplets is respectively 183, 122, 95 Hz, inversely 242 related to their size. For low frequencies, the motion of droplets is in phase with the 243 plate due to the relatively higher surface tension ( $\varphi = 0$ ). Then, a phase shift  $\varphi$  starts to appear from a certain frequency (defined as starting frequency  $f_s$ ).  $\varphi$  continues to 244 245 increase rapidly until the input frequency reaches a sufficiently high value (defined as inversion frequency  $f_i$ ). Beyond  $f_i$ , the droplet is completely in opposite oscillation 246 247 compared with the plate ( $\varphi = \pi$ ). Fig. 4a shows similar phase shift trends for the three droplet sizes. For each droplet, the starting and inversion frequencies,  $f_s$  and  $f_i$ , are close 248 249 before and after their corresponding resonance frequency. Comparing experimental and 250 theoretical results, the maximum theoretical phase shift  $\phi_t$  is slightly smaller than found 251 experimentally. This is due to the deformation of droplet at high frequencies, which is not considered in the theoretical model. Globally, the way its trend and slope change 252 253 with the input frequency in good agreement with the experiment, validates the 254 theoretical approach.

255 Fig. 4(b) shows the amplification ratio of droplets compared with the plate oscillation

amplitude. Both theoretical and experimental data show peak amplifications at around

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resonance frequency (183, 122, 95 Hz). Under our conditions, the highest value of *AR* is about 6, implying much stronger drop motion than the plate. The amplification ratio

remains higher than 5 at a bandwidth of  $\sim 10$  Hz around the resonance frequency. The high *AR* at the large-bandwidth frequency is a valuable condition to achieve drop

In sum, experimental results fit well with theoretical ones in terms of amplification ratio

and phase shift between single droplets and plate. Considering the consistency between

the theoretical and experimental data, the theoretical model can be used to predict the

motion of droplets in broader conditions. The following results are based on the

shedding or coalescence. We discuss about it later with two droplets dynamics.

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droplets to merge. In addition, the slope of the curve rises with the increase of diameter

in the opposite phase. This provides condition (necessary but not sufficient) for adjacent

278 because damping ratio *p* decreases for larger drop sizes.

279 Fig. 5(b) gives the theoretical phase shift between pairs of two droplets, using the 1.00 mm one as a reference. The phase shift between two droplets,  $\Delta \varphi$ , starts to be significant 280 281 for larger droplets who have lower resonance frequencies. Then,  $\Delta \varphi$  reaches its peak value before decreasing at higher frequencies. The frequency corresponding to the peak 282 phase shift is the optimal frequency fopt to promote drop merging. For smaller droplets 283 that are closer to the reference size, the phase shift frequency range is narrower. 284 Moreover, the peak phase shift value decreases: it is  $(3/4)\pi$  for the largest droplets pair 285 286 (1.40 mm and 1 mm, near opposite motion) while only  $(1/3)\pi$  for the pair 1.10 mm and 287 1mm. We define the continuous frequency plateau where at least 90%  $\Delta \varphi_{\text{max}}$  is satisfied as bandwidth. For example, between two droplets (1.10mm and 1.40mm),  $f_{opt} = 133$  Hz. 288 The  $\Delta \varphi$  around  $f_{opt}$  is a relatively large plateau. The whole range of [121, 146] Hz allows 289 at least 90% maximum phase shift  $\Delta \varphi_{max}$ . The larger of this bandwidth corresponds to 290 291  $\pm 12.5$  Hz or  $\pm 9.4\%$  f<sub>opt</sub>. Compared to the resonance-induced shedding, our technique offers a larger bandwidth frequency at which two neighboring droplets could coalesce. 292 293 For example, Sun et al. investigates droplet detachment on a superhydrophobic surface at resonant vibration.<sup>20</sup> At the lowest amplitude, frequency range should be kept within 294 295  $\pm 1\%$  of its resonance frequency ( $\leq \pm 1$  Hz). Out of this range, the vibration surface should be amplified by +10% of the lowest amplitude. The operational bandwidth of 296 297 our study is thus much larger.

To generalize the theory in search of the best operational condition for different drop sizes, we determine the theoretical optimal frequency  $f_{opt}$ . At this frequency, the phase shift reaches its peak value  $\Delta \varphi_{max}$ , (c.f. Fig. 5(b)). Ideally,  $\Delta \varphi_{max}$  between two droplets would be  $\pi$ :

302 
$$\left| \arctan \frac{-2\omega_{opt}p}{\omega_1^2 - \omega_{opt}^2} - \arctan \frac{-2\omega_{opt}p}{\omega_2^2 - \omega_{opt}^2} \right| = \pi$$
(22)

303 The optimal frequency  $f_{opt} = \omega_{opt} / 2\pi$  can be obtained by:

304 
$$\omega_{opt} = \sqrt{\frac{d_1\omega_1^2 + d_2\omega_2^2}{d_1 + d_2}}$$
 (23)

305 The theoretical optimal frequencies of 22 drop size combinations are given in (Fig.

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306 6(a)). The corresponding droplet sizes are  $d_1 \in [1.2, 1.5]$  mm and  $d_2 \in [0.8, 1.4]$  mm. One 307 take-away message is that the optimal frequency lies between the two resonance 308 frequencies for each droplet. Furthermore, phase shift between two droplets ( $\Delta \varphi$ ) at corresponding optimal frequency decreases as the sizes of the two droplets get similar, 309 as listed in Tab.1. It is thus difficult to achieve an absolute opposite movement for 310 droplets with similar sizes. It is worth noting when  $d_1=d_2$ , there is no way to force them 311 into contact by the proposed vibration technique, since they are strictly in phase 312 313 whatever the frequency is.

314 In addition to  $\Delta \varphi$ , we also need a high enough AR to promote droplets merging. Shown in Fig. 6(b) is the spectral distribution of AR for four typical drop sizes. Peak AR values 315 correspond to the resonance frequency of one specific drop size, and are generally 316 around 6 with our surface wetting conditions. According to the size combination, the 317 318 optimal working frequency will not be at one specific resonance frequency but in 319 between the two resonance ones. Consequently, the AR is always lower. Fortunately, 320 the operating frequency can be adjusted within the 90% optimal frequency range 321 without strong disturbance to the relative phase shift  $\Delta \varphi$ . For instance, Fig. 6(a) 322 indicates an optimal frequency between a 1.2 mm drop and a 1.5 mm one should be at 118 Hz and the corresponding 90% optimal frequency range is from 108 Hz to 129 Hz. 323 324 At this frequency, Fig. 6(b) gives ARs of around 3.2 and 2.3 for the two droplets respectively. If we fix the operating frequency at 108 Hz, the two ARs will be 2.4 and 325 326 3.9, which holds greater potential for droplet merging.

Eventually, we can define the scope of application of the strategy in terms of size limit and frequency limit. The study by P. B. Weisensee et al. points out that water droplets grow mainly *via* coalescence instead of direct condensation once their diameter is larger than the effective transition one at ~20  $\mu$ m.<sup>34</sup> With the current surface condition, theoretically it corresponds to a frequency limit of 65 kHz. The upper limit drop size can be obtained by balancing the gravitational and surface tension forces.<sup>35</sup> The average drop departure size for our surface is estimated to be ~1.60 mm. The corresponding

334 frequency (lower frequency limit) is ~90 Hz.

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FIG. 6. (a) The optimal frequency in favor of droplet coalescence according to drop size combination. Shaded area shows the bandwidth around the optimal frequency satisfying 90%  $\Delta \varphi_{\text{max}}$ . (b) The frequency dependence of amplification ratio (AR) for different droplets.

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340 TAB. 1. Maximum phase shift between two droplets ( $\Delta \varphi_{max}$ ) at corresponding optimal frequency.

| $\frac{d_1(\mathbf{mm})}{d_2(\mathbf{mm})}$ | 1.20  | 1.30  | 1.40  | 1.50  |
|---|-------|-------|-------|-------|
| 0.80  | 0.82π | 0.85π | 0.87π | 0.89π |
| 0.90  | 0.75π | 0.80π | 0.84π | 0.86π |
| 1.00  | 0.63π | 0.73π | 0.78π | 0.82π |
| 1.10  | 0.40π | 0.60π | 0.71π | 0.77π |
| 1.20  |       | 0.38π | 0.58π | 0.69π |
| 1.30  | 0.38π |       | 0.34π | 0.56π |
| 1.40  | 0.58π | 0.34π |       | 0.34π |

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342 C. Application of the new operating strategy

343 We use a syringe to create droplets with controllable diameters and relative position.

Specifically, a smaller droplet with a diameter of 1.12 mm and a larger one with 1.39 344

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mm diameter are attached to a plate. They are aligned along the vibrating direction. The relative position of them in the x direction is 1.50 mm (center-to-center). Sinusoidal vibrations of different frequencies and amplitudes are applied to the plate and the corresponding motions of the two droplets are visualized. Slow-motion videos showing the behaviors of the two droplets at different frequencies can be found in Fig. 7 (Multimedia available online).

The resonance frequencies fr for the two droplets are 154 Hz and 112 Hz respectively. 351 352 Our theoretical model gives an optimal frequency  $f_{opt}$  at 132 Hz and a 90% satisfactory 353 range from 121 Hz to 143 Hz. During the experiment, we adopt three excitation frequencies at 80 Hz, 140 Hz and 260 Hz. Theoretically, the 140 Hz one is within the 354 optimal range, while 80 Hz and 260 Hz would be either too low or too high. For  $f_{\rm P}$  =140 355 Hz and 260 Hz, the amplitude of plate  $X_p$  is set to 0.042 mm. For  $f_p = 80$  Hz, the 356 357 amplitude of the plate  $X_p$  is set to 0.085 mm. Based on the amplitude and frequency of 358 the plate, the motion profiles of the two droplets are given in Fig. 7 (Multimedia 359 available online). We distinguish the upper and lower limits of each droplet as well as 360 their centers of gravity. The motion of the plate is also given. For merging purpose, 361 special attention should be paid to the lowest limit of the upper droplet (larger one in our case) and the highest point of the bottom droplet (smaller one). The experimental 362 363 keyframes are inset in the corresponding figures. At  $f_p = 80$  Hz, the movements of the 364 two droplets are almost synchronous with the movement of the plate. Therefore, the 365 blank region between the two droplets indicate that they are in phase and cannot merge. 366 Even if increasing the amplitude  $X_p$ , it has little effect on the blank region. At  $f_p = 260$ 367 Hz, the responses of the two droplets to the vibration of plate are weak. The blank region still exists due to gentle fluctuation of two droplets and small phase shift  $\Delta \varphi$ . 368 369 When  $f_p = 140$ Hz, the two droplets are in totally reversed phase. The highest point of

the smaller droplet and the lowest point of the larger droplet may come into contact when the plate is near the peak. The reason is that the larger droplet nearly asynchronous to the plate moves downwards while the smaller one nearly synchronous to the plate moves upwards. This produces a contrary motion which brings them together. Once the

two droplets encounter each other, they merge rapidly and in an irreversible way.<sup>32</sup>

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375 The process of contact is also observed in the experiment, as shown in the keyframe in

contact represents the virtual motion because the two droplets coalesce into a new one.

376 Fig. 7(c) (Multimedia available online). It should be noted that the dashed line after



FIG. 7. Theoretical motion curves of two droplets and experimental keyframes at different
frequencies. Shaded region represents the droplet projection on the plate and dash-dotted line
represents the motion curve of centers of gravity. Red lines represent the motion curve of the plate.
(Multimedia available online)

383 In addition to choosing the right frequency band, high enough amplitude is also a condition to achieve droplet merging. Fig. 8 (Multimedia available online) shows the 384 385 motion of the two droplets  $(x_d)$  driven by the plate  $(x_p)$  at three different amplitudes. 386 The frequency is fixed and within the optimal frequency band. As illustrated, the motion 387 of the larger droplet (Drop 1) is nearly opposite to the plate motion while the smaller 388 one (Drop 2) is nearly synchronous to the plate. This property does not change as long as the frequency is fixed. At low vibration amplitude  $X_p = 0.022$  mm, the responses are 389 390 so mild that the two droplets remain separated. As  $X_p$  increases to 0.033 mm, the 391 minimum distance between the two droplets reduces significantly but the two droplets 392 still do not contact. When  $X_p$  increases to 0.042mm, their reactions further intensify so 393 that they merge into one droplet spontaneously at the moment of approaching.

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FIG. 8. Theoretical motion curve of the two droplets at different excitation amplitudes from the plate. For drop motion x<sub>d</sub>, only the close border lines are shown, i.e., lower limit of Drop 1 and upper limit of Drop 2. The dashed lines are virtual as merging already occurs. (Multimedia available online)

#### 400 V. Conclusion

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401 The dynamics of droplet coalescence driven by vibration on a vertical surface are experimentally and theoretically investigated. A phenomenological equation is 402 403 introduced into a mass-spring-damper theoretical model to solve two problems: strong 404 external force and additional shear force due to unpredicted slip of droplet footprint 405 area. Results show phase shift  $\varphi$  occurs from size-dependent starting frequencies  $f_s$  to inversion frequencies  $f_i$ . This leads to specific optimal frequencies at which two droplets 406 407 of different sizes move in opposite directions. Coupled with a high enough amplitude, these droplets may furtherly coalesce. The working frequency bandwidth is at the order 408 409 of  $\pm 10$  Hz or higher, offering more chances to promote coalescence of adjacent droplets. 410 Our findings could be helpful for the coalescence and removal of droplets. Given the 411 droplets' size on the vertical surface, the optimal frequency can be obtained and then 412 the vibration at the large-bandwidth frequency can be applied on the surface. The

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droplets coalesce and form a larger droplet which could detach from the surface. During
the sliding, the droplet entrains other drops, acting as wipers. Therefore, larger
uncovered surface is made available for an intensified condensation effect. Furthermore,
no special hydrophobic requirement is necessary for the surface. With certain
frequencies applied on the surface, the droplets of different sizes can coalesce and
detach much easier. Potential applications are promising in condensation enhancement,
window cleaning, photovoltaic panel cleaning, etc.

Although the whole study is based on a vertical surface driven by sinusoidal vibration, it can be extended to other situations. First, by simply replacing  $F_g$  by  $F_g = m_d g sin \alpha$ , the developed model remains valid for inclined surfaces with an inclination angle  $\alpha$ . Second, other forcing functions, like triangle, square, or sawtooth, can also induce droplet coalescence as they can be treated as a series of sinusoidal functions by Fourier expansion.

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- 436 AUTHOR DECLARATIONS
- 437 **Conflict of Interest**
- 438 The authors have no conflicts to disclose.

#### 440 DATA AVAILABILITY

441 The data that support the findings of this study are available from the corresponding

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442 author upon reasonable request.

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