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Conical Interfaces between Two Immiscible Fluids Induced by an Optical Laser Beam

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We demonstrate the existence of conical interface deformations induced by a laser beam that are similar to Taylor cones in the electrical regime. We show that the cone morphology can be manipulated by fluid and laser parameters. A theory is proposed to quantitatively describe these dependences in good agreement with experimental data obtained for different fluid systems with low interfacial tensions. Counterintuitively, the cone angle is proved to be independent of the refractive index contrast at leading order. These results open a new optofluidic route towards optical spraying technology—an analogue of electrospraying—and more generally for the optical shaping of interfaces.

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One hundred years ago, in a pioneering work, Zeleny observed the destabilization of a suspended conducting liquid drop submitted to a sufficiently strong electric field [1,2]. The interface takes a conical shape, followed by a jet that usually breaks up into a spray of tiny droplets, a key phenomenon for electrospraying and electrospinning technologies [3–5]. Such conical menisci were theoretically understood by Taylor [6] and are now commonly termed as “Taylor cones.” Beyond the surprising and fascinating elegance of such a simple conical solution for a complex mathematical problem involving deformable boundaries, this Taylor cone is important for applications. Indeed, the finite angle of the cone is a key parameter determining the size of the emitted jet, and thus of the resulting droplets [7].

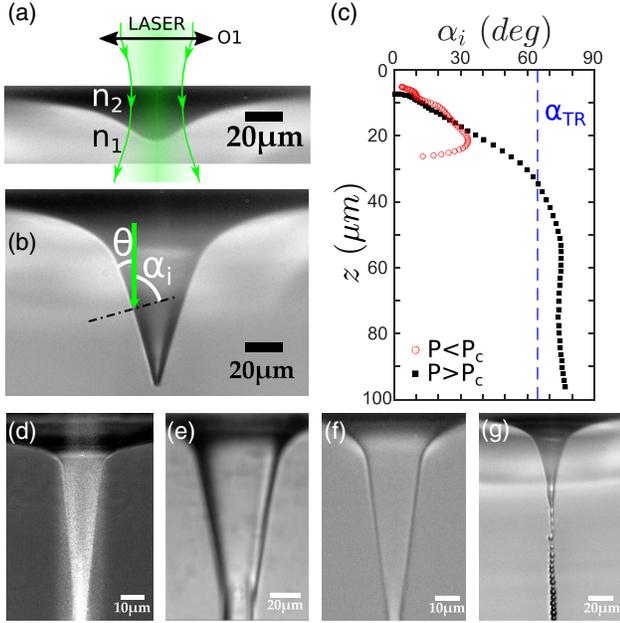
Taylor cones are thus an essential component in processes as varied as the emission of monodisperse droplets [8], ink jet printing [3,5], the design of nanostructures [9] and encapsulation techniques [10]. Taylor cones were naturally generalized to electrically or magnetically induced deformations of interfaces between fluids presenting different conductivities, dielectric constants or magnetic susceptibilities [11]. Depending on these properties, but also on the nature of the field (either ac or dc) [12], the cone angle can vary over a wide range. Furthermore, conical shapes of fluid interfaces seem even more general, since portions of cones naturally appear in situations as varied as drops stretching [13], viscous breakup of pendant drops [14], tip streaming by Marangoni stress [15], or inertial jet eruption [16]. This suggests that various types of excitatory fields are able to induce conical deformations, as earlier suggested by Taylor himself [13]. In this context, considering the developments on the manipulation of fluids by light [17,18] and previous studies [19–21] where conical shapes could be suspected, a natural and surprisingly unresolved question is whether or not cones can be induced by light as well.

The goal of this Letter is to demonstrate the emergence of conical shapes in the optical regime and to characterize

their geometry. Using very different fluid systems, we show indeed that above a critical radiation pressure exerted by a continuous laser wave, soft interfaces deform and adopt a conical shape. We propose a theory that correctly predicts the cone angles for a wide range of fluid and excitation parameters. Counterintuitively, we show that the cone angle does not depend on the refractive index contrast, while it is at the origin of the radiation pressure that induced the conical deformation.

To observe optically induced cones, we consider a continuous Gaussian laser wave that impinges a soft fluid interface from the liquid of higher refractive index as shown in Fig. 1(a). The laser beam is focused on the interface using standard optical elements that can be adjusted to vary the beam waist w_0 at the interface. At low power, the interface is gently deformed into a bell-shaped profile by optical radiation pressure [Fig. 1(a)]. This is due to the transfer of optical momentum of photons to the interface, as previously described [22,23]. Above a critical beam power P_c , the interface profile lengthens and sharpens, and a conical deformation emerges [Fig. 1(b)].

To characterize the geometry of the interface, we represent the local angle α_i in Fig. 1(c) as a function of the height z , i.e., the distance to the undeformed interface. This curve clearly exhibits a plateau region that is absent in the low power regime. This plateau demonstrates the existence of an optically induced conical deformation and defines its angle. To get insight in the mechanism at the origin of the cone formation, we image the optical path of the laser wave using specific optical filters [Fig. 1(d)]. The intense reflection of the laser beam at the cone interface tends to show that light is totally reflected inside the conical structure, which thus acts as a self-induced funnel guide. This is further confirmed in Fig. 1(c) by the fact that the incident angle α_i is always larger than the total reflection angle [$\alpha_i > \alpha_{\text{TR}} = \arcsin(n_1/n_2)$] in the plateau region. We anticipate that this total reflection condition is important to



F1:1 FIG. 1. (a) Sketch of the experiment: a laser beam ($\lambda = 532$ nm
 F1:2 in vacuum) is focused at the interface with the objective O1
 F1:3 (Olympus x10) and deforms this interface by radiation pressure.
 F1:4 Deformation of the interface for $P < P_c$ (Winsor toluene S1b
 F1:5 for $w_0 = 12.9 \mu\text{m}$, $P = 1.53$ W). (b) Conical deformation by
 F1:6 radiation pressure for $P > P_c$ (Winsor toluene S1b for
 F1:7 $w_0 = 12.9 \mu\text{m}$, $P = 1.55$ W). (c) Typical variation of the incident
 F1:8 angle α_i as a function of the height of deformation for $P < P_c$
 F1:9 and $P > P_c$. Note that the curve for $P > P_c$ exhibits a clear
 F1:10 plateau. α_{TR} is the total reflection (TR) incident angle. (d) Light
 F1:11 path revealing the total reflection mechanism inside the conical
 F1:12 deformation (microemulsion S3). (e)–(g) Conical deformations
 F1:13 for various experimental systems: (e) Winsor heptane S2 for
 F1:14 $w_0 = 12.2 \mu\text{m}$, $P = 2.25$ W, (f) microemulsion S3e for $w_0 =$
 F1:15 $9.0 \mu\text{m}$ and $P = 2.89$ W, and (g) jet and drop emission at the tip
 F1:16 of the cone for Winsor toluene S1b system with $w_0 = 8.8 \mu\text{m}$ and
 F1:17 $P = 1.06$ W.

88 explain the formation of optically-induced cones in our
 89 conditions.

90 To test the generality of optical liquid cones, we consider
 91 three main experimental fluid systems that are transparent at
 92 the used optical wavelength (optical absorption smaller than
 93 $3 \times 10^{-4} \text{ cm}^{-1}$) and based on Winsor phases (toluene:
 94 S1a–S1b, heptane: S2) and quasicritical microemulsions
 95 (S3a–S3e). By varying chemical composition or tempera-
 96 ture, we obtain in the end eight subsystems denoted by
 97 S1a–S1b, S2, S3a–S3e (see Supplemental Material [24–28]
 98 for details). This enables us to vary the refractive index
 99 contrast involved in the radiation pressure ($\Delta n = n_2 -$
 100 $n_1 = 0.0129 - 0.1449$) and the interfacial tension involved
 101 in the restoring capillary pressure ($\gamma = 2 \times 410^{-7} - 1 \times$
 102 310^{-5} N/m) over more than one order of magnitude.
 103 Refractive indexes for various systems were measured by
 104 standard refractometry methods, while interfacial tensions
 105 were determined by analyzing the viscous breakup dynamics

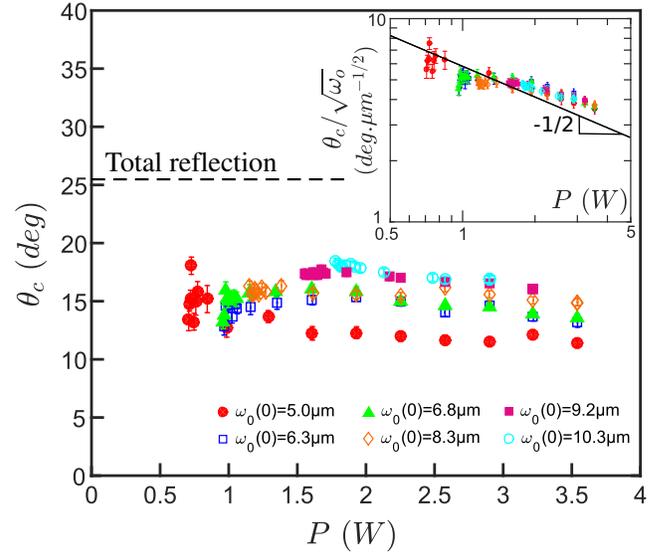


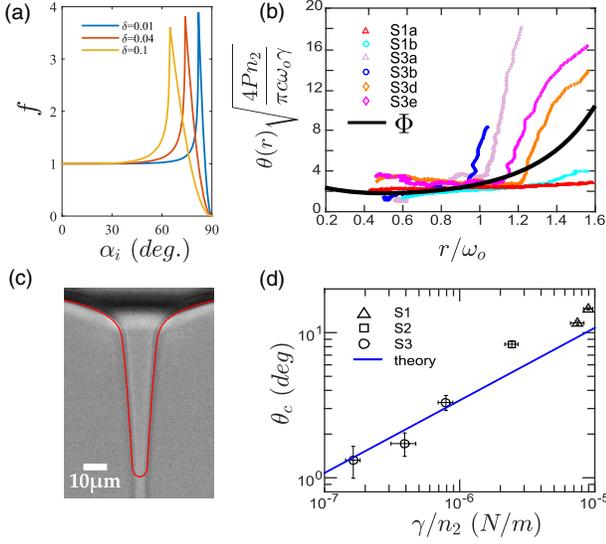
FIG. 2. Semiangle θ_c of the cone for Winsor toluene system
 S1b as a function of the power P and the beam waist w_0 of the
 laser. $w_0(0)$ represents the laser beam waist extrapolated at zero
 power (see the Supplemental Material [24]). The dashline
 indicates the total reflection value $\pi/2 - \alpha_{\text{TR}}$. Inset: Cone semi-
 angle rescaled by $\sqrt{w_0}$ versus laser power P in log-log scale.

of liquid thread [29]. As illustrated in Fig. 1, stationary
 optically induced cones are generated for all experimental
 systems. Similar to electrified interfaces, these conical
 shapes are very stable and robust for both turbid (S1, S3)
 and nonturbid (S2) fluid systems. Remarkably, the conical
 structure often emerges together with a jet that emits droplets,
 as illustrated at the bottom in Fig. 1(g). Importantly, as shown
 in Fig. 1, we observe that the cone angle is specific to each
 fluid system, indicating that fluid properties are important to
 define the cone morphology.

We now quantify the effects of the laser parameters on the
 cone angle. The edge of the cone is detected by a homemade
 image analysis program that measures the cone semiangle
 $\theta_c = \pi/2 - \alpha_i$ in the plateau region [see Fig. 1(c)] after
 averaging over several stationary profile pictures. In Fig. 2,
 we show how θ_c depends on the incident laser power P at
 various waists w_0 for system S1 as an example. To be as
 accurate as possible, we note that increasing P of our laser
 also results in an increase of the waist $w_0 = g(P, w_0(0))$ via a
 function g , which is fully characterized in the Supplemental
 Material [24], with $w_0(0)$ as the extrapolated waist at zero
 power. As shown in Fig. 2, the cone semiangle increases with
 the beam waist w_0 at a given power and slightly decreases
 with the applied power. This indicates that laser parameters
 are crucial for controlling the cone morphology.

To understand the physical mechanism at the origin of
 the conical deformation, it is useful to start with the force
 balance equation for an axi-symmetric stationary profile
 [22,30]:

$$\gamma\kappa(r) - \Delta\rho gh(r) = \Pi_{\text{rad}}(r), \quad (1)$$



F3:1 FIG. 3. (a) Variation of the f function with the incident angle α_i
 F3:2 for different relative index contrasts $\delta = 2\Delta n/(n_1 + n_2)$.
 F3:3 (b) Variation of the normalized cone angle as a function of the
 F3:4 normalized radial position r/w_0 for various liquid systems. The
 F3:5 solid line indicates the theoretical prediction $\Phi(\cdot)$ [see Eq. (6)].
 F3:6 (c) Comparison between a theoretical cone deformation (red line)
 F3:7 and an experimental deformation for microemulsion system
 F3:8 S3b for $P = 0.5$ W and $w_0 = 5.8$ μm . (d) Cone angle as a
 F3:9 function of γ/n_2 for various systems and for a given ratio
 F3:10 $w_0/P = 4.57$ $\mu\text{m}/\text{W}$. The line indicates the theoretical prediction
 F3:11 [Eq. (7)].

136 where both the Laplace pressure $\gamma\kappa(r)$ and buoyancy
 137 $\Delta\rho gh(r)$ balance the optical radiation pressure $\Pi_{\text{rad}}(r)$.
 138 Here, r is the radial distance to the beam axis, h is the
 139 height of the profile, g the earth acceleration, and κ is
 140 the local curvature. The optical radiation pressure is given
 141 for a continuous Gaussian wave (mode TEM_{00}) by the
 142 following:

$$\Pi_{\text{rad}}(r) = \frac{n_2}{c} \frac{2P}{\pi w_0^2} e^{-\frac{2r^2}{w_0^2}} \delta f(\alpha_i), \quad (2)$$

143 where c is the light celerity, $\delta = 2\Delta n/(n_1 + n_2)$ is the
 144 relative index contrast between the two phases, and f is
 145 a geometric function that describes the variation of the
 146 radiation pressure with the local incident angle α_i ,
 147 $f(\alpha_i) = \cos^2(\alpha_i)\{1 + R(\alpha_i) - [\tan(\alpha_i)/\tan(\alpha_t)]T(\alpha_i)\}/\delta$,
 148 R and T being the reflexion and transmission Fresnel
 149 coefficients and α_t the refracted angle. This function f is
 150 plotted in Fig. 3(a). Importantly, it displays a decreasing
 151 behavior above the total reflection angle α_{TR} , which means
 152 that the more inclined is the interface the less efficient is the
 153 radiation pressure. Therefore, above α_{TR} , the intensity of
 154 the radiation pressure is directly related to the local
 155 inclination of the interface, which will be the determinant
 156 to set the value of the cone angle.
 157

We first describe the interface deformation at moderate 158
 beam power. As the optical Bond number (defined by 159
 using the beam waist as the characteristic length scale) 160
 $\text{Bo} = (\Delta\rho g w_0^2/\gamma) \approx 0.001\text{--}0.2$ is small, buoyancy can be 161
 neglected in first approximation. Furthermore, as the 162
 relative index contrast δ is also a small parameter, 163
 the radiation pressure can be considered as constant over 164
 a large range of inclination angles [i.e., $f \approx 1$, see Fig. 3(a)]. 165
 With these approximations, the force balance Eq. (1) at low 166
 powers becomes this: 167

$$\frac{\gamma}{r} \frac{\partial}{\partial r} (r \cos \theta) \approx \frac{2P\Delta n}{\pi c w_0^2} e^{-2r^2/w_0^2}. \quad (3)$$

This equation is readily integrated for a closed profile, 168
 leading to $\cos \theta = (P\Delta n/2\pi c \gamma w_0)(1 - e^{-2u^2}/u)$ where 170
 $u = r/w_0$. The self-consistency condition that $\cos \theta$ 171
 remains lower than unity for all r leads to the definition 172
 of a critical power 173

$$P_c \approx 2.2 \frac{\pi c w_0 \gamma}{\Delta n}, \quad (4)$$

above which one should observe strongly deformed inter- 174
 faces, with inclination angles of the order of the total 175
 reflection angle. This condition is compatible with previous 176
 analyses [21,31] and is also in good agreement with the 177
 critical power values measured in our experiments (see 178
 Fig. S3 in the Supplemental Material [24]). 179
 180

Above the critical power P_c , a new region appears where 181
 total reflection conditions hold, so that $f \approx 2\theta^2/\delta$, indicat- 182
 ing that the radiation pressure depends on the local profile 183
 slope. Moreover, in this region the opening angles θ are 184
 small compared to one, a condition which is satisfied in all 185
 our experiments. Hence, in this region the force-balance 186
 equation can be considerably simplified and becomes 187

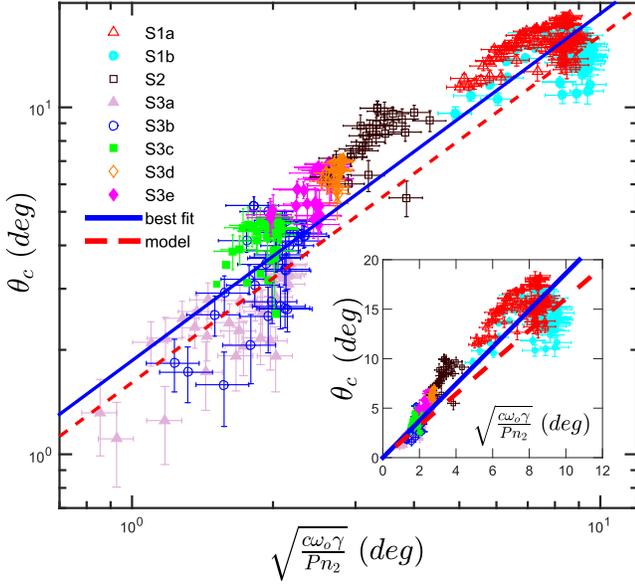
$$\frac{\gamma}{r} = \theta^2 \frac{4n_2 P}{\pi c w_0^2} e^{-2r^2/w_0^2}, \quad (5)$$

leading to: 188

$$\theta(r) = \sqrt{\frac{\pi c \gamma w_0}{4P n_2}} \Phi\left(\frac{r}{w_0}\right), \quad \Phi(X) \equiv \frac{e^{X^2}}{\sqrt{X}}. \quad (6)$$

The local angle in the total reflection region is therefore 190
 proportional to the dimensionless function $\Phi(\cdot)$, which is 191
 plotted in Fig. 3(b). It exhibits a clear plateau characterizing 192
 the conical deformation in the range $r/w_0 \approx 0.3\text{--}1$. To fully 193
 predict the cone angle variation with physical parameters, 194
 we characterize the minimal half-opening angle in Eq. (6), 195
 which is obtained for $r/w_0 = 0.5$. We find this: 196
 197

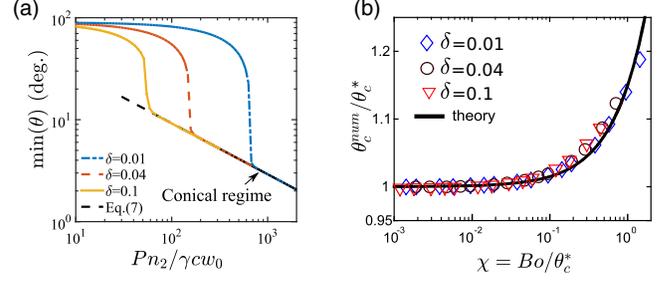
$$\theta_c^* = \beta \sqrt{\frac{c w_0 \gamma}{P n_2}}, \quad (7)$$



F4:1 FIG. 4. Experimental cone angles versus the characteristic cone
 F4:2 angle $\sqrt{cw_0\gamma/Pn_2}$ for all the investigated experimental systems.
 F4:3 The best fit is $\theta_c = 1.86(cw_0\gamma/Pn_2)^{0.5}$ whereas the dashed line
 F4:4 refers to Eq. (7). Inset: same plot in linear scales.

199 with $\beta = e^{1/4}\sqrt{(\pi/2)} \approx 1.61$. Importantly, this expression
 200 predicts that the cone angle θ_c decreases with the applied
 201 power P and increases with the waist w_0 as observed
 202 experimentally.

203 Corresponding scalings in w_0 and P are experimentally
 204 confirmed in the inset of Fig. 2 where all the data are
 205 rescaled by $\sqrt{w_0}$ and collapse into a single master curve.
 206 In Fig. 3(b), comparisons of the renormalized angle profiles
 207 for various fluid systems show a good agreement with the
 208 theory even if experimental profiles are more extended than
 209 theoretical ones. Note that experimental angle profiles are
 210 limited to $r/w_0 \geq 0.4$ because a jet usually forms at the
 211 cone tip, contrary to theoretical modeling which only
 212 considers closed deformations. As explained in the
 213 Supplemental Material [24], a discussion on the jet that
 214 forms at the tip of the conical deformation is beyond the
 215 scope of the present work, but we note that breakup and
 216 drop formation are not expected to significantly affect the
 217 profile in the conical region. For higher r/w_0 , experimental
 218 profiles also display slope discontinuities, corresponding in
 219 the theory to the switching point where total reflection is no
 220 longer satisfied (see Fig. S5 in the Supplemental Material
 221 [24]). We then numerically calculate the full height profile
 222 $h(r)$ from Eq. (1), and superimpose it with the experimental
 223 measurements in Fig. 3(c). In this example, despite the
 224 difference at the cone tip, the theoretical profile fits
 225 reasonably well the experimental deformation (without
 226 any fitting parameters). This demonstrates the ability of
 227 our model to describe the radial variation of the cone angle.
 228 We then test the scaling with interfacial tension in Fig. 3(d),
 229 where we compare predictions to the experimental cone



F5:1 FIG. 5. (a) Theoretical minimal angle of the deformation
 F5:2 without gravity effects for various refractive index contrast
 F5:3 (b) Rescaled cone angle $\theta_c^{\text{num}}/\theta_c^*$ numerically obtained compared
 F5:4 with the perturbative result $\theta = \theta_c^*(1 + \nu\chi)$ as a function of the
 F5:5 $\chi = Bo/\theta_c^*$ parameter for $P/P_c = 1.35$.

angles for all the systems at a given ratio w_0/P .
 Remarkably, the model is also in good agreement with
 the experimental data over almost two decades in interfacial
 tension. This strongly supports that the characteristic cone
 angle is given by $\sqrt{cw_0\gamma/Pn_2}$ as suggested by Eq. (7).

To further confirm this model, we plot in Fig. 4 the cone
 semiangle θ_c for the eight experimental systems investi-
 gated here for all the experimental conditions as a function

of the characteristic cone angle $\sqrt{cw_0\gamma/Pn_2}$. Over more
 than one decade (see also the same data in linear scale in
 the inset of Fig. 4), all the data collapse into a single master
 curve despite some inherent dispersion of data, in particular
 close to the critical power P_c where the interface sensitivity
 to excitation is the largest. The best fit is $\theta_c =$
 $1.86\sqrt{cw_0\gamma/Pn_2}$, which is very close to the model pre-
 diction $\theta_c = 1.61\sqrt{cw_0\gamma/Pn_2}$ [see Eq. (7)]. The agree-
 ment is even reinforced considering that no adjustable parameter
 is used in the model. Consequently, conical deformations
 can be fully controlled with both fluid properties and
 excitation parameters.

Counterintuitively, the model predicts that the cone angle
 θ_c does not depend on the relative index contrast δ [see
 Eq. (7)]. Indeed, as shown in Fig. 5(a), as soon as the
 critical power is reached, the minimal deformation angle
 $\min(\theta)$ switches to a single behavior independent of δ .
 This is due to the independence of the radiation pressure
 with the refractive index contrast in the total reflection
 regime [see Eq. (5)], as opposed to the normal incidence
 case. However, the refractive index contrast Δn remains
 essential to set the critical power P_c to observe a cone.

We now investigate whether gravity effects could be
 responsible for deviations between experimental data and
 theory. Gravitational effects can be evaluated by forming
 the ratio between the buoyancy $\Delta\rho gh$ and the characteristic
 Laplace pressure γ/w_0 . Since $h \sim w_0/\theta_c^*$ in the conical
 region, the relevant dimensionless parameter is $\chi =$
 $\Delta\rho gw_0^2/(\gamma\theta_c^*) = Bo/\theta_c^*$. Intuitively, increasing gravita-
 tional effects should flatten the deformation and thus increase
 the cone angle θ_c . As explained in the Supplemental Material
 [24] by a perturbation analysis, the cone semiangle is

270 expected to vary as $\theta = \theta_c^*(1 + \nu\chi)$, where $\nu \simeq 0.14\text{--}0.18$ is
 271 a weakly varying parameter. These results are confirmed in
 272 Fig. 5(b) by comparing with the complete numerical reso-
 273 lution of the force balance equation [Eq. (1)] for various
 274 index ratio. The numerical results in Fig. 5(b) collapse into a
 275 single master curve, validating this perturbation analysis. We
 276 evaluate the deviations from the analytical results without
 277 gravity [Eq. (7)] to be at most 30% for the largest values of
 278 χ in our experiments ($\chi \approx 10^{-2} - 2$), confirming that gravity
 279 can be neglected at leading order.

280 To conclude, we have experimentally and theoretically
 281 demonstrated the existence of optically induced conical
 282 deformations. The cone morphology is controlled by the
 283 fluid properties and laser parameters. The analytical and
 284 numerical analyses quantitatively predict an optical cone
 285 semiangle in good agreement with measurements over a
 286 wide range of parameters for several liquid systems. Such
 287 cones can be considered as “optical analogues” of Taylor
 288 cones, in the sense that the structure of the electromagnetic
 289 field near the interface results from its interference with
 290 refracted ray and is strongly coupled to its deformation due
 291 to total reflection conditions. As already demonstrated for
 292 Taylor cones [7], we anticipate that the properties of these
 293 static optical cones will be a key parameter to control the
 294 hydrodynamic jet at its tip as suggested by Fig. 1(g). Our
 295 results quantitatively establish the first step towards opto-
 296 spraying and a new optical control of interfacial properties
 297 and interfacial morphologies. This Letter also advances a
 298 new example showing that conical shapes corresponds to a
 299 universal form when liquid interfaces are stretched beyond
 300 linearity [13].

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