

Leidenfrost hydrogels: Elastic surface-wave instability and sustained bouncing

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(Received 23 September 2025; accepted 19 May 2026; published 17 June 2026)

Leidenfrost hydrogel spheres exhibit interfacial oscillations that inject energy and drive sustained bouncing. Using a computational model, we identify these oscillations as an evaporation-driven instability of elastic surface waves. The resulting dynamics conform to a classical engine cycle [S. R. Waitukaitis *et al.*, *Nat. Phys.* **13**, 1095 (2017)], even in the absence of vapor trapping at the solid surface previously proposed as the injection mechanism.

DOI: [10.1103/xzg7-4gzy](https://doi.org/10.1103/xzg7-4gzy)

The original focus of research into the Leidenfrost effect was on the levitation of liquid drops above “hot” solids that are supported by an evaporation-driven cushion of vapor [1–6]. Subsequently, an array of other Leidenfrost configurations have been investigated, including “inverse” configurations for liquid drops [7,8] and stiff sublimating solids [9]. Our interest here is on recent experiments showing that soft solids, namely, hydrogel “drops,” also display Leidenfrost dynamics [10–14]. These centimetric drops are predominantly (~99%) water, but behave like incompressible elastic solids when subjected to external stress (Young’s modulus ~10 kPa [10,12]).

As with liquid drops, hydrogels can display a “floating regime” [11], which has been theoretically analyzed in Ref. [13] for a steady state in which elasticity replaces surface tension as the dominant force. The floating regime enables horizontal motion, mediated by the frictionless vapor cushion, from very small external forcing, suggesting applications in self-propulsion (using a ratcheted surface [1,15,16] or thermal gradient [17,18], for example). However, the most dramatic effect, discovered in Ref. [10], is that there is also a dynamic “bouncing regime,” in which hydrogel spheres perform sustained jumps from the hot solid while emitting a strange “screeching” noise. The bouncing regime has been observed from experiments in which the hydrogel is dropped very close to the hot solid [11], suggesting that, depending on parameters such as the substrate temperature, its initiation does not always require an impact stage. Although this phenomenon is similar to the trampolining behavior observed for liquid drops [6,19,20], the typical bounce height in the hydrogel case is larger (order of centimeters rather than millimeters).

Optical observations of the bouncing regime in Ref. [10] suggest that the screeching is generated by high-frequency oscillations (2–3 kHz) of the deformed underbelly of the drop. It is suggested that these oscillations are the result of the cyclical trapping and release of vapor by the hydrogel’s underbelly. Furthermore, the corresponding oscillations in the pressure and volume of the cushioning vapor differ in phase, meaning that each oscillation is a work cycle, enclosing a nonzero area on a pressure-volume diagram. Therefore, Leidenfrost hydrogel spheres act as heat engines, converting thermal energy from the substrate into mechanical bouncing energy. This behavior unlocks potential applications, such as in the harvesting of waste thermal energy from hot surfaces [14]. Aside from the simplified model in Ref. [10], thus far there have been no efforts to capture these phenomena within a detailed theoretical framework.

Here, we develop a computational modeling approach to understand the physical mechanisms underpinning the bouncing regime and discover the trigger for the floating to bouncing transition, which is shown to be the growth of standing waves on the drop-vapor interface.

Modeling. We consider a three-dimensional axisymmetric description of the drop coupled to a lubrication equation for the evaporation-driven vapor flow. This approach has successfully described liquid drop dynamics for isothermal collisions/impacts [22–24] and static/dynamic Leidenfrost states [4,25,26], as well as elastic drop dynamics for isothermal impacts [27] and (steady-state) floating regime behavior [13]. Model details for the elastic Leidenfrost drop, described as a neo-Hookean near-incompressible solid, may be found in the Supplemental Material [21]. Of particular note are the vapor lubrication equation and the resulting surface traction, which under the assumption of axisymmetry are

$$\frac{\partial h}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left[r \left(-\frac{h^3}{12\mu_v} \frac{\partial p_v}{\partial r} + u_s \frac{h}{2} \right) \right] - \frac{k_v \Delta T}{\rho_v L h} = 0, \quad (1a)$$

$$\boldsymbol{\tau}_v = \left[-p_v \mathbf{I} + \left(\frac{h}{2} \frac{\partial p_v}{\partial r} + \frac{\mu_v u_s}{h} \right) (\mathbf{e}_r \mathbf{e}_r + \mathbf{e}_z \mathbf{e}_z) \right] \cdot \mathbf{n}. \quad (1b)$$

In the above, $h(r, t)$ and $u_s(r, t)$ denote the surface height and radial velocity of the vapor at the interface, respectively,

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while $p_v(r, t)$, μ_v , and ρ_v denote the vapor pressure, viscosity, and density, respectively. The thermal parameters ΔT , k_v , and L are the superheat (i.e., the substrate-hydrogel temperature difference), the thermal conductivity of the hydrogel, and the latent heat of vaporization, respectively, and are associated with the crucial vapor flux into the film, which is generated by heat conduction across the vapor driving evaporation. The effect of evaporation on the hydrogel mass is assumed to be negligible over the duration of an impact, an assumption that is justified in the Supplemental Material [21]. The vector quantities $\boldsymbol{\tau}_v$, $\mathbf{n}$, and $\mathbf{e}_{r(z)}$ denote the surface traction, surface normal, and radial (vertical) unit vectors, respectively. $\mathbf{I}$ is the identity tensor. Note that this framework assumes the substrate to be sufficiently smooth that direct contact between the drop and the substrate is never made, even in the isothermal case. This is not representative of hydrogel impacts in general; brief contacts may be observed depending on the impact conditions [11]. However, our simulation results suggest that near-contact events are short-lived and highly localized to the “neck” region of the hydrogel underbelly. In the early stages of the transition from the floating regime to the bouncing regime, which is a key focus of this work, an absence of contact is expected as the Leidenfrost oscillations are not yet large enough for contact to occur. The inclusion of contact, which may require the consideration of additional physics such as van der Waals forces and gas kinetic effects, has been reserved for future work. We also assume that the drop is held at its boiling temperature throughout impact, a strong assumption but one that is consistent with previous approaches for liquid Leidenfrost drops [4].

In the elastohydrodynamic lubrication literature, it is common to represent the solid as a linearly elastic half-space, allowing for formulation in terms of the linear elastic Green’s function [13,28,29]. In Ref. [29], this approach was used to model impact dynamics, assuming the dynamics of the drop to be quasistationary. Such an approach would not be appropriate here as the elastic waves visible in Fig. 1(a) from our new computational model would not be captured.

The computational model utilizes an interface-tracking finite element method that enables high-accuracy resolution of surface and bulk effects, with adaptive time stepping and mesh refinement necessary to resolve the high-frequency oscillatory phenomena observed. This was achieved using the open-source finite element library oomph-lib [30], with the code developed provided open source in Ref. [31]. A full account of the numerical challenges encountered and developments required, as well as the benchmark tests, can be found in the Supplemental Material [21].

Simulations predicting the bouncing regime. Figure 1 shows the results of our impact simulation for a representative case of a sphere dropped from a height of one radius ($a = 7.5$ mm), with an initial velocity of zero and under standard gravity. For clarity, we note that the isothermal case in the context of this work is the scenario in which both the substrate and the hydrogel are at a temperature of 100°C . All simulation parameters, which are summarized in the Supplemental Material [21], are taken from the experiments presented in Ref. [10]. In consistency with the recent experimental discoveries, panels (a) and (b) show that the Leidenfrost effect generates oscillations, which are absent in the isothermal case, that emanate from the

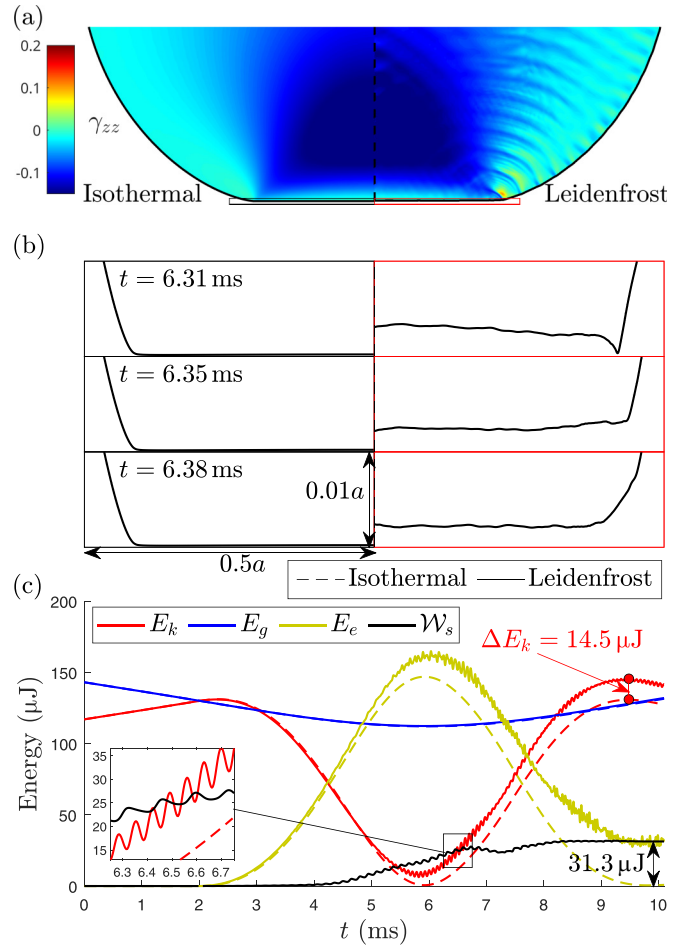


FIG. 1. Isothermal vs Leidenfrost dynamics. (a) Strain colormaps, for which bulk waves initiated at the solid-vapor interface are visible only in the Leidenfrost case. (b) Interface plots at three different times (over the course of roughly half an oscillation period in the Leidenfrost case). Note that there is a scaling factor of 50 between the x and y axes in these plots. (c) Plots of the kinetic, gravitational, and elastic energy as well as the cumulative work done by surface forces (denoted by E_k , E_g , E_e , and $\mathcal{W}_s$, respectively) against time ($t = 0$ is the time of release). The inset shows oscillations in the E_k and $\mathcal{W}_s$ dependencies. For all plots, the initial drop height was one sphere radius, with the remaining parameters taken from Ref. [10], as described in the Supplemental Material [21]. See Movies S1 and S2 in the Supplemental Material [21] for animations of the full impact.

surface and propagate into the bulk to engulf the entire drop. While the interfacial oscillations were found using high-speed imaging [10], the fascinating bulk waves that fill the drop are a fresh insight that would be difficult to observe experimentally.

Figure 1(c) shows how the kinetic, gravitational, and elastic energy, denoted by E_k , E_g , and E_e , respectively, as well as the work done by surface forces, $\mathcal{W}_s$, evolve over the impact. Most importantly, comparing the difference between profiles in the Leidenfrost and isothermal cases, we see how our model naturally predicts the energy injection discovered in Ref. [10]. In the simulation shown, of the total energy injected $\mathcal{W}_s = 31.3 \mu\text{J}$, the drop leaves the substrate with an additional kinetic energy of $\Delta E_k = 14.5 \mu\text{J}$ compared to the start of impact. This is of the same order of magnitude as that

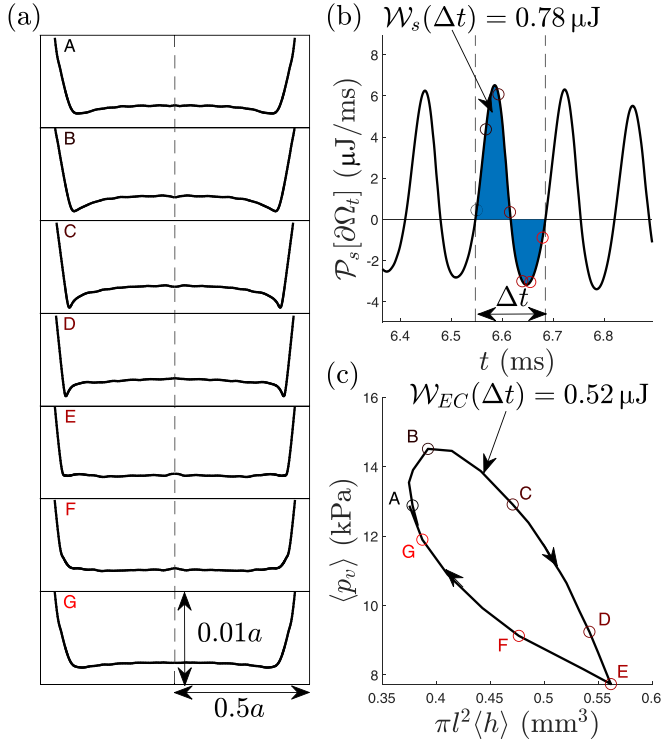


FIG. 2. (a) Interface profiles at times shown in panel (b). (b) The power due to surface forces, $\mathcal{P}_s[\partial\Omega_t]$, plotted against time during oscillations. The shaded area shown is $\mathcal{W}_s(\Delta t)$, the exact amount of energy injected over one oscillation period, Δt . (c) The area-averaged vapor pressure, $\langle p_v \rangle$, plotted against the volume of the vapor cushion, $\pi l^2 \langle h \rangle$. The area enclosed by the curve, $\mathcal{W}_{EC}(\Delta t)$, provides an estimate for the energy injection, assuming the mechanism to be analogous to a classical engine cycle.

seen in the experiment ($\Delta E_k \approx 38 \mu\text{J}$) and precise agreement is unlikely given “significant bounce-to-bounce fluctuations” observed there. Although the scope of our simulation is limited to a single bounce only, it is expected that over the course of multiple bounces the energy gain is balanced by dissipation due to damping forces, leading to sustained bouncing with a constant maximum bounce height, in line with experimental findings [10]. The mean oscillation frequency over the course of impact was approximately 7 kHz, within the same order of magnitude as the value quoted experimentally (2–3 kHz).

Leidenfrost engine. We now consider the hypothesis of Waitukaitis *et al.* [10] that the oscillations create a Leidenfrost engine. Figure 2(a) isolates one cycle of this behavior: In (A)–(C), the hydrogel spreads along the vapor cushion, forming a “neck” that partially traps a vapor pocket. According to the hypothesis in Ref. [10], this trapping leads to a monotonic increase in pressure until a substantial gap is opened for pressure release. Here, however, we observe a pressure decrease before the gap opens in (C)–(E). This may be due to the fact that direct contact between the hydrogel and the substrate is never made in our simulations, so the vapor pocket is never truly sealed. In (E)–(G), the solid recoils elastically, starting the cycle over again.

The “engine” can then be understood from thermodynamic arguments: First, we note that the energy injection over the

course of a single oscillation may be calculated exactly by integrating the surface power, $\mathcal{P}_s[\partial\Omega_t]$, through time:

$$\mathcal{W}_s(\Delta t) = \int_{\Delta t} \mathcal{P}_s[\partial\Omega_t] dt, \quad (1)$$

$$\mathcal{P}_s[\partial\Omega_t] = \int_{\partial\Omega_t} \boldsymbol{\tau}_v \cdot \mathbf{u}_s dA, \quad (2)$$

where Δt denotes the oscillation period and $\partial\Omega_t$ denotes the time-dependent hydrogel surface (the quantity $\mathcal{W}_s$ discussed earlier is the same as the one defined here, except that the integration is now over a single oscillation period). A second method of estimating energy injection is as follows: Plotting the mean pressure in the vapor against its volume over the course of one oscillation, as done in Fig. 2(c), we see that a closed loop is formed. The area enclosed by this loop is

$$\mathcal{W}_{EC}(\Delta t) = \oint \langle p_v \rangle dV, \quad (3)$$

Here, $\langle \cdot \rangle = (2/l^2) \int_0^l \cdot r dr$ denotes an average across the vapor cushion and the subscript stands for “engine cycle.” The quantity $\mathcal{W}_{EC}(\Delta t)$ provides an estimate of work done in analogy with a classical heat engine. These two metrics are equal if the pressure is spatially uniform across the vapor cushion, which is approximately the case even when Leidenfrost oscillations are present (see Movies S2 and S3 in the Supplemental Material [21]), and if shear stress contributions can be neglected. Agreement between $\mathcal{W}_s(\Delta t)$ and $\mathcal{W}_{EC}(\Delta t)$, then, serves as a confirmation that the energy injection mechanism seen here can be described as a classical engine cycle. As an example from our simulations, taken from a work cycle roughly midway through impact, we find $\mathcal{W}_s(\Delta t) = 0.781 \mu\text{J}$ and $\mathcal{W}_{EC}(\Delta t) = 0.521 \mu\text{J}$ [see Figs. 2(b) and 2(c) for reference]. Discrepancy between these values is likely due to averaging error, arising from the fact that $\langle p_v h \rangle \neq \langle p_v \rangle \langle h \rangle$ in general, as the vapor pressure profiles can sharply increase around the neck front at certain stages of an oscillation. Interestingly, the shape of the loop observed here is different from that suggested by Waitukaitis *et al.* [10]. In fact, work cycles similar in shape to Fig. 2(b) can also be seen for small-amplitude Leidenfrost oscillations, as we discuss below.

From floating to bouncing. Given the results above, it is natural to ask under which conditions we expect to see the Leidenfrost oscillations that produce sustained bouncing. To address this, we turn our attention to stationary states that are found from our simulations using an artificial damping force. We propose that the large oscillations seen in impact simulations ultimately arise due to an interfacial instability that is only present in the Leidenfrost case, a hypothesis that can be tested by removing the damping force once a steady state has been attained and monitoring the subsequent dynamics of the interface. Figure 3(a) shows which of the various stationary states, obtained by varying the superheat, ΔT , and the ratio of the neck radius and the initial drop radius, l/a , which is primarily varied by changing a itself (see the Supplemental Material [21] for more details), were found to be unstable. We see that increasing either of these quantities past a threshold (shown by the red line) leads to an instability. A similar trend is seen for impact simulations in Fig. 3(b), where ΔT is

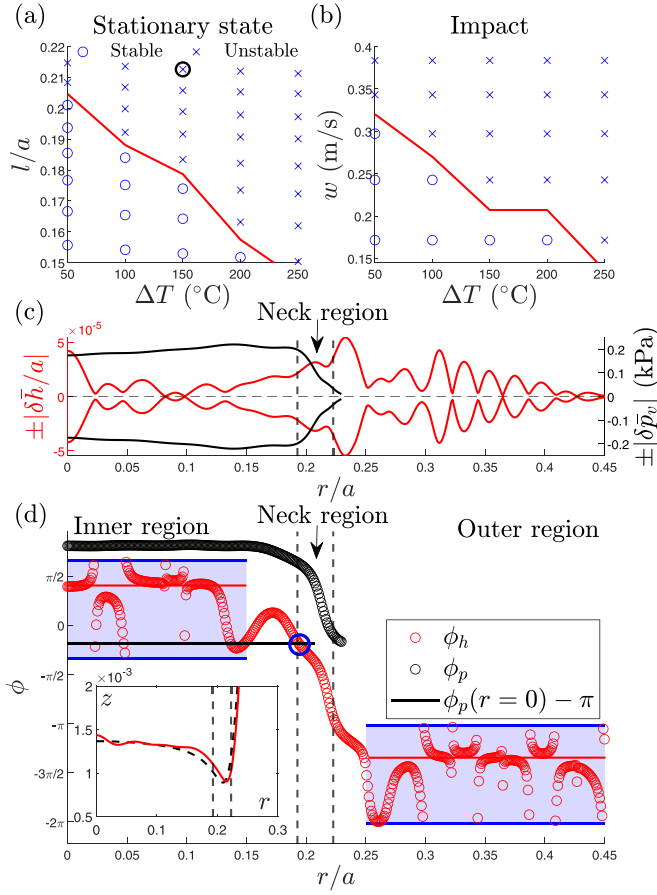


FIG. 3. (a), (b) Stability of Leidenfrost stationary states and impacts, determined through direct simulation. In each case, a red line is plotted to show the stable/unstable threshold. The black circle in panel (a) indicates the parameter set used in panels (c) and (d). (c) Plots of the perturbation amplitude for displacement and pressure, denoted by $|\delta\bar{h}|$ and $|\delta\bar{p}_v|$, respectively, as a function of r (negative amplitudes are also plotted to aid visualization). (d) The phase of the displacement (red) and pressure (black) oscillations, denoted by ϕ_h and ϕ_p , respectively. In this plot, ϕ_h is cast into the interval $[-\pi/2, \pi/2]$ for $r < 0.2$, so for standing waves a roughly constant phase can be seen despite the expected jumps by π at the nodes of the standing waves. The profile is then allowed to vary continuously for $0.2 < r < 0.25$ before being cast into the interval $[-\pi, 0]$ for $r > 0.25$ to allow for visualization of standing waves outside the neck. Red lines are plotted in the standing wave regions to guide the eye. The two dashed vertical lines, also plotted in the inset that shows the steady state and perturbed interface profiles, indicate the vicinity of the neck. The solid black line shows the average pressure phase over the region $0 < r < 0.15$.

instead plotted against the impact speed w , which has a positive correlation with the area of contact.

To investigate the unstable dynamics, we shall focus on the perturbed pressure and vertical displacement profiles, given by $\delta p_v = p_v - p_{v,s}$ and $\delta h = h - h_s$, respectively, where $h_s(r)$ and $p_{v,s}(r)$ are steady-state profiles. Information on the character of the oscillations can be obtained from a temporal Fourier transform of these perturbed quantities. Letting $\delta\bar{p}_v$ and $\delta\bar{h}$ denote the complex-valued coefficients of the dominant mode in this transform, we are interested

in the amplitudes $|\delta\bar{p}_v|$, $|\delta\bar{h}|$ and phases ϕ_p , ϕ_h that satisfy $|\delta\bar{p}_v| \exp(i\phi_p) = \delta\bar{p}_v$ and $|\delta\bar{h}| \exp(i\phi_h) = \delta\bar{h}$. These quantities are plotted as a function of the radial coordinate r in Figs. 3(c) and 3(d). As is clear from the amplitude plots in panel (c), $\delta\bar{h}$ exhibits “nodes” at which the oscillation amplitude falls near to zero. These nodes are located away from the neck region, which, in this case, is at $r = l \approx 0.21a$. Looking at the phase plots in panel (d), the phase is roughly constant in the sections between these nodes. Together, these observations imply the existence of standing waves on either side of the neck (these waves are also visible in Movie S3 of the Supplemental Material [21]). In the vicinity of the neck, nonzero amplitude and r -dependent phase imply a propagating wave initiated by interaction of the unstable mode with the neck.

A different picture is apparent for the pressure perturbation: for this, we see from Figs. 3(c) and 3(d) that both $|\delta\bar{p}_v|$ and ϕ_p are nearly constant in the “inner region” (between $r = 0$ and the first dashed vertical line). Most importantly, however, is that this constant phase is different from the displacement phase. This is because the pressure phase is mainly determined by the displacement phase at the neck, where we observe a phase difference of $\phi_p(r=0) - \phi_h \approx \pi$. This can be seen in Fig. 3(d) by following the black line, which corresponds to $\phi_p(r=0) - \pi$, to the intersection point in the “neck region” with the displacement phase profile. With the pressure phase in the inner region set by the displacement phase at the neck, there is a phase difference of $\phi_p - \phi_h \sim 0.4\pi$ in the inner region. This ultimately gives rise to a loop in a PV diagram and net positive work (see Movie S3 of the Supplemental Material [21]), much like what is observed for the large Leidenfrost oscillations in impact simulations. Understanding how the interaction of certain elastic surface modes with the neck causes this effect is likely central to understanding the origin of the instability and should be the subject of future work in this area. We note that, although Figs. 3(c) and 3(d) only consider a single parameter combination, similar dynamics were observed in all simulations that exhibited a floating to bouncing transition.

For small perturbations to the interface, such as the ones considered here, the vapor trapping effect proposed by Waitukaitis *et al.* [10] and briefly discussed above is not relevant. Thus, we see that, while vapor trapping may play an important role for large Leidenfrost oscillations, it is not necessary for the existence of work cycles. As energy is injected for each oscillation of the unstable mode, the system deviates further from the stationary state. We suspect that this continuous energy injection will eventually give rise to the spontaneous bouncing phenomenon observed by Pham *et al.* [12], which has a counterpart in the liquid Leidenfrost case [19].

Conclusions and future directions. The results in this work confirm the hypothesis of Waitukaitis *et al.* [10] that energy injection and thus sustained bouncing can be understood from the perspective of a classical engine cycle. Notably, however, we find that the proposed mechanism of energy injection, where the drop makes direct contact with the heated solid, sealing off a pocket of vapor, is not necessary. This is especially the case in the early stages of the instability, where work cycles are still observed for small-amplitude oscillations.

Since Eq. (1) makes the standard assumptions of incompressibility and negligible inertia for vapor [4,13,26,27], neither of these factors is key to observe the Leidenfrost instability. An obvious direction for future work would be to lift one or both of these assumptions to see how, if at all, the character of the Leidenfrost oscillations changes. Of particular interest would be the influence of vapor compressibility on the resulting PV diagram. We also hope to explore larger velocity impacts with the code developed here, which will likely require a suitable modification of the numerical scheme to account for the increasing complexity of the wave structures in the drop at the late stages of impact.

With a link now established between the large oscillations and the instability of standing wave perturbations to the stationary state, an analytical treatment of the drop's stability would also be desirable. Progress in this direction may be possible by drawing on the existing theory of surface waves in elastic solids [32], as well as the analytical techniques used to assess the stability of the air-liquid interface during liquid drop impacts [22,33].

Finally, we note that oscillatory dynamics and sustained bouncing are not unique to hydrogels; the behavior observed here bears a strong resemblance to the “trampolining” instability reported for Leidenfrost droplets [6,19,20], in which oscillations of the vapor pressure are coupled to the capillary waves on the drop surface. Our findings here reveal a similar feedback mechanism, but with elastic surface-wave modes replacing capillary ones. This difference in restoring force alters the amount of energy that can be stored and released over the course of a single oscillation, potentially explaining the higher bounce heights observed for hydrogel spheres (order of centimeters rather than millimeters). Given the parallel between liquid and hydrogel Leidenfrost bouncing, our findings here may also provide insight into related Leidenfrost configurations.

Acknowledgment. This work was supported by the Engineering and Physical Sciences Research Council (EPSRC) under Grants No. EP/W031426/1, No. EP/S029966/1, No. EP/P031684/1, and No. UKRI3193.

Data availability. The data that support the findings of this article are openly available [31], embargo periods may apply.

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