Can a liquid drop on a substrate be in equilibrium with saturated vapor?

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It is well known that liquid and saturated vapor, separated by a flat interface in an unbounded space, are in equilibrium. One would similarly expect a liquid drop, sitting on a flat substrate, to be in equilibrium with the vapor surrounding it. Yet, it is not: as shown in this work, the drop evaporates. Mathematically, this conclusion is deduced using the diffuse-interface model, but it is also reformulated in terms of the maximum-entropy principle, suggesting model independence. Physically, evaporation of drops is due to the so-called Kelvin effect, which gives rise to a liquid-to-vapor mass flux if the boundary of the liquid phase is convex.

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Introduction. The diffuse-interface model (DIM) was proposed by Korteweg in 1901 [1], and later developed by Ginzburg [2] and Cahn [3]. Since then, it has been used in numerous problems including nucleation and collapse of bubbles [4–7], phase separation in polymer blends [8,9], contact lines [10–17], etc.

It was also used in Ref. [18] to prove the nonexistence of solutions describing static two-dimensional (2D) drops on a substrate. This result, however, gives rise to numerous follow-up questions. If a drop cannot be static, how exactly does it evolve? Is it spreading out, while decreasing in thickness—or perhaps it acts as a center of condensation for the surrounding vapor and, thus, grows? In addition, the 2D proof of Ref. [18] was not applicable to the most interesting case, that of 3D axisymmetric drops. Does this mean that they can be static—or their nonexistence can still be proved using a different method?

In the present work, the approach of Ref. [18] is modified to prove the nonexistence of static 3D drops on a substrate. This result is then reformulated in terms of the maximum entropy principle and interpreted via the Kelvin effect [19–26], with both suggesting that drops *evaporate*. This conclusion agrees with, and explains, the evaporation of drops observed in numerical simulations [27].

Formulation. There are two different versions of the diffuse-interface model (DIM): one assuming the fluid velocity to be solenoidal [28,29] and another based on the full equations of compressible hydrodynamics [30,31]. For static solutions, however, the two models coincide.

Even though the results below are applicable to an arbitrary nonideal fluid, they are easier to present using the van der Waals equation of state (say, with parameters a and b). Introduce, also, the Korteweg constant K characterizing the intermolecular attractive force [1,28-31].

The following nondimensional variables (marked with the subscript $_{nd}$) will be used:

$$r_{nd} = \frac{r}{l}, \quad z_{nd} = \frac{z}{l}, \quad \rho_{nd} = b\rho, \quad T_{nd} = \frac{RTb}{a},$$

where r is the horizontal (polar) radius, z is the vertical (axial) coordinate, ρ is the density, T is the temperature, R is the specific gas constant, and $l = (K/a)^{1/2}$ is the characteristic interfacial thickness. Physically, l is on a nanometer scale and, thus, will be referred to as "microscopic."

According to the DIM, a static axisymmetric distribution of a van der Waals fluid satisfies (the subscript _{nd} omitted)

$$\frac{1}{r}\frac{\partial}{\partial r}\left(r\frac{\partial\rho}{\partial r}\right) + \frac{\partial^{2}\rho}{\partial z^{2}}$$

$$= T\left(\ln\frac{\rho}{1-\rho} + \frac{1}{1-\rho}\right) - 2\rho - \mu, \tag{1}$$

where μ is a constant. The physical meaning of this equation will be explained later.

As illustrated in Fig. 1, Eq. (1) requires four boundary conditions.

Let the drop be horizontally and vertically localized. The latter implies that far above the substrate, the density tends to a constant—say, ρ_{∞} . If ρ_{∞} exceeds the density ρ_{v} of saturated vapor, the setting under consideration becomes physically meaningless (because an infinitely large volume of *over*saturated vapor—with or without a liquid drop—is thermodynamically unstable). If, in turn, $\rho_{\infty} < \rho_{v}$, the problem becomes trivial, as evaporation of drops surrounded by *under*saturated vapor is evident without proof. Thus, assume

$$\rho \to \rho_v \quad \text{as} \quad z \to \infty.$$
 (2)

The saturated-vapor density ρ_v can only be defined together with the matching liquid density ρ_l (in what follows, the latter will not be involved). They are both determined by the Maxwell construction, comprising the requirements that the vapor's pressure and chemical potential match those

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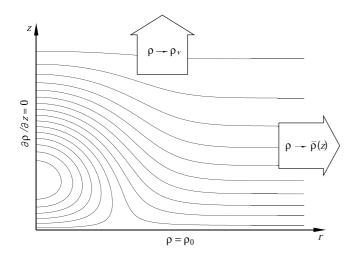


FIG. 1. A schematic illustrating the setup of boundary conditions for, and a typical solution of, Eq. (1).

of the liquid. For the van der Waals fluid, these requirements amount to

$$\frac{T\rho_{v}}{1-\rho_{v}} - \rho_{v}^{2} = \frac{T\rho_{l}}{1-\rho_{l}} - \rho_{l}^{2}, \tag{3}$$

$$T\left(\ln\frac{\rho_{v}}{1-\rho_{v}} + \frac{1}{1-\rho_{v}}\right) - 2\rho_{v}$$

$$= T\left(\ln\frac{\rho_{l}}{1-\rho_{l}} + \frac{1}{1-\rho_{l}}\right) - 2\rho_{l}. \tag{4}$$

It can be shown that if T < 8/27 (subcritical temperature for the van der Waals fluid), Eqs. (3) and (4) admit a unique solution such that $\rho_l > \rho_v$ and

$$\frac{T}{(1-\rho_v)^2\rho_v} > 2, \quad \frac{T}{(1-\rho_l)^2\rho_l} > 2.$$
 (5)

Conditions (5) guarantee that both liquid and vapor are stable (the pressure grows with density). For supercritical temperatures, only the trivial solution ($\rho_l = \rho_v$) exists, so interfaces do not.

Observe that boundary condition (2) and Eq. (1) entail

$$\mu = T \left(\ln \frac{\rho_v}{1 - \rho_v} + \frac{1}{1 - \rho_v} \right) - 2\rho_v. \tag{6}$$

Physically, μ is the specific chemical potential of the van der Waals vapor (or, to be precise, differs from that by a constant).

Let the fluid be bounded below by a substrate located at z = 0, in which case the DIM implies [31,32]

$$\rho \to \rho_0 \quad \text{as} \quad z \to 0,$$
 (7)

where ρ_0 is a constant characterizing the liquid-substrate interaction. In this Letter, the substrate is assumed to be neither perfectly hydrophilic nor perfectly hydrophobic, with the implication that $\rho_v < \rho_0 < \rho_l$ [31].

To clarify the physical meaning of condition (7), consider the intermolecular forces exerted on a fluid molecule in an infinitesimally thin layer near the substrate: the solid pulls the molecule toward the substrate, while the fluid outside the layer pulls it in the opposite direction. Since the former force is fixed, whereas the latter grows monotonically with the near-substrate density, the balance is achieved when the density assumes a certain value—which is what condition (7) reflects. Furthermore, the main conclusion of this Letter (evaporation of all sessile drops) would not change even if (7) were replaced by the Neumann or mixed boundary conditions (assumed phenomenologically and used, for example, in Refs. [3,33] and [8,9], respectively). From a physical viewpoint, evaporation occurs at the liquid-vapor interface, so the fluid-substrate interaction does not affect it much.

The fact that the drop is localized horizontally implies that at large r, the substrate is dry—i.e., the vapor and solid are adjacent to one another, with no liquid in between. In terms of the DIM, a solid-vapor interface corresponds to a microscopic boundary layer where the density field is homogeneous horizontally, but changes vertically from ρ_0 at the substrate toward ρ_v far above it. The profile $\bar{\rho}(z)$ of this layer satisfies the one-dimensional reduction of the boundary-value problem (1), (7), and (2),

$$\frac{d^2\bar{\rho}}{dz^2} = T\left(\ln\frac{\bar{\rho}}{1-\bar{\rho}} + \frac{1}{1-\bar{\rho}}\right) - 2\bar{\rho} - \mu,\tag{8}$$

$$\bar{\rho} = \rho_0 \quad \text{at} \quad z = 0, \tag{9}$$

$$\bar{\rho} \to \rho_v \quad \text{as} \quad z \to \infty.$$
 (10)

Thus, horizontal localization of the drop assumes that

$$\rho \to \bar{\rho}(z)$$
 as $r \to \infty$. (11)

To ensure that $\rho(r, z)$ is analytic at r = 0, require

$$\frac{\partial \rho}{\partial r} = 0$$
 at $r = 0$. (12)

Finally, let

$$\left| \int_0^\infty (\rho - \bar{\rho}) r dr \right| < \infty, \tag{13}$$

which implies that the excess mass between any two horizontal planes is finite, and so is the drop's net mass.

Properties of boundary-value problem (1)–(13). Mathematically, Eqs. (1)–(13) have a lot in common with their 2D counterparts examined in Ref. [18]. In what follows, the properties of the former will be briefly outlined with references to the latter.

(i) As $z \to \infty$, Eq. (1) can be linearized against the background of ρ_v and thus becomes a Helmholtz equation,

$$\frac{1}{r} \frac{\partial}{\partial r} \left[r \frac{\partial (\rho - \rho_v)}{\partial r} \right] + \frac{\partial^2 (\rho - \rho_v)}{\partial z^2} - \left[\frac{T}{(1 - \rho_v)^2 \rho_v} - 2 \right] (\rho - \rho_v) = 0.$$
(14)

By virtue of (5), the second expression in the square brackets is positive—which implies that all solutions of Eq. (14) are either exponentially decaying or exponentially growing as $z \to \infty$. The latter is ruled out by boundary condition (2)—hence,

$$(\rho - \rho_v)z^n \to 0$$
, $\frac{\partial \rho}{\partial z}z^n \to 0$ as $z \to \infty$, (15)

for all n.

The above argument can be reworked into a formal proof similar to that for 2D drops in Ref. [18]. One only needs to replace in the latter the Fourier transformation with the Hankel transformation.

(ii) As $r \to \infty$, Eq. (1) can be linearized against the background of $\bar{\rho}(z)$ and written in the form

$$\frac{1}{r}\frac{\partial}{\partial r}\left[r\frac{\partial(\rho-\bar{\rho})}{\partial r}\right] - \hat{A}(\rho-\bar{\rho}) = 0, \tag{16}$$

where the operator

$$\hat{A} = -\frac{\partial^2}{\partial z^2} + \left[\frac{T}{(1 - \bar{\rho})^2 \bar{\rho}} - 2 \right] \tag{17}$$

is self-adjoint. As before, (16) and (17) form a Helmholtz equation, but this time the expression in the square brackets in (17) can be negative for some z. As a result, the exponential decay of the solutions of Eq. (16) as $r \to \infty$ is not obvious, but still follows from the fact that the operator \hat{A} is positive definite (see Lemma 4 of Ref. [18]). Thus,

$$(\rho - \bar{\rho})r^n \to 0, \quad \frac{\partial \rho}{\partial r}r^n \to 0 \quad \text{as} \quad r \to \infty$$
 (18)

for all n.

(iii) Assume that $\varrho(z)$ is a smooth function such that $\varrho(0) = \rho_0$ and $\varrho(z) \to \rho_v$ as $z \to \infty$, and introduce the following functional:

$$\begin{split} F[\varrho(z)] &= \int_0^\infty \left[\frac{1}{2} \left(\frac{d\varrho}{dz} \right)^2 \right. \\ &+ \left. T\varrho \ln \frac{\varrho}{1 - \varrho} - \varrho^2 - \mu\varrho + p \right] \! dz, \end{split}$$

where μ is given by (6) and

$$p = \frac{T\rho_v}{1 - \rho_v} - \rho_v^2$$

is, physically, the pressure of saturated van der Waals vapor.

As shown in Ref. [18], F reaches the absolute minimum when $\varrho = \bar{\rho}(z)$. Thus, if (1)–(13) admit a nontrivial ($\rho \neq \bar{\rho}$) solution, it satisfies

$$F[\rho(r,z)] > F[\bar{\rho}(z)] \quad \forall r \in (0,\infty). \tag{19}$$

Note that the integrals involved in $F[\bar{\rho}(z)]$ converge due to (15).

Nonexistence of static drops. The nonexistence of solutions of the boundary-value problem (1)–(13) will be proved by contradiction.

Assuming that a solution exists, multiply Eq. (1) by $r^2 \partial \rho / \partial r$ and integrate from z = 0 to $z = \infty$. By integrating by parts and using conditions (15) to interchange differentiation with respect to r and integration with respect to z, one obtains

$$r^{2} \frac{\partial F[\rho(r,z)]}{\partial r} - \frac{1}{2} \frac{\partial}{\partial r} \int_{0}^{\infty} \left(r \frac{\partial \rho}{\partial r} \right)^{2} dz = 0.$$

Integrate this equality with respect to r from r = 0 to $r = \infty$ and, replacing in the first term

$$\frac{\partial F[\rho(r,z)]}{\partial r} \to \frac{\partial (F[\rho(r,z)] - F[\bar{\rho}(z)])}{\partial r},$$

integrate by parts. Recalling conditions (18), one obtains

$$\int_0^\infty r\{F[\rho(r,z)] - F[\bar{\rho}(z)]\}dr = 0.$$

Given (19), this last equality is incorrect—hence, the contradiction.

Thus, the only existing steady-state solution is the trivial one (describing dry substrate).

The maximum entropy principle. The steady-state equations (1)–(13) can be reformulated as a problem of maximization of the net entropy subject to the net energy and mass being fixed.

To this end, introduce the specific (per unit mass) entropy $s(\rho, T)$ and the specific internal energy $e(\rho, T)$. They are not entirely arbitrary, as they are supposed to satisfy the fundamental thermodynamic relation,

$$\frac{\partial e}{\partial T} = T \frac{\partial s}{\partial T}.$$
 (20)

Introduce also the fluid's chemical potential,

$$G = \frac{\partial}{\partial \rho} [\rho(e - Ts)]. \tag{21}$$

For the van der Waals fluid, for example, one has (nondimensionally)

$$e = c_V T - \rho$$
, $s = c_V \ln T - \ln \frac{\rho}{1 - \rho}$,

$$G = T \left(\ln \frac{\rho}{1 - \rho} + \frac{1}{1 - \rho} \right) - 2\rho + c_V (1 - \ln T),$$

where c_V is the nondimensional heat capacity at constant volume.

The van der Waals equation (1) can now be extended to the general case, in the form

$$\nabla^2 \rho = G - \mu',\tag{22}$$

where ∇^2 is the axisymmetric Laplace operator and μ' is related to its van der Waals counterpart by $\mu' = \mu + c_V(1 - \ln T)$.

Next, introduce the net excess mass M, the net excess entropy S, and the net excess full energy E,

$$M = \int (\rho - \bar{\rho})dV, \quad S = \int (\rho s - \bar{\rho}\bar{s})dV,$$

$$E = \int \left\{ \rho e - \bar{\rho}\bar{e} + \frac{1}{2} \left[\left(\frac{\partial \rho}{\partial r} \right)^2 + \left(\frac{\partial \rho}{\partial z} \right)^2 \right] \right\} dV, \quad (23)$$

where $\bar{s} = s(\bar{\rho}, T)$, $\bar{e} = e(\bar{\rho}, T)$, $dV = 2\pi r dr dz$, and the integrals are evaluated over the semispace z > 0. The derivative terms in expression (23) represent the energy of the intermolecular attraction as the DIM describes it.

All of the results obtained earlier for the van der Waals equation (1) can be readily reproduced for the general equation (22). Even more importantly, the latter can be used to find out the physical meaning of the nonexistence of solutions describing sessile drops.

Observe that Eq. (22) follows from the requirement that the net entropy be maximum subject to the net mass and energy

be fixed, i.e.,

$$\delta(S + \eta M + \lambda E) = 0,$$

where η and λ are the Lagrange multipliers. Carrying out variation in the above equation and recalling boundary conditions (2) and (7), which imply

$$\delta \rho = 0$$
 at $z = 0$,

$$\delta \rho \to 0$$
 as $z \to \infty$.

one obtains

$$\begin{split} \int \left[\frac{\partial (\rho s)}{\partial \rho} + \lambda \frac{\partial (\rho e)}{\partial \rho} - \lambda \nabla^2 \rho + \eta \right] \! \delta \rho dV \\ + \int \left[\frac{\partial (\rho s)}{\partial T} + \lambda \frac{\partial (\rho e)}{\partial T} \right] \! \delta T dV = 0. \end{split}$$

Setting $\lambda = -1/T$, one can make the second term in this equation vanish subject to identity (20), whereas $\eta = \mu'/T$ makes the first term vanish subject to condition (21) and the steady-state equation (22).

Thus, since $\bar{\rho}(z)$ (describing the solid-vapor interface) is the only solution of Eq. (22), it corresponds to the maximum entropy and all other solutions evolve towards it. This means evaporation of the liquid phase, with the excess mass and energy spreading out to infinity.

The only DIM-specific part of the above variational problem is the intermolecular part of energy (23). It seems unlikely that another form of this term would fundamentally change the properties of the functionals involved. Hence, one could conjecture that drops on a solid substrate evaporate in *any* model conserving mass and energy, and conforming to an H-Theorem—such as, for example, the Enskog-Vlasov kinetic equation for dense fluids [34–39] (which is generally viewed as a much more accurate model than the DIM).

Physical interpretation. The nonexistence of drops on a substrate can be explained through the so-called Kelvin effect [19–23,25,26], which gives rise to a mass flux through a liquid-vapor interface provided it is curved. The direction of the flux depends on the sign of the interfacial curvature: for a volume of liquid with a convex boundary, the flux is directed from the liquid toward the vapor, and vice versa. Since the boundary of a drop on a flat substrate is convex, it comes as no surprise that it evaporates. One can further conjecture that drops floating in saturated vapor evaporate too [40]. As shown in Ref. [41], the only kind of drops that do not evaporate are those in a sufficiently acute corner, so that their surface is concave—hence, vapor is condensating on it.

Finally, the quantitative theory of the Kelvin effect can be used to show that depending on the drop size and temperature, the timescale of evaporation ranges from several seconds to several days. This estimate (to be published separately) suggests that the evaporation of drops into saturated vapor can be observed experimentally and it should certainly be observable numerically via molecular dynamics.

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