THE ORIGIN OF SURFACE TENSION

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ABSTRACT
In passing from the molecular description of matter to the continuum scale, many material properties and physical quantities emerge that do not exist at the molecular scale. They account for the way we observe lumped effects of molecular properties. So, they are linked to molecular properties and molecular constitution of materials. One such continuum property is surface tension and/or interfacial tension, a property we observe at the interface between two immiscible phases at continuum scale. How surface tension is related to molecular properties and the molecular description of materials is important. Unfortunately, the explanations provided in much of the literature are wrong and/or incomplete. Often, it is linked to the forces of cohesion between molecules of a liquid, which is only one of the intermolecular forces in a fluid; a force which is commonly almost negligible within a fluid. Also, it is said to be due to the “tendency of liquid surfaces at rest to shrink into the minimum surface area” (6), which is not really a physical principle.

In this treatise, a rigorous explanation of the origin of surface tension is provided, based on intermolecular forces and the concept of upscaling from the molecular to the continuum scale. A full account of these intermolecular forces is given, along with an explanation of how these forces differ for molecules inside a liquid compared to those on its surface. It is explained that there exists a transition region with a finite thickness at the molecular scale, which is replaced by a sharp surface of discontinuity in material properties at the continuum scale. It is demonstrated that while the state of stress inside a liquid is compressive and isotropic, it becomes anisotropic in the interfacial region. Additionally, it is noted that while there is a compressive force in the direction normal to the interface, a less compressive or even tensile force exists in the tangential direction. It is this pressure deficit that is experienced as surface tension.

KEYWORDS
Surface tension, Interfacial tension, From the molecular to continuum scale, Intermolecular forces

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1. INTRODUCTION

Interfacial tension is one of the most important properties controlling the flow of two or more immiscible phases in a porous medium. It is a continuum-scale property that gives the state of stress, and is defined at a sharp interface between two immiscible phases. Surface tension is the interfacial tension between a solid or liquid phase and its own vapor. Therefore, the surface tension is defined at the surface of a phase (solid or liquid), whereas the interfacial tension is defined at the sharp interface between two liquids or a solid and a liquid. While surface tension is an intrinsic property of a phase, interfacial tension is an intrinsic property of two phases. In general, the interfacial tension between a phase and a gas is almost the same as the surface tension of the phase because the presence of another gas in its vapor phase does not affect its surface tension. In this paper, the discussion will be limited to surface tension for a liquid, and only at the end will interfacial tension be discussed.

Surface tension is a concept that must be introduced when passing from molecular scale to the continuum scale. It accounts for the lumped effect of subscale information that gets lost as a result of upscaling. Like any other property, it originates from the molecular interactions of the two fluids (liquid and its own vapor or a gas). Thus, one should be able to explain the concept of surface tension in terms of molecular properties. Such a link to the molecular interactions is indeed provided in the literature when explaining surface tension. This is found in the popular literature, such as Wikipedia, as well as the specialized or more technical literature, such as lectures notes and/or scientific documents posted on websites of universities or research institutes. However, the explanations provided in the popular literature and a large portion of the technical literature are either incorrect or partially incorrect. For example, consider the Wikipedia page on “Surface Tension” (6), where it is stated that “Surface tension is the tendency of liquid surfaces at rest to shrink into the minimum surface area possible.” This is not correct. What is correct is that all systems have the tendency of attaining a minimum energy state under equilibrium conditions. The liquid surface is part of a larger system and does not always attain the minimum area. For example, the surface of an open body of water is flat, whereas when a capillary tube is inserted, the air-water surface becomes curved inside the capillary tube; that is, the air-water surface area increases. This is because two other interfaces with different properties are involved (namely, air-solid interface and water-solid interface), and all three interfaces will have to go to a lower energy state. This was illustrated in the experiments conducted by Aslannejad et al. (1), and is discussed there in detail. Another example is that if we place a droplet of water, which has a finite surface area, on a perfectly hydrophilic surface, it will spread as thinly as possible, and a very large air-water surface is created. Again, this occurs because the entire system tends to go to a lower energy state. In any case, the concept of surface tension is unrelated to the tendency of liquid surfaces to attain a minimum area, a tendency that does not exist in general. Furthermore, one finds the following statement on the aforementioned Wikipedia page (6): “There are two primary mechanisms in play. One is an inward force on the surface molecules causing the liquid to contract. Second is a tangential force parallel to the surface of the liquid.” In support of this statement, there is the drawing shown in Figure 1, which originates from this Wikipedia page (6). This figure, showing the intermolecular forces for molecules of a liquid, is flawed for three reasons. First, the interface is shown as a sharp monomolecular surface, whereas the interfacial region has a finite (albeit very small) thickness; it is a three-dimensional region at the molecular level, as explained later. Second, we know that the result of all forces acting on any object (in this case, molecules of a liquid) must be zero at equilibrium. Although this holds true for the two molecules

![Figure 1: Diagram showing the forces acting upon water molecules inside a liquid and located on its surface (6).](https://via.placeholder.com/150)
shown inside the liquid, it is clearly not the case for the two molecules lying on the liquid surface. Based on this figure, these two molecules must fly inward, as there is a net force acting on each of them, and the water surface should collapse. We know that this does not occur at the equilibrium. The third flaw in this drawing is that the state of the stress within the liquid is tensile. However, it is known that attraction forces among molecules of a liquid are typically negligible, and the state of stress within fluids is compressive. Similar explanations and drawings are found on many other websites and even in lecture notes, books, and scientific documents posted on websites of universities or research institutes; many examples can be found via Google search page for “molecular surface tension.” What is missing in these explanations is a full picture of the intermolecular forces within fluids and a proper interpretation of the concepts of interface and surface tension. Moreover, this inaccurate and incomplete model of molecular interactions cannot explain many observations related to surface tension, such as why surface tension decreases with increasing temperature.

In this treatise, a rigorous explanation of the origins of surface tension based on intermolecular forces and the concept of upscaling from molecular to the continuum scale will be provided. To this end, the necessary background will be provided by starting at the molecular level and by explaining the concept of a sharp interface. Also, the concept of excess properties associated with such a sharp interface will be introduced. Next, a full account of the intermolecular forces within fluids and a proper interpretation of the concepts of interface and surface tension. Moreover, this inaccurate and incomplete model of molecular interactions cannot explain many observations related to surface tension, such as why surface tension decreases with increasing temperature.

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2. THE NATURE OF AN INTERFACE AT MOLECULAR AND CONTINUUM SCALES

The behavior of materials is entirely determined by their molecular properties and interactions. For example, water and alcohol behave differently because of the differences in the properties of and interactions among their molecules. Ice, liquid water, and water vapor behave differently solely because of the difference in the spacing of H₂O molecules and the corresponding interactions among them. Therefore, the identification of what constitutes an interface must be based on its molecular makeup and spacing. In fact, what one observes and models as a sharp interface between two bulk domains is a manifestation of their molecular makeup as well as upscaling from the molecular scale to the continuum scale. In the classical treatment of thermodynamics of interfaces, Gibbs (3) considers an interface to be a thin three-dimensional region at the molecular scale, and employs a “dividing surface” of discontinuity.
Consider two immiscible phases $a$ and $b$ filling a closed space (this molecular picture is based on the presentation by Murdoch & Hassanizadeh [4]). For the sake of this discussion, it is assumed that the solubility of each phase in the other phase is zero, and that there are no chemical reactions. The total number of molecules in each phase filling the space is time independent. Thus, the space is filled by the total material system $\mathcal{M}$ (i.e., the total collection of molecules considered), which consists of fixed sets of $a$ and $b$ molecules, denoted by $\mathcal{M}_a$ and $\mathcal{M}_b$, respectively. Because the two phases are immiscible, they fill mutually exclusive parts of the space, except for a narrow transition zone where both molecules may be found. The domains filled by the two bulk phases and the transition region at the molecular scale are shown in Figure 2.

As mentioned above, the properties of a bulk phase (on the continuum scale) are determined by the makeup of its molecules. The corollary to this statement is that for a pure phase, a molecule belongs to that phase if, and only if, it “feels” and interacts with the molecules of that phase; otherwise, it will exhibit a different behavior than that phase. Therefore, in Figure 2, any phase-$a$ molecule that comes across and interacts with phase-$b$ molecules does not belong to phase $a$ or to phase $b$; it belongs to the transition region, to which we also refer as the interfacial region. For molecules of any material, an interaction distance $\delta$ can be identified beyond which the molecular interactions are negligible. We denote the sets of molecules of the $a$ and $b$ phase that interact only with their own type by $\mathcal{M}_a^a$ and $\mathcal{M}_b^b$, respectively. These are disjoint systems. In other words, $\mathcal{M}_a^a(\tau)$ consists of $a$-molecules that, within a distance $\delta^a$ of each of them, only $a$-molecules can be found. $\mathcal{M}_b^b$ is defined similarly (see Fig. 2).

In addition to the zones filled by bulk-phase molecules, we can identify a transition zone where two other sets of molecules are present.

- $\mathcal{M}_b^a$ is the set of $a$-molecules that within a distance $\delta^a$ of each of them, at least one $b$ molecule is to be found.
- $\mathcal{M}_a^b$ is similarly defined (with the roles of $a$ and $b$ molecules interchanged).

Clearly, these two sets of molecules together form the interfacial region with a thickness of $\delta^{ab} = \delta^a + \delta^b$, which we denote by $\mathcal{J}^{ab}(\tau)$, also shown in Figure 2. Therefore, we have:

$$\mathcal{J}^{ab}(\tau) = \mathcal{M}_a^a(\tau) \cup \mathcal{M}_b^b(\tau)$$

When performing molecular simulations, all regions are modelled as three-
dimensional domains. However, when one studies such a system at the continuum level, the thickness of interfacial region is far too small for it to be modelled as a three-dimensional domain, and must be treated as a two-dimensional sharp surface with its own continuum properties. The question is where (in the molecular domain) such a two-dimensional surface should be located, and how its properties should be defined. First, one should define the properties of the bulk phases. To do so, one needs to define a representative elementary volume (rev), and a representative averaging time period over which average quantities should be defined. The rev should have a size much larger than molecular dimensions and much smaller than dimensions of the domain of interest. In general, its size will be different for different phases. Here, let us assume the rev to be a sphere of radius $d$ for both phases. One then assigns such a sphere to every point within the bulk phase, and defines the average quantities. For a proper definition of average properties of a phase, such a sphere should be placed at spatial points such that it will contain molecules of that phase only. This means that its center cannot be placed closer than a distance $d$ from the boundaries of the domain occupied by the phase or from the interface region. Therefore, the macroscopic properties of the bulk phases can be defined for domains that are smaller than the molecular domains, as shown in Figure 2. These domains are shown in solid colors in Figure 3. Evidently, there is a gap between the two macroscopic domains, whose width $\delta$ is equal to $2d + \delta_{ab}$. This domain cannot be ignored when modelling this system at the continuum level, not so much because of its size and dimension, but mainly because it has a different behavior than bulk domains. Therefore, at the continuum scale, the two phases are assumed to fill the gap and border each other at a sharp discontinuity surface, as shown in Figure 4. This sharp interface has macroscopic properties that are distinctly different from those of bulk phases.

3. INTERFACIAL EXCESS PROPERTIES

The question is, which average properties should be assigned to this continuum-scale interface? The guiding principle is the conservation of mass, momentum, and energy for the entire system. Let us consider the conservation of mass under the equilibrium conditions. As explained above, to arrive at the continuum picture, the various thermodynamic quantities were averaged over a rev. Therefore, for example, the average mass density of each phase at a given spatial point is equal to the mass of all molecules of that phase found within a rev that is assigned to that point divided by the volume of rev. Such an averaging operation under equilibrium conditions will result in a mass density variation similar to that shown in Figure 5, where phase $a$ is assumed to be much lighter than phase $b$. As can be seen, the mass density varies steeply over the transition region. Such a variation over such a small distance (of molecular dimensions) cannot possibly be modelled in continuum-scale studies. For homogeneous phases, a constant density on the continuum scale with a step change at the interface is expected. That is, the mass densities $\rho^a$ and $\rho^b$ are assigned to the entire continuum space filled by the two phases. This means that for the situation shown in Figure 5, compared to the real mass density curve, one assigns too much mass to the phase $b$ domain (on the right-hand side of the interface) and too little mass to the phase $a$ domain (on the left-hand side of the interface). The exact mass imbalance is equal to Equation 1 where $l^a$ and $l^b$ denote parts of the interfacial region assigned to the continuum phases $a$ and $b$, respectively.

$$\int_{\delta \text{int}} \rho dl - \rho^a l^a - \rho^b l^b = \Gamma^{ab}$$

(1)
Clearly, $\delta_{\text{int}} = l^a + l^b = 2d + \delta^{ab}$. In writing Equation 1, it is assumed that the system is homogeneous in the direction normal to the screen. Note that the position of the sharp surface is chosen arbitrarily. Depending on the chosen position of the sharp surface, this mass imbalance $\Gamma^{ab}$ can be positive, negative, or zero. In any case, to restore mass conservation for the whole system, this mass imbalance is assigned to the interface. This concept was introduced by Gibbs (3). He referred to this mass imbalance $\Gamma^{\text{int}}$ as the *excess mass*, which can be negative, positive, or zero, as we just explained. Note that the interfacial excess mass, $\Gamma^{\text{int}}$, has the dimension of mass per unit area. In cases that a surfactant is present in one of the bulk phases, it will accumulate in the interfacial region, and the excess mass of the interface is always positive. Similarly, the excess momentum and excess energy must be defined and assigned to the interface. This leads to the concepts of surface tension and interfacial tension, as explained in the following section.

### 4. STATE OF STRESS WITHIN A FLUID AND AN INTERFACE

#### 4.1. Molecular nature of pressure within a fluid

As explained in the last section, molecular interactions determine the observed or measured properties of a material. Take the state of stress in a fluid. Under a vast range of no-flow conditions, one experiences an isotropic compressive stress within the largest majority of fluids, whose magnitude is called *pressure*. This is the continuum-resultant of forces that molecules exert upon each other, to which one refers to as the momentum exchange. Molecules exchange momentum with each other in two different ways: via attraction/repulsion forces and via impulse (i.e., hitting each other). The attraction/repulsion forces are a function of the molecular separation distance $r$. Assuming molecules to be approximately spherical, and if they are apolar or weakly polar, attraction/repulsion forces can be quantified by the Lennard-Jones potential (cf. Chapter 2 in [2]), where $\varepsilon$ is the depth of the “potential well” (minimum value of potential function) and $\delta$ is the separation distance at which total potential $\phi$ is zero (representative of the molecular diameter) (Eq. 2).

$$\phi = 4\varepsilon \left[ \left( \frac{\delta}{r} \right)^{12} - \left( \frac{\delta}{r} \right)^6 \right] \quad (2)$$

The corresponding intermolecular attraction/repulsion force is equal to the derivative of the potential function with respect to $r$; as shown in Equation 3.

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**Figure 6:** Variation of intermolecular potential and intermolecular force for carbon tetrachloride (due to Berg [2]). The typical separation distance of molecules in liquids and gases are indicated.
\[ F_{a/r} = \frac{d\phi}{dr} = \frac{24\varepsilon}{\delta} \left[ 2 \left( \frac{\delta}{r} \right)^7 - \left( \frac{\delta}{r} \right)^{13} \right] \]  

(3)

In Figure 6, both \( \phi \) and \( F_{a/r} \) are plotted for carbon tetrachloride (for which \( \delta = 5.881 \text{ Å} \), and \( \varepsilon = 4.514 \times 10^{-14} \text{ erg} \)), under normal conditions. Similar plots are found for most liquids and gases. It is evident that the net attraction/repulsion forces are negligible for gases and a negligibly small positive force for liquids under equilibrium conditions. So, why does one experience pressure in fluids and gases? This is due to the exchange of momentum between the molecules as they continuously hit each other at high velocities.

To elucidate these interactions, let us consider a small cylinder containing a thin gas. The cylinder is closed by means of a frictionless piston (see Fig. 7), which needs to be held in place by the normal force \( F_n \) in order to counteract forces of molecules hitting it.

Each molecule that hits the piston and returns exerts a net force on the piston, as given by the first law of Newton (Eq. 4) where \( m^i \) is the mass of molecule and \( \Delta v_n^i \) denotes the change in velocity of molecule in the direction normal to the piston surface.

\[ F_n^i = m^i \frac{dv_n^i}{dt} \approx m^i \frac{\Delta v_n^i}{\Delta t} \]  

(4)

This velocity goes from \( v_n^i \) to 0 and then back to \(-v_n^i\); this is a velocity change of \( \Delta v_n^i = -2v_n^i \). Each molecule must traverse the length of the cylinder, \( L \), back and forth, before it can hit the piston again; the time needed for this is \( \Delta t = 2L/v_n^i \). Therefore, according to Equation 4, the force exerted by one molecule on the piston is equal to \( F_n^i = -m^i(v_n^i)^2/L \). If there are \( N \) molecules in the cylinder, one needs to sum over all molecules in order to obtain the total force \( F_n \) (Eq. 5), where one assumes that all molecules have the same velocity, and \( \rho = \sum_{i=1}^{N} m^i/AL \) denotes the molecular mass density of the gas, with \( A \) being the cross-sectional area of the cylinder.

\[ F_n = -\sum_{i=1}^{N} \frac{m^i}{L}(v_n^i)^2 = -\rho Av_n^2 \]  

(5)

If instead of the piston, one considers a virtual plane in an arbitrary direction within the cylinder, it can be shown that exactly the same net force, as delineated above, will be exchanged among the molecules that meet each other on that plane. Of course, the molecules are also exerting attraction/repulsion forces on each other as given by Equation 3. Thus, the net force exchanged between molecules across a surface is equal to the algebraic sum of these two forces, \( F_{a/r} + F_n \). However, as explained earlier, the attraction and repulsion forces are negligible for the fluids. Thus, the dominant state of stress within a fluid is generally compressive, to which one refers to as pressure, denoted by \( p \) (Eq. 6):

\[ (F_{a/r} + F_n)/A = (F_{a/r} - \rho v_n^2)/A \approx -\rho v_n^2 = -p \]  

(6)

It is clear that the fluid pressure is related to the kinetic energy of the molecules per unit volume, also referred to as the thermal agitation energy.

The foregoing explanation explains why most fluids exhibit negligible tensile strength under normal conditions. This is not the case for liquids that exhibit strong intermolecular interactions such as ionic bonds, metallic bonds, or hydrogen bonds on top of van der Waals interactions. For those fluids, the effect of attractive intermolecular forces \( (F_{a/r}) \) can be significant and must be taken into account. That is why some liquids, such as molten salt or liquid metals exhibit a finite tensile strength.

In the case of flowing fluids (with the piston in Figure 7 moving with average flow velocity), Equation 4 has to be modified, and the velocity of each molecule must be replaced by its velocity relative to the flow velocity.
The foregoing exposition of intermolecular forces also reveals that any action that leads to a decrease in the molecular separation distance or an increase in the molecular velocity causes an increase in pressure. This explains why most of the liquids are highly incompressible. If one attempts to compress a fluid, the intermolecular separation distance must decrease. However, this leads to very large negative repulsion forces, as shown in Figure 6b (i.e., \( F_{a/r} \) becomes negative and significant), which contributes to the negative state of stress on top of the thermal agitation component. Thus, very high pressures are required to compress a liquid. Increasing the temperature of a fluid at constant volume will cause the pressure to rise; this is because the velocity of molecules increases, which means that, according to Equation 5, \( F_n \) increases and leads to a larger pressure.

4.2. Molecular nature of pressure within an interface

When one moves away from inside a fluid and approaches its boundary, sharp changes in intermolecular forces appear, as discussed next. Consider a pure liquid in a container in contact with a vacuum. Obviously, the liquid will evaporate, and its vapor will fill up the vacuum. There are two major differences in the molecular features of the liquid and its vapor. Molecules of the vapor move much faster, and the distance between them is much larger (Fig. 8). This means that in Equation 5, while the mass density of the vapor is much smaller than that of the liquid, the square of the velocity of the molecules is much larger. These two effects balance each other such that in the absence of external forces, the vapor pressure and liquid pressure are equal. In fact, under equilibrium conditions, because there is no flow,
the pressure must be constant in each direction. Owing to the symmetry conditions inside the bulk phase, the pressure is the same in all directions, i.e. the stress tensor is fully isotropic.

However, in the interfacial region, we do not have full symmetry because the molecular makeup is different in the normal and tangential directions, as schematically shown in Figure 8a. Moreover, as we move away from the bulk liquid, the molecular velocities and, more importantly, the molecular separation distance increase. Therefore, different forces act in these two directions and the resulting stress tensor is anisotropic.

An increase in the molecular separation distance indicates that the attraction force among molecules increases drastically, as shown in Figure 6. This force reaches a maximum as one moves away from the bulk liquid phase, and then decreases to zero as one approaches the bulk vapor phase. This means that a large attractive force exists between molecules in the interfacial region, and thus the pressure in the interfacial region, given by \( F_{\text{attr}} \), is much smaller than the bulk pressure of phases. It can even be a positive force, but not necessarily. The variation of pressure in the interfacial zone is shown in Figure 8b, decreasing to a negative pressure (i.e., tension).

As explained earlier, at the continuum scale, the interfacial region is experienced as a sharp surface of discontinuity (see Fig. 8c) and the pressure is found everywhere to be equal to the bulk pressure, as shown in Figure 8d. However, this means that the conservation of momentum is violated when passing from the molecular picture to the continuum description. As this never happens in nature, we experience a positive force at the sharp surface of discontinuity, as shown by \( \sigma \) in Figure 8d, which we refer to as surface tension. The magnitude of this tension must be such that the resultant of lateral forces acting on a vertical plane AB will be the same at both the molecular and continuum scales. Thus, \( \sigma \) can be found from Equation 7:

\[
\sigma = \int_{\text{int}} (p - p_{\text{bulk}}) dl
\]

Therefore, the surface tension is a pressure deficit that exists within the interfacial region. Note that the dimension of surface tension is force per unit length.

The sharp interface is also called the “surface of tension.” For a flat interface, the value of the lateral pressure deficit (i.e., surface tension \( \sigma \)) is independent of the location of surface tension in the transition zone. This is also approximately true for a curved interface provided that its mean radius of curvature is large relative to the thickness of the transition zone. Otherwise, the surface tension can be a function of the curvature (5). Because the typical transition zone has a thickness on the order of only a few angstroms, the dependence on curvature will be negligible, except for interfaces formed within nano-sized pores.

If the domain filled by the vapor phase contains air or other gases, the description of the intermolecular interactions given above still holds. Even the value of the surface tension of the liquid is not expected to be significantly affected. However, if one of the bulk phases is another liquid, then the value of the pressure deficit \( \sigma \) will be different, and we refer to it as the interfacial tension, instead of the surface tension. Interfacial tension is a property of two liquids.

The foregoing exposition can help explain many phenomena related to surface tension in terms of the type and strength of the intermolecular forces that prevail within various fluids. For example, some liquids have high surface tension because, in addition to van der Waals interactions, there are other intermolecular interactions, such as ionic bonds, metallic bonds, and hydrogen bonds. For these liquids, \( p \) in Equation 7 has a much larger deviation from \( p_{\text{bulk}} \) leading to a larger value of \( \sigma \).

Additionally, one may explain why the surface tension of a given liquid decreases with increasing temperature. At higher temperatures, the portion of the pressure component due to thermal agitation increases linearly with temperature, while the forces due to intermolecular attractive forces remain
essentially constant. Thus, as temperature increases, the difference between $p_{\text{bulk}}$ and $p$ in the interfacial zone diminishes, and surface tension decreases in an approximately linear fashion (7).

5. CONCLUSIONS

The explanation of origin of surface tension found in the literature is erroneous, as it is based on an incomplete picture of intermolecular forces. This explanation is based solely on the attraction/repulsion forces among molecules. However, molecules also exert forces on each other because of collisions. The interplay between attraction/repulsion and collision forces determines the state of stress within the bulk fluids and their interfacial regions. The difference in these forces between the interfacial regions and inside the bulk domains is known as surface tension. This thorough and rigorous description allows us to explain various phenomena related to the surface tension.

STATEMENTS AND DECLARATIONS

Conflicts of Interest

There are no conflicts of interest to declare.

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