

Characterization of Nontoxic Liquid-Metal Alloy Galinstan for Applications in Microdevices

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Abstract—We have obtained interfacial properties of Galinstan, a nontoxic liquid-metal alloy, to help replace mercury in miniature devices. To prevent formation of an oxide skin that severely hinders the fluidic behavior of small Galinstan droplets and leads to inaccurate property data, we performed our experiments in a nitrogen-filled glove box. It was found that only if never exposed to oxygen levels above 1 part per million (ppm) would Galinstan droplets behave like a liquid. Two key properties were then investigated: contact angles and surface tension. Advancing and receding contact angles of Galinstan were measured from sessile droplets on various materials: for example, 146.8° and 121.5°, respectively, on glass. Surface tension was measured by the pendant-drop method to be 534.6 ± 10.7 mN/m. All the measurements were done in nitrogen at 28 °C with oxygen and moisture levels below 0.5 ppm. To help design droplet-based microfluidic devices, we tested the response of Galinstan to electrowetting-on-dielectric actuation. [2011-0124]

Index Terms—Electrowetting-on-dielectric (EWOD), Galinstan, Galinstan contact angle, Galinstan oxidation, Galinstan surface tension, liquid-metal.

I. INTRODUCTION

FOR THE excellent conductivities of a metal and the fluidic properties of a liquid, metals are often employed in their liquid state: mercury, gallium (alloy), aluminum, tin, etc. At room temperature, however, the choice is significantly limited; mercury, used commonly for thermometers and electromechanical relays, remains the only liquid metal as an element. Gallium and many of its alloys are a liquid near but above room temperature. Although some laboratories may have produced some in-house, a room-temperature liquid metal has not become available until recently. Galinstan, an alloy of 68.5% Ga, 21.5% In, and 10.0% Sn, is a commercially available (from Geratherm Medical AG in Germany and distributed by RG Medical Diagnostics in the U.S.) nontoxic liquid metal that replaces mercury mostly in thermometers [1]. Physical properties of Galinstan and mercury [1], [2] are summarized in Table I for comparison. Like mercury, Galinstan retains its

liquid state well below 0 °C. Its nontoxic nature and ultralow vapor pressure ($< 10^{-6}$ Pa at 500 °C) make Galinstan a better substitute for mercury in many liquid-metal applications, e.g., electromechanical relays [3], coolants [4], and ion sources [5]. In microelectromechanical systems (MEMS), Galinstan has already been used [6], [7] or is in line to replace mercury [8], [9]. It has, however, failed to produce many successful results in MEMS as initially anticipated, and mercury is still the common choice of liquid metal in research at room temperature. The main reason for the slow adoption in microdevices is the fast oxidation of Galinstan, as it was for gallium [10], making any study or development involving Galinstan below millimeters difficult. In addition, several parameters that are important for microdevices are unavailable for this relatively new material. Even worse, some information available on the Internet (e.g., that Galinstan wets glass) lacks rigor and often misleads.

Due to the dominance of the capillary effect at microscale, contact angles and surface tension are two key properties of a liquid used in MEMS, especially the droplet-based microdevices. For Galinstan, however, no systematic study on contact angles has so far been reported, and surface tension data are either limited (from Geratherm Medical AG in Germany [1]) or scattered (between 0.501 and 0.718 N/m [11], [12]) in the literature. The major challenge of characterizing Galinstan lies in the fact that its surface oxidizes instantaneously in air. The oxidation effect is rather profound at microscale, where the surface-to-volume ratio is large, making small droplets of Galinstan behave like a gel rather than a normal liquid. To obtain genuine properties not obscured by the surface oxidation, we performed all the tests in this paper inside a glove box of high-purity inert environment. Keeping Galinstan free of oxidation will stay as a main issue in developing microdevices that involve free surfaces of Galinstan.

II. THEORIES

A. Contact Angle Measurements

The contact angle is an experimentally observable quantity that describes the wetting property of a liquid in contact with a solid surface and surrounded by another immiscible fluid (most commonly a gas), as shown in Fig. 1(a). This macroscale measurement ignores the nanoscopic region where molecular interaction between the phases (i.e., liquid, gas, and solid) is significant. Intrinsic contact angle free of hysteresis is expected only on an ideal solid surface, while in reality, the contact angle of an advancing meniscus exceeds that of a receding meniscus. The advancing and receding contact angles define the upper and lower limits of the contact angle of a static liquid,

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TABLE I
COMPARISONS OF PROPERTIES OF GALINSTAN AND MERCURY

Property	Galinstan [®] [1]	Mercury [2]
Color	Silver	Silver
Odor	Odorless	Odorless
Boiling point	> 1300°C	356.62°C
Melting point	-19°C	-38.83°C
Vapor pressure	<10 ⁻⁶ Pa at 500°C	0.1713 Pa at 20°C
Density	6440 kg/m ³	13533.6 kg/m ³
Solubility	Insoluble	Insoluble
Viscosity	2.4×10 ⁻³ Pa·s at 20°C	1.526×10 ⁻³ Pa·s at 25°C
Thermal conductivity	16.5 W·m ⁻¹ ·K ⁻¹	8.541 W·m ⁻¹ ·K ⁻¹
Electrical Conductivity	2.30×10 ⁶ S/m	1.04×10 ⁶ S/m

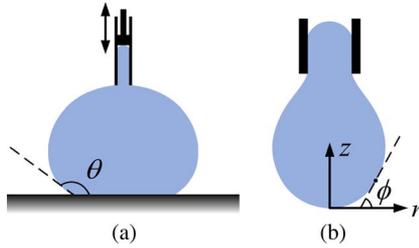


Fig. 1. Measurements of contact angles and surface tension in this paper. (a) Advancing and receding contact angles are measured by adding or subtracting volume to or from the sessile drop via the top needle until the contact line starts to move. (b) Surface tension is measured by the pendant-drop method using the cylindrical coordinate shown.

and their difference is referred to as contact angle hysteresis. Since deviations from the ideal surface, such as roughness and chemical inhomogeneity, are believed to cause the hysteresis and affect its magnitude, complete contact angle data should include information about the solid surface as well. As the surface cleanliness (chemical homogeneity) can be reasonably well maintained, in this paper, we measure roughness of the solid surface along with the contact angles.

B. Surface Tension Measurement

The surface tension of a liquid is the boundary tension at the interface between a liquid and an immiscible fluid (e.g., gas) due to the imbalanced molecular attraction from two sides of the interface. Because it is vital information in any capillary study, a series of methods of measuring surface tension has been established, among which the du Noüy ring method (e.g., du Noüy ring tensiometer), the Wilhelmy slide method (e.g., Wilhelmy plate tensiometer), and the pendant-/sessile-drop method (goniometer) are most widely used. For surface tension measurement of molten metals, in particular, the maximum bubble pressure method is also favored, as well as the pendant-/sessile-drop method [13]. Considering the limited space inside a glove box to handle Galinstan and perform the entire measurement, we have excluded the du Noüy ring method and the Wilhelmy slide method. Also excluded was the maximum bubble pressure method, which requires precise pressure measurement equipment and a tank of Galinstan. In contrast, the pendant-/sessile-drop method uses a simple optical system (i.e., light source and camera with pendant/sessile drop in-between) and only a few microliters of sample liquid (< 2 μ L in our case).

The pendant-/sessile-drop method extracts the surface tension from the shape of the drop when the forces due to gravity and surface tension are comparable. Following the Young–Laplace equation, the pressure difference across a meniscus (liquid–air interface) is given by

$$\Delta p = \gamma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) \quad (1)$$

where γ is the surface tension and $(1/R_1 + 1/R_2)$ is twice the mean curvature of the free surface. Note that R_1 and R_2 are not necessarily the principal radii of curvature as long as their planes are mutually orthogonal [13]. As a pendant/sessile drop is axisymmetric, the two radii of curvature R_1 and R_2 are equal, i.e., $R_1 = R_2 = R$, at the apex. Placing the coordinate origin at the apex, as seen in Fig. 1(b), pressure difference at the apex is given by $\Delta p(0) = 2\gamma/R$. Hydrostatic pressure due to gravity will distort the shape of the drop, resulting in a pressure difference at height z from the apex to be

$$\Delta p(z) = \Delta p(0) - \Delta \rho g z = \frac{2\gamma}{R} - \Delta \rho g z \quad (2)$$

where $\Delta \rho = \rho_{\text{liquid}} - \rho_{\text{gas}}$ and g is the gravitational acceleration. Combining (1) and (2) together yields

$$\gamma \left(\frac{1}{R_1} + \frac{1}{R_2} \right) = \frac{2\gamma}{R} - \Delta \rho g z \quad (3)$$

which will serve as the governing differential equation of the droplet profile after substituting R_1 and R_2 with the variables that are proper for the coordinate system of interest. However, there is no analytical solution to this differential equation, and only a numerical solution exists. Equation (3) can be rewritten into a set of three arc-length-based first-order equations

$$\begin{aligned} \frac{d\phi}{ds} &= \frac{2}{R} - \frac{\Delta \rho g z}{\gamma} - \frac{\sin \phi}{r} \\ \frac{dr}{ds} &= \cos \phi \\ \frac{dz}{ds} &= \sin \phi \end{aligned} \quad (4)$$

where s is the arc length and ϕ is the angle between the r axis and the tangent at a point on the meniscus [Fig. 1(b)]. Using these equations, one can generate a series of theoretical profiles of a pendant/sessile drop by varying surface tension γ and the radius at the apex R as two parameters. By comparing

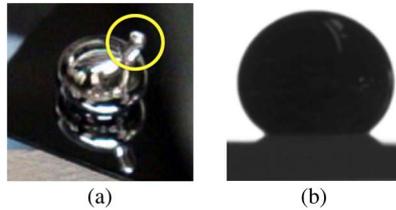


Fig. 2. Oxidation effect on a Galinstan droplet. (a) Angled view of a Galinstan droplet (~ 2 mm in diameter) oxidized while being dispensed from a pipette (in less than 0.25 s), forming a nonspherical shape (i.e., the hornlike shape marked with a circle), even when the oxygen content was only $\sim 0.2\%$. (b) Side view of a Galinstan droplet (~ 1.5 mm in diameter) that formed a spherical shape and behaved as a liquid, only when the oxygen content was kept below 1 ppm.

the data points obtained from experiment (i.e., drop profile on an image) to the theoretical profiles, the surface tension can be determined [14].

In practice, in an effort to reduce the time of calculation and comparison, initial values of surface tension (or a parameter involving surface tension, e.g., Bond number) and the radius at the apex are estimated using empirical tables or polynomials and two data points from the experimentally obtained droplet image (viz., the selected-plane method [14], [15]). Then, an accurate value of surface tension is obtained by varying surface tension and radius at the apex around the initial estimated values to minimize the sum of squares of the normal distance between the experimental data and the theoretical profile. We avoided the more complex and detailed optimization process used by Lin *et al.* [14], which includes two more variables to locate the actual location of the apex (r_0, z_0) , by introducing a plumb bob in our experiment. The plumb bob reveals the true axis of symmetry and corrects the data before they are sent to the computer program for calculation.

III. EXPERIMENTAL PROCEDURE

A. Prevention of Oxidation

As soon as Galinstan is exposed to ambient air, its surface gets oxidized instantaneously (well below 1 s), turning the color from a shiny silver into a dull gray. Such an oxide skin influences the behavior of microscale droplets significantly due to their large surface-to-volume ratio. Fig. 2 vividly shows the fast oxidation of a 2-mm-diameter droplet of Galinstan dispensed from a plastic pipette (Gilson PIPETMAN P1000) in a mere 0.2% oxygen environment—100 times less than in ambient air. The surface was already oxidized during the dispensing action (< 0.25 s), forming a droplet of a pronounced nonspherical shape. When dispensed in ambient air, Galinstan was pinned at the pipette tip by instantaneous oxidation, requiring a tap to the pipette to free the droplet.

In order to prevent the oxidation, Galinstan should be kept in an inert environment (e.g., nitrogen [16] or argon [17]) or covered by another aqueous solution (e.g., diluted HCl [11], [12]), similarly to other gallium alloys. Following the procedures described in [16], we purged a glove box (VAC 101965 with oxygen and moisture sensors) with prepurified nitrogen (99.998%) and had the built-in purification system continue to absorb oxygen and moisture. In addition, we controlled

TABLE II
BEHAVIOR OF GALINSTAN DROPLET AT DIFFERENT
OXYGEN TRACE LEVELS

Oxygen	Observation
0.2-20.9%	Droplet is distinctively non-spherical by instantaneous surface oxidation even at the moment of dispensing
~ 20 ppm	Droplet is spherical Droplet behaves like gel rather than true liquid
< 1 ppm	Droplet behaves like true liquid
< 0.5 ppm	Droplet behaves like true liquid The condition for all data in this report

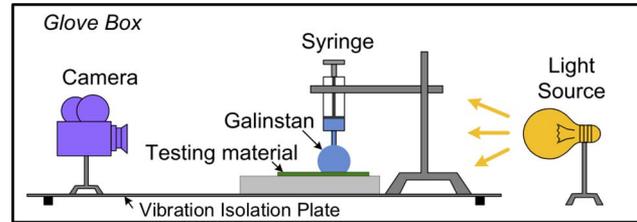


Fig. 3. Schematic of the experimental setup in a nitrogen-filled glove box, drawn for the measurement of contact angles on a sessile droplet. To measure the surface tension with a pendant drop, the testing material and its base substrate were removed, and a plumb bob was added.

the flow rate of nitrogen (or air in the highest oxygen level case) in order to observe the behavior of Galinstan at different levels of oxygen. The summary of our observations is listed in Table II. Note that even at ~ 20 parts per million (ppm) (i.e., 0.002%) of oxygen, in which the dispensed droplets displayed a spherical shape, the droplet behaved as if a gel droplet rather than a normal liquid droplet; it was easily distorted to an irreversible nonspherical shape, if shear was applied on the droplet. Only when the oxygen content was reduced to below 1 ppm (0.0001%) did Galinstan behave like mercury or other normal liquids, recovering a spherical shape at the removal of external shear on the droplet. To ensure that Galinstan is free of oxidation, all of our characterization measurements were performed in nitrogen with both oxygen and moisture contents below 0.5 ppm. Also note that all the data in this paper were collected at 28 °C (measured by Ever-Safe N16B glass organic-filled MCT classical thermometer).

B. Vibration Isolation

Both contact angle and surface tension measurements in this paper involve an image analysis at the interfaces. Therefore, accuracy of the measurement highly depends on the quality of the pictures. In order to capture clear pictures of sessile or pendant drops, many aspects should be considered and controlled well when designing the experiments, such as vibration, lighting, camera tilt, and pixel errors from small droplets. Vibration, which leads to a blurred droplet profile, would cause large errors. To eliminate the vibration, we have placed the entire experiment setup, except the backlight, on a vibration-isolation plate (Vistek VIP Series 320) inside the glove box, as shown in Fig. 3. The backlight used had a significantly larger beamwidth than the camera's field of view and hence did not require vibration isolation.

TABLE III
SURFACE PREPARATION OF DIFFERENT MATERIALS
TESTED FOR CONTACT ANGLES

Type	Material	Thickness	Preparation Method
Conductor	Tungsten	200 nm	Sputtered on silicon wafer
Insulator	Silicon nitride	1 μm	Low-stress LPCVD on silicon wafer
	Glass	1 mm	Degreased soda-lime microscope slide (Fisherfinest [®])
Polymer	Parylene	1 μm	Vapor deposition on microscope slide
	Teflon [®]	560 nm	Spin-coated on PECVD silicon nitride on ITO-coated glass
Mica	Phlogopite	500 μm	Cleaved from its natural form provided by Cogebi, Inc.
	Muscovite	400 μm	Cleaved from its natural form provided by Cogebi, Inc.

C. Contact Angle Measurements

Imitating the commercial instruments (e.g., First Ten Angstroms FTÅ200) which measure contact angles using drop shape analysis, we have developed a setup inside the glove box for contact angle measurements of Galinstan, as schematically shown in Fig. 3. A syringe with liquid-metal-compatible needle (Hamilton 50- μL syringe 1705RN and removable needle 7770-02 RN NDL with 0.41-mm inner diameter) was used to generate sessile droplets of Galinstan on various solid surfaces.

The experiments proceeded as follows. First, a captive Galinstan drop was created, as shown in Fig. 1(a), by dispensing it manually from a syringe through the needle. The droplet was made much larger than the needle tip to minimize the surface distortion by the tip [18]. Second, the sessile droplet was expanded or contracted by pumping the liquid in and out of the syringe continuously and slowly ($\sim 0.25 \mu\text{L/s}$) to maintain quasi-static states, while recording the side views of the droplet with a video camera (Edmund Optics EO-0413C) in audio video interleave file format at 28 frames per second. Slow pumping of Galinstan was more difficult than aqueous solutions (e.g., water), because liquid metals have much larger surface tension and do not wet the needle material. Fresh Galinstan was used for every test to prevent potential contamination. Finally, after the experiment was done, the video was inspected frame-by-frame, and advancing and receding contact angles were obtained from the snapshot right before the contact line moved.

We measured the contact angles of Galinstan on several solid materials commonly used in microdevices. Among metals, almost all of which chemically react with gallium and gallium alloys [19], tungsten was chosen for its chemical resistance. Mica was chosen for their well-known surface smoothness, despite its rare use in MEMS. Methods of surface preparation for all the materials tested are summarized in Table III. After completing contact angle measurements, surface roughness was measured for all the tested materials using atomic force microscopy (AFM).

D. Surface Tension Measurement

For surface tension measurement by the pendant-drop method, the testing material and its substrate were removed from the contact angle measurement setup of Fig. 3. Added, instead, was a plumb bob that provides a reference vertical line in the camera view. The quasi-static growth of the pendant droplet, which took around 80 min, was video-recorded. After extracting snapshots (bitmap image files, viz., BMP file format)

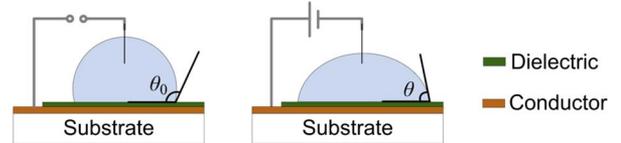


Fig. 4. EWOD actuation of a sessile liquid droplet.

from the video, the tilted images were rotated to align with the vertical line of the plumb bob (i.e., the true axis of symmetry for the droplet profile) prior to the profile analysis. A larger droplet elongates more by gravity and fills the picture frame fully, providing a droplet profile with more pixels and improving the measurement accuracy. For our case, the capillary diameter to maximize the number of pixels on the droplet profile in the picture frame of our USB camera was calculated to range between 0.72 and 2.44 mm for surface tension between 501 and 718 mN/m [11], [12], respectively. Instead of the stainless-steel needle used in the contact angle measurement, we chose a glass capillary for the pendant-drop method because of its availability in the aforesaid desired diameter and its transparency that allows observation of the meniscus movement inside the capillary. Recalling that Galinstan is nonwetting on glass, i.e., it does not spread on the glass surface outside the capillary, the diameter calculated earlier should be the inner diameter of the capillary; we chose 1.22 mm for our measurements.

E. EWOD

The wetting conditions of a liquid on a solid can be modulated by various methods. In general, external energy (e.g., electrical and thermal) enters the system and calls for a configuration that would minimize the total energy. Among electrical methods, the mechanism of electrowetting on dielectric (EWOD) has been widely adopted, as shown in Fig. 4. Relations between the applied voltage and the resulting contact angle are described by the Lippmann–Young equation [20]

$$\cos \theta = \cos \theta_0 + \frac{1}{2\gamma} cV^2 \quad (5)$$

in which V is the voltage applied across the dielectric layer (between the liquid and the conductor underneath the dielectric), c is the capacitance per unit area of the dielectric, θ_0 is the initial contact angle without voltage applied, and θ is the contact angle when applying V .

Hermetic feedthrough enabled EWOD tests of Galinstan inside the glove box while placing other apparatuses, e.g., the dc

TABLE IV
CONTACT ANGLES OF GALINSTAN ON DIFFERENT MATERIALS

Type	Material	Contact Angles			Surface Roughness			
		$\theta_a/^\circ$	$\theta_r/^\circ$	$(\theta_a - \theta_r)/^\circ$	R_q/nm	R_a/nm	R_{max}/nm	Surfaces Area Ratio
Conductor	Tungsten	161.3	119.6	41.7	3.31	2.54	27.27	0.13%
Insulator	Silicon nitride	147.0	126.1	20.9	1.38	1.09	13.43	0.37%
	Glass	146.8	121.5	25.3	2.05	1.38	30.2	0.30%
Polymer	Parylene	146.3	112.6	33.7	13.48	9.02	105.37	0.85%
	Teflon [®]	161.2	144.4	16.8	2.04	1.62	21.03	0.59%
Mica	Phlogopite	148.0	124.5	23.5	1.97	1.59	13.57	0.23%
	Muscovite	163.6	148.1	15.5	1.02	0.82	7.64	0.11%

power source (Keithley 2425) and the amplifier (Trek PZD700), outside the glove box. We chose to use a tungsten probe tip for its chemical inertness with Galinstan to electrically connect the droplet and the power source. Note that the synchronization of electrical activation and snapshots in our experiment allowed us to quickly capture the droplet spreading and minimize the effect of dielectric charging by reducing the total application of high voltages to less than 1 min.

IV. RESULTS AND DISCUSSION

A. Contact Angle Measurements

Results of contact angle measurements of Galinstan on different solid surfaces are summarized in Table IV along with the roughness information of the surfaces obtained by AFM measurements. The contact angles were calculated with sub-pixel accuracy [21]. The roughness was quantified by different parameters interpreting the vertical deviations of the roughness profile from the mean line: R_q being the root mean square of the deviations, R_a being the arithmetic average of the absolute values, R_{max} being the maximum difference between the peaks and valleys, and surface area ratio being the ratio between the increased surface area (due to the roughness) and the perfectly flat surface. Galinstan was found nonwetting on all surfaces tested.

However, we found that, once Galinstan gets oxidized, the oxide shell, though thin, sticks on the surface and gives the illusion that Galinstan wets almost all materials. To emphasize the detrimental impact of oxidation on measuring the wetting properties of Galinstan, we measured its advancing and receding contact angles on glass in ambient air ($\sim 20.9\%$ oxygen) as well, as shown in Fig. 5. The experimental setup remained exactly the same, except that the glove box was vented to air. For example, the oxide shell was pinned on the glass surface during receding until most of the Galinstan liquid has been pumped back into the syringe, as shown in Fig. 5(b). Such pinning effect resulted in incorrect advancing and receding contact angles of 165° and 7° , in contrast to the accurate angles of 146.8° and 121.5° , respectively, in Table IV. The receding angle would have been even lower than 7° if the measurement had not been ended prematurely by the liquid disconnection during the suction back.

The contact angle information of Galinstan is rare at this point. Kocourek [11] reported that the contact angles of Galinstan on glass are 122° in air and 166° in 6% hydrochloric

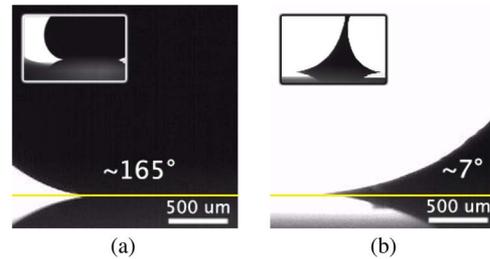


Fig. 5. Three-phase contact regions of Galinstan on glass in ambient air ($\sim 20.9\%$ oxygen). (a) Advancing angle. (b) Near receding angle. Pinning of the oxide skin makes the receding angle appear very low. Both (a) and (b) include an inset showing the larger field of view. The measurement of receding contact angle ended prematurely when the droplet was separated from the top needle.

acid. Their result in air stays within the wide range that we observed in air, i.e., 7° and 165° . However, a direct comparison is inadvisable, because Kocourek reported only one contact angle instead of advancing and receding angles and did not provide additional information such as roughness of the glass surface and details of the measurement procedure.

B. Surface Tension Measurement

For our pendant-drop measurement, one experiment consisted of the formation, quasi-static growth, and dropping of a droplet, taking about 90 min. Frames of every second over the quasi-static growth (~ 80 min), i.e., ~ 4800 pictures, were collected from the video recording and provided enough data for statistical analysis. All pictures were analyzed using a computer program written to follow (1)–(4) [22]. In addition to the Canny edge detection algorithm used in [16], the Sobel edge detection algorithm was also used for comparison. Fig. 6(a) shows the surface tension results with box plots, a quick graphical approach to examine data without any assumption of the underlying statistical distribution, using the two algorithms. Note that multiple outliers might overlap together, giving the appearance of one outlier in the plots. To assess whether the data have an approximate normal distribution, the normal probability plot of the two sets of data is drawn in Fig. 7. We can see that the calculated results from the Sobel edge detection algorithm not only tend to be larger but also deviate more from the normal distribution, which is commonly assumed as observational errors. This is understandable, considering that an edge is a human-defined perception; different definition (i.e., different edge detection algorithms) might lead to different

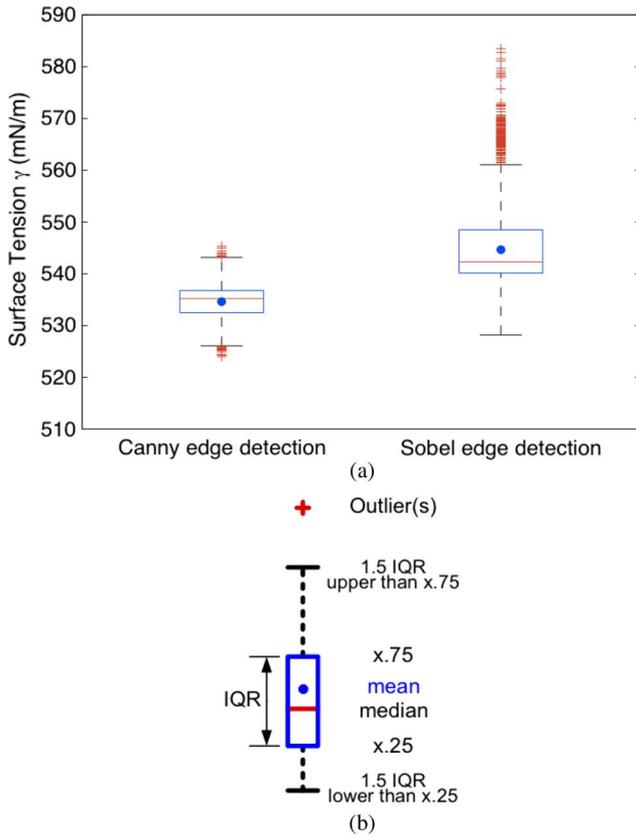


Fig. 6. Surface tension data of Galinstan obtained. (a) Box plot of the surface tension measured by two edge detection algorithms. (b) Meaning of the box plot. Interquartile range (IQR) excludes the highest and lowest 25% of data.

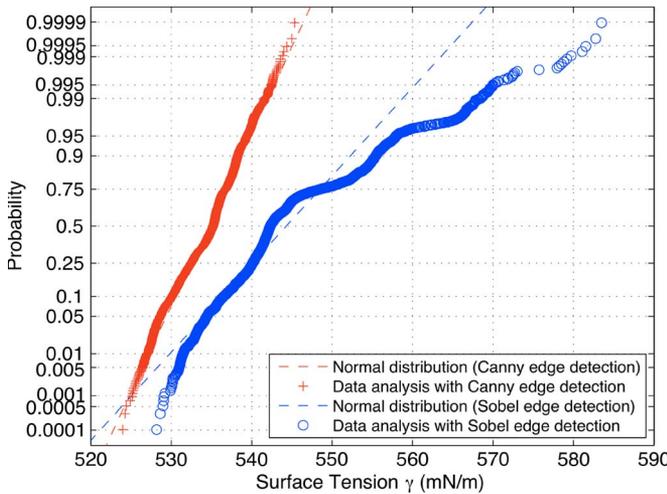


Fig. 7. Normal probability plots of the Galinstan surface tension measured by two edge detection algorithms. The standard deviations of the Canny and Sobel algorithms are 3.2093 and 7.7184 mN/m, respectively.

results. It is commonly believed that Sobel edge detection is more sensitive to noise and thus inaccurate when compared to Canny edge detection, which reduces the noise by smoothing the image [23]. Therefore, based on the distribution of the results in Fig. 7 and information in [23], we conclude that the results from Canny edge detection are more reliable and use them for discussions in the remainder of this paper.

Following the procedures described previously, we have obtained the surface tension of Galinstan as 534.6 ± 10.7 mN/m.

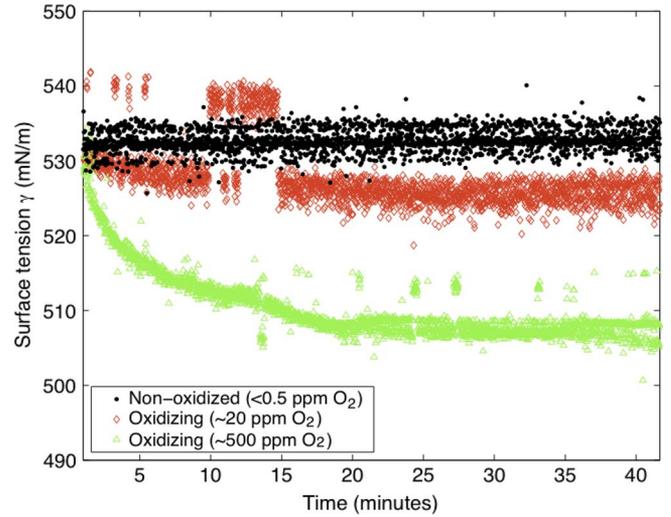


Fig. 8. Surface tension of Galinstan measured during the first 40 min of the quasi-static droplet growth. The Canny edge detection algorithm was used for calculation of surface tension every second. The surface tension in this paper (solid black circles for $O_2 < 0.5$ ppm) displays virtually constant values, showing no effect of oxidation. In contrast, the values in two slightly oxidizing conditions (red hollow diamonds for $O_2 \sim 20$ ppm and green hollow triangles for $O_2 \sim 500$ ppm) show the effect of oxidation before the measurements were completed.

Because oxidation affects the surface tension measurement, the reading would drift as a freshly formed pendant droplet gets oxidized over time. As shown in Fig. 8, the main data (in $O_2 < 0.5$ ppm) stayed constant, confirming that the obtained value was free of any oxidation effect. To compare, we have run a pair of controlled experiments in slightly higher oxygen levels (in $O_2 \sim 20$ and 500 ppm), which clearly reveal the oxidation effect on the surface tension measurement. The scatters in the data were caused mostly by the shocks and vibrations from the environment not filtered out by the current vibration-isolation plate. Despite the excessive scatters in the data of 20-ppm oxygen during 10–15 min, the overall trend is still clear.

In the pendant-drop method, the fact that the capillary tip shares the focal plane with the hanging drop due to axisymmetry allowed us to obtain the scale factor without resorting to additional tools. In contrast, the sessile-drop method has to use a separate “ruler” to obtain the scale factor, and a tiny mismatch between the focal plane of the “ruler” and that of the sessile drop will linearly bias the results of surface tension [24]. Accordingly, we believe that the pendant-drop method used in this paper provides more accurate results than the sessile-drop method in measuring surface tension. Our result (534.6 mN/m in nitrogen) is comparable to but different from that of Kocourek [11] (517 mN/m in air), who used a sessile-drop method.

C. Response to EWOD Actuation

The result of EWOD actuation on Galinstan is shown in Fig. 9, with the inset explaining the device used for the test. The EWOD device was prepared by depositing an ~ 4500 -Å-thick silicon nitride on an ITO-coated glass substrate using PECVD, followed by spin coating a ~ 1700 -Å-thick Teflon AF 1600. The experimental data shown in Fig. 9 represent a mean of

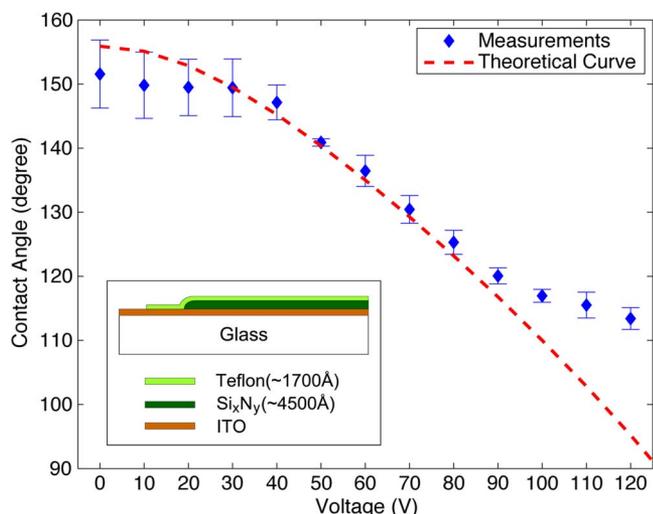


Fig. 9. Response of Galinstan to EWOD actuation. The theoretical curve was drawn using the initial contact angle measured without a probe tip connection (155.9°) and the surface tension obtained in this paper (534.6 mN/m). The total capacitance (per unit area) of the dielectric layers (Teflon in series with silicon nitride) was calculated to be $\sim 61 \mu\text{F/m}^2$. The inset shows the device configuration used for the EWOD test.

five experiments, and the error bar indicates standard deviation (instead of the mean standard error in [16]) of the data. The EWOD actuation was confirmed; contact angles changed with applied voltage following the Lippmann–Young equation (5) very well until contact angle saturates. The EWOD testing of liquid metals is not as easy as regular liquids because their large surface tension and nonwetting nature make it difficult to insert the probe into the liquid metal and form a stable electrical connection. The first few contact angles ($V = 0\text{--}20 \text{ V}$) in our measurements were affected by contact angle hysteresis because the droplet spread out slightly during the insertion and receded back after the insertion of the tungsten probe tip, which is highly nonwetting to Galinstan. Therefore, the theoretical curve in Fig. 9 was generated using the initial contact angle obtained without the probe tip (155.9°). Excluding the saturation region, a contact angle decrease of $\sim 30^\circ$ has been achieved by applying 90 V. Such a contact angle change was sufficient for MEMS applications, such as the microswitch developed in [8].

V. CONCLUSION

We have characterized the interfacial properties of Galinstan that are important for the design of microdevices: contact angles and surface tension. We overcame the strong oxidation of Galinstan by performing the entire experiments inside a glove box of high-purity inert environment; its droplets finally behaved like a typical liquid when the levels of oxygen and moisture were brought down below 1 ppm (i.e., 0.0001%) and 0.5 ppm (i.e., 0.00005%), respectively. Signs of oxidation, such as color changes and gellike behavior, were not found for more than 2 h, providing us with sufficient time to measure the contact angles and surface tension of Galinstan. The contact angles of Galinstan were measured by sessile droplets on several materials commonly used in MEMS devices, and the surface roughness of the materials was also reported. For example,

the advancing and receding contact angles of Galinstan on glass were measured to be 146.8° and 121.5° , respectively. In comparison, we also tested the contact angles in ambient air and confirmed that the illusion of Galinstan wetting solid surfaces, including glass, was caused by the oxide skin sticking on the surfaces. The surface tension of Galinstan was measured to be $534.6 \pm 10.7 \text{ mN/m}$, using a pendant-drop method. In addition, the EWOD actuation of oxide-free Galinstan was tested and found matching the Lippmann–Young equation. As a high-level hermetic packaging of MEMS devices becomes available to prevent oxidation, Galinstan is expected to eventually substitute mercury for microscale applications.

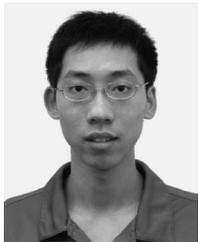
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