

# Liquid Metal Micro- and Nanodroplets: Characteristics, Fabrication Techniques, and Applications

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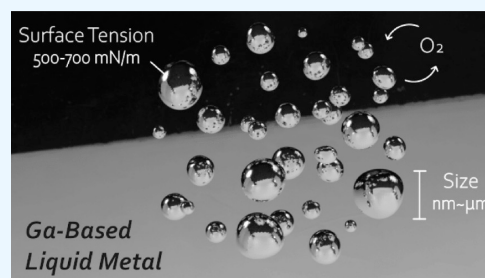
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**ABSTRACT:** Gallium-based liquid metal micro- and nanodroplets are being extensively explored in innumerable emerging technologies. Although many of these systems involve the interfaces of liquid metal with a continuous phase liquid (e.g., microfluidic channels and emulsions), the static or dynamic phenomena at the interface have been scarcely discussed. In this study, we begin by introducing the interfacial phenomena and characteristics observed at the interface between a liquid metal and continuous-phase liquids. Based on these results, we can employ various methods to fabricate liquid metal droplets with tunable surface properties. Finally, we discuss how these techniques can be directly applied to a wide range of state-of-the-art technologies including microfluidics, soft electronics, catalysts, and biomedicines.



## 1. INTRODUCTION

Metals that exist in the liquid state near room temperature belong to a class of materials known as liquid metals. Liquid metals exhibit superior electrical and thermal conductivities compared with nonmetallic liquids. Their liquid properties can be employed for the design of interconnections, whereas external deformations such as bending or stretching are prone to damage solid materials. Liquid metal-based structures can overcome this lack of durability while simultaneously maintaining their electrical conductivity.<sup>1</sup> There are several types of liquid metals with low melting points: mercury, alkali metals, and gallium-based alloys. Mercury is known for its high toxicity and low vapor pressure. Alkali metals (e.g., sodium, rubidium, cesium, and francium) ignite spontaneously and react vigorously with water. Rubidium and cesium are highly explosive, while francium displays radioactivity. Meanwhile, gallium is a nontoxic and environmentally friendly alternative that opens the door to a wide range of industrial applications. In particular, gallium-based micro- and nanodroplets have been employed for the design of systems that require both conductivity and flexibility.<sup>2,3</sup> In this review, we summarize and discuss the interfacial properties, fabrication processes, and technological applications of gallium-based liquid metal droplets.

The metallic bonding forces of liquid metals contribute to their extremely high surface tension (500–700 mN/m).<sup>4,5</sup> Although high-surface-tension liquids generally bead up into spheres, gallium-based liquid metal droplets tend to exhibit nonspherical geometries. This can be attributed to the effect of surface oxidation,<sup>5,6</sup> which alters the mechanical and rheological properties of liquid metal. It is common for wrinkles to develop on the oxidized membrane, even for

microscale liquid metal droplets prepared by ultrasonication.<sup>5</sup> While oxide-free liquid metal exhibits Newtonian behaviors with constant viscosity, oxidized liquid metal is known to display shear-thinning characteristics. However, different viscosities may be reported for liquid metal exposed in air, as the measured viscosity is strongly affected by the presence of the oxide skin. Furthermore, the oxide membrane makes it difficult to measure the interfacial energy of liquid metal via conventional techniques such as contact angle measurements or the pendant droplet method. While these methods depend on the equilibrium of a droplet's geometry at the liquid–liquid or liquid–gas interface, liquid metal surrounded by an oxide membrane can no longer be considered as an ideal liquid droplet. Since these droplets exhibit irregular geometries and nonconstant curvature profiles, it is difficult to obtain accurate values of the corresponding interfacial properties.<sup>4,5</sup>

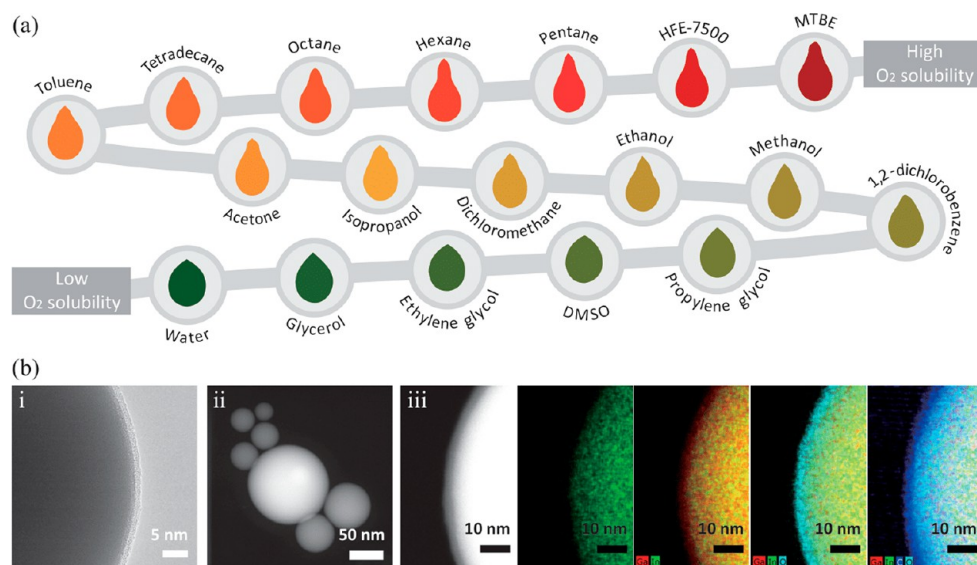
Therefore, investigating the various phenomena taking place on the surface of liquid metal micro- and nanodroplets is crucial to utilizing their full potential. Identifying the effects of ambient oxygen solubility<sup>5</sup> and electric fields (e.g., electrocapillarity, continuous electrowetting, electrowetting-on-dielectric, and electrochemically controlled capillarity)<sup>7</sup> on liquid metal droplets provides insight into how the metallic surface interacts with its environment. The interface between a liquid

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**Figure 1.** (a) Schematic of various bulk-scale pinch-off geometries of Galinstan droplets immersed in different liquids, in the order of high to low oxygen solubility. Reprinted with permission from ref 5. Copyright 2022 Elsevier. (b) Characterization of EGaIn (eutectic alloy of Ga and In) nanoparticles. (i) Transmission electron microscope (TEM) image for EGaIn coating. The black core is the liquid metal, and the thin and bright part is the gallium oxide layer. The surface of the oxide layer can be coated by an additional carbon layer. (ii) Scanning transmission electron microscope (STEM) image of EGaIn nanodroplets. (iii) Element mapping of the EGaIn coating on the leftmost picture, with elements colored respectively as Ga (red), In (green), O (cyan), and C (blue). From left to right, the four pictures show the mapping results for the EGaIn nanodroplet, adding the contribution of each element in sequence. Ga and In exist near the core of the droplet, which are surrounded by an oxide layer. The oxide layer is surrounded by an additional layer of carbon. Reprinted with permission from ref 4. Copyright 2015 John Wiley & Sons.

metal droplet and its surroundings can be further modulated by applying various surface functionalization techniques.<sup>6,9–11</sup> When it is required to merge the droplets into continuous networks, we may introduce additional processes of thermal, laser, or mechanical sintering. Based on these characteristics, researchers have sought a number of different methods to tailor the design of liquid-metal droplets. A liquid metal stream traveling through microfluidic channels can be separated into individual droplets of particular dimensions or geometries. Droplets with tunable properties can also be achieved by performing ultrasonication or mechanical agitation. We can incorporate these liquid metal droplets to design structures with a higher degree of complexity, such as microfluidic systems, soft robots/electronics, catalyst particles, and biomedicines/drug delivery.<sup>1–4</sup>

## 2. INTERFACIAL PROPERTIES OF LIQUID METAL DROPLETS IN FLUID SYSTEMS

In gallium-based liquid metal, the oxide thickness depends on the ambient condition of oxygen or water vapor concentration, as well as the chemical environment.<sup>4,5</sup> The surface properties of liquid metal droplets can be tuned by selecting an appropriate surrounding fluid,<sup>5</sup> by the means of electrochemical approaches,<sup>7</sup> or by introducing different types of surface functional groups.<sup>6,9–11</sup>

**2.1. Oxygen Solubility of the Ambient Fluid.** The morphology and surface energy of gallium-based liquid metal droplets exhibit a strong correlation with the mole fraction of dissolved oxygen in the ambient fluid. In the case of bulk liquid metal, the droplet's eccentricity (i.e., eccentricity of an ellipse with a moment of inertia identical to the two-dimensional profile of the droplet) increases sigmoidally with respect to the logarithm of the oxygen solubility. Compared with droplets present in liquids with low oxygen solubility (e.g., water),

droplets immersed in liquids with high oxygen solubility (e.g., alkanes and fluorinated liquids) tend to be encapsulated by a solid-like oxide membrane.<sup>5</sup> Since liquid metal immersed in oxygenated environments readily forms oxide layers, the resulting droplets display pear-shaped geometries (Figure 1a).<sup>5</sup>

In the case of liquid metal emulsions in organic solvents, the oxygen solubility of the ambient fluid may additionally lead to varying degrees of molecular adsorption.<sup>5</sup> While metallic components exist in the core of the drop, the surrounding oxide mainly consists of gallium and oxygen.<sup>4,6</sup> High-resolution transmission electron microscope (TEM) images and element mapping results have suggested that liquid metal micro- and nanoparticles sonicated in certain organic solvents tend to form an additional carbon coating surrounding the oxide layer (Figure 1b).<sup>4</sup> This implies that liquid metal droplets immersed in oxygen-rich environments can additionally facilitate the adsorption of organic solvent molecules.<sup>5</sup>

Likewise, adsorption is known to occur on liquid metal droplets dispersed in many types of solvents, regardless of the polarity of the liquid media. When liquid metal microdroplets are sonicated in different solvents, the carbon/oxygen and carbon/gallium atomic percent ratios on the surface of the oxide layer are known to increase with the solvent's oxygen solubility.<sup>5</sup> For instance, alkanes facilitate a large fraction of dissolved oxygen molecules, which are conducive to the formation of the surface oxide. Therefore, Galinstan–alkane emulsions contain large percentages of carbon atoms despite the relatively weak interactions of alkanes with the surface groups (e.g., –OH) on the oxide layer. On the other hand, acetone has a lower oxygen solubility compared with alkanes. In spite of the stronger interactions between acetone and the oxide layer, the relative atomic percentage of carbon is lower than what is measured for Galinstan–alkane systems.<sup>5</sup> This suggests that the degree of oxidation is the decisive factor in secondary adsorption, where the oxide layer serves as an

anchor for organic solvent molecules. Furthermore, the liquid metal layer's nonpolar surface energy displays a logarithmic relationship with the oxygen solubility of the solvent used to treat the liquid metal surface.<sup>5</sup>

**2.2. Electrical Control of Interfacial Tension.** Voltage-driven control methods (e.g., electrocapillarity, continuous electrowetting, electrowetting-on-dielectric, and electrochemically controlled capillarity) can be implemented to manipulate liquid metal droplets at sub-millimeter length scales. In these techniques, charged species or chemical species are driven toward the interface to lower the interfacial tension between liquid metal droplets and their surrounding environment. Electrical approaches are advantageous in that they require low power and are scalable to microsystems. Since one can control the magnitude of the applied voltage, electrical methods facilitate the precise regulation of interfacial tension.<sup>7</sup>

Introducing either a positive or negative electrical potential at the boundary between two fluids results in a change in interfacial tension. This is a method known as electrocapillarity, which is a classical approach for tuning the effective interfacial tension of a liquid metal droplet within an inert electrolyte. By applying a small electrical potential ( $\sim 1$  V) to the droplet relative to a counter electrode, we can change the density of charge in the electrical double layer at the metal–solution interface. The surface area of liquid metal increases to reduce the capacitive energy at the surface, concomitantly changing the effective interfacial tension. The presence of an electrical potential flattens down the droplet's geometry, as gravitational effects come into play.<sup>7</sup>

The basic principles from electrocapillarity can be extended to electrowetting, which refers to the change in wettability between a given liquid (e.g., liquid metal) and a specific material. When the material is fluidic, we observe a continuous change in the wetting properties (i.e., continuous electrowetting). Let us conceive of a microchannel framework containing an aqueous solution, where a thin liquid layer exists between the metal droplet and the capillary walls. Applying an external electric field to the liquid metal droplet allows continuous electrowetting to take place; if we introduce a voltage difference between each end of the microchannel, a potential drop develops through the liquid layer. Due to the asymmetry in the electrical double layer, a differential in surface tension builds up across the surface of the droplet. Although continuous electrowetting is an effective method for the modulation of liquid metal–liquid interfaces, applying an excessive voltage leads to electrolysis reactions. Electrolysis accompanies the formation of hydrogen bubbles at the cathode, which can be problematic for microfluidic systems. High voltages may additionally cause liquid metal droplets to split up inside of microchannels.<sup>7</sup>

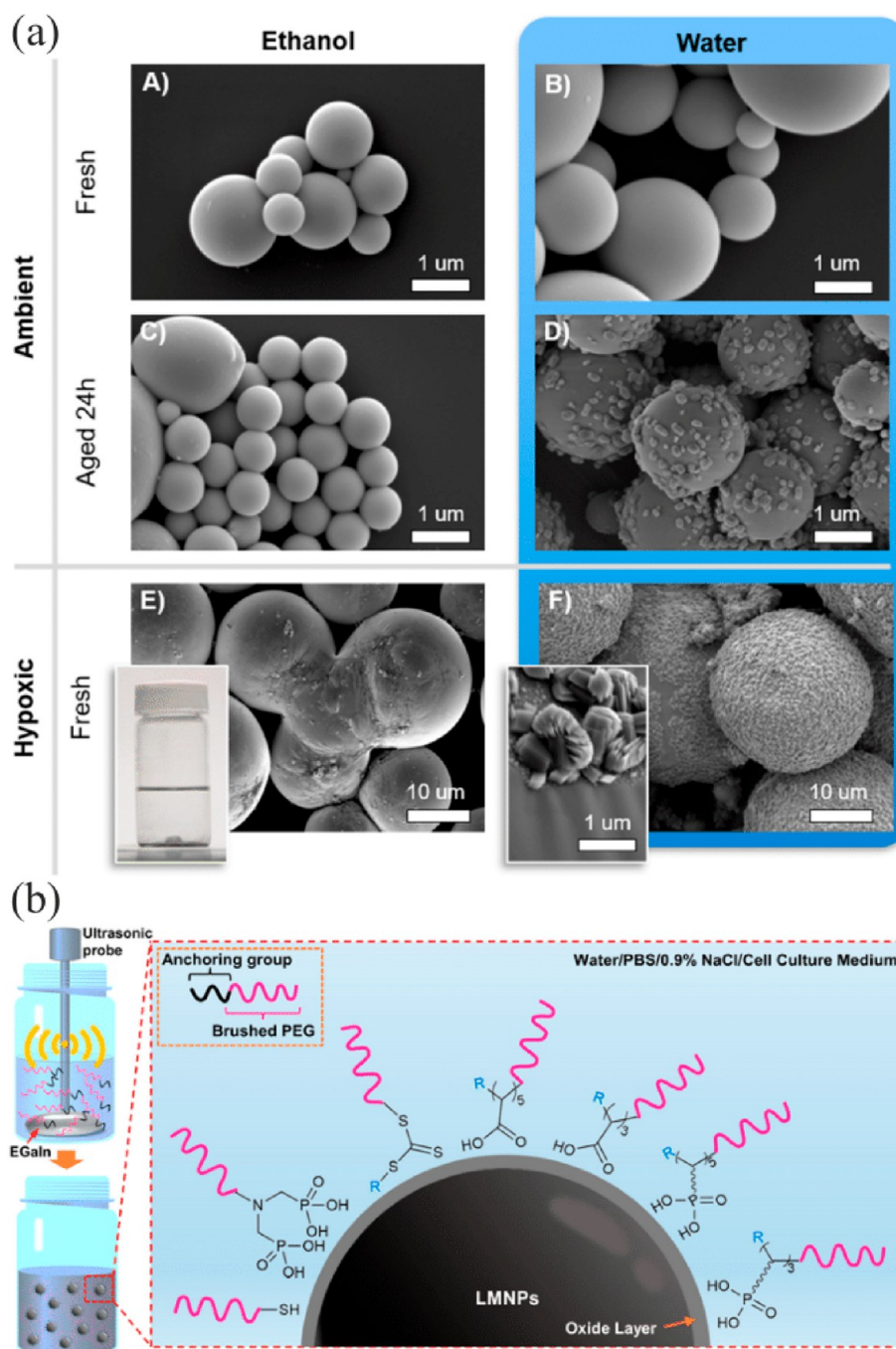
On the other hand, we can consider the case where electrocapillarity changes the wetting properties between a liquid and a solid. This phenomenon is known as electrowetting-on-dielectric. For electrowetting-on-dielectric techniques, an insulating layer is added between the liquid and a conductive substrate. Unlike electrocapillarity, continuous electrowetting, and electrochemically controlled capillarity, electrowetting-on-dielectric techniques do not require the use of electrolytes. Moreover, while the three other methods only require a small electric potential, a large voltage ( $>100$ s of volts) must be supplied to overcome the large surface tension of liquid metal. Applying a large voltage can theoretically change the interfacial tension of a liquid metal droplet on a

substrate, thereby allowing the active control of its equilibrium geometry. However, dielectric breakdown effects limit the magnitude of the electric field that can be applied to the system. Another issue is that liquid metal droplets with oxide layers cannot be effectively controlled via electrowetting-on-dielectric methods. Since conductive materials should not be introduced in electrowetting-on-dielectric techniques, it is also undesirable to remove the oxide via aqueous acids/bases.<sup>7</sup>

Electrochemical approaches (e.g., galvanic replacement reactions) can be adopted to reduce the surface tension of liquid metal. Electrochemically controlled capillarity is one method that uses surfactants to inject/withdraw liquid metal from electrolyte-filled capillaries to selectively grow and remove the oxide membrane. This method is unique in that it utilizes chemical reactions to modify the surface properties of liquid metal micro- and nanodroplets. Notably, the interfacial energy between liquid metal droplets and the fluid environment can be significantly reduced by electrochemically depositing oxide species. During the deposition process, redox reactions in sodium hydroxide have been shown to effectively dissolve oxide species.<sup>7</sup> Using this idea, microfluidic systems composed of liquid metal droplets can control the flow rate by adjusting the percentage of surface oxides. However, injecting liquid metal inevitably leads to the formation of a surface oxide, which necessitates the continuous introduction of acids/bases to the system. Neutral electrolytes also lead to a buildup of the oxide membrane, which hinders the flow of the liquid metal droplet. In addition, electrochemical reactions on the surface of liquid metal droplets accompany half reactions at counter electrodes. This leads to the formation of bubbles that can be undesirable for practical applications.<sup>7</sup>

**2.3. Surface Functionalization.** We have discussed how the oxide layer on the surface of a liquid metal serves as an anchor for organic solvent molecules.<sup>5</sup> Likewise, specific functional groups can be anchored to liquid metal droplets to alter their interfacial properties. For example, thiolated molecules can functionalize the surface of liquid metal nanodroplets. Thiols tend to compete with oxygen atoms on the empty sites of metallic surfaces. This property can be used to suppress the growth of the oxide layer on the surface of liquid metal. We may also modulate the thermodynamics of oxide growth by choosing different types of thiols to functionalize the surface.<sup>6</sup>

Aggregation in colloidal systems occurs because the Gibbs free energy is reduced by decreasing the total surface area. Such an aggregation of particles may be inhibited by electrostatic repulsion or steric hindrance. While the zeta potential (i.e., electrostatic repulsion between adjacent droplets in fluid media) of liquid metal droplets is affected by numerous factors (e.g., pH, temperature, and concentration of charged particles), a substantial portion of the colloidal stability is governed by steric effects.<sup>4,5,10</sup> The native oxide skin on the surface of liquid metal is known to prevent the merging of droplets. In the case of liquid metal droplets immersed in solvents with high oxygen solubility, it has been suggested that the carbon layer plays a role similar to that of a surfactant; the thin solid layer acts to further stabilize the liquid metal emulsions in the liquid medium.<sup>4,5</sup> On the other hand, liquid metal droplets dispersed in water often fail to maintain colloidal stability for longer periods of time. This can be partially attributed to the relatively low oxygen solubility of liquid metal–water systems, as well as their incapability to form carbon adsorbates. Moreover, dispersing liquid metal in

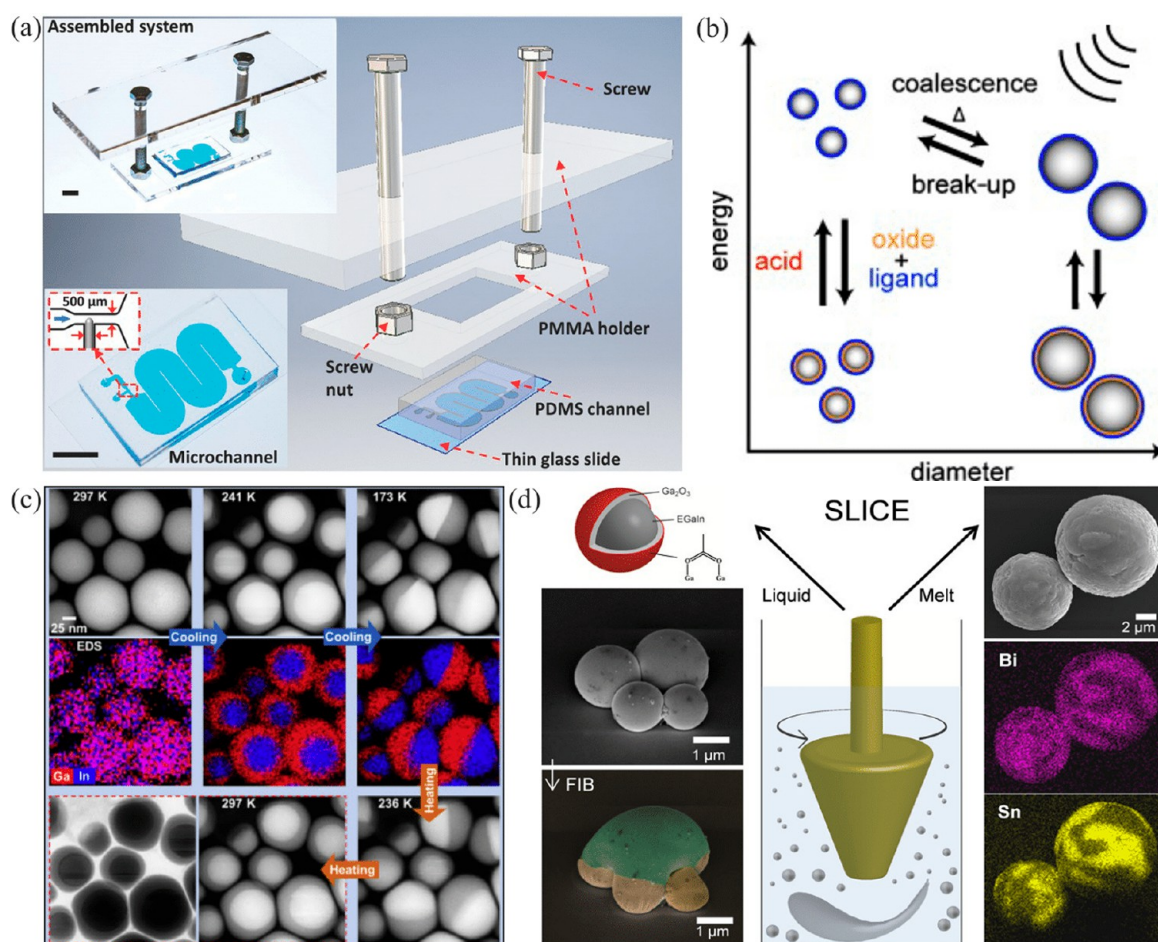


**Figure 2.** (a) Demonstration of solvent effects on liquid metal droplets. Suspensions of EGaIn liquid metal droplets, synthesized in ethanol or water under different conditions. Reprinted with permission from ref 8. Copyright 2020 American Chemical Society. (b) Production of polymer-grafted liquid metal nanodroplets in polymer solutions via ultrasonication. The colloidal and chemical stabilities of liquid metal nanodroplets grafted by brushed polymers (bPEG) bearing a variety of anchoring groups were systematically investigated in biological buffers. Reprinted with permission from ref 11. Copyright 2022 American Chemical Society.

water for longer periods of time may lead to additional chemical reactions. Since gallium is a strong Brønsted acid, the metal is easily deprotonated and exists as an ion in water. As oligomerization occurs, metallic species can precipitate as hydroxide crystals.<sup>8</sup> This may significantly reduce the stability of liquid metal–water systems in the long term, as presented in Figure 2a.<sup>8</sup>

Polymer brushes are macromolecular structures with polymer chains that use bond linkages to tether to various surfaces. The addition of polymer brushes (e.g., polymeric

surfactants or hydrophobic molecules) can further enhance the colloidal and chemical stability of liquid metal micro-/nanodroplets dispersed in various solvents.<sup>9</sup> Bulk droplets generated by ultrasonication or mechanical agitation initially rupture and break down into smaller droplets. During this stage, the juvenile surfaces of liquid metal droplets tend to constantly disintegrate and redevelop. The addition of polymeric or molecular surfactants allows anchoring groups to attach in situ during the fabrication process. In this process,



**Figure 3.** (a) Exploded schematic representation of the platform for on-chip production of liquid metal nanodroplets. The upper inset shows the assembled system, while the lower inset shows the microfluidic chip with a T-junction. Reprinted with permission from ref 13. Copyright 2018 John Wiley & Sons. (b) An ultrasonication method for reversibly tuning the size of gallium nanodroplets. Reprinted with permission from ref 15. Copyright 2015 John Wiley & Sons. (c) Phase separation behavior of gallium-based liquid metal nanodroplets. The particles transform into solid Janus nanoparticles after temperature cycling. Reprinted with permission from ref 16. Copyright 2019 Elsevier. (d) Synthesis of liquid metal core-shell particles via shearing liquids into complex particles (SLICE). Reprinted with permission from ref 17. Copyright 2014 American Chemical Society.

the simplest method to enhance the colloidal stability of the droplets is to increase the number of anchoring groups.<sup>9</sup>

In general, high reaction temperatures are required to anchor polymer brushes to the surface of metallic particles. However, liquid metal micro- and nanodroplets allow similar interactions to occur at much lower temperature regimes. This means that the polymer–metal coordination is weaker than that of colloidal systems based on solid metal particles.<sup>9</sup> Especially in the case of aqueous solvents, water molecules display a strong affinity toward the metal cations present in the system. When there is an excess of water in the liquid medium, the surface coating of stabilizing polymers may be affected by the attraction between water and metal ions.<sup>8,9</sup> To resolve these issues, various methods have been established for the stabilization of liquid metal droplets in the emulsion state. In a recent study, researchers introduced hydrochloric acid and polyvinylpyrrolidone (PVP) to a water–ethanol binary mixture containing liquid metal droplets.<sup>10</sup> While the acid partially removed the oxide membrane, PVP could effectively suppress the coalescence of liquid metal droplets dispersed in the mixture. During solvent evaporation, the presence of PVP enhanced the steric forces between individual liquid metal droplets, which subsequently dried into a uniform layer.<sup>10</sup>

Attaching low concentrations of well-defined diblock copolymers to the oxide skin can form polymer brushes that are highly effective for the stabilization of liquid metal nanodroplets.<sup>11</sup> Polymer brushes are especially useful in that blocks with multiple coordination sites can attach to the surface of liquid metal droplets. These diblock copolymers can be prepared via atom-transfer radical polymerization to establish flexible brush architectures, controllable molecular weight, and narrow molecular weight distribution. Introducing even a small concentration of polymer brushes can effectively prevent the agglomeration of nanodroplets.<sup>9–11</sup> Compared with small-molecule surfactants, block copolymer brushes at the surface of liquid metal droplets are known to significantly improve the colloidal stability as well as the yield of liquid metal droplets.<sup>11</sup>

Reversible addition–fragmentation chain transfer is a common polymerization method used to incorporate a wide variety of anchoring groups into brush polymers (Figure 2b).<sup>9</sup> Polymers bearing phosphonic acids can promote chemical stability by passivating the surface of liquid metal droplets.<sup>9,11</sup> When introduced in aqueous systems, they are also capable of maintaining the morphology of liquid metal droplets at room temperature.<sup>9</sup> Brushes with multidentate phosphonic acid

groups can effectively functionalize liquid metal interfaces with complex fluids, such as those used for biological applications.<sup>11</sup> Liquid metal nanodroplets grafted with nonphosphonic acid anchoring groups (e.g., trithiolcarbonates, thiols, and carboxylic acids) are also widely used to stabilize liquid metal based colloidal systems. For instance, brushes based on carboxylic acids use multiple coordination interactions to firmly attach onto oxides at the surface of a liquid metal droplet. However, such anchoring groups may be unable to prevent the spontaneous shape transformation of liquid metal droplets at room temperature.<sup>11</sup>

### 3. FABRICATION OF LIQUID METAL DROPLETS

Fabricating liquid metal micro- and nanodroplets is crucial for the design of liquid metal-based polymer composites,<sup>1–3,12</sup> microfluidic systems,<sup>13,14,18</sup> and flexible/stretchable or wearable devices.<sup>1–3,12</sup> In this section, we present the various methods that can be exploited for the fabrication of liquid metal micro- and nanodroplets.

**3.1. Microfluidics.** A microfluidic platform can be used to mass-produce nanoparticles with tunable size distributions on a chip (see Figure 3a).<sup>13</sup> Notably, monodispersed spherical liquid metal droplets have been achieved by introducing a three-channel F-junction generator. By employing this setup for dielectrophoresis, one of the branch channels could provide a flow of sodium hydroxide to improve maneuverability.<sup>7,14</sup> Considering that liquid metal-filled microchannels serve as noncontact electrodes, the electrical field created throughout the channel could automatically sort the liquid metal droplets.<sup>14</sup> After the liquid metal droplets are produced, such droplets can be hydrodynamically transferred through a liquid-to-liquid interface into other fluidic components within a microelectromechanical system (MEMS) chip.

**3.2. Ultrasonication.** Ultrasonication uses sound to generate vapor cavities which can nebulize bulk liquid metal within the aqueous media (see Figure 3b).<sup>15</sup> The competition between breakup and coalescence of liquid metal droplets may be controlled by precise monitoring of the fabrication process. Droplets of diverse geometries can be obtained by controlling the chemical environment, temperature, and power supply during sonication.<sup>15</sup> While droplet atomization continues to occur, the surface oxide layer on liquid metal droplets can be removed by adding a small concentration of acids or bases. On the other hand, polymeric/molecular surfactants or rheology additives (e.g., colloidal clays such as laponite) interact with the metallic surface to stabilize the droplets from coalescence during ultrasonication.<sup>10,15</sup>

Ultrasonication generates a substantial amount of heat, which may act as a double-edged sword. The use of a highly volatile solvent or poor sealing can lead to evaporation of the liquid medium during sonication. To minimize the effects of excess heat, it is generally required to turn on a chiller to modulate the temperature of the liquid media. However, in certain instances, inducing a change in temperature can be used as a viable alternative to precisely control the fabrication process.<sup>16</sup> A temperature cycling process during ultrasonication was reported as an effective method to transform gallium-alloy-based nanodroplets into solid Janus nanoparticles (see Figure 3c). Such results could be achieved by inducing the phase separation of gallium and other metallic species within the eutectic alloy.<sup>16</sup>

**3.3. Mechanical Agitation.** The droplet emulsion technique has been introduced as a tunable method for the

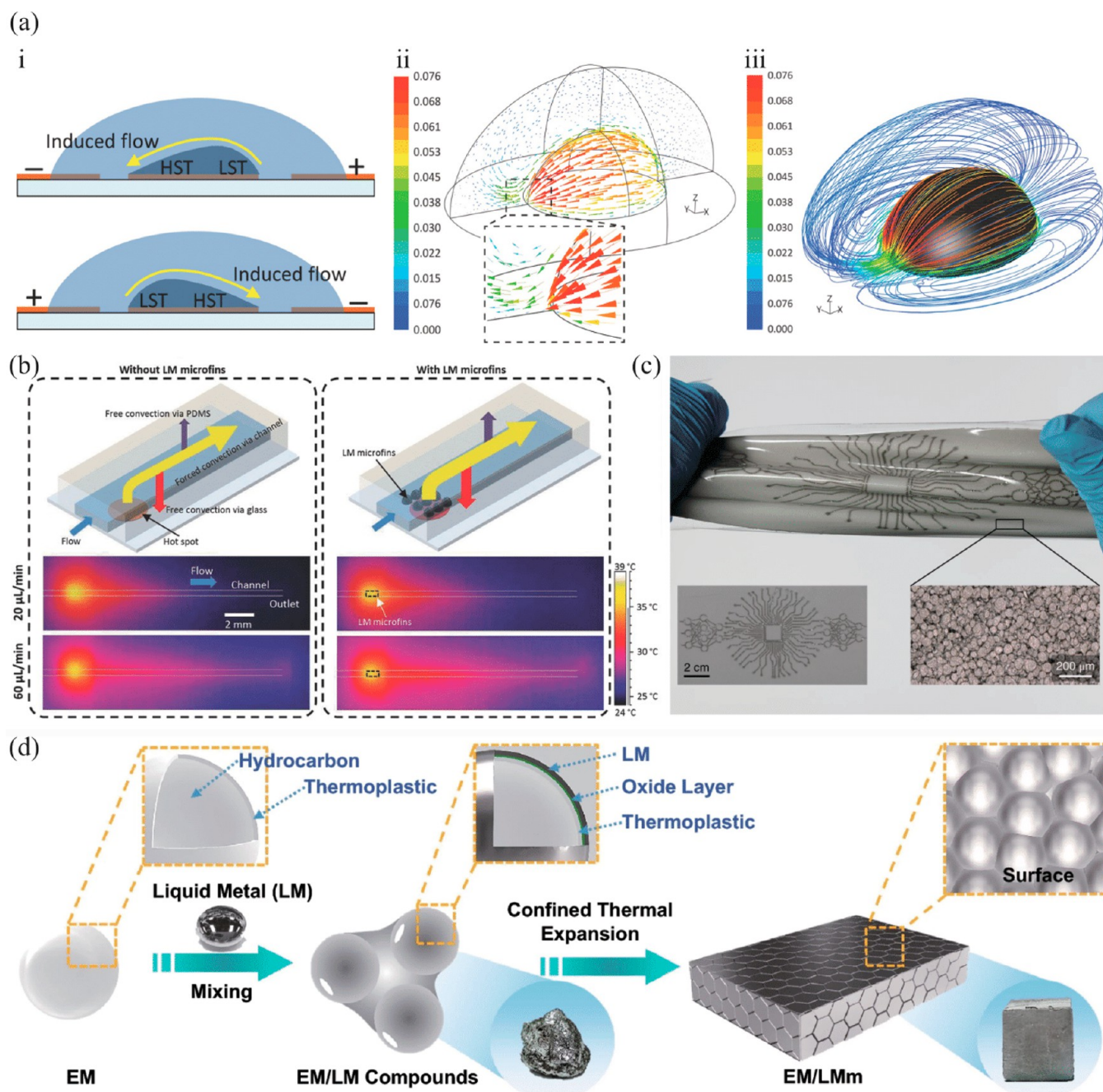
design of metallic particles. By shearing molten metals in the presence of a carrier fluid, researchers have been able to design a variety of complex micro- and nanoparticles (e.g., core–shell, hard/soft, and smooth/patchy).<sup>17</sup> As a column of liquid metal is locally elongated, it breaks up into uniform satellite droplets, where oxidation and additional functionalization produces droplets with tunable morphologies (see Figure 3d). When emulsion shearing is performed within an acidic carrier fluid environment, surface tension effects drive the phase segregation of the elements constituting the liquid metal alloy.<sup>17</sup> By controlling the process of phase separation, it is possible to fine-tune the surface composition and morphology of droplets. Optimal shearing speeds and carrier fluids may be chosen to design particles of desired sizes or shapes.<sup>17</sup>

### 4. APPLICATIONS OF LIQUID METAL DROPLETS

In the previous sections, we have investigated systems involving the interfaces of micro- and nanoscale liquid metal droplets with a continuous phase, as can be seen in the case of microfluidics, emulsions, or droplets with surface modification. Such droplets are being widely explored for the fabrication of state-of-the-art devices, such as flexible/stretchable electronics,<sup>1,4</sup> polymer composites,<sup>1–3</sup> and soft robotics.<sup>2</sup> Liquid metal droplets can also serve as the basic building blocks for the design of novel materials, such as catalytic particles or nanocarriers in biomedical applications. Owing to their high surface-to-volume ratios, micro- and nanodroplets can be utilized as a platform to fabricate catalytic materials directly from the liquid phase.<sup>22,23</sup> The low toxicity of liquid metal droplets allows them to be incorporated in biological applications, including biomedicines or drug delivery systems.<sup>24,25</sup>

**4.1. Microfluidics.** While bulk-scale liquid metal (i.e., scales of several millimeters or centimeters) can be utilized for device applications, liquid metal in the form of micro- and nanodroplets accompanies a variety of merits. To begin with, the ultrahigh surface tension (500–700 mN/m)<sup>4,5</sup> and oxide membrane makes it challenging to control metallic liquids at the bulk scale. Liquid metal streams ejected through nozzles readily destabilize due to oxidation and surface tension effects. For patterning or microfluidic/MEMS applications, it is difficult to fit bulk-scale liquid metal through smaller grooves or channels. Despite the high surface tension of liquid metal, micro- or nanodroplet suspensions may alleviate the difficulties of fitting bulk liquid metal into the grooves of microchannels.<sup>18</sup> In the form of micro- or nanodroplets, it is easier to control volumes of liquid metal with higher degrees of precision and controllability.<sup>18–20</sup>

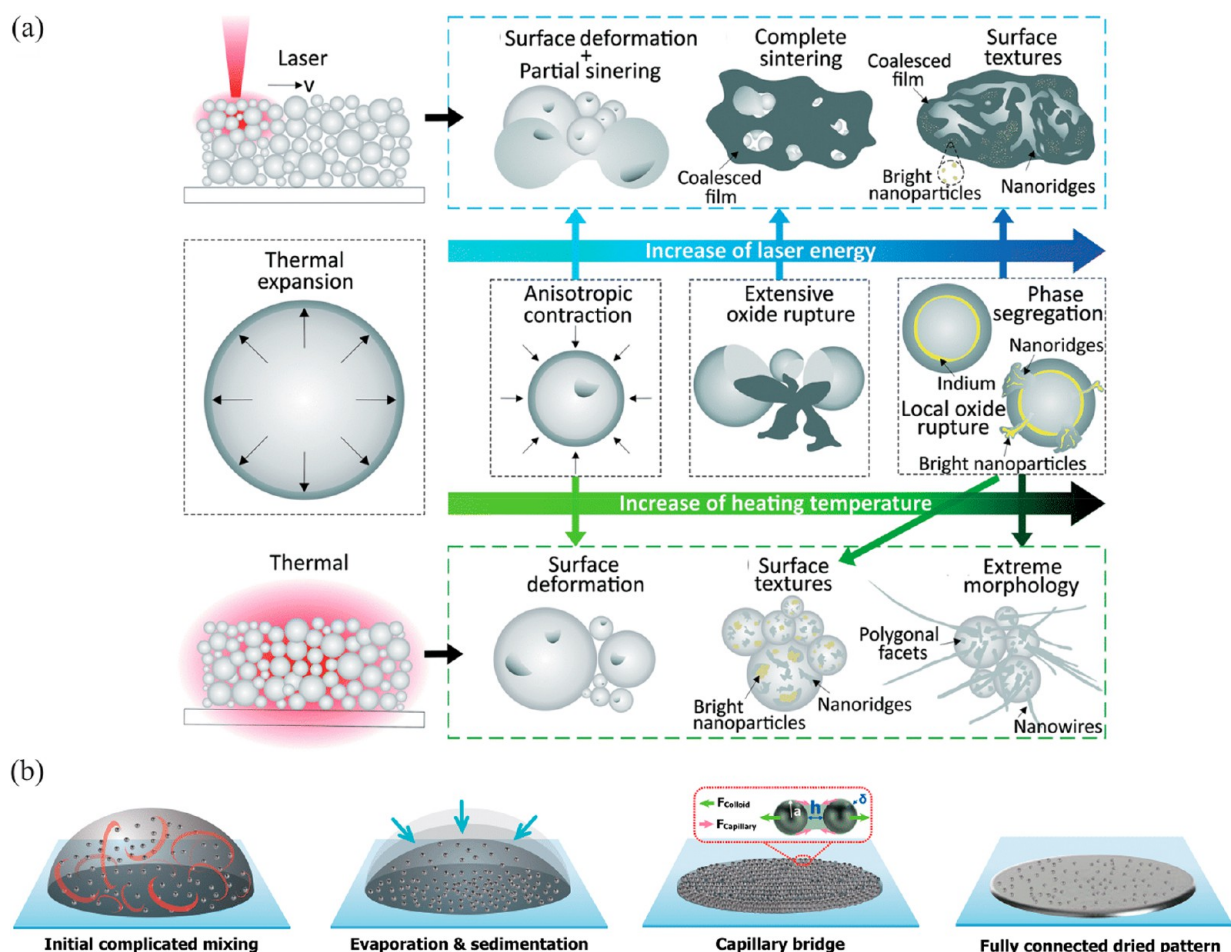
Micro- and nanoparticle-laden systems are effective carrier fluids due to their high thermal and electrical conductivity.<sup>18</sup> By modulating the oxide layer, we can also tune the interfacial and rheological properties of liquid metal droplets for miniaturized microfluidics (e.g., pumps, valves, heaters, and electrodes).<sup>18–20</sup> For instance, inducing a charge gradient on the surface of the liquid metal droplet is known to generate surface-tension-driven Marangoni flows. Since the metal spheres are fixed in space, it is possible to create a pump when electrolytes travel across the spherical surface.<sup>19</sup> One study succeeded in creating harmonic Marangoni flows by inputting a sinusoidal signal into a solution with liquid metal droplets. By generating vortices, the resulting chaotic advection allowed a highly efficient mixing process within the microfluidic system, as shown in Figure 4a.<sup>19</sup>



**Figure 4.** Applications using liquid metals for microfluidics and soft electronics. (a) (i) Liquid metal actuator. (ii) Flow velocity vectors (m/s) along the Galinstan surface. (iii) Formation of vortices within the surrounding liquid colored by magnitude of flow velocity (m/s). Reprinted with permission from ref 19. Copyright 2014 John Wiley & Sons. (b) Three-dimensional (3D) Galinstan microstructures for heat dissipation from a hot spot. Comparison of temperature contours for the microfluidic platform in the channel with liquid metal components. Reprinted with permission from ref 20. Copyright 2015 John Wiley & Sons. (c) Liquid metal–elastomer composites being stretched and twisted with an intricate design of electrically conductive traces. The lower left inset shows the undeformed sample and the lower right inset is an optical micrograph showing the liquid metal microdroplets in the elastomer. Reprinted with permission from ref 2. Photograph courtesy of Eric Markvicka. Copyright 2018 Springer Nature. (d) Illustration of the fabrication process of expandable microsphere/liquid metal monoliths for electromagnetic interference (EMI) shielding. Reprinted with permission from ref 3. Copyright 2022 Springer Nature.

Liquid metal droplets can also be used to trap suspended particles or enhance convective heat transfer processes within a microfluidic channel. In one study, dielectrophoresis was used to immobilize Galinstan microstructures on planar microelectrode pads.<sup>20</sup> This method allowed researchers to fabricate patterns of three-dimensional microstructures of various dimensions and aspect ratios, which could be further tuned to trap nanoparticles flowing through a microfluidic channel. In

a separate demonstration, three-dimensional liquid metal microfin arrays could be placed over a hot spot to dissipate heat within the channel (see Figure 4b).<sup>20</sup> The microstructures could enhance the convective heat transfer process by directing more heat into the liquid stream traveling through the channel.<sup>20</sup> Such microfluidic systems can be integrated into mechanically adaptive materials for the design of soft diodes, transient circuits, and biomedical sensing chips.

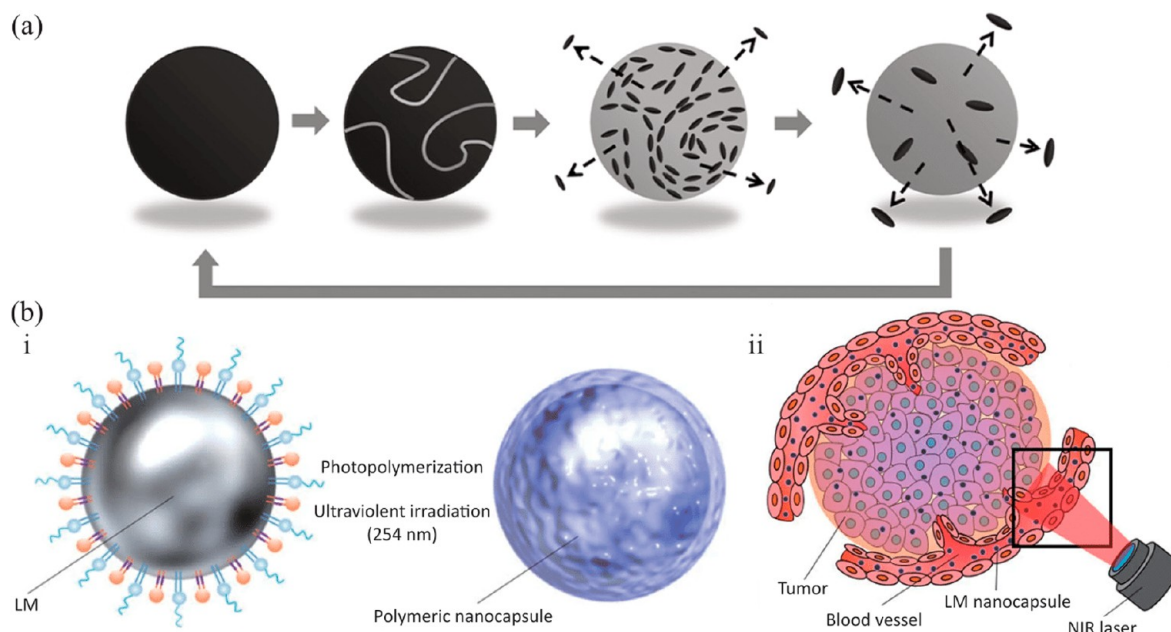


**Figure 5.** (a) Schematic for mechanisms of laser and thermal sintering liquid metal nanoparticle films. Reprinted with permission from ref 21. Copyright 2019 Royal Society of Chemistry. (b) Formation of a self-sintered uniform liquid metal pattern using sintering-free ink. The dried pattern is fully connected after the avalanche coalescence of liquid metal particles due to capillary adhesion. Reprinted with permission from ref 10. Copyright 2022 John Wiley & Sons.

**4.2. Soft Electronics and Robotics.** Introducing liquid metal into a system does not degrade the general properties (e.g., stretchability, elastic compliance, and dielectric breakdown strength) required for soft-matter engineering.<sup>1,2,4,10</sup> Since liquid metal exhibits both electrical conductivity and fluidity, it is possible to construct conductive networks of liquid metal droplets with dynamic responses. While external deformations such as bending or stretching are prone to damage solid materials, the use of liquid metal can overcome this lack of durability.<sup>1,4</sup> This means that liquid metal-based structures can undergo continuous changes in cross-sectional area, maintaining their electrical conductivity at the same time. Likewise, liquid metal-based electrodes patterned over a polymer structure can resist considerable strain without destabilizing into shorter ligaments.<sup>2,10</sup> Postfabrication treatments allow liquid metal droplets to further merge into percolated networks.<sup>4,12</sup> Based on these micro- and nanoscale building blocks, we can design complex systems involving the interfaces of liquid metal with a continuous phase (e.g., liquid metal based inks, emulsions, and soft composites). It is possible to configure a wide range of material properties from the different combinations of liquid metal–solvent or liquid metal–polymer interfaces that can exist; this feature highlights the versatility of liquid metal micro- and nanodroplets.

In this regard, liquid metal–elastomer nanocomposites with dielectric properties are highly promising for flexible and stretchable devices. Conventional liquid metal–elastomer systems (e.g., microfluidic circuits, liquid metal embedded elastomers through dispersions, drop-cast films, and backfilled cellular structures) may often lead to electrical shorting due to unintended activation. Employing liquid metal droplets dispersed in an elastomeric phase allowed researchers to design high-performance electric circuits/soft robotic devices with autonomous and instantaneous self-healing characteristics (see Figure 4c).<sup>2</sup> By dispersing liquid metal droplets dispersed in an elastomeric phase, the composite exhibited a significant level of stress shielding that was effective in preventing electrical shorting issues. Even when subject to spatially random damage, the self-healing architecture helped the electrical wiring to maintain its functionality. An additional elastomeric sealing layer or selective patterning techniques could be applied to protect the liquid metal circuit and inhibit inadvertent activation between adjacent pathways. Liquid metal droplets have also gained much attention as composite fillers for electromagnetic interference (EMI) shielding applications.<sup>3</sup> In order to construct conductive networks, expandable microspheres with thermal self-expanding properties were incorporated with liquid metal droplets (see Figure 4d). A mechanical mixing process continuously generated a





**Figure 6.** Applications using liquid metals for catalysts and biomedicines. (a) Sketch of transformation of liquid metal/metal oxide spherical structures during sonication. Gray shapes, black shapes, and black ovals stand for Galinstan spheres, oxide layers, and nanoplatelets, respectively. Reprinted with permission from ref 22. Copyright 2014 John Wiley & Sons. (b) (i) Polymeric core–shell structure encapsulating liquid metal photopolymerized using functional phospholipids with 254 nm ultraviolet irradiation. (ii) Schematic for laser-triggered liquid metal transformation in a blood vessel model. Reprinted with permission from ref 25. Copyright 2017 Springer Nature.

new oxide membrane, which allowed the microspheres to be coated with liquid metal. The self-healing properties of these networks are expected to resolve the leakage issues of conventional electromagnetic sealing interfaces.<sup>3</sup>

However, the oxide membrane can act as an insulating barrier between individual liquid metal droplets. This means that additional manufacturing processes may be required to create electrically conductive pathways between individual droplets. Various sintering methods have been proposed for the design of soft and deformable circuit interconnects based on liquid metal inks.<sup>4</sup> Since liquid metal droplets can be easily merged under sufficient pressure, mechanical sintering allows the fabrication of self-healing electronic devices. For example, we can consider a polymer composite containing droplets of liquid metal. When this composite is sliced in half, the droplets will initially be smeared along the side walls, later creating a conductive path to restore their intrinsically high conductivity.<sup>12</sup> These droplets can rupture to form new connections with neighboring droplets and effectively reroute electrical signals.<sup>2,12</sup>

Thermal and laser sintering methods have also been proposed to rupture the oxide membrane and reconnect the liquid metal droplets (see Figure 5a).<sup>21</sup> In the case of thermal sintering, only a small portion of the oxide layer is affected by thermal stress. This local rupture is followed by a strong phase segregation process, which contributes to the formation of solid-like particle films.<sup>21</sup> Meanwhile, laser sintering accompanies an extensive rupture of the oxide membrane. This allows the cores of liquid metal particles to interconnect and coalesce into conductive networks.<sup>21</sup> In recent years, sintering-free liquid metal inks have been successfully prepared by adding a certain volume of acids and polymers into the continuous phase.<sup>10</sup> Such inks can be suspended in soft elastomers to design composites with spontaneous self-repairing characteristics.<sup>10</sup> Even when the matrix is significantly

damaged, liquid metal droplets can rupture to form interconnections with neighboring droplets. Without additional manual reapplying or heating processes, electronic signals can be reconfigured to achieve an electronically robust network as shown in Figure 5b.<sup>2,10</sup> This percolated path is created throughout the elastomeric matrix due to the merging of liquid metal droplets.<sup>4</sup>

**4.3. Catalysts.** Compared with bulk materials, micro- and nanoparticles exhibit high surface areas and high surface-to-volume ratios. This makes small-scale structures particularly useful for catalyst applications that tend to be highly dependent on surface reactions. Liquid metal micro- and nanodroplets are effective platforms for designing such catalytic materials. Liquid metal-based particles are especially unique in that they originate from the liquid phase; the methods for controlling their surface morphologies and catalytic reactions are different from those applied to conventional solid particles.<sup>22,23</sup> Micro- and nanosized liquid metal droplets can be combined with nanosized metal oxides to create liquid metal/metal oxide frameworks. Studies have shown that it is possible to actively control the diameter and stoichiometry of these spherical structures. When metallic droplets are prepared in the form of liquid suspensions, we can specifically tune the plasmon resonance wavelength of the catalytic frameworks to be designed. The resulting spheres display high sensitivity toward heavy-metal ions and excellent photocatalytic properties.<sup>22</sup>

In one study concerning photochemical catalysts, researchers focused on the photocatalytic performance of stoichiometric gallium oxides ( $\text{Ga}_2\text{O}_3$ ) that could decompose different pollutants and organic dyes under ultraviolet (UV)/visible-light irradiation.<sup>23</sup> The researchers developed a novel fabrication method to directly synthesize the oxide from liquid-state gallium droplets. The self-limiting oxide skin of liquid gallium was implemented as a precursor for suspensions

of gallium oxide nanoparticles. After sonicating gallium in water, annealing the liquid metal–water suspension produced rods/nanoflakes of gallium oxide as presented in Figure 6a. The harvested nanoflakes were shown to contain trap states under the conduction band minimum, thereby effectively reducing the energy band gap. Under irradiation of solar light, such structures could be used as photocatalysts for the decomposition of organic model dyes.<sup>23</sup>

**4.4. Biomedicines.** In the field of biomedicines and drug delivery, many types of inorganic nanocarriers exhibit systemic toxicity while lacking in biodegradability. Moreover, it is difficult to tailor their physicochemical properties to satisfy the conditions required for target delivery and efficient elimination. Due to their low toxicity, gallium-based liquid metal micro- and nanodroplets are being implemented for various healthcare applications.<sup>24,25</sup> In addition, the transformable and degradable nature of liquid metal nanocapsules enables them to effectively regulate the drug release process.<sup>24</sup> In this regard, novel methods are being explored to attach biopolymers and enzymes onto the activated surfaces of liquid metal nanodroplets.

Nanomedicines based on liquid metal droplets carry distinct advantages: droplets are simple to fabricate, allow the bioconjugation of functional molecules, and are capable of fusing/degrading in mildly acidic environments. One group of researchers created a liquid metal-based drug delivery platform for anticancer therapy.<sup>24</sup> By using ultrasonication, drug-loaded nanodroplets of liquid metal could be formed via ligand-mediated self-assembly. In mildly acidic cellular environments, these liquid metal nanodroplets were shown to easily degrade by fusing with one another. After cellular internalization, the loaded chemical could be released at the opportune moment. Data from X-ray images also suggested that the fused nanospheres could serve as theranostic reagents.<sup>24</sup> Additional properties intrinsic to liquid metal droplets (e.g., mechanical flexibility, electrical conductivity, and the ability to dissolve other metallic species) may be investigated for the development of new materials/scaffolds for drug delivery and tissue engineering.

With proper functionalization, liquid metal droplets can facilitate the design of nanocapsule configurations. Photopolymerized liquid metal droplets prepared with functional phospholipids were shown to produce nanocapsules with low toxicity and high dispersibility in water.<sup>25</sup> Under near-infrared (NIR) light, these capsules were capable of generating heat, simultaneously producing reactive oxygen species. NIR laser irradiation could disrupt the surface of these capsules, which consequently transformed their equilibrium geometry.<sup>25</sup> This configuration allowed the contactless release of loaded drugs with exceptional controllability. The study further demonstrated the chemotherapeutic capability of liquid metal nanodroplets functionalized with a tumor-targeting ligand (Figure 6b).<sup>25</sup> By inhibiting the formation of tumors, these droplets are expected to serve as nontoxic theranostic drug delivery systems. Such liquid metal nanodroplets have been utilized as markers for spatiotemporal targeting for the X-ray-enhanced imaging of biological organs. These nanostructures can also be beneficial when optical manipulation is required for microfluidic blood vessel systems. Moreover, the researchers demonstrated that liquid metal droplets could be used to eliminate cancer cells and control the calcium ion flux between cells.<sup>25</sup>

## 5. FUTURE OUTLOOK

Liquid metal is a truly unique material in that it exhibits a metallic nature while simultaneously displaying fluidic properties. Unlike conventional liquids, the metallic nature of liquid metal droplets gives rise to a number of unique characteristics. The metallic bonding forces in liquid metal contribute to extremely high values of surface tension.<sup>1,5</sup> This surface is prone to interact with oxygen/adsorbate molecules, which subsequently alter the geometry, chemistry, and surface energy of liquid metal droplets.<sup>5</sup> Therefore, a thorough understanding of liquid metal surfaces is critical in designing droplets with tunable interfacial, morphological, and rheological properties. Liquid metal droplets of specific geometries or surface wettabilities can be achieved by subjecting droplets to acid/base treatment,<sup>3,10</sup> making use of electric fields,<sup>7</sup> or performing experiments in vacuum environments. Such droplets can be further incorporated into electrodes/microchannels that are elemental for the fabrication of various emerging technologies. For example, liquid metal micro- and nanodroplets are promising materials for future-generation soft composites,<sup>1–3</sup> printable circuits,<sup>4</sup> micro-/nanofluidics,<sup>13,14,18</sup> catalysts,<sup>22,23</sup> and biomedicines.<sup>24,25</sup> Despite the multitude of papers focused on the potential applications of liquid metal droplets, much remains to be explored about the fundamental physicochemical aspects of liquid metal interfaces. The behavior of liquid metal droplets in different fluid environments<sup>5</sup> and surface-functionalization conditions<sup>9,11</sup> have only recently been investigated. Employing these characteristics, researchers must constantly look out for novel approaches to fabricate liquid metal droplets in various environments.

In order to fully appreciate the nature of liquid metal droplets, contemporary researchers should develop a comprehensive perspective across a wide range of fields. For one, chemists must seek effective methods for the production of spherical liquid metal droplets and functionalization of liquid metal-based nanoconjugates. The next milestone will be to improve the synthesis and chemical modification procedures of future-generation structures and complexes based on liquid metal micro- and nanodroplets. At the same time, one may find it thought-provoking to review the classical papers in the field of metallurgy; the well-known material/physical properties of molten metals of historic importance can help gain insight into room-temperature liquid metal alloys. From the viewpoint of rheology and fluid dynamics, we should further explore the non-Newtonian properties of metallic inks/suspensions prepared at different shear rates, temperatures, and chemical conditions. Understanding the dynamics of liquid metal droplets and their flow characteristics in various microchannel configurations will also be essential for practical applications. This directly connects to the role of mechanical and electrical engineers, who are to devise effective MEMS platforms and soft/stretchable circuits that can embody these novel materials. By incorporating the self-healing properties and sintering methods of liquid metal droplets, we must continue to design liquid metal-based soft electronic systems with electrical stability. For biologists, it would be critical to examine the behavior of liquid metal-based nanocomplexes in physiological environments through assays and imaging techniques. Evaluating the biocompatibility, spatiotemporal controllability, and bioactive functionalization of these novel materials remains a quest for liquid metal droplets in biomedical/healthcare applications.

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### Notes

The authors declare no competing financial interest.

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