# Liquid-Phase Gallium–Indium Alloy Electronics with Microcontact Printing

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**ABSTRACT:** Liquid-phase electronic circuits are patterned on an elastomer substrate with a microcontact printer. The printer head dips into a pool of a liquid-phase gallium—indium alloy, e.g., eutectic gallium—indium (EGaIn) or gallium—indium—tin (Galinstan), and deposits a single drop on a silicone elastomer substrate. After patterned deposition, the liquid-phase circuit is sealed with an additional layer of silicone elastomer. We also demonstrate patterned deposition of the liquid-phase GaIn alloy with a molded polydimethylsiloxane stamp that is manually inked and pressed into an elastomer substrate. As with other liquid-phase electronics produced through needle injection or masked deposition, the circuit is elastically deformable and can be stretched to several times its natural length without losing electronic functionality. In contrast to existing fabrication techniques, microcontact printing and stamp lithography can be used to produce circuits with any planar geometric feature, including



electrodes with large planar area, intersecting and closed-loop wires, and combs with multiple terminal electrodes. In air, the surface of the coalesced droplets oxidize to form a thin oxide skin that preserves the shape of the circuit during sealing. This first demonstration of soft-lithography fabrication with liquid-phase GaIn alloy expands the space of allowable circuit geometries and eliminates the need for mold or mask fabrication.

# INTRODUCTION

Liquid-phase electronic circuits are composed of thin sheets of silicone elastomer embedded with microfluidic channels of conductive fluid. In contrast to conventional electronics, these circuits are intrinsically soft and remain functional when stretched to several times their natural length. Early examples of liquid-phase electronics include the Whitney strain gauge,<sup>1</sup> a mercury-filled rubber tube that changes electric resistance in response to hyperelastic stretch, and mercury switches used to measure tilt and rolling. Presently, liquid-phase electronics are produced with nontoxic alternatives to mercury, such as eutectic gallium-indium (EGaIn),<sup>2,3</sup> gallium-indium-tin (Galinstan), and other GaIn alloys that are liquid at room temperature.<sup>4</sup> Figure 1a presents an example of an elastically soft capacitor that is composed of silicone elastomer (Ecoflex 0030; Smooth-On, Inc.) embedded with interdigitated channels of Galinstan. As demonstrated in panels b and c of Figure 1, the capacitor is intrinsically soft and can be stretched or elastically deformed into a non-developable shape.

In this paper, we report a new method to produce liquidphase GaIn electronics that incorporates microcontact printing and stamp lithography. The microcontact printer is composed of a print head mounted to a three-axis Cartesian robot. As illustrated in Figure 2a, patterned circuits are produced by depositing successive droplets of liquid alloy on an elastomer substrate. After deposition and insertion of external wiring, the circuit is sealed with an additional layer of silicone elastomer (Figure 2b). In air, the surface of the GaIn alloy oxidizes and forms a thin oxide skin that preserves the shape of the



Figure 1. (a) Elastically soft capacitor composed of liquid-phase Galinstan wires embedded in Ecoflex 0030 silicone elastomer. The liquid-phase circuit can be (b) stretched and (c) elastically deformed into a non-developable shape without losing electronic functionality.

deposited features during wire insertion and sealing. We also attempt to produce liquid GaIn circuits with stamp lithography. Rather than print circuits droplet by droplet, entire patterns may be transferred with a molded stamp that is manually inked in a pool of GaIn alloy and pressed into an elastic substrate.

# BACKGROUND

Elastomer-sealed circuits of liquid-phase GaIn alloy are currently produced using the casting and needle-injection

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**Figure 2.** (a) Deposition of liquid-phase GaIn with microcontact printing ( $\mu$ CP) and (b) sealed circuit with external copper wires.

steps presented in Figure  $3.^{5-12}$  First, silicone elastomer is cast into a micropatterned mold that is produced with photo-



Figure 3. Fabrication of soft-matter circuit with casting and needle injection.<sup>4</sup> First, (a) a 3D printed mold is (b) filled with uncured silicone elastomer. After the silicone is cured and (c) sealed, the embedded channels are (d) filled with EGaIn using a needle and syringe. Lastly, (e) external wires are inserted into the circuit terminals.

lithography or a rapid prototyping tool [e.g., laser engraver, CNC mill, and three-dimensional (3D) printer] (Figure 3a). Next, the cured elastomer is bound to a sealing layer of additional elastomer (Figure 3b), and the embedded microchannels are filled with liquid GaIn alloy using a needle and syringe (Figure 3c). Lastly, external wires are inserted into the circuit terminals, and the connections are sealed with droplets of silicone elastomer. This fabrication technique represents an extension of methods used in soft microfluidics to embed polydimethylsiloxane (PDMS) with microchannels of fluid.<sup>13–15</sup> In addition to functioning as a stretchable electrical wire<sup>16,17</sup> or strain gauge,<sup>1,5,11</sup> microfluidic channels of liquidphase gallium-indium alloy also function as elastically tunable antenna,<sup>6,7</sup> soft-matter capacitors and inductors,<sup>10,12</sup> and a hyperelastic pressure<sup>8,11</sup> or curvature sensor.<sup>9</sup> Potential applications include soft and stretchable electronics for wearable technologies that monitor human  $motion^{18-20}$  and electronic skin for biologically inspired soft robots.<sup>21</sup> When embedded in a silicone elastomer, microfluidic channels of GaIn alloy function as electrical circuits that are as elastically soft as natural human tissue and as much as 10× stretchable.

While reliable for producing simple circuits with a single inlet and outlet, replica molding and needle injection cannot be used to produce complex patterns involving multiple intersections and terminals or terminal electrodes that cover a large planar area. Moreover, this current fabrication technique is labor- and time-intensive, requiring hours to produce a mold with SU-8 photolithography or rapid prototyping tools and to cast and bond layers of silicone elastomer prior to needle injection. To improve manufacturing scalability and speed, researchers have focused on alternative fabrication methods that could significantly increase speed and allow for automation and scalable production. This includes masked deposition (i.e., stencil lithography), illustrated in Figure 4, in which the GaIn



**Figure 4.** Fabrication of the soft-matter circuit with stencil lithography. (a) A stencil is produced by laser engraving a thin film mask and placed on an elastomer. Next, (b) liquid GaIn alloy is spread over the stencil. Lastly, (c) the stencil is removed, and copper wires are inserted prior to sealing. (d) Unsealed Galinstan heater on VHB elastomer produced with stencil lithography.

alloy is selectively deposited on an elastomer substrate using a patterned stencil mask. This method for producing GaIn circuits was independently discovered by Majidi et al.<sup>22</sup> and Wu et al.<sup>23</sup>

Stencil lithography broadens the range of allowable circuit geometry and enables rapid and potentially automated manufacturing. However, each new circuit design requires its own stencil, and the stencil cannot be used to make circuits with closed-loop features. To deposit liquid alloy into any arbitrary planar geometry, masked deposition must be performed with a sacrificial mask that is produced with photolithography.<sup>24</sup> Promising alternatives to mask deposition include jetting (i.e., inkjet printing), stamp lithography, and microcontact printing. Jetting and microcontact printing are particularly attractive because they are completely automated and can rapidly deposit liquid alloy into any pattern.

With printable inks, conventional inkjet printing produces droplets with diameters as small as  $10-20 \ \mu$ m. Higher resolution is possible with electrohydrodynamic jetting (e-jet printing), which uses an electric field to produce features with dimensions as small as  $1 \ \mu$ m.<sup>25</sup> Although inkjet and e-jet printing are possible with liquid GaIn alloys, jetting must be performed in an oxygen-free environment to prevent surface oxidation. Attempts by the authors and their collaborators (see the Acknowledgments) to jet in air (Microfab MJ-AB inkjet; Microfab Technologies, Inc.) lead to the accumulation of the liquid alloy at the nozzle tip. Nonetheless, pendant drop experiments performed in a nitrogen-filled glovebox suggest that liquid GaIn jetting should be possible in an oxygen-free environment.<sup>26</sup>

The microcontact printing ( $\mu$ CP) technique that we introduce here represents an extension of transfer printing methods commonly used in soft lithography.<sup>13,14</sup> Current applications of  $\mu$ CP and stamp lithography range from patterning self-assembled monolayers to the selective transfer of microtissue on PDMS substrates for biohybrid actuators. While slower than jetting,  $\mu$ CP is a versatile and inexpensive technique that can be used to deposit liquid GaIn alloy in air. Microcontact printing is relatively easy to implement with inks that readily wet the silicone elastomer but is challenging with

liquid-phase GaIn alloys. Surface oxidation interferes with wetting to the print head and elastomer substrate and causes the alloy to form a large droplet that requires force to rupture.<sup>2,3,24</sup> Although surface oxidation interferes with  $\mu$ CP deposition, it has the advantage of preserving the shape of deposited structures during external wiring and sealing.<sup>3</sup>

#### FABRICATION METHOD

As shown in Figure 5, the microcontact printer is composed of a print head mounted to a computer-controlled three-axis Cartesian robot.



**Figure 5.** Microcontact printer is composed of a PDMS print head mounted to a three-axis Cartesian robot. The motors are controlled through a user-defined Matlab script that commands the robot to dip the print head into a pool of liquid GaIn alloy and transfer droplets to an elastomer substrate.

The robot is assembled from an open-source 3D printer kit (Hadron) and contains four motors, two for *Z* height control and one each for planar *X* and *Y* motion. The motor drivers are commanded by a user-defined script in MATLAB (R2012a; The Mathworks, Inc.) to sequentially ink the print head in a pool of Galinstan (Rotometals, Inc.) or EGaIn (99.99%, Sigma-Aldrich Co. LLC) and deposit an individual droplet on an elastomer substrate. The substrate is prepared by spin-coating liquid silicone (Ecoflex 0030; Smooth-On, Inc.) and then curing the elastomer at room temperature. Print head rewetting and contact deposition are repeated until the circuit is complete.

The print head is made by casting PDMS (Sylgard 184, 10:1 base/ catalyst ratio; Dow Corning Corporation) in a 3D printed mold (VeroWhitePlus polymer resin, Objet24 Desktop 3D Printer; Stratasys, Ltd.) to form a 750  $\mu$ m diameter needle that is 2 mm tall and has a hemispherical tip. The Cartesian robot uses four stepper motors (NEMA17 1.6 A) to provide three-axis position control. We control position with two stages, the XZ stage, which supports the print head, and the Z stage, which supports the substrate and GaIn alloy reservoir. Each motor is operated with a Big Easy Driver board (Sparkfun Electronics) that interfaces with a personal computer (PC) through an Arduino Uno USB microcontroller board (ATmega328; Arduino). A custom script in MATLAB establishes a three-dimensional coordinate system with a reference origin, in which the spatial position of the print head, relative to the substrate, can be controlled through the execution of pre-defined print commands.

Figure 6 presents the steps for depositing a wire of liquid-phase GaIn alloy on an Ecoflex 0030 substrate. For each droplet, deposition begins with the print head positioned above a pool of GaIn alloy (Figure 6a). Next, the head is dipped (Figure 6b) into the pool and immediately retracted (Figure 6c). During immersion and retraction, EGaIn wets the tip of the print head and forms a suspended bead. Next, the wetted print head is positioned above the elastomer substrate (Figure 6d) and lowered (Figure 6e) into contact. The elastomer substrate is produced by spin coating (SCS 6812; Specialty Coating Systems, Inc.) layers of Ecoflex 0030 elastomer onto a glass disc. As the hemispherical tip is retracted, some of the liquid alloy transfers to the flat substrate and forms a droplet. Droplets that are



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**Figure 6.** Single droplet deposition: (a and b) an elastomeric print head is dipped into a pool of EGain; (c) a droplet of EGaIn wets the tip as the print head is lifted up; next, (d) the droplet is transferred to the elastomer substrate and (e) deposited as the print head is pressed into contact. (f) Liquid-phase EGaIn wire produced by coalescing droplets printed with 202.6  $\mu$ m spacing.

deposited with a spacing less than the droplet diameter coalesce and form a solid line (Figure 6f).

For stamp lithography, a liquid GaIn alloy is deposited with an elastomer stamp that is produced by casting PDMS (Sylgard 184) in a 3D printed mold (Objet 24). In contrast to single droplet deposition with a motorized print head, stamp lithography eliminates the need for repeated contact and can be performed manually. To assess the feasibility of stamp lithography with liquid-phase GaIn alloys, we produced masters with a variety of patterns and surface textures. These include solid lines and filled rectangles with smooth or dimpled surfaces. The stamp features protrude 2 mm from the stamp base, and the textured dimples have 0.75 mm diameter and are spaced 1 mm apart. A paint brush is used to ink the stamps with a coat of liquid-phase GaIn alloy.

After the circuit is printed, it must be wired and sealed with an additional layer of elastomer. Figure 7 presents all of the steps



**Figure** 7. Complete device fabrication. (a) Silicone elastomer is repeatedly spin coated onto a glass disc to create the substrate. (b) The liquid alloy circuit is printed onto the elastomer substrate. (c) A frame is placed around the circuit, controlling the thickness of the sealing layer. (d) A syringe is used to deposit droplets of liquid alloy at the terminal ends of the circuit. Copper wire is then inserted into these terminal ends. (e) The circuit is frozen and covered with uncured elastomer. (f) Upon curing, the liquid alloy device is completely sealed and can be safely manipulated.

necessary for device fabrication, starting with substrate preparation (Figure 7a) and microcontact printing (Figure 7b). Before sealing, a plastic 3D printed frame (VeroWhitePlus) is placed over the circuit (Figure 7c) and metal wires are inserted into the terminal ends of the circuit, where large droplets of EGaIn are deposited using a syringe (Figure 7d). To preserve the soft elastic functionality of the circuit, these external wires must be located at the edge of the device. To help protect the shape of the pattern, the printed circuit may be placed in a freezer or on a cold plate and cooled to below the melting point of the alloy. The frozen sample is then covered with uncured silicone elastomer (Figure 7e). Upon curing, the circuit is completely sealed and can be deformed in its room-temperature liquid state without smearing or leaking (Figure 7f).



Figure 8. (a and b) Unsealed 340  $\mu$ m diameter droplets of EGaIn on a silicone elastomer substrate (scale bar = 1 mm). (c) Solid line (1 mm ruler markings) and (d) filled square of coalesced droplets spaced 202.6  $\mu$ m apart (scale bar = 5 mm). (e) Unsealed comb capacitor (scale bar = 10 mm). (f) EGaIn wire sealed in silicone rubber with 0.07  $\Omega$  of electrical resistance. (g) Sealed comb capacitor with 2.5 pF of capacitance (h) remains functional when stretched.

# RESULTS

Following the procedures described above, we produced a series of test structures that demonstrate the reliability and versatility of microcontact printing with liquid-phase GaIn alloy. As shown in Figure 8a, the print head deposits individual droplets with a diameter of approximately 340  $\mu$ m. The droplet spacing can be precisely controlled in 202.6  $\mu$ m increments, although modifications to the motor driver connections can allow for much higher resolution, with the potential to control spacing in 12.6625  $\mu$ m increments. For example, the droplets in panels a and b of Figure 8 are spaced 810.5 and 607.9  $\mu$ m apart. In contrast, the droplets in panels c-e of Figure 8 are spaced 202.6  $\mu$ m apart, allowing them to coalesce and form a solid line and filled square. After wiring and freezing, the circuits are sealed and tested to measure electrical resistance or capacitance. The liquid-phase EGaIn wire in Figure 8f has a resistance of 0.07  $\Omega$ , while the capacitor in Figure 8g has a capacitance of 2.5 pF. As shown in Figure 8h, the devices are elastically deformable, such that stretching and squeezing may alter their resistance or capacitance. Although here we only print lines and right-angle intersections, the printer may also be programmed to deposit liquid alloy into any arbitrarily curved or intersecting pattern.

For stamp lithography, we attempted to transfer a solid line and filled rectangle onto an elastomer substrate using both flat stamps and textured stamps. Although this transfer method is similar to existing approaches in  $\mu$ CP, we were not successful in extending this technique to liquid-phase GaIn alloy. As shown in Figure 9, stamped patterns of Galinstan on Ecoflex 0030



Figure 9. Unsuccessful attempt to deposit Galinstan with smooth and textured stamps.

have non-uniform thickness and uneven surface wetting. The Galinstan does not wet the elastomer in some regions and coalesces into droplets in others. In the case of the textured stamp, droplets deposited by each dimple do not coalesce uniformly. This uneven wetting may result from non-uniform wetting during "inking", when the stamp is initially coated with Galinstan, or from non-uniform contact pressures during transfer. Nonetheless, these attempts demonstrate that liquid GaIn alloy can wet the elastomer and provided the original inspiration for exploring droplet-based deposition.

We performed tensile tests on the comb capacitor in Figure 8g to determine its electrical response to stretching. The

capacitance was measured with a BK Precision 889B Bench LCR/ESR Meter, and the sample was stretched with two linear actuators. We performed four sets of measurements, two in which the stretch direction was parallel to comb capacitor electrodes and two in which the stretch was perpendicular to the electrodes. As expected, the normalized change in capacitance of the capacitor increased linearly when stretched along its electrodes (Figure 10c) and monotonically decreased when stretched in the direction perpendicular to the electrodes (Figure 10d).



**Figure 10.** Comb capacitor (a) before and (b) after applying a stretch of  $\lambda \approx 2.5$ . Relative change in the capacitance of the comb capacitor as a function of uniaxial stretch are plotted in panels c and d for both experimental measurements (markers) and theoretical<sup>12</sup> predictions (solid lines).

# DISCUSSION

The results presented in Figure 8 demonstrate the potential of extending  $\mu$ CP to the fabrication of soft-matter circuits with liquid-phase alloy. These circuits are composed of a conductive microfluidic channel alloy embedded in a sheet of soft elastomer. The channels are filled with EGaIn and Galinstan, alloys that are liquid at room temperature and have an electrical resistivity of approximately  $3 \times 10^{-7} \Omega/m$ , about  $^{1}/_{20}$  the conductivity of copper. When exposed to air, the alloy forms a Ga<sub>2</sub>O<sub>3</sub> oxide layer that interferes with wetting and jetting but allows individual droplets to be moldable and deposited with a motorized print head.

Liquid GaIn circuits are presently produced with needle injection (Figure 3) and masked deposition (Figure 4). Although reliable, these fabrication techniques restrict the circuit patterns to a limited class of geometries. To produce any arbitrary planar geometry, the liquid alloy must instead be deposited with inkjet printing or  $\mu$ CP. However, attempts to jet GaIn alloy in air were unsuccessful because the alloy would oxidize and accumulate into a large droplet below the nozzle tip. We were also unsuccessful with stamp lithography, which, as demonstrated in Figure 9, results in uneven wetting. Nonetheless, attempts to print circuits with a textured stamp demonstrated that  $\mu$ CP is possible when the liquid GaIn is deposited as individual droplets. On the basis of these preliminary results with stamp lithography, we have instead decided to focus our efforts on developing a computercontrolled microcontact printer (Figure 5) that sequentially deposits individual droplets of GaIn alloy (Figure 6) to produce liquid-phase electronic circuits.

As shown in panels c-e of Figure 8, droplets deposited next together coalesce to form lines and filled geometries. Because of surface oxidation, these printed features are structurally stable and can be sealed with elastomer without losing their shape. The circuit may also be frozen (EGaIn has a melting point<sup>3</sup> of 15 °C) to prevent the alloy from spreading out during spin coating. Once connected with external wiring and sealed (panels f and g of Figure 8), the liquid alloy functions as a stretchable circuit that conforms to the elastic deformation of the surrounding elastomer (Figure 8h). Elastic deformation causes the length and cross-sectional area of the liquid channels to change. Dependent upon how the circuit is designed, this can lead to corresponding changes in electric resistance and capacitance. Liquid-phase circuits with elastically tunable resistance or capacitance function as hyperelastic deformation sensors that measure stretch,<sup>11,12</sup> contact pressure,<sup>8,10,11</sup> bending,<sup>9,19</sup> and shear.<sup>22</sup> As shown in Figure 10, an elastomer-sealed EGaIn comb capacitor produced with  $\mu$ CP exhibits electrical responses to stretch that are consistent with theoretical predictions. As previously shown by Fassler and Majidi, stretching causes the capacitance to increase by a relative amount  $\Delta C/C_0 = \lambda - 1$  when the capacitor is stretched parallel to the electrodes and  $\Delta C/C_0 = 1/(\lambda^{1/2}) - 1$  when stretched perpendicularly to the electrodes.<sup>12</sup>

The dimensions and spacing of the printed circuit features are governed by the resolution of the print motors, size and shape of the print head, contact pressure during deposition, and the wetting angle between the liquid alloy and elastomer substrate. Using a hemispherical 750  $\mu$ m diameter needle PDMS print head, we have demonstrated circuits with 340  $\mu$ m feature resolution. While this is adequate for millimeter-scale circuits and sensors, it is not sufficient for soft-matter microelectronics and sensors with micrometer-scale dimensions. To improve feature resolution, the  $\mu$ CP print head could be replaced with a motorized syringe that dispenses individual GaIn droplets. Chiechi et al. have previously demonstrated that a millimeter diameter drop of EGaIn dispensed on Ag film with a 26s-gauge needle (127  $\mu$ m inner diameter) will pinch at the top, leaving a ~0.05  $\mu$ L cone of EGaIn on the needle tip.<sup>2</sup> Subsequently, pressing the needle tip into an elastomer substrate would leave a droplet with a radius of less than 300  $\mu$ m. However, the cone tip has a radius of less than 1  $\mu$ m, suggesting that a smaller droplet might be possible with a higher gauge needle. For example, with an inlet gauge pressure of 89 kPa, EGaIn can be dispensed through a 20  $\mu$ m wide microfluidic channel<sup>3</sup> and may form <100  $\mu$ m radii droplet when deposited. Nonetheless, the current printer would need to be redesigned for motorized syringe dispensing, and additional experiments would have to be performed to identify the smallest allowable droplet size.

The printer hardware can also be redesigned to improve printing speed with faster motors or a mechanism for rewetting the PDMS print head. This could potentially eliminate the need for the print head to travel back and forth between the substrate and pool of liquid alloy. For example, the pool could be mounted on the Cartesian robot along with an additional motor to swivel the print head between the pool and the elastomer substrate. Alternatively, the print head could be

rewetted with a motorized brush that alternately brushes against the print head and a palette coated with liquid alloy.

# CONCLUSION

We have introduced a technique to produce liquid-phase GaIn electronics with microcontact printing. An elastomer print head is mounted on a three-axis Cartesian robot and deposits individual droplets of EGaIn or Galinstan on an elastomer substrate. After the liquid circuit is printed, external wires are inserted in the terminals and the circuit is sealed with an additional layer of elastomer. The current printer is designed to produce 340  $\mu$ m diameter droplets that, when spaced approximately 200  $\mu$ m apart, coalesce to form lines and filled shapes. In contrast to current needle-injection methods, microcontact printing is a rapid and fully automated method to produce liquid-phase circuits with arbitrary planar geometries.

In this study, we have limited our efforts to demonstrating microcontact deposition of liquid-phase EGaIn and Galinstan on Ecoflex 0030. However,  $\mu CP$  should also be possible with other elastomer substrates, such as PDMS and soft polyurethanes, and liquid-phase alloys, including low-melting-point solder deposited with a heated print head or substrate. For these combinations of materials, further studies will be required to determine the appropriate print head design and print settings. Future efforts should also focus on hardware modifications to improve printing resolution and speed. This includes a new print head with a rewetting mechanism that is mounted to the Cartesian robot as well as a careful examination of the influence of tip shape, composition, and contact pressure on droplet size and shape. Lastly, additional work is required to explore printing with a motorized syringe or an inkjet that operates in an oxygen-free environment to prevent alloy oxidation.

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#### Notes

The authors declare no competing financial interest.

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