Laser-based Rapid Prototyping Techniques for Liquid Metal Circuits

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Laser-based Rapid Prototyping
Techniques for Liquid Metal Circuits

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Abstract

Stretchable electronics has been a rapidly developing technology which allows devices to be deformed dramatically. Compared with conventional technologies, with which devices are made of rigid materials, stretchable electronics enables devices to be stretched, twisted, and bent robustly. Such features contribute to a greater functionality and make the devices suitable for applications such as wearable devices that need to adapt to complicated geometries. Additionally, compared to conventional electronics, a wearable device that consists of stretchable electronics is safer for users. The existing materials and fabrication methods, however, lack the capability of creating practical and precise stretchable electronics rapidly at a reasonable cost.

In this work, liquid metals (EGaIn, Galinstan), along with conductive polymers (cPDMS, etc), are patterned with an innovative prototyping method based on two different laser systems (CO$_2$ and UV). With the methods developed, stretchable circuits are successfully fabricated in minutes. Experiments have been performed to show the excellent electrical properties of the laser-patterned circuits.

Besides rapid prototyping, an innovative interfacing method between liquid metal and solid-state circuit components is also developed. This method is based on magnetic conductive particles embedded in polymer. The particles are aligned in a magnetic field so the composite is only conductive through its thickness. Compared with existing techniques, this method significantly enhanced the reliability of the interface between liquid metal and solid-state components and can be potentially applied on stretchable integrated circuits.

A series of applications, including tactile and strain sensors, are presented to validate the developed methods.
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Chapter 1

Introduction

Circuits that are soft and stretchable can conform to irregularly shaped and deformable surfaces. They can be integrated into clothing, medical dressing, robotic systems, and inflatable structures without introducing mechanical rigidity or kinematic constraints [7–9] As with conventional flexible printed circuit boards (PCBs), these stretchable circuits should be thin, lightweight, and interfaced reliably with surface-mounted microelectronic chips and packaged electronics. This requires circuit trace widths on the order of 0.1-1 mm and volumetric electrical conductivities that equal or exceed $10^4$ S/m. However, unlike flex PCBs, stretchable circuits must be extensible and exhibit limited mechanical resistance to tensile loading. Like natural skin or elastene fabrics (e.g. Spandex®), this requires an elastic strain limit of above 100% (preferably as high as 200-1000%) and a tensile elastic modulus of $\sim$0.1-1 MPa (see Figure. 1.1). Without these properties, the circuit may introduce mechanical constraints that could limit the motion of the host, i.e. human, robot, or inflatable.

A variety of materials, physical architectures, and fabrication methods have been used for producing stretchable electronics, with some techniques dating back to the late 1940s. For certain specialized applications, these approaches have been successful in replacing rigid and flex PCBs. Examples range from the mercury-based Whitney strain gauge for measuring muscle contraction[10] to an epidermal “electronic tattoo”
for physiological health monitoring[3]. However, the broad application of soft stretchable circuits to personal electronics and robotics is currently limited by a number of technical challenges and bottlenecks. While there has been progress, more work is required in engineering elastomers and rubbery polymers that exhibit the mechanical properties of soft biological tissue and the electronic properties of more rigid crystalline, semi-crystalline, and glassy materials. Another challenge is the development of versatile fabrication methods that enable automated, rapid, and high-throughput patterning of circuits and their reliable integration with surface mounted chips, PCBs, and flat flexible cable (FFC) interfaces.

My dissertational work addresses the latter need by introducing novel techniques for soft electronics rapid prototyping. I have chosen to focus on laser micromachining since it is compatible with the broadest range of circuit materials: insulating carrier films, conductive elastomer composites, liquid-phase metals, solid thin metal films, ceramics, and glasses. While some of these materials can also be patterned with additive manufacturing techniques (e.g. extrusion-based 3D printing, stereolithography), they represent only a limited subset that do not exhibit the mechanical and electronic properties necessary for all applications. In contrast, CO$_2$ and UV laser micromachining can be used to pattern virtually any organic or inorganic film (thickness $\sim$ 0.01-1mm). Moreover, planar feature sizes can be as small as 25 $\mu$m, which is more than adequate for most PCB applications.

A secondary focus is the development of methods for interfacing a laser-patterned circuit with rigid and flex electronics. These interfaces should have low electrical contact resistance and be mechanically robust under stretching, bending, and twisting. Although this aspect of stretchable electronics has gotten relatively less attention than materials selection and circuit patterning, it is nonetheless critical to progress in the field. What makes this problem particularly challenging are the large stress con-
centrations and mechanical failures that can occur when applying load to interfaces where there is a mechanical impedance mismatch. This is especially pronounced for interfaces that contain liquid metal alloy, which can seep through interfacial cracks or leak out of tears. While still an area of ongoing research, I will present some efforts to address this with a special class of “z-axis” anisotropically conductive films.

1.1 Material Requirements

Like their rigid and flexible counterparts, stretchable circuits are composed of both electrically conductive and insulating materials.[11] Fig. 1.2 presents an overview of conductors and insulators that are used for conventional PCBs or stretchable electronics.[2]

1.1.1 Electrical Properties

The conductors function as electrical wiring and typically have the following dimensions: width $\sim 0.1$-1mm; thickness $\sim 10$-100 $\mu$m. While thinner or larger wiring is occasionally used, this range is typical in existing PCB and electronic textile applications. In order to preserve the digital functionality of a circuit, the electrode resistance should be moderate (i.e. well bellow 1k$\Omega$). For $\sim 1$-10 mm long traces, this requires volumetric resistivities below $10^{-4}$ $\Omega$·m (conductivity $> 10^4$ S/m). However, for some applications like capacitive tactile sensing, simple circuit functionality can be achieved with traces and electrical transducers that exhibit higher resistance.

The insulator is used as a carrier film or substrate that supports the circuit and can bond to a host surface (e.g. clothing, human skin, balloon, etc.). In Figure. 1.2, only PDMS is shown to be functional as an insulator due to its negligible conductivity.
Figure 1.1: Young’s modulus of some common natural and synthetic materials. Adapted from the work of Prof. Wanliang Shan, et al.[1].
Figure 1.2: Volumetric conductivity vs. mechanical rigidity for various materials. Credits: Andrew Fassler, et al. [2]
Other ways to characterize the insulating properties of the film is by its electrical permittivity, breakdown strength under high electrical field, and loss tangent under various electrical frequencies and voltages. In addition to PDMS, soft polyacrylates and polyurethane films are also popular as carrier films for stretchable electronics. These elastomers all share insulating properties that are similar to the plastics (e.g. polyimide) used in flex PCBs.

1.1.2 Mechanical Properties

In order to be compatible with natural human tissue and fabrics, the circuit must be able to support elastic strains on the order of 100%. Of course, there are plenty of applications or loading conditions where the strain won’t be this extreme. For example, in many wearable applications, the strain could be only 10% or smaller depending on where the circuit is placed on the body. However, as a general-purpose technology that can be extended to a broad range of computing, healthcare, and robotics applications, the ultimate goal is to have a robust material and architecture where mechanical extensibility is not a practical limitation in where/how the circuit is integrated. In addition to being mechanically extensible, the circuit should also be elastic – i.e. when released from loading, the circuit should spring back to its original shape.

For solid materials, Young’s modulus is usually used as a measure of mechanical compliance under small elastic deformation. Figure 1.1 shows the Young’s modulus of common natural and synthetic materials[1]. The Young’s modulus of the insulating materials selected for stretchable elastic must be comparable with biological materials so that the stretchable electronic devices are fully compliant to biological tissue. The materials with a Young’s modulus of greater than 10 MPa are usually considered to be rigid materials[9] while the ones with Young’s modulus of 1 MPa or less are
considered to be soft materials. For example, poly(dimethylsiloxane) (PDMS), which is a silicone rubber widely used in stretchable electronics research, is a soft material that falls in the desired modulus range.

Mechanical compliance can also be defined with Shore hardness. Shore hardness is usually used to measure the hardness of polymers, elastomers, and rubbers. There are different types of Shore hardness and in any type a material’s Shore Hardness is a value between 0 and 100. The higher the value is, the harder the material is. PDMS, for example, has a Shore A hardness of about 50[12]. It’s important to note that this measure of compliance is not based on elastic deformation and is only based on mechanical resistance to an approach indentation. For soft polymers and highly viscous matter, a Shore hardness value could include resistance from inelastic response. In the case of fluids, the mechanical resistance to loading is best captured by shear (dynamic) viscosity, which is shown in Figure. 1.2 for some conductive matters.

1.2 Existing Materials and Architectures

The prototyping techniques introduced in this dissertation were developed to be compatible with all of the materials currently used for stretchable electronics. As described below, these materials share a complementary set of features that makes it advantageous to have a “universal” fabrication method in which they can all be patterned and integrated.

1.2.1 Elastomers & Elastomer Composites

Elastomers are generally used as the substrate/matrix material for stretchable electronic devices. Stretchability differs among elastomers. Depending on the polymer chain lengths and cross-linking density, the elastic strain limit of Pt-catalyzed sil-
icicles can range from 100%[13] to as much as 900%. The high deformability of elastomers makes them suitable for stretchable electronics, especially for applications such as wearable devices because they can naturally conform to human body and deform with skin, which is typically strained to 30%[11]. Another important feature of elastomers like silicone is that they are bio-inert, nontoxic, and biocompatible.[14] For this reason, stretchable electronics with a PDMS carrier film have the potential to interface with biological tissue and be used for medical treatments, wound healing, and implants.

In order to become conductive, elastomers are typically embedded with a percolating network of conductive nano- or microparticles. Typical conductive fillers include carbon black, silver particles[15][16][4], and carbon nanotube[17][18]. Common matrix materials (also known as the dispersion medium) include PDMS[16], PU[19], and elastomeric fluoropolymers[20]. Figure. 1.3b shows an example of a stretchable PCB with silver-filled PDMS as conductor. This type of conductive composite is inherently stretchable and easy to pattern with stencil lithography or screen printing. However, they are relatively low in electrical conductivity compared with metals and consequently their application is limited. For example, for silver-filled PDMS, with a silver content of 84 wt%, its electrical conductivity is less than 10 S/m[15]. Besides their low conductivity, the filler particles in polymer can significantly increase the stiffness of the composite, which reduces the composite’s stretchability and softness.

In general, elastomers are ideally suited as an insulating carrier film. When filled with a percolating network of rigid conductive particles, the composite is conductive but volumetric resistivity is typically above the $10^{-4} \Omega \cdot m$ threshold for circuit wiring. Moreover, when stretched, this resistivity will exponentially increase due to the separation of the particles and disintegration of the percolating network as the surrounding elastomer elongates. Similar degradation in conductivity during stretch
Figure 1.3: (a) A stretchable circuit using wavy patterned metal [3]. (b) A circuit made with silver-filled PDMS, scale bar: 10 mm [4]. (c) A tactile keypad embedded with LM microchannels [5]. (d) A circuit with ionic fluid microchannels embedded [6].

has also been shown with conductive block copolymer elastomers, in which polyaniline or ionomers are grafted to the copolymer.[21] Nonetheless, as shown in Chapter 3, such composites can still be useful as wiring and transducers for simple capacitive and voltage divider circuits for human tactile sensing.

1.2.2 Metals & Wavy Electronics

Although not inherently soft, metals can be patterned into wavy or spring-like shapes that are extensible in certain loading directions. These so-called “deterministic ar-
chitectures” have been demonstrated in a broad range of systems, from embroidered conductive textiles[22] to thin-film microelectronics[3, 7]. The metals used are typically copper, silver, gold, or aluminum.[11] Examples of wavy circuits produced with photolithography are presented in Figure. 1.3a.[3]. Such microfabricated circuits includes both planar (2D) and out-of-plane (3D) deterministic architectures and are capable of strains of up to 200%[23]. There are also techniques that simply embed rigid materials in polymer film to get electronics that are flexible but not stretchable [24].

For metals with deterministic structures, there are several factors that can potentially limit their universal applicability. Because the material is intrinsically rigid, there will always be length scales or loading direction under which the meso- or macroscale compliance will no longer be exhibited. Also, the mechanical impedance mismatch between the metal and carrier film can result in stress concentrations that lead to circuit delamination. Lastly, miniaturized circuits are currently produced with clean-room lithographic techniques that are not amenable to rapid prototyping and can be expensive to scale up for high throughput production.

1.2.3 Ionic Fluids and Hydrogels

Ionic fluids are usually salt solution in water and have become increasingly popular in stretchable electronics[11, 25, 26]. The conductivity of ionic fluids is determined by its composition and concentration of the salt. Ionic fluids can be injected into microchannels [6, 27, 28] and have been shown to remain conductive when stretched up to 700%[29]. An example of an ionic fluid inside of microchannel is shown in Figure. 1.3d.

Ionic hydrogels are a hydrophilic network of polymer chains that is highly absorbent and contains a large volume of electrolytic solution (up to 90%). Because
of the high fluidic volume, the ionic solution forms continuous conductive pathways and enables the gel to be conductive. Once challenge with ionic hydrogels is water evaporation, which causes it to dry out over time and lose its mechanical and electrical properties.[11, 30] Nonetheless, there has been recent success with creating stretchable electronic devices using ionic hydrogels[31] and recently there have been attempts to improve stability in air[32].

Soft microfluidics with ionic solution are promising due to their high stretchability (>100%) and low elastic modulus (<10 kPa).[32] However, their electrical conductivity of \( \sim 1 \text{ S/m} \) is inadequate for most stretchable electronics applications. Instead, they’re best suited for specialized applications where they can be used as transparent electrodes for capacitive elements and electrostatic transducers.[31]

### 1.2.4 Liquid Metals

Liquid-phase metal alloys have the unique advantage that they are both low viscosity fluids and also exhibit the electrical and thermal properties of a metal. They are naturally compliant to any deformation and have much higher conductivity than conductive polymers. Starting with the Whitney strain gauge, mercury was used in liquid metal electronics but has since been abandoned due to its toxicity. In that original implementation, Hg was filled into a rubber tube and used as a stretchable wire and strain sensor to measure the limb motion and muscle contractions during physiological studies.[10] More recently, Hg has been replaced with alloys of gallium and indium. Eutectic Ga-In (EGaIn; 75 wt% Gallium and 25 wt% Indium) and Ga-In-Sn (Galinstan) are particularly popular due to their non-toxicity, high electrical conductivity \( (3.4 \times 10^6 \text{ S/m}) \), and low viscosity \( (\sim 2 \text{ mPa-s}) \) at room temperature.

As with an ionic solution, the low viscosity of EGaIn and Galinstan allows them to be injected into soft microfluidic channels, as shown in Figure. 1.3c), and remain
intact when the surrounding elastomer is stretched. Another feature of Ga-based liquid metal (LM) alloys is that it forms a self-passivating coating (0.5-2 nm) of Ga$_2$O$_3$ when exposed to oxygen (e.g. in air or O$_2$ saturated PDMS) in < 1s. Ga$_2$O$_3$ is an n-type semiconductor with a band gap of approximately 4.85 eV. This coating assists with wetting to elastomer by increasing the adhesion between GaIn and substrate and also allows free standing droplets to hold their shape because of the high surface tension. As discussed below, these wetting and “moldability” properties can be exploited for a variety of fabrication techniques. There’s also been recent interest in voltage-controlled EGaIn oxidation and reduction of droplets inside of an electrolytic bath.

Despite their excellent electrical properties and intrinsic deformability, Ga-In alloys are corrosive to other metals and should not come in direct contact with human skin. Therefore, the LM circuit must be sealed, which introduces challenges with packaging and electrical interfacing with surface mounted electronics. Typically, contact is formed by inserting thin solid wiring into the ends of the LM channel. This method is reliable for larger circuit features but cannot be extended to packaged microchips and other small surface-mounted components that have flat pins. Moreover, wire insertion is often done manually and is therefore not appropriate for automated or large scale fabrication. Reliable interfacing between LM circuits and rigid electronics remains an open research challenge and will be a topic of focus in Chapter 4.

1.3 Prototyping Methods of LM Circuits

Prototyping methods for liquid metal circuits include injection filling, stencil lithography, photolithography, microcontact printing, etc. These methods, while able to make devices of different dimensions, are limited by different disadvantages such as
being time-consuming, high cost, or requiring advanced or customized equipments. Some of the typical prototyping methods are discussed below.

### 1.3.1 Injection Filling

Injection filling is the first and most common technique for producing liquid metal circuits. A needle and syringe is used to inject the LM alloy into microfluidic channels that are molded into the elastomer. The channels are produced with replica casting using molds that are produced using 3D printing\cite{44} or photolithography\cite{45}. Inlet and outlet holes must be included in the microchannels so that syringes can insert and inject liquid metal\cite{33, 34, 45}. 3D printing with extrusion or jetting can produce molds that have feature sizes as small as $\sim200 \, \mu\text{m}$. For smaller features, laser-based techniques based on stereolithography or two-photon polymerization are required.

Molds with small feature sizes ($\sim1$-$10 \, \mu\text{m}$ dimensions) are typically produced with SU-8 photolithography.\cite{45}. Photolithography is the technique which utilize selective exposure of a light sensitive polymer to UV light to define certain patterns\cite{46, 47}. This polymer, which is called photoresist, is first applied on a substrate. A photomask, which is a transparent substrate patterned with opaque features, is placed over the photoresist. The UV light go through the photomask and break down the polymer chains of photoresist in the exposed areas (areas with no opaque feature above). Consequently, the photoresist in exposed areas gets more soluble in certain chemical solution. After removing exposed photoresist, the unexposed photoresist is left on the substrate with desired patterns. The photoresist remained can be used as masks with which materials such as polymers, metals, etc., can be patterned. Subsequently, the remained photoresist is removed with chemicals like acetone. The patterning resolution of photolithography can be as small as $2 \, \mu\text{m}\cite{48}$. However, despite its advantages in terms of pattern resolution, photolithography is very time/labor-consuming and
not amenable to rapid prototyping. It also requires specialized training and access to a clean room.

The major advantage of injection is its overall simplicity, especially when performing replica casting using a 3D printed mold. However, key disadvantages are the limited geometries that can be produced with a single inlet and outlet and the labor intensive nature of the process. Especially in the case of samples produced with a microfabricated mold, the fabrication process typically takes 1-2 days and is full of manual steps during which failures occur. While adequate for early demonstrations of LM electronics, injection filling is not commonly accepted as a viable method for rapid prototyping or production-level fabrication.

1.3.2 Stencil Lithography

As presented in Chap. 2, LM stencil lithography was only recently discovered independently by my collaborators and I in the Soft Machines Lab (SML; PI: Majidi) and by a group at Uppsala University led by Dr. Zhigang Wu. It has since become a popular technique for patterning liquid metal. Because it does not involve elastomer casting, it can be performed much faster and with fewer manual steps. Details of the fabrication method are described in the next chapter. In summary, a mask(stencil) with desired patterns is applied to an elastomer substrate and then a film of LM is deposited over the mask. The LM selectively wets to the exposed portions of the substrate, leaving a patterned circuit once the stencil is removed.

There are different techniques to create the masks based on the feature sizes. For circuits with mm-scale features, the mask can be printed with a 3D printer[49] or patterned through milling[50] or laser machining[51, 52]. For smaller circuit features, the mask can be produced using photolithography.[53] As with injection filling, stencil lithography is a simple process that can be performed with commercially available...
equipment. However, it is also limited by the types of planar geometries that can be produced. For example, it cannot contain self-intersecting traces or other patterns with closed-loop features. In principle, such geometries could be produced with screen printing, but attempts with LM have been inconclusive due to clogging of the screen pores.

1.3.3 Microcontact Printing and Direct Writing

Microcontact printing is the method which utilizes a polymer stamp to get patterns of certain materials. For liquid metal, the stamp is usually a round tip made with PDMS[54]. This tip is mounted on an automated stage. It dips into a reservoir of liquid metal and becomes wetted with a droplet of fluid. Subsequently it transfers the droplet to a substrate, which binds to the liquid and pulls it off of the tip. This process repeats and the liquid metal droplets are deposited onto the substrate one by one. The droplets are closely lined up and coalesce to form a circuit. For microcontact printing, the feature size of the printed pattern is directly related to the size of the tip, although this method has only been demonstrated at the mm-scale with a 0.75 mm diameter tip.

Direct writing is similar to microcontact printing in that a customized system with a motorized stage or print head is needed. Direct writing is the method that uses a nozzle to deposit liquid metal in forms of droplets or lines[41, 43, 55]. For Gallium-Indium alloys, the oxide layer on the surface helps maintain its shape. The nozzle can be simply a needle[41, 55] or a ballpen-like structure[43]. Based on the structure of the stage, the liquid metal can be patterned into 2-D or 3-D structure. The feature size can be as small as 100 μm[55].

Both techniques here are promising and can eventually be incorporated into 3D printers for multi-material or high throughput multi-nozzle deposition. However, the
range of materials that existing 3D printer technologies can deposit are currently limited. For example, such techniques cannot be used by themselves to print “hybrid” circuits composed of liquid-phase and solid metal films. Moreover, not all elastomers and composites can be deposited with 3D printing.

1.4 Laser-based Prototyping

While appropriate for specific applications, the above techniques are not compatible with all the materials that could go into an LM-based circuit. Just as in conventional prototyping, the most versatile technique is to use machining and computer-controlled milling. In the case of soft elastomers and fluids, the most promising technique is to use laser-based milling. This can be accomplished with either a CO$_2$ laser engraver or UV laser micromachining system. Between the two, these lasers can be used to ablate and pattern a broad range of organic and inorganic materials, including thin films of Ga-based LM alloy.

More generally, laser have been widely used in areas such as metal processing[56] and soft-tissue surgery.[57] When interacting with a laser, materials absorb photonic energy and may ablate thermally or chemically depending on the frequency of excitation and nature of materials[58]. Lasers have also been used in stretchable electronics to create wavy metal interconnects.[59] Researchers have used laser to create masks for conductive polymer deposition[37]. Furthermore, lasers have been used for welding at the nano scale to create metal network for stretchable electronics[60]. In those applications of stretchable electronics, laser demonstrates its advantages such as high energy and high precision in patterning materials. These advantages has shown that laser has the potential to be applied in patterning soft conductive materials.
1.5 Objectives and Overview of Dissertation

Stretchable electronics have shown their potential in applications such as next-generation wearable computing. Conductive and insulating elastomers and LM alloys are especially promising as candidate materials due to their intrinsic compliance and deformability. However, most of the current methods to pattern and integrate these materials with rigid electronics are time-consuming and/or labor-intensive processes, require specialized equipment, and are limited in the specific materials they can process. Instead, what’s required is a universal fabrication approach that can be used to rapidly pattern and integrate all classes of materials that are of interest for stretchable electronics. Such an approach should be repeatable, have high yield, have precise alignment between patterned layers, and have a limited number of manual steps. Of the existing prototyping techniques discussed above, only laser patterning satisfies all of these requirements.

My dissertation is focused on extending laser-based rapid prototyping to the domain of soft-matter LM electronics. The research tasks and accomplishments presented in the following chapters are guided by the following objectives:
1. Demonstrate and examine the ability to pattern films of Ga-based liquid metal and conductive elastomers and produce soft-matter circuits using laser ablation.

1a. Develop methods to produce LM circuits with both CO$_2$ and UV laser patterning.

1b. Validate rapid prototyping methods with representative circuits and sensors that remain electrically functional during elastic deformation.

2. Explore methods to engineer a mechanically robust electrical interface between the terminals of the soft-matter circuit and the pins of rigid electronic components.

I begin in Chapter 2 by introducing a method to pattern Galinstan using a conventional stencil lithography technique. This is demonstrated on a variety of substrates, including silicone and polyacrylate (Sec. 2.1). The polyacrylate (VHB tape, 3M. Inc.) is a highly stretchable and adhesive elastomer that is directly patterned with a CO$_2$ laser engraver (VLS 3.50; Universal Laser Systems) to create open channels for depositing the Galinstan. The liquid metal channel is combined with a phase-transition material (Field’s metal) that melts when heated by current passing through the LM circuit. When this phase transition occurs, the rigidity of the composite decreases dramatically. The same fabrication method is also used to make a rigidity tunable composite with a layer of thermally-responsive shape memory polymer (SMP). Details of the fabrication method and experimental study are presented in Sec. 2.2. Details of the theoretical analysis are presented in [51] and [61] and have been omitted from the dissertation for the sake of brevity.

In Chapter 3, I present a more versatile fabrication method based on direct CO$_2$ laser patterning of conductive elastomers and Ga-based LM alloy. Direct laser pat-
tering is generally preferable to stencil lithography because it provides a higher precision and is capable of producing a broader range of planar geometry. The method of using CO$_2$ laser to pattern conductive polymers and liquid metal is introduced in Section 3.1. The patterning quality is examined in Section 3.2. Some applications of CO$_2$ laser patterning are also shown (Section 3.3, 3.4).

For a functional soft-matter circuit, it is not enough to have a method for materials patterning. A reliable method is also required to interface the conductive elastomer or LM circuit with rigid, surface-mounted components. In Chapter 4, I present one potential method for creating a mechanically robust electrical interface between liquid-phase metal traces and solid metal pins. The approach is based on coating the LM circuit with a PDMS film that’s embedded with an array of vertically-aligned columns of magnetic particles (Ag/Ni particles). The processing steps for producing this film and methodology for circuit characterization are presented in Sec. 4.1. The results of the measurements and circuit implementation are described in Sec. 4.2. This materials provides a new direction of solving the interfacing issue between liquid metal and solid-state electronic components.

Although rapid and reliable, CO$_2$ laser patterning isn’t capable of creating planar features with dimensions smaller than 200 $\mu$m for LM. For circuits with $\sim$0.05-1mm feature dimensions, a UV laser micromachining system (Protolaser U3; LPKF) is used. As described in Chapter 5, the UV laser enables a smaller spot size and higher photon energy and can be used to directly ablate liquid metal. LM circuits are made with UV laser micromachining and their electrical properties are verified by experiments, as described in Secs. 5.1 and 5.2).

Lastly, in Chapter 6, I close by summarizing the achievements and discuss some of the remaining challenges related to the techniques that I developed. I suggest areas for further scientific exploration and propose several improvements that could
potentially enhance the performance of these methods. I also suggest novel ways in which LM-based soft-matter electronics can be applied, particularly in the areas of wearable computing and healthcare.
Chapter 2

Stencil Lithography with Liquid Metal and Its Application

Stencil-based patterning and screen printing are common techniques for selective depositing inks, uncured polymers, and other fluids onto a substrate. The term ”stencil lithography” is occasionally used when extending this method to microfabrication and soft lithography[62]. In contrast to microcontact printing, injection filling, and other soft litho techniques that are dependent on elastomer casting, stencil lithography does not require labor-intensive steps in the clean room to produce a microfabricated mold. Instead, a stencil can be rapidly produced with laser ablation of a semi-rigid film or sacrificial mask layer. To date, a broad range of materials have been patterned with stencil lithography, from photoresist[15] to polymers like PDMS[63], PMMA[64], and PET[65].

Here, I show that stencil mask deposition can be further extended to the patterning of Ga-based liquid metal alloys. This adaption of stencil lithography form LM-electronics fabrication exploits the unique wetting properties of EGaIn and Galinstan that were described in the previous chapter. It should be noted that while developing
Figure 2.1: The method that uses paper stencil (Credits: James Wissman, SML)
Figure 2.2: (a) Fabrication of a Galinstan heating element embedded in an acrylic VHB elastomer: (left) VHB film and liner are patterned with a laser engraver and bonded to a VHB substrate; (middle) sample is coated with Galinstan, which wets to the exposed portions of the substrate; (right) the liner mask is removed and the patterned surface is sealed with a VHB film. (b) Liquid-phase Galinstan circuit embedded in VHB elastomer. (c) A 3-axis capacitive tactile sensor with liquid metal electrodes embedded.

This technique, the application of stencil lithography to LM patterning was independently discovered by a research group at Uppsala University led by Dr. Zhigang Wu [66].


2.1 General Patterning Technique

Referring to Figure 2.1, selective LM deposition can be performed by using a paper stencil that is patterned with a CO$_2$ laser engraver.[67] First, a silicone substrate is prepared by coating a plastic 9 cm diameter Petri dish with a non-stick spray (Ease Release 200; Mann Release Technologies, Inc.) and then depositing un-crosslinked Pt-cured silicone elastomer (Ecoflex 0010 or Ecoflex 0030; Smooth-On) with a spin coater (KW-4A, SPI, Inc.) at 500 RPM for 9 seconds followed by 2000-4000 RPM for 20 seconds. Prior to curing, the elastomer is degassed in a desiccation chamber (01823-AB, SPI, Inc.) for approximately 15 minutes in order to remove gas bubbles. Next, the stencil mask is created by patterning plain printer paper (998067, Georgia-Pacific) with a 30W VLS3.50 engraver from Universal Laser Systems. The mask is then placed on the cured silicone elastomer and a film of EGaIn (495425-25G, Sigma-Aldrich, Inc.) is brushed over the paper using a customized application pen. The pen consists of a 3D printed (Objet24, Stratasys) plastic tube with a 1 cm diameter rounded piece of Ecoflex 0030 on the end. The EGaIn is wetted to the Ecoflex tip and applied to the required areas using dabbing motions. Because EGaIn wets more willingly to the elastomer than to the paper, minimal liquid metal transfers to the mask, requiring little to no cleanup after the mask is removed.

Stencil lithography can also be used to pattern LM circuits on a commercial polyacrylate adhesive that is soft and elastic (VHB Tape; 3M). Referring to Figure 2.2, the CO$_2$ laser engraver is used to cut planar patterns in a sheet of the tape and its liner film. After removing the excess material, the patterned sheet is bonded to a second layer of VHB tape. Next, liquid-phase Galinstan alloy is deposited using the application pen. Removing the liner mask leaves liquid alloy only in the exposed surface features. Lastly, we attach copper shim wires and seal the liquid by bonding a
third layer of the tape. As demonstrated in Figure. 2.2b,c, this method allows an LM
circuit or electrodes to be embedded in a thin sheet of the polyacrylate elastomer.

2.2 Application to a Rigidity Tuning Composite

Stencil lithography is applied to produce an LM-based Joule heating element used
in a thermally-activated rigidity-tuning composite. This work was performed in co-
operation with Dr. Wanliang Shan, who is currently an Assistant Professor at the
University of Nevada, Reno. Referring to Figure. 2.3(a), the composite is composed of
a thermally responsive layer (e.g. Field’s metal, wax, thermoplastic, or shape memory
polymer) covered by the heating element and a sealing layer. When current is applied
to the heater, the thermal layer melts or softens and reduces the tensile rigidity of
the composite. Because the heater is made with a liquid metal trace, it can deform
with the surrounding material as the composite is stretched.

The effective Young’s modulus $E'$ of the composite before and during activation
is measured with a motorized tensile machine (Instron 5969). When Field’s metal
(RotoMetals, Inc.) is used as the thermal layer, 6A of current (3.6 W) is required
to heat up the composite and cause the metal to melt ($T_m = 60 ^\circ C$). For shape
memory polymer (Gemini; SpinTech LLC), electrical activation causes the thermal
layer to heat up above its glass transition temperature ($T_g = 62 ^\circ C$) and soften. In
contrast to existing methods for tunable elastic rigidity, these soft-matter composites
are electrically-powered and do not rely on external pneumatics, hydraulics, elec-
tromagnets, or motors to operate. The composites may be scaled and patterned to any
size or shape and can function in applications ranging from an artificial muscle for
biologically-inspired robots to a wearable brace for human motor assistance and in-
jury prevention. Moreover, the composites contain no rigid or inextensible elements
when fully activated. This soft-matter functionality is especially important for me-
2.2.1 Fabrication

As illustrated in Figure 2.3a, the rigidity tunable composites consist of two functional components: the Joule heater and a thermally-responsive bottom layer containing either a thin strip of low-melting-point Field’s metal embedded in VHB tape or an SMP film. The heater, which is essentially a liquid metal circuit, is fabricated with the method introduced in last section. Since the heater is composed of VHB tape, it readily bonds to the SMP under pressure-sensitive adhesion. The Field’s metal strip is 1 mm deep and 7 mm wide and embedded in VHB tape. The strip is produced by melting the Field’s metal at 100 °C on a hotplate and then depositing it on a laser-patterned sheet of VHB tape using the same fabrication steps (Figure 2.2a) used to produce the liquid-phase heater. The heater and thermally-responsive layers are then bonded together through the pressure-sensitive adhesion of the VHB tape.
In addition to the Galinstan channel and Field’s metal strip, these test specimens contain 3D-printed plastic attachments (Verowhite\textsuperscript{TM}; Objet Ltd.) that are bolted to the ends of the Field’s metal strip and allow the specimen to be fixed to a tensile testing machine.

The Field’s metal composite is $h_c = 4.5$ mm thick, $w_c = 15$ mm wide, and 50 mm long. It contains a rectangular strip of Field’s metal that is $h_F = 1$ mm thick and $w_F = 7$ mm wide. The composite also contains a serpentine channel of Galinstan that is $w_G = 1.27$ mm wide, $h_G = 0.5$ mm deep, and 218.9 mm long. Each turn is approximately $L_t = 10$ mm long and spaced $s = 1.23$ mm apart. Referring to the section of Experimental Testing, the Young’s modulus of the VHB elastomer and Field’s metal are $E_V = 128$ kPa and $E_F = 9.25$ GPa, respectively. The shape memory polymer (SMP)-based composite has an identical Galinstan heater but in the thermally-responsive bottom layer, the embedded Field’s metal strip and VHB seal are replaced with a 1 mm thick sheet of SMP.

2.2.2 Experimental Testing

The effective Young’s modulus ($E'$) of the composite was measured with a motorized tensile tester (Instron\textsuperscript{TM}, Norwood, MA), as shown in Figure 2.4b. The specimens were loaded with a 1 mm/s extension rate to a final extension of 7 mm (14% strain). As shown in Figure 2.4a, the composite specimens have dog-bone shape with 4.5 mm $\times$ 15 mm rectangular cross sections and a gauge length of 50 mm. The samples also have wider rectangular shaped ends with through holes to allow for rigid clamping. We first performed measurements at room temperature, when the Field’s metal is solid and rigid. Next, we melted the Field’s metal by supplying the composite with 6 A of electrical current and performed additional tensile measurements. The modulus $E'$ is obtained by fitting linear regressions to the experimental stress-strain plots and
Figure 2.4: Non-activated specimen and an electrically activated specimen that is stretched to twice its natural length.

calculating the slope.

For Field’s metal, dog-bone specimens of different sizes were used to determine the Young’s modulus. Five of these specimens had a size of 3 mm × 5mm × 50 mm and the remaining four had dimensions of 3 mm × 5mm × 91 mm. The Young’s modulus of the Field’s metal was measured to be 9.25 ± 0.60 GPa (N = 9). Similarly, the Young’s Modulus of VHB\textsuperscript{TM} tape was measured to be 128.4 ± 24.3 kPa (N = 5).

Temperature is measured with a Fluke 62 Mini IR thermometer (Fluke Corp. Everett, WA) as a function of time starting from when 6 A of current is supplied to the composite. The composite starts at room temperature and is suspended on a mechanical stage to allow for free air convection. These samples are either clamped at the ends to fixtures or suspended in air with the clamped ends removed and replaced.
by thin VHB strips (for the purpose of minimizing heat loss through contact). Next, the specimen is supplied with 6 A of electrical current using an external power supply. An IR thermometer and a camera are used to record the temperature around the center of the bottom composite surface until the temperature reaches \( \sim 90 \, ^\circ C \). The temperature vs. time profile is then extracted from the video. This profile is used to evaluate the time needed to complete the alloy phase change.

### 2.2.3 Results and Discussions

Following the experimental methods presented above, we measured the effective elastic modulus \( E' \) of the Field's metal composites. For specimens in the non-active and active states. The effective Young’s modulus \( E' \) is obtained by calculating the slope of the curve. At room temperature, the Field’s metal is solid and \( E'_{\text{off}} = 0.86 \pm 0.10 \) GPa (sample number, \( N = 9 \)). When external power is supplied, the Field’s metal melts and the effective Young’s modulus of the composite reduces by four orders of magnitude to \( E'_{\text{on}} = 95.2 \pm 10.5 \) kPa (\( N = 6 \)). A total of twelve samples were tested, three of which were tested in both the activated and non-activated states.

The Galinstan channel in the Joule heating layer is 218.9 mm long, 1.27 mm wide, and 0.5 mm deep. Galinstan has a resistivity of \( \rho = 2.9 \times 10^{-7} \) \( \Omega \).m, so the total resistance is \( R = 0.10 \) \( \Omega \). The heater is activated with \( i = 6 \) A of current and thus consumes approximately \( P = i^2R = 3.6 \) W of power. Therefore, the composite is expected to require \( t = U/P = 102.8 \) seconds to be supplied with the \( U = 370 \) J of energy necessary to melt the Field’s metal. This prediction is roughly consistent with the results presented in Figure. 2.5, which demonstrates that the heated alloy undergoes a phase change after approximately 130 seconds. This phase change corresponds to an inflection in the temperature profile – the rate at which the temperature increases slows down as \( T \) approaches \( T_m \) and then increases again when \( T > T_m \).
There are several potential reasons for why the theory underestimates the actual activation time. One factor is that the semicircular region of the serpentine channel is wider than the rest of the channel and this will reduce the electrical resistance and power output of the Joule heater. Another factor is that when the Field’s metal melts, the temperature of the heater will be much higher than that of the Field’s metal layer. Therefore, the assumption that internal temperature is uniform will be violated and the actual energy required to complete the melting will be greater than predicted. Nonetheless, as demonstrated in Figure 2.5, the theory provides a reasonable estimate of the time it takes for the temperature $T$ of the composite to reach the melting point $T_m = 62^\circ\text{C}$ of Field’s metal.

The measurements performed with a tensile tester demonstrate that soft-matter composites composed of liquid-phase and phase changing metal alloys are capable of dramatic changes in elastic rigidity. According to the stress-strain measurements performed with the Instron, the effective Young’s modulus $E'$ decreases by four orders of magnitude when the composite is electrically activated: $E'_\text{off} = 0.86 \pm 0.10 \text{ GPa}$ versus $E'_\text{on} = 95.2 \pm 10.5 \text{ kPa}$. Likewise, as shown in Figure 2.6, activating the Joule heating element in the SMP-based composite causes the SMP to soften and the composite to become significantly more flexible.

### 2.3 Summary

In summary, I have introduced a rapid prototyping method for liquid metal circuits based on stencil lithography. A CO$_2$ laser engraver is used to pattern a acrylic tape to obtain planar features in which liquid metal is embedded. The acrylic tape is inherently adhesive and this makes it possible to make multi-layer structures. Besides the acrylic
Figure 2.5: Time versus temperature for specimens activated with 6 A of current. Note that removing the clamping ends of the specimens reduced the time needed to induce phase change.

Figure 2.6: (a), (b) Rigidity tunable composite with embedded Galinstan heater and SMP.
tape used in this work, other materials, such as PDMS, can also be patterned with this method and used as channels for liquid metal.

As an application of this fabrication method, we have also demonstrated reversible elastic rigidity control with phase-changing alloy (Field’s metal) and shape memory polymer using a soft-matter Joule heater. The heater and rigidity-tunable material form a composite that becomes elastically soft and stretchable when activated with electrical current. With 6A of current, the composite transforms from a rigid solid that resists stretching and bending to a soft elastomer that easily deforms. Because the heater does not contain any rigid or inextensible parts, it can accommodate the extreme elastic deformation of the stiffness-tunable layer without breaking. Electronic functionality allows the composite to operate with battery power and on-board electronics and eliminates the need for pneumatic, fluidic, or motorized external hardware. The experimental results are in reasonable agreement with theoretical predictions derived from elementary composite mechanics and heat transfer.

Stencil lithography is proven to be an effective method for liquid metal patterning at mm scale. However, for sub-mm scale, the deposition of EGaIn may be influenced by factors like surface tension of liquid metals. Moreover, stencil lithography cannot be used for producing closed or self-intersecting circuit features. For these reasons, a prototyping method that can directly pattern liquid metal at a lower scale is needed.
Chapter 3

Rapid Prototyping with a CO\textsubscript{2} Laser

In this chapter, I present a method for producing soft-matter electronic circuits by patterning thin films of cPDMS and liquid-phase EGaIn alloy with a CO\textsubscript{2} laser (10.6 μm wavelength). I also briefly discuss the role of EGaIn oxidation and moldability to enable patterning without direct photophysical ablation. Figure 3.1 presents examples of how CO\textsubscript{2} laser patterning is used to produce soft-matter sensors for electronic skin applications. These include a fingertip mounted carbon-based cPDMS sensor for detecting skin contact (Figure 3.1a), a miniaturized multi-layered carbon-based cPDMS sensor array (Figure 3.1b), and inclusions of PEDOT:PSS embedded in PDMS (Figure 3.1c) and urethane (Figure 3.1d). The test patterns in Figure 3.1e are produced by using the CO\textsubscript{2} laser to pattern a thin layer of EGaIn alloy sandwiched between two layers of PDMS. After the excess EGaIn and elastomer are removed, the patterned film is sealed in additional PDMS. Figure 3.1f, g present an integrated circuits composed of serpentine EGaIn wires and cPDMS pads for mounting an LED and connecting to an external power supply.

CO\textsubscript{2} laser ablation has been previously used to produce microfluidic channels in
Figure 3.1: (a) Resistive tactile sensor composed of laser-patterned conductive poly(dimethylsiloxane) (cPDMS). (b) Sensor array composed of overlapping strips of cPDMS insulated by non-conductive elastomer. (c) Laser-patterned inclusions of PEDOT:PSS embedded in PDMS. (d) Laser-patterned PEDOT:PSS embedded in polyurethane. (e) Laser-patterned eutectic Gallium-Indium alloy (EGaIn) embedded in PDMS. (f) Integration of a serpentine EGaIn wire and cPDMS electrodes in a PDMS-sealed circuit. (g) LED-embedded circuit composed of laser-patterned cPDMS and EGaIn.
Figure 3.2: (a) Fabrication steps for a resistive tactile sensor composed of a multiple layers of laser-patterned cPDMS. (i) For the bottom half of the sensor, cPDMS is firstly applied on a brass substrate. After curing, it is patterned with the CO$_2$ laser with desired features. (ii) Excess material is removed with the desired features left on the substrate. (iii) The features are sealed with non-conductive PDMS and the bottom half is done and peeled from the substrate after curing. (iv) The top half starts with creating a bi-layered structure of cPDMS and PDMS. (v) This layer is then patterned with CO$_2$ laser. (vi) Before peeling off excess material, a layer of PDMS is applied on top of this bi-layered structure as bonding agent. (vii) Before this layer of PDMS cures, the excess material is removed and the features are left on the brass substrate. (viii) The bottom half of the multi-layered structure that made from steps (i) to (iii) is then attached to the features left on brass substrate. (ix) The multi-layered sensor done after curing. (b) Fabrication steps for laser-patterned EGaIn. (i) A PDMS is applied as the substrate. After curing, EGaIn film is applied on top of it. (ii) an extra layer of PDMS is then applied on top of the EGaIn. (iii) This tri-layered structure is then patterned with laser. (iv) The excess material is removed and (v) the features are sealed in PDMS. (vi) The fabrication process is done after curing.
PMMA [68] and PDMS. Polymers are typically patterned through a combination of vaporization and the displacement of molten polymer. Molten polymer is displaced when the pressure of the escaping vapor (recoil force) exceeds the surface tension of the liquid. We postulate that thin films of PDMS filled with conductive microparticles can be similarly patterned with a CO$_2$ laser. This includes carbon based cPDMS, such as PDMS filled with acetylene carbon black, and PDMS filled with silver and nickel microparticles.

In contrast to elastomers, liquid-phase GaIn metal alloys cannot be ablated with a CO$_2$ laser. As with other metals, EGaIn would require a high-energy UV radiation source for photochemical ablation. Instead, we postulate that EGaIn is removed with the same liquid metal expulsion mechanisms used in CO$_2$ laser beam welding (LBW). However, whereas stainless steel, titanium, and other solid metals require a high power to initiate melting (>1 kW for CO$_2$ lasers), GaIn alloys are already molten at room temperature and, as with molten polymer, can be displaced by pressure from the vapor escaping the PDMS beneath. In addition to being in a liquid state, GaIn alloys form a surface oxide of Ga$_2$O$_3$ that prevents the liquid from flowing back into the laser-patterned region. In other words, this property of moldability enabled by the oxide skin [35, 71–73] allows the liquid to retain its shape after it has been patterned. Since they have both low fluidic viscosity and a high surface tension, GaIn alloys can simultaneously behave like a solid and a liquid at sub-millimeter length scales. This unique combination of properties enables the rare ability to pattern a metal alloy with an inexpensive laser engraving system that operates at relatively high wavelengths (10.6 $\mu$m) and low power (1-30 W).
3.1 Method

I use a 30 W CO$_2$ laser (VLS 3.50; Universal Laser Systems, Inc.) to pattern thin films of electrically conductive cPDMS and EGaIn and produce elastic, soft sensors and circuit elements. The detailed procedures are described below.

3.1.1 Patterning cPDMS

The methods for patterning cPDMS and a multi-layered circuit are presented in Figure. 3.2a. As discussed in the Experimental Section below, carbon-based cPDMS is composed of 84 wt% 20:1 PDMS (Sylgard 184; Dow Corning, Inc.) mixed with 16 wt% acetylene carbon black powder (Alfa Aesar). We have also patterned films of cPDMS containing either 84% silver microparticles (2-3.5 μm; Sigma Aldrich, Inc.) or a combination of 65% nickel and 25% silver (NuSil R-2637; NuSil Technology LLC). First, a thin film of cPDMS is applied by spin coating the uncured elastomer on a brass sheet (700 and 2000 RPM, for 9 and 25 s, respectively) and laser-patterning it with 0.6 W, 5% speed, and 1000 PPI (repeat for 3 times) after curing (130 °C for 10 min) (i), followed by removal of the excess cPDMS (ii). The brass sheet is finally coated with a sealing layer of non-conductive PDMS (10:1) before peeling the cured film from the brass cutting surface (iii). For this sequence of steps, which was used to produce the tactile sensor in Figure. 3.1a, the embedded cPDMS circuit is exposed and flush with the surrounding PDMS. However, to produce multi-layered circuits, such as the sensor array in Figure. 3.1b, additional layers of conductive and non-conductive PDMS must be added. For a multilayered circuit, the bottom layer is patterned and sealed as before (iii). After removal of this sheet, the cutting surface is coated with a new layer of cPDMS and subsequently PDMS after curing (iv). The two layers are then patterned with the laser (v) before a final coating of PDMS (vi) and removal of excess material (vii). The uncured PDMS deposited after patterning the two sheets
(vi) acts as an adhesive film for bonding the top layer with the previously fabricated bottom layer (viii). Lastly, the completely cured multi-layered circuit is peeled from the cutting surface (ix).

3.1.2 Patterning Liquid Metal

Films of liquid EGaIn alloy are patterned using the steps presented in Figure. 3.2b. I begin by spin coating (1000 RPM for 10 s) a layer of PDMS on a brass cutting sheet. Next, an elastomeric roller (PDMS) is used to deposit a 10-20 μm thin layer of EGaIn and the excess EGaIn is removed by using a spin coater at 2000RPM for 10 s (i); a minimum thickness of 10 μm has been measured with optical profilometry (Zygo NewView 7300 Optical Profilometer). An extra layer of PDMS is spun-coat on the GaIn alloy (700 and 1700 RPM, for 9 and 5 s, respectively) and cured (130 °C for 10 min) (ii) before patterning the alloy and underlying PDMS substrate with a CO₂ laser (0.6 W, 5% speed, and 1000 PPI. Repeat for 3 times) (iii). This extra layer of PDMS serves to prevent the EGaIn from oxidizing and getting coated in debris during laserpatterning. Next, we remove the excess material (iv) and seal the patterned circuit (v). After the sealing layer has cured, we peel the circuit from the brass cutting sheet (vi). This approach to EGaIn patterning exploits the alloys liquid state and strong surface tension, which allows it to hold its shape after being cut with a CO₂ laser. It should be noted that the spin coating parameters for EGaIn removal were selected based on the idea that the excess EGaIn should be removed at the spin rate that is higher than that used for the deposition of the PDMS protection layer, so the thickness of EGaIn can stay constant when deposit the PDMS protection layer. The relationship between thickness of EGaIn and spin rate still needs to be studied.
3.2 Results and Discussion

Figures 3.3b,c demonstrate CO$_2$ laser machining of single and double-layers of cPDMS with laser powers of 0.6 and 1.2 W, respectively. For a single layer of cPDMS, a minimum line-to-line spacing of 260 $\mu$m is possible (Figure 3.3c). This maximum resolution is limited by the laser spot size and pulse rate. Smaller feature sizes can be achieved by a lower wavelength laser with a smaller beam diameter and higher pulse rate to prevent heating and melting of the surrounding material. The samples presented in Figures 3.3ac are produced with carbon-based cPDMS although this same technique can also be used to pattern silver/nickel- (Figure 3.1f) and silver-based (Figure 3.3d) cPDMS. Because of their higher viscosity, thin layers of these metal-based conductive elastomers may be prepared with a thin film applicator (ZUA 2000; Zehntner Testing Instruments) instead of spin coating. As demonstrated in Figure 3.4, a CO$_2$ laser can also be used to pattern poly(3,4 ethylenedioxythiophene) poly(styrenesulfonate) (PEDOT:PSS; Sigma Aldrich, Inc.) and embed it in either PDMS (Figure 3.4a) or soft polyurethane (Ure-Bond II; Smooth-On, Inc.) (Figures 3.4b,c).

The samples presented in Figures 3.5 a-e demonstrate the versatility of this approach to rapidly pattern liquid GaIn with millimeter-scale resolution over a large area. Figure 3.5f presents an integrated circuit composed of laser-patterned EGaIn and conductive elastomer electrodes (NuSil R-2637) used to mount an LED and connect the circuit to an external power supply. This sample demonstrates the ability to pattern multiple conductive materials and maintain conductivity.

I have demonstrated that thin films of cPDMS and EGaIn can be patterned with a CO$_2$ laser to produce intrinsically soft sensors and circuits that are elastically compatible with human skin. The steps presented in Figure 3.2 require a relatively inexpensive commercial laser engraving system and can be performed in minutes. As with other polymers and elastomers, we expect that the cPDMS is patterned through
Figure 3.3: (a) Test sample of a single layer of cPDMS patterned with a 0.6 W laser power. (b) Test sample of a double-layer cPDMS patterned with 1.2 W. (c) Patterned carbon-based cPDMS with a minimum line-to-line spacing of 260 μm. (d) Patterned silver-based cPDMS.

Figure 3.4: (a) PDMS embedded with laser-patterned inclusions of PEDOT:PSS. (b),(c) Urethane embedded with laser-patterned PEDOT:PSS.
Figure 3.5: (a),(b) PDMS embedded with laser-patterned inclusions of EGaIn. (c),(d) Large area sample. (e) Miniaturized features: 2 mm × 2 mm squares spaced 0.25, 0.5, and 1 mm apart; concentric rings with diameters of 1.5, 3, and 5 mm and widths of 0.25, 0.35, and 0.5 mm, respectively; circles with diameters of 0.5, 1, 1.5, and 2 mm; rectangular strips with widths of 0.3, 0.7, and 1 mm. (f) LED circuit containing laser-patterned (Ag/Ni)cPDMS and EGaIn.
a combination of photothermal ablation and liquid expulsion. However, the ability to
pattern EGaIn with a CO\textsubscript{2} laser is surprising since GaIn is a metal alloy that does not
ablate at 10.6 \( \mu \text{m} \) and will not vaporize at 1-30 W to produce the vapor recoil force
necessary for liquid expulsion. Instead, we postulate that the EGaIn is displaced by
the recoil force of the PDMS vapor generated below the liquid metal film.

As illustrated in Figure. 3.6a, the liquid alloy is deposited between two thin layers
of PDMS. When exposed to the laser, the sample is locally heated (Figure. 3.6b) and
PDMS in the top and bottom layers is ablated (Figure. 3.6c). We then postulate
that the vaporized polymer exerts sufficient pressure on the EGaIn film to penetrate
through and escape (Figure. 3.6d). As in conventional laser beam welding (LBW),
displacement occurs when the recoil force from this pressure exceeds the surface force
supported by the surface tension \( \gamma_0 \) and yield stress \( \tau_0 \) of the liquid. [35] These values
are estimated by pendant droplet measurements on EGaIn, which suggest \( \gamma_0 \approx 0.445 \)
J m\(^{-2}\) and \( \tau_0 \approx 119 \) Pa. [74] Interestingly, pendant droplet experiments on Galinstan
in pure nitrogen (\(<0.5 \) ppm O\textsubscript{2} ) show greater surface tension (0.535 J m\(^{-2}\) ) than in
an oxygenated (500 ppm O\textsubscript{2} ) environment (0.510 J m\(^{-2}\) ).[73] In practice, the laser
has a cutting radius of approximately \( r_c \approx 50 \) \( \mu \text{m} \), which implies that the vaporized
polymer exerts pressure on a circular region of GaIn with a circumference \( \eta = 2\pi r_c = 314 \) \( \mu \text{m} \). For a film thickness \( h = 10 \mu \text{m} \) and assuming \( \gamma_0 \approx 0.5 \) J m\(^{-2}\) the force required
to puncture the oxide skin and displace the liquid alloy is approximately \( F = \eta (\gamma_0
+ \tau_0 h) \approx 157 \mu \text{N} \). This corresponds to a difference of 20 kPa between the local vapor
pressure and the atmospheric pressure.

For comparison, the recoil pressure of vapor under ambient conditions is estimated
to be \( p_r \approx 0.55 p_s (T_s) \), where \( p_s \) is the saturation vapor pressure and \( T_s \) is the
surface temperature of the melting substrate.[75] An approximate estimate for \( p_s \) is
obtained from the Clausius-Clapeyron relation and Troutons rule (valid for many
organic liquids): \( \ln(p_s/p_{atm}) \approx -10.6(T_v / T_s - 1) \), where \( p_{atm} = 101.3 \text{ kPa} \) is the atmospheric pressure and \( T_v \) is the boiling point of the molten polymer. [76] Assuming \( T_v = 473 \text{ K} \) and a surface temperature \( T_s = 523 \text{ °C} \), that is, \( T_s \) the recoil pressure will be approximately 152 kPa, which corresponds to a pressure difference of 50.7 kPa exerted on the liquid alloy film. Another value for comparison is the direct radiation pressure exerted by the laser beam itself. For a maximum power of \( P=30 \text{ W} \), this is approximately \( p = nP/\pi cr_c^2 = 24.1 \text{ Pa} \), where \( n = 1.89 \) is the refractive index of the Ga\(_2\)O\(_3\) oxide surface [77] and \( c = 3 \times 10^8 \text{ m/s} \) is the speed of light. This is orders of magnitude less than the estimated pressure necessary to displace liquid GaIn and suggests that force from the beam alone is not sufficient for patterning.

Referring to Figure. 3.3, linewidths of 118 \( \mu \text{m} \) are accomplished with a cutting power of 0.6 W, PPI (pulses per inch) 1000, and speed 20%. Although not at the length scale necessary for microelectronics, 0.1-1 mm linewidths are adequate for a variety of sensing applications, including the contact detection sensors presented in Figure. 3.1a,b. The sensor in Figure. 3.1a consists of a central electrode and four surrounding electrodes. When the fingertip makes simultaneous contact between the inner circle and one of the outer triangles, an electrical connection is formed. Contact between different peripheral electrodes can be used to signal an “up”, “down”, “left”, or “right” command. Similarly, for the sensor in Figure. 3.1b, the location of the contacting fingertip can be determined by measuring the electrical resistance between electrode rows and columns at each node.

In this study, I have focused on elastically soft circuits with 0.11 mm linewidth circuit features that can be produced with a CO\(_2\) laser. For microelectronics with 20-100\( \mu \text{m} \) linewidths, a lower wavelength laser is required. The fundamental advantage of UV laser micromachining is the greater focal depth \((F)\), and hence cutting depth, that can be achieved for a prescribed beam diameter (i.e., waist size, \(w_0\)). The VLS
Figure 3.6: Proposed mechanism for expulsion of a thin film of liquid GaIn alloy on a PDMS substrate. (a) The liquid alloy is coated with a thin layer of PDMS to prevent oxidation and limit exposure to debris. (b) When exposed to the laser, the sample is locally heated and (c) PDMS from the top and bottom layers vaporize. (d) When the pressure difference between the vaporized polymer and atmosphere exceed the surface tension of the liquid film (including its Ga$_2$O$_3$ skin), the vapor will puncture the liquid film and escape. The sides of the GaIn traces are still exposed.
3.50 CO₂ laser engraver used for fabricating these devices has a beam diameter of 100 μm.[78] For a Gaussian beam, \( F = 2\pi w_0^2/\lambda \), where \( w_0 \) is the minimum diameter of the beam (“waist size”) and \( \lambda \) is the wavelength of the laser. This implies that, in theory, a 355 nm Nd:YAG laser should have on the order of 30× the cutting depth of a 10.6 μm CO₂ laser. Also, the lower wavelength of a UV laser allows for a much higher pulse rate (e.g., \( \approx 100 \) kHz). This prevents heat from accumulating outside of the beam path, which would lead to melting and burning in the surrounding material and hence result in wider cuts.

### 3.3 Tactile Sensors with Skin as Conductor

Here, I introduce several ways to use CO₂ laser patterning to produce tactile sensors composed of insulating and conductive PDMS. When filled with acetylene carbon-black, PDMS is conductive (cPDMS) [15] and can transmit voltage signals transferred through human skin during tactile data entry. In these examples, a sensor array is connected to an external data acquisition board (DAQ) through a flat flexible cable (FFC), which can be bonded to the cPDMS terminals with a Z-tape acrylic-based pressure sensitive adhesive elastomer (3MTM ECATT 9703) that is only conductive through its thickness (Z-Axis). Examples of wearable sensors produced with this technique are presented in Figure. 3.7.

Successive layers of insulating PDMS and conductive cPDMS are patterned with a CO₂ laser[16] and bonded together to form multi-layer soft circuits. Successful laser patterning has also been demonstrated with poly(3,4-ethylene dioxythiophene): poly(styrene sulfonate) (PEDOT:PSS) conductive polymer, conductive nickel and silver-filled PDMS elastomer (e.g. AgPDMS), and liquid-phase gallium-indium metal alloy (e.g. EGaIn). Depending on their architecture and integration with external DAQ hardware, these soft-matter circuits can be designed to detect surface pressure
Figure 3.7: Tactile sensors made with cPDMS by laser patterning: (a) single layer arrow pad with coplanar electrodes: (b) multi-layer number input pad.

and sliding friction, stretch, and bending curvature. Presently, this is accomplished by detecting changes in resistance\[79\] or capacitance\[24, 44, 49\] induced by mechanical deformation or electrical contact with skin. However, in the case of resistive sensing, this requires moderate power to supply current to the circuit and can lead to Joule heating. Although capacitive sensing requires significantly less power, it requires supporting electronics to filter noise generated by parasitic capacities from the environment \[80\]. In this study, I instead focus on an alternative sensing method that detects fingertip contact through a binary voltage response. This low-power, low-noise approach to tactile sensing only detects the location and duration of finger contact and cannot be used to measure the intensity of applied pressure.
Figure 3.8: When any of the surrounding electrodes is connected to the central electrode, the voltage on it jumps from zero to a certain value. Fingertip is equivalent to a resistor in series. (b) When fingertip contacts the three electrodes at an intersection node, its equivalent to two resistors in parallel followed by a resistor in series.

**3.3.1 Sensing Principles and Designs**

The two sensor architectures are presented in Figure 3.8 and referred to as Sensor A and Sensor B. These correspond to the co-planar and multi-layer (grid) designs, respectively. Fingertip contact is detected through a jump in voltage delivered to the DAQ through a signal electrode. This requires a counter (or reference) electrode that is always supplied with a fixed voltage $V_0$ (12V, as shown in the illustration). When the fingertip is in contact with both the reference and signal electrodes, its natural conductivity transfers the voltage signal between the pair of electrodes, forming and electrical connection between the power supply to the DAQ. Because of the large resistivity of the cPDMS electrodes ($R_e$) and skin ($R_s$), the electrical power drawn from the power supply ($P = V_0^2/(R_e+R_s)$) will be low. Moreover, in contrast to resistive sensing, voltage-controlled tactile sensors will only consume power during contact and will not change temperature significantly through Joule heating.
Figure 3.9: Designs of (a) game controller, (b) number input pad, and (c) a zoom-in view of design details of number input pad.
Referring to Figures 3.8a and 3.9a, Sensor A is composed of a single layer of co-planar electrodes. Implementation is demonstrated below in Sec. 3.3.2 with a wearable arrow pad that can be interfaced with software to move objects on a screen. The arrow pad consists of a central reference electrode and four surrounding cPDMS signal electrodes that are all exposed and flush with the surrounding insulating PDMS. When a fingertip is in simultaneous contact with the reference and a signal electrode, the voltage on the signal electrode immediately increases and contact is detected by the DAQ board and software. As illustrated, a four-key arrow pad requires five channels (one for each signal electrode and one for the reference). These are connected to the DAQ via an FFC, which is bonded to the ends of the cPDMS traces through either pressure fitting or Z-tape.

Sensor B, shown in Figures 3.8b and 3.9b uses similar principles for tactile sensing. The sensor array is composed of an overlapping grid of electrode rows and columns. This layout is similar to the tactile sensor reported in but the sensing principle is different. To demonstrate the multi-layer sensing system we designed a multi-layer number input pad (Figure 3.9b). The bottom layer includes four row electrodes and a reference electrode connected to the power supply. The top layer contains three column electrodes. When a fingertip touches any of the intersections, three electrodes one column, one row, and the reference are connected. By monitoring the change in voltage for each of the seven signal electrodes, it is possible to register the location of fingertip contact by registering which row and column is activated. As with Sensor A, we use $V_0 = 12$ V for the reference voltage.

The central electrode of Sensor A has a diameter of 20 mm and is spaced a distance of 0.48 mm from the surrounding electrodes. For Sensor B, the outer diameter of each column and row electrode is 7 mm and 5 mm, respectively. The spacing between the row and power electrode is 0.35 mm. The insulating layer has the same planar
dimensions as the column electrodes. The keys in Sensor B are spaced 9 mm apart in both the horizontal and vertical directions. For both sensors, the terminals connected with FFC have a trace width of 0.4 mm and are spaced 0.15 mm apart.

### 3.3.2 Sensor Fabrication

Both types of sensors are composed of insulating and conductive PDMS. The conductive PDMS is prepared by mixing PDMS (Sylgard 184; Dow Corning, Inc), carbon black powder (100

Figure. 3.10 shows the fabrication steps for producing Sensor A. First, conductive PDMS is spun coated on a piece of brass sheet (McMaster-Carr, Inc.). The spin coating includes two stages. In each stage, the spinning speed is 700 and 2000 rpm and the duration is 9 and 25 seconds, respectively. After spin coating, the brass sheet is placed on a hot plate at 130°C for 10 minutes and the cPDMS is cured. Layers of different thickness can be obtained by adjusting the spin coat speed and timing accordingly. Layer thickness can also be controlled by the viscosity and composition of the uncured cPDMS.

After curing, a laser engraver (VLS 3.50; Universal Laser System, Inc) is used to pattern the conductive PDMS. A high definition lens is used for patterning. The settings are power of 0.6 W, laser head speed of 0.0125 m/s, 1000 PPI, and 0.3 mm in Z-axis. The patterning is repeated three times in order to ensure sufficiently clean and deep cuts. Greater power and/or slower speed would allow for less passes but also result in greater heat accumulation, which can lead to burning of material around the ablation zone. After laser patterning, the excess material is manually removed, leaving only the patterned electrodes on the brass substrate. Next, the electrodes are sealed with an insulating layer of PDMS (Sylgard 184; Dow Corning; 10:1 base-to-catalyst ratio). The PDMS is deposited with a spin coater and, as with cPDMS, its
Figure 3.10: Fabrication methods for single layer sensor (Sensor A) and multi-layer sensor (Sensor B)
thickness can be controlled adjusting the settings of spin coating. After heating with the same hot plate settings, the PDMS is cured and the sensor can be removed from the brass substrate.

The multi-layer circuit for Sensor B requires additional fabrication steps. The bottom layer, which contains the row and reference electrodes, is fabricated in the same way as shown for Sensor A. The top layer, which contains the column electrodes, is produced using the steps shown in Figure 3.10. First, a thin layer of cPDMS is spun coat on a brass cutting substrate. After curing, a layer of PDMS (10:1) is spun coat on the top of cPDMS. In both steps, the settings for spin coating are the same as for Sensor A. After curing on hot plate at 130°C for 5 minutes, laser engraving is used to pattern the PDMS coated electrodes. Before removing the excess material, a very thin layer of PDMS (10:1) is spun coated on the patterned PDMS-cPDMS layer. For stages 1 and 2 of the spin coating, the spinning speeds are 700 and 3500 rpm and durations are 12 and 30 seconds, respectively. Next, the excess material is removed and top layer is attached to the bottom layer. Finally, the composite is cured for 10 minutes at 130°C and then peeled from the brass sheet.

### 3.3.3 Results and Discussion

Reliable, low-power, low-noise tactile sensing is successfully accomplished using the architectures presented in Figure 3.8. Moreover, samples are produced rapidly with a commercially-available CO₂ laser, eliminating the need for customized printing hardware. As shown in Figures 3.11 and 3.12, the sensors are designed to function as keypads that can be placed on the skin and record tactile inputs. Because the keypads are made entirely out of soft elastomer, they can bend, stretch, and conform to skin without irritation or introducing mechanical constraints.

Application demos are performed to validate the functionality of the two types
Figure 3.11: Tactile output from a co-planar arrow pad sensor (Sensor A).
of tactile sensors. The sensors are connected to a DAQ (USB-1208fs, Measurement Computing Corp) through a breakout board and FFC (GP-2106, SparkFun, Inc.). The traces in the FFC is either pressure fitted to the aligned cPDMS terminals using a staple or bonded with Z-tape (3M™ ECATT 9703). Z-tape has the unique advantage of matching the elasticity of cPDMS and only exhibiting conductivity through its thickness. This prevents shorting between non-adjacent electrodes and eliminates the need for additional patterning and alignment.

The four signal electrodes in the arrow pad function as up, right, down, and left keys. They are connected to the DAQ through four analog input channels. The center
electrode is connected to a DC power supply (Model DIGI 360, Electro Industries, Inc.) which is set at 12 V. A script is created in MATLAB R2014a to display the tactile input as XY motion of a cursor. As shown in Figure 3.11, pressing the keys of the arrow pad moves a black square around the screen. The path of motion is traced in gray.

Sensor B is implemented as a number pad (Figures 3.9 and 3.12), with four row and three column electrodes intersecting to function as twelve keys (‘0’-‘9’, ‘*’, and ‘#’). The signal electrodes are connected to the DAQ through seven analog input channels. By touching any intersection node, the voltage in a corresponding pair of row and column electrodes will be detecting and the corresponding number or symbol can be told. In our experiment we test the number pad in the following sequence: 1-4-7-*-0-8-5-2-3-6-9-#. As shown in Figure 3.12, the voltage inputs for the activated row and column electrodes are consistent with our input.

Because they respond to binary voltage input, the sensors exhibit virtually no noise and are sensitive to light finger pressure. When placed on a digital weight scale (SP2001, Ohaus, Corp.), 98 mN of minimum applied force is measured as necessary to activate a key on the arrow pad. Food coloring was used to determine a fingerprint contact area of 0.73 cm². This corresponds to approximately 1.342 kPa of applied finger pressure which is comparable with pressure required by a common keyboard.

It should be noted that although the sensors are operational, the voltage delivered to the DAQ through the sensing electrodes are far less than the voltage on the reference electrode. For the arrow pad, the voltage change in each channel is about 0.2 V while for the number pad, the voltage change in row electrodes and column electrodes 1 and 2 is about 0.2 V while the voltage change in column electrode 3 is no higher than 0.1 V. Multiple prototypes have been produced for both sensor types and in each case we do not observe signal voltages of greater than 0.5 V. This dramatic drop
in voltage is primarily attributed to electrical grounding through human skin but can also be caused by the poor conductivity of the cPDMS electrodes. Fluctuations or changes in cPDMS resistance over time may degrade functionality. It should also be noted that the sensitivity of the sensors is related to the condition of fingertip. For example, we observe that the sensors are more responsive to sweat-tipped fingers.

3.3.4 Summary

In summary, I present two novel architectures for tactile sensing. As with the touchscreens used in personal electronics, these sensors use the natural conductivity of human skin. Moreover, because they are lightweight and made entirely out of soft and elastic material, they can be worn on the body without altering the natural mechanics of skin. Lastly, sensor arrays can be produced rapidly (in minutes) using a variety of fabrication methods, including patterning with a commercial CO$_2$ laser engraver.

Keypad implementations for both sensor architectures demonstrate sufficient accuracy for tactile data entry. The pressure required to activate the keys is comparable to that used in conventional keyboard typing. This is consistent with predictions from an idealized model based on Hertzian contact theory. The major limitation of this technique is the long-term reliability issue due to the surface contamination and this can potentially be improved by increasing the conductivity of the sensing and reference electrodes. Possible alternatives include AgPDMS, EGaIn, [52] PEDOT:PSS [81, 82] or other easily printable/patternable material that exhibits significantly more conductivity than cPDMS.

In addition to this, the resolution of the sensors is limited. While seeking methods and materials to make thinner electrodes, we will also work on a flexible touch pad which has only four leads but theoretically infinitely high resolution. Graphene is a
candidate as conductive materials. Additionally, by using transparent conductive materials such as PEDOT:PSS, sensors with transparent electrodes can hopefully be fabricated for applications like soft-matter touch screen.

3.4 iSkin: Flexible, Stretchable and Visually Customizable On-Body Touch Sensors for Mobile Computing

The human skin is recognized as a promising input surface for interactions with mobile and wearable devices. However, it has been difficult to design and implement touch sensors that can be placed directly on the skin. With the techniques developed from the tactile sensors presented in last section, a series of flexible, stretchable and visually customizable on-body touch sensors, iSkin, have been co-developed with Martin Weigel at Max-Planck Institute in Saarbrucken, Germany for mobile computing applications. iSkin is a mobile human-computer interface that can be in direct contact with human skin. Thanks to the materials and fabrication method introduced in Section 3.1, iSkin is highly customizable and suitable for different applications. Figure 3.13 shows several customized implementations. One example is a very thin, yet large input area, which can be wrapped around a finger and is stretchable enough to be worn over joints. It features three touch buttons and one linear slider with five elements. It allows for fast and direct control of mobile devices using touch input even when the hands are busy.

iSkin relates to the emerging stream of on-body interaction, e.g.[84–92]. It is technically based on advances in electronic skin (e-skin) and soft-matter electronics [93–96], an active research field in robotics and material science. To our knowledge, our work is the first investigation into how electronic skin can be used for on-body
Figure 3.13: iSkin is a thin, flexible, stretchable and visually customizable touch sensor that can be worn directly on the skin. We present three novel classes of on-body devices based on iSkin: (a) FingerStrap, exemplified here with a strap on the index finger for fast, one-handed control of incoming calls; (b) Extensions for wearable devices, exemplified here with a rollout keyboard attached to a smart watch; and SkinStickers, exemplified here with (c) an input surface for a music player attached to the forearm, (d) a click wheel on the back of the hand and (e) a headset control behind the ear.
interactions to control mobile computing devices; this includes interactive scenarios, sensing principles, form factors and device types as well as aesthetics[97].

In the materials science community, first proofs of concept have shown the technical feasibility of sensor overlays for touch sensing on the body [16, 79, 98]. Kramer et al. presented a pressure sensitive skin overlay composed of PDMS embedded with microfluidic channels of eutectic gallium-indium (EGaIn) alloy [79]. In Section 3.3, I also demonstrated the feasibility of patterning cPDMS with a laser engraver to produce an alternative to EGaIn for soft-matter sensors and circuits. This allows for tactile sensing through direct electrical contact between the circuit and human skin. However, sensing with exposed electrodes is not suitable for quantitative sensing because skin conductance heavily varies between users and electrodes might become stained or worn through skin contact. Woo et al. demonstrated pressure and strain measurements using a compressible layer of EcoFlex [98]. While the measurement is continuous, the sensor is unable to differentiate between pressure and strain. Furthermore, this approach relies on microcontact printing and cleanroom microfabrication and therefore lacks possibilities for rapid customization. Our sensor adds to this body of research by capturing two levels of normal force independently of how much it is stretched, an important requirement for robust on-body input. Moreover, it is compatible with rapid prototyping in a simple lab environment and can be visually customized for an aesthetic appearance.

The HCI community has presented several sensing techniques to capture input on the skin for interaction with mobile devices. Prior work can be split into five sensing approaches. One is camera-based sensing, with RGB cameras [92, 99] or depth cameras [86, 87, 100, 101]. This allows for interacting directly on the skin, but requires direct line-of-sight to a camera and is susceptible to lighting conditions. Second, bio-acoustic sensors [88] work on many body locations and do not require a sensor overlay,
Figure 3.14: iSkin device types highlighting various forms of attachment: (a) wrapping around a body location, (b) attaching iSkin to a body-worn device or accessory, (c) sticking on skin with a skin-friendly adhesive.

at the cost of low spatial sensing resolution and single-touch-only sensing. A third approach consists of magnetic sensing [84] using magnets and Hall-effect sensor grids attached to the body. Those approaches do not require a skin overlay, but fall short in detecting precise touch-down and touch-up events. We designed iSkin to be thin and soft enough to retain much of the sensory stimulation that occurs naturally and upon touch. Finally, photo-reflective sensing [102] is based on two armbands with infrared reflective sensors. These are able to recognize deformations of skin, but do not recognize touch, which has been suggested to be the most important on-skin modality[103].

iSkin also relates to flexible sensors for smart clothing and wearables [89, 104, 105]; those support interaction in close proximity to, but not on, skin. In addition, iSkin is related to flexible touch sensors for use on objects [106–108]. These are well suited for rapid prototyping of input on non-planar surfaces. However, these approaches were not intended to be used on skin and thus are not elastic, bio-compatible and robust enough for skin interaction.
3.4.1 Design Goals for iSkin

As a human-computer interface, iSkin must satisfy several requirements. Firstly, it must be compatible with human skin. To be specific, it needs to be thin, stretchable (around 20% [95]), lightweight, and non-toxic. Secondly, iSkin must be able to fit different body locations (Figure 3.14) in order to have different application, such as palm [100], finger [84], arm [109] [103], and head [110]. We demonstrate that iSkin can be worn on these locations. In addition, its flexibility allows for exploration of other on-body locations as well as use underneath or on top of clothing. Thirdly, iSkin needs to be easy to attach or detach and robust to movement and wear. Fourthly, since visual appearance is very important for iSkin, the patterns need to be customizable.

3.4.2 Materials, Design, and Sensing Principles

This section introduces the materials, design, and sensing principles of iSkin. The sensing designs and principles were proposed by Martin Weigel and I selected materials and performed the implementation.

3.4.2.1 Materials

iSkin is made of multiple layers of thin, flexible and stretchable silicone. The matrix material is PDMS ancPDMS is used as elastic conductor elastic conductor. The carbon particles make the material appear black and opaque. PDMS and cPDMS are permeable to oxygen, but cPDMS does not oxidize at room temperature. Therefore the electrical resistance of the electrodes remains fairly stable over time. Compared with other elastic conductors, such as liquid phase conductors [79], conductive meshes [111], or AgPDMS [16], cPDMS is very cheap, can be realized in a thinner form factor and neither encapsulates nor exposes harmful substances.
The cost of material for a letter-sized sheet is about 1 USD. Therefore the sensor patch can be designed for one-time use, if desired. Alternatively, it can be used for a longer time without problems, as the material is robust, can be cleaned with water and can even be disinfected for hygienic reuse.

3.4.2.2 Electrode Design

Both sensing techniques share the same physical structure, illustrated in Figure. 3.15: two embedded electrodes are overlaid and held apart with a spacing layer. The embedded conductive traces and electrodes are realized with cPDMS. We use solid layers of PDMS on top and on the bottom to seal off the electrodes from contact with skin and the environment. PDMS is also used for the spacing layer in between both electrodes. The spacing layer is solid at areas where no electrodes are located; it is permeable in between electrodes, to allow for pressure creating a conductive connection. At areas where no electrodes or wires are laid out, only the transparent base layer needs to be realized. The sensor is very thin: from 190 μm at areas where no electrodes or wires are laid out to approximately 700 μm at locations where all layers are realized. Given the high resistance of cPDMS, conductive traces need to be fairly wide. We identified the minimal width of a trace for robust conductive connection to be 100 μm.

3.4.2.3 Fabrication

PDMS is produced by mixing a silicone elastomer base with a silicone elastomer-curing agent (both from Sylgard 184; Dow Corning, Inc.) in a weight ratio of 10:1. For cPDMS, 13% (by weight) of acetylene carbon black powder (Alfa Aesar) is added to the uncured mixture of PDMS (weight ratio 20:1). The material can be formed to thin films using a spin-coater or thin-film applicator. We found that it helps to make
the cPDMS film very thin (∼ 100 μm), as this reduces sedimentation of the carbon black powder to the bottom of the film during curing, which would result in a lower conductivity.

The functional sensor is produced with laser-patterning using a method introduced in [16]. Figure. 3.15a shows the composite structure of our sensor, which is composed of PDMS and cPDMS layers. Before application, the layers are laser-patterned to create conductive lines and electrodes (in cPDMS) or insulating areas (in PDMS). We use a 30 W laser engraver from Universal Laser Systems (VLS 3.50) for patterning. Each layer is bonded to the composite by adding a very thin layer of uncured PDMS as connective glue. As soon as the PDMS is cured, the layers are firmly attached. To increase breathability, the final sensor could be perforated as described in [112].

3.4.2.4 On-Body Touch Sensing

Sensing touch input on the body with cPDMS faces multiple challenges. First and foremost, cPDMS is a very poor conductor. Its resistivity is as high as 100 Ω.m [15] and further decreases when it is being stretched (it even takes several hours to go back to its initial resistance). Secondly, both capacitive and resistive sensing exhibit unique
challenges: permanent contact with human skin results in added capacitive coupling, while the curvature of the body disallows using the standard approach for inter-layer spacing in resistive touch sensing. In the following, we address these challenges and show how to implement robust touch sensing. We present a soft-matter electrode design that supports both projected capacitance and resistive touch sensing. Both techniques give precise real-time information about touch down and up events. Both modes combined allow for distinguishing between two levels of touch pressure. In contrast, resistive sensing alone is less prone to accidental input, as more pressure is required to trigger a touch down event.

**Capacitive Sensing for Light Touch Contact**  
Projected capacitive sensing uses capacitive coupling between the two electrodes (Figure. 3.15b). The bottom electrode connects to a 5 V square wave signal of 1.000 kHz generated by a wave generator (Agilent 33210A). If the sensor contains several separate touch-sensitive areas, the top electrodes are time-division multiplexed to sequentially measure the transmitted signal on each of them. The received signals are processed by a PC oscilloscope (PicoScope 6402A). Bringing a finger near the surface of the sensor changes the local electric field, which reduces the mutual capacitance. Therefore the signal amplitude decreases (Figure. 3.15c). After calibration, the sensor can reliably detect touch events despite the high resistance of the conductor. The sensor needs to be calibrated after it is applied to the skin. It reacts on very slight touch contact, as known from commercial capacitive touch sensors.

**Resistive Sensing for Firm Pressure**  
Resistive touch sensing relies on pressure to create a contact through the permeable spacing layer between both electrodes. A firm touch physically closes the circuit (Figure. 3.15d). In this case, the waveform of the received signal on the upper electrode is changing, serving as a reliable indicator
for firm touch.

To ensure both layers are reliably spaced apart even when they are curved or stretched, our solution uses uniform tiling with a hexagon pattern, similar to honeycombs. This improves the robustness against deformations occurring on the body while minimizing the required spacing material to decrease the required pressure for touch detection. For our prototypes we used a hexagon diameter of 1.5 mm.

**Sensing of Two Levels of Touch Pressure** Combined projected capacitive and resistive sensing enables sensing of two levels of normal force: capacitive sensing detects light touches, while resistive sensing detects firm touches. The sensing techniques use the same physical electrode structure, the same sensing circuit and are performed in the same sensing cycle. Therefore, the frame rate of sensing is not reduced. Figure. 3.15b–d shows an example of the values captured for light and firm touches.

Results from our technical evaluation below show that this approach is capable of reliably distinguishing between both pressure levels, independently of how much the sensor is stretched or bent. We consider this robust detection to be a very important requirement for successful on-body interfaces. While continuous normal force could in principle be captured using a force-sensitive resistor approach, sensor readings would be corrupted by large changes in resistance that result from stretching, which naturally occurs during use on human skin.

**Interactive Widgets** The electrode design of both techniques allows for flexibly shaped interactive areas and senses precise touch down and touch up events. This allows for designing more complex widgets, such as sliders or click wheels. An example of a five-element slider is implemented in the FingerStrap (Figure. 3.13a) and the EarSticker (Figure. 3.13e), the click wheel as a WheelSticker (Figure. 3.13d).
3.4.2.5 Interfacing and Data Processing

The flexible sensor patch is tethered with a ribbon cable to a Arduino-compatible microcontroller (Teensy 3.1), which is processing the data and driving the sensor. Signal measurements are time-division multiplexed with a frequency of 17 kHz. For interfacing, the sensor has a connector area, on which all pins are exposed. A custom-made rigid connector board is attached to these pins using z-axis conductive tape. Its other side is connected to wires leading to the micro-controller. An additional transparent adhesive tape on the top of the connector further stabilizes it and avoids lateral shifting of the connector board on the sensor. The connector area can be laid out anywhere on the sensor patch where no interactive area is located.

3.4.3 Application Examples

iSkin enables several classes of interaction and supports various application scenarios. We present prototypes of three novel on-body device classes. They support a wide variety of body locations demanding different sizes and shapes, different sensor designs and various degrees of flexibility and stretchability. They are organized in three groups, highlighting the flexibility in attachment of the sensor on the human skin: wrapping around body parts, attaching to on-body devices, and sticking onto the skin using bio-compatible adhesives.

**FingerStrap**

The FingerStrap (Figure. 3.13a) is a touch-sensitive film wrapped around the middle segment of the index finger to support microinteractions. Compared to ring-like devices, the strap increases the input space by covering a larger area without preventing movements. It features three buttons and a touch slider with 5 sensitive areas, all integrated in a tattoo-like visual design. It uses resistive sensing to avoid accidental
activation by requiring a certain limit of pressure. FingerStrap is especially useful when the hands are busy with a primary task, for instance while driving a car. It supports eyes-free input. Simple finger movements such as a slide of the thumb on the index finger can activate a command. It can also be used for casual interactions such as discreetly rejecting a call during a meeting or controlling a stopwatch during sports activities.

**Extension for Wearable Objects**

This prototype of a rollout keyboard can be attached to a smartwatch (Figure 3.13b). It enlarges the input space by letting the user interact on skin in the vicinity of the watch. The keyboard can be fully rolled in to be portable and can be pulled out on demand to overlay the skin of the forearm, as shown in Figure 3.13b. It provides a large input area for entering text using a full QWERTY keyboard with 30 keys. This highlights the possibility of sensing many interactive areas using a grid-like structure and time-division multiplexing.

**SkinStickers**

This class of interaction devices is useful for fast and direct selection of one or several frequent operations. While a SkinSticker can be attached virtually anywhere on the body, the forearm is suggested as a convenient location for quick and direct access [103, 109]. To attach the sensor patch onto skin, we use mastic. This is a medical-grade adhesive for use on skin. It is inexpensive (less than 0.40 USD), can be easily applied, and is fully compatible with use on skin. After use, the sensor can be easily peeled off without hurting the skin and without tearing out body hair. Previous work reported successful use of ADM Tronics Pros-Aide medical grade adhesive [113]. We show three SkinStickers for different functionalities:
MusicSticker  MusicSticker supports several functionalities in a visually aesthetic design, as shown in Figure. 3.13c. It contains five interactive areas for controlling a music player: Play, Previous, Next, Vol+ and Vol-.

ClickWheel  We have implemented a ClickWheel sticker (Figure. 3.13d). It captures circular rotation gestures. Moreover, touching and pressing on a segment differentiates between two commands.

EarSticker  Inspired by Earput [89], EarSticker (Figure 3.13e) can fully exploit the flexibility, stretchability and the affordances for input on the back of the ear and the earlobe. It supports input related to audio, such as adjusting the volume. A technical experiment evaluated stretchability and bendability of the sensor in a controlled setup.

3.4.4 Evaluation

A series of experiments have been conducted by Martin Weigel on the sensors that I produced to evaluate performance and reliability in a controlled setup.

3.4.4.1 Stretchability and Bendability

Methodology  Testing was performed with a rectangular sensor (8.5 cm × 4 cm) with an electrode diameter of 1.5 cm, conductive trace width of 1 mm and a thickness of 700 μm. This reflects the properties of the sensors in our application scenarios. The sensor was stretched by 0%, 10%, 20% and 30%. Moreover, it was bent around 3D printed cylinders of four radii: 0.5 cm, 1 cm, 2 cm, and 3 cm. These situations cover typical deformations when worn on the body. In each condition, 10 consecutive touch contacts were created using a circular shape of fingertip-sized diameter 8 mm in resistive sensing and with a human fingertip for capacitive sensing. For each
Figure 3.16: Results of the technical evaluation: (a) relative change in resistance, (b) measured voltage \( \hat{U} \) for capacitive touch contact and no touch, (c) required pressure for resistive touch contact.
Results  Figure 3.16 depicts the results for each condition. This includes changes in resistance of the circuit, the average peak-to-peak voltage readings for capacitive sensing of slight touch contact as well as the average normal pressure required for creating a contact using resistive sensing. First and foremost, the data shows that in all test conditions the sensor remains functional and sensitive to touch on both pressure levels. Secondly, as indicated by the dotted line in the capacitive sensing chart, touch vs. no touch can be classified without knowledge of how much the sensor is currently stretched or bent. Thirdly, the pressure required for resistive sensing of touch remains fairly stable. A smaller bend radius decreases the required force since bending reduces the distance between the electrodes.

While resistive sensing requires a firm touch, it can be a useful mode to support in addition to capacitive sensing, which reacts to very light touches. The required pressure can be tuned by changing the diameter of the hexagon pattern.

3.4.4.2 Reliability Across Body Locations

To investigate the accuracy and robustness of the touch sensor when worn on various body locations, a small-scale user study was conducted by Martin Weigel and the other collaborators at Max Planck Institute.

Methodology  6 voluntary participants (2f, 4m; median age 27y) were recruited for the study. The same sensor design as described above was used. The sensor was affixed on the participant’s body, directly on skin. My collaborators evaluated touch contact (capacitive sensing) and firm pressure touch (resistive sensing) on three body locations, reflecting the main locations identified in the design goals section: the fore-
arm, the back of the hand, and the index finger. The order of body locations was randomized. In each condition, the task consisted of repeatedly touching the sensitive area in 1.5s intervals, as accurately as possible. The participant was guided by an auditory metronome and received additional auditory feedback when the sensor was detecting touch contact. Participants were allowed to practice until they felt comfortable with the task. Touch events were logged on a computer with a PC oscilloscope (PicoScope 6402A). A total of 2160 touch inputs (360 per participant) were collected. Each session took approximately 15 min. The analysis identified correct touch events (exactly one touch recorded within the 1.5s interval), false negatives (no touch recorded), and false positives (more than one touch recorded within the interval).

Results The average accuracy was 92.0% (SD=8.6) for touch contact and 98.0% (SD=2.6) for firm pressure touch. For touch contact, it was highest on the finger (94.2%), followed by the forearm (91.1%) and the back of the hand (90.8%). For firm pressure touch, accuracy was also highest on the finger (98.9%), followed by the back of the hand (98.1%) and the forearm (96.7%). It is noteworthy that the task was more challenging than typical real-life scenarios due to the timing requirement, giving us a conservative estimate of accuracy. The lower accuracy of capacitive sensing compared to resistive sensing can be explained by the simple classification method we have used, which was merely based on measuring raw peak-to-peak voltage and did not make use of any signal conditioning. It seems quite safe to assume that a dedicated processing unit for capacitive touch sensing will lead to higher accuracies. We conclude that these results provide a lower bound, showing acceptable (90.8%) to very good (98.9%) results despite the proof-of-concept level processing of sensor data. This provides first evidence for suitability of the sensor for practical on-body input tasks.
3.4.5 Summary

The laser patterning technique that I developed has contributed the design of a novel class of “iSkin” skin-worn touch sensors. The underlying technical solution of iSkin builds on and extends recent advances in research on electronic skin. iSkin is a proof of concept of on-skin touch sensing that bears some promise over rigid sensors and computer vision based solutions. iSkin supports touch input on the skin. While depressing the skin for input does not offer the dynamic properties of physical buttons, it does retain some of the known benefits for tactile feedback and supports eyes-free input on the body through proprioception. Compared with the tactile sensors presented in the first half of this chapter, iSkin has no issues with surface contamination since all electrodes are sealed in polymer.

The technical solution allowing this is based on embedded carbon-doped electrodes and combined capacitive and resistive sensing. A technical evaluation showed that this solution supports bending around radii of 5 mm and stretching by 30% and is therefore well suited for on-body interaction. Due to its nature of being flexible and different wearing location, however, the limitation of iSkin is that it cannot measure pressure precisely.

The design of iSkin supports the customization of touch sensors to specific applications and body locations, desired visual appearance and button layouts. This has allowed us to create applications to very challenging body parts, like the index finger or the back of the ear. Moreover, while my collaborators and I are far from a generic controller for mobile devices, we can already cover some essential aspects like unidimensional selection tasks and text entry. We hope that this study will stimulate future research on electronic skin for mobile human-computer interaction, leading to multimodal input-and-output skins that improve interaction with mobile computing devices in a wide variety of tasks and activities.
3.5 Summary

Using a commercial 30 W CO\textsubscript{2} laser, I am able to rapidly produce elastically soft electronic circuits composed of patterned cPDMS, PEDOT:PSS, and liquid-phase EGaIn alloy embedded in a thin film of PDMS. These circuits contain features with 0.1-1 mm linewidths and function as elastically stretchable wiring and sensors for measuring stretch, surface pressure, and friction. In the case of EGaIn, we postulate that patterning is accomplished with a mechanism that is analogous to the liquid expulsion that occurs in high power (> 1 kW) laser beam welding (LBW). However, in contrast to LBW, recoil force does not come from the vaporized metal but is instead produced by vaporization of the underlying elastomer substrate. Moreover, because the liquid GaIn rapidly oxidizes in air, it forms a thin oxide skin that allows the patterned film to hold its shape after laser cutting and during sealing.

In Chapter 5, I will focus on further miniaturization with a commercial UV laser. Its smaller beam size (\sim 15 \mu m) and higher pulse rate (\sim 200 kHz) will allow for linewidths as small as 20 \mu m. Moreover, at the UV wavelengths (e.g., \lambda = 355 nm), EGaIn and other liquid metals can be directly patterned through photochemical ablation. This will eliminate the need to rely on the liquid expulsion mechanism presented in Figure. 3.6 and instead allows the liquid alloy to be patterned on any substrate, including metal. UV laser micromachining will also allow us to pattern thin films of metal-based cPDMS without burning. As with EGaIn, silver microparticles should vaporize through photochemical ablation when excited with \lambda = 355 nm radiation. Ultimately, I hope to have a universal protocol for using commercial laser micromachining to rapidly produce soft-matter electronics and hence eliminate the need for customized hardware or cleanroom fabrication.
Chapter 4

Soft Anisotropic Conductors as Electric Vias for Ga-based Liquid Metal Circuits

Although promising for applications in wearable computing and soft microfluidics, further progress in Ga-based LM electronics depends on robust methods for sealing.

Figure 4.1: (a) PDMS embedded with an array of vertically-aligned columns of Ag-Ni microparticles that support conductivity only through the thickness of the film, i.e. along its z-axis; (b) Ag-Ni columns function as electrical vias between embedded EGaIn circuit terminals and surface mounted electronics. (c) EGaIn traces attached to LEDs via a commercial z-axis conductive film (ECATT 9703; 3M). PDMS/Ag-Ni z-axis film (“z-film”) functions as a transparent seal and electrical interface for tactile sensing in an (d) arrowpad and (e) 5×6 node touchpad.
and electrical interfacing. Sealing is essential to prevent the liquid from leaking, staining skin and clothing, or reacting with other metals through corrosion (e.g. Al) or alloying (e.g. Cu). At present, the terminals of LM circuits are typically accessed by manually inserting thin metal wires. While reliable for prototyping and proof-of-concept demonstrations, these connections are not mechanically robust and introduce fluidic-rubbery-rigid boundaries that are susceptible to leaking, changes in electrical contact resistance, or even wire pull-out. Increasing the rigidity of the elastomer near the terminal can reduce the interfacial stress concentration between the polymer and solid wiring to prevent tearing and leaking[114]. However, this method is not applicable to tactile sensing or stretchable circuits that rely on electrical contact with skin or preservation of elastic compliance. Another approach is to seal the terminals with conductive elastomers such as polydimethylsiloxane (PDMS) filled with carbon (cPDMS) and Ag powder (AgPDMS)[16]. When embedded above the terminal points, they function as electrical vias for direct electrical contact with skin or external electronics without altering the integrity or elasticity of the surrounding microfluidic circuit. However, conductive elastomers typically exhibit isotropic conductivity and therefore need to be patterned and aligned in order to prevent shorting between adjacent circuit terminals. This can be performed with a CO$_2$ laser engraver, which allows for simultaneous patterning of liquid metal and both conductive and insulating elastomer[16]. However, other fabrication methods such as stencil lithography, microcontact printing[54] and additive fabrication[41] require separate patterning and alignment of the liquid metal circuit and the conductive elastomer vias. While possible, the additional alignment step requires specialized hardware and remains to be demonstrated.

In this chapter, I introduce a robust and versatile alternative for LM interfacing using an elastomer film that is only conductive through its thickness (z-axis)
An anisotropically conductive “z-film” simultaneously prevents leakage while creating electrical vias for accessing LM without the need for alignment. It is composed of polydimethylsiloxane (PDMS) embedded with vertically-aligned columns of conductive Ag-coated Ni microparticles. As shown in Figure. 4.1a, the ferromagnetic particles are magnetically aligned to form percolating networks that support electrical conductivity only through the thickness. Z-axis anisotropic conductors were first reported in 1988 by researchers at AT&T[115] and have since been used for touch-sensitive screens[116], tactile shear sensing[117], solder replacement for surface mount technology (SMT)[118], and electronics packaging[119, 120]. In this study, we extend the use of z-axis conductors to LM circuits and demonstrate compatibility for illustrative examples presented in the Figure. 4.1 overview. These include surface mounted electronics (Figure. 4.1b and 4.1c), tactile sensing (Figure. 4.1d and 4.1e), and joint proprioception, which are described in greater detail in Sec. 4.2. In all cases, stretchable functionality is preserved by the elasticity and compliance of the soft z-film composite.

4.1 Experimental Methodology

4.1.1 Synthesis of Z-axis Anisotropic Conductive Material

Z-axis anisotropic conductors are produced by first mixing PDMS (Sylgard 184, 10:1 base-to-catalyst ratio; Dow Corning) with 15 μm diameter Ag-coated Ni microparticles (69.5%Ni-30.5%Ag, by wt; SN15P30, Potters Inc.). Next, the uncured polymer/particle mixture is deposited on a substrate by spin coating (Model KW-4A; SPI Supplies) or with a thin-film applicator (ZUA 2000 Universal Applicator; Zehntner GmbH). Prior to deposition of the mixture, the substrate may be covered with layers of conductive paper (CN-3490; 3M) and insulating PDMS that will bond to the z-film.
after curing. For mixtures with 40% Ag-Ni powder (by wt), spin coating at 600, 1200, and 1800 rpm for 10s results in the following film thicknesses: 173, 107, and 69 μm, respectively. The sample is then placed on top of a flat magnet (2” × 2” × 1/4” NdFeB; K&J Magnetics, Inc.) and cured in an oven at 100°C for 25 minutes. The magnetic field aligns the particles as the elastomer cures, as has been previously discussed in the literature[121, 122]. During or prior to curing, surface mounted electronics may be deposited on the z-film.

### 4.1.2 Fabrication of the Tactile Sensor

A tactile sensor is produced using the laser patterning techniques previously introduced in Ref. [16] and summarized in Figure. 4.2. First, the cutting substrate is prepared by covering one edge with CN-3490. Next, z-film is deposited and cured using the steps described in the previous paragraph. EGaIn is then deposited on the cured z-film using a roller (e.g. a PDMS cylinder or lint roller) followed by a layer of insulating PDMS applied with spin coating or a thin-film applicator. After the PDMS seal is partially cured on a hotplate or in a curing oven (at 100°C for 5 minutes), the sample is patterned with a CO2 laser (VLS 3.50; Universal Laser Systems), as illustrated in Figure. 4.2a. After the excess material is removed (Figure. 4.2b), a second layer of insulating PDMS is deposited and cured Figure. 4.2c). Lastly, the patterned CN-3490 traces are clipped into a ribbon cable adapter for external wiring (Figure. 4.2d).

### 4.1.3 Measurement of Anisotropic Conductivity

Anisotropic electrical conductivity is measured by inserting the probes of a multimeter (34401A; Agilent Technologies) into EGaIn droplets deposited above masked portions of the z-film (see Figure. 4.3). Beneath the PDMS-based z-film, is a strip
Figure 4.2: Fabrication of tactile sensor. (a) A CO\textsubscript{2} laser (VLS 3.50; Universal Laser Systems) is used to substrate covered with (from bottom) conductive paper (CN-3490; only along edge of sample as represented by the dark gray), z-film, a 10 \( \mu \text{m} \) layer of EGaIn, and a film of insulating PDMS. (b) Excess material is removed and the sample is (c) coated with an additional layer of PDMS. (d) A ribbon cable adapter is clipped into the conductive paper traces to connect the sensor with an external microcontroller (Arduino Uno).

Figure 4.3: (a) Layup for testing electrical resistance of z-film. Image of measurement setup with z-film on top of (b) CN-3490 (3M) conductive paper or (c) copper traces.
of conductive paper (CN-3490; 3M) patterned with a CO$_2$ laser (VLS 3.50; Universal Laser Systems) or copper traces that are cut with a UV laser micromachining system (Protolaser U3; LPKF). The conductive traces are insulated with a mask (Scotch Tape, 3M) such that only square terminals (and the associated z-film) are exposed for EGaIn contact. The resistance of the z-film is calculated by subtracting the resistance of the test strips with and without the z-film covering and dividing by two. The values presented in Figure 4.7 are only estimates since they include both the volumetric resistivity of the bulk as well as the contact resistance between the z-film and EGaIn.

We also tested the resistance of z-film with strain and cyclic loading. The experimental setup is shown in Figure 4.4. The test sample was fixed on the linear actuators for stretching (Figure 4.4a). Except for a 9 mm$^2$ opening where conductivity was measured, the film was coated with a very thin layer of PDMS ($\sim$10 $\mu$m).
The sample was then placed on top of a reservoir containing EGaIn with the tested region in complete contact with EGaIn. A drop of EGaIn was also placed on top of the test region. Wires were then inserted into the EGaIn to measure electrical resistance through the film thickness (Figure. 4.4b). The resistance of the test region was isolated by subtracting the resistance measured by inserting the metal wires into a single pool of EGaIn.

4.2 Results & Discussion

The z-axis conductive films are composed of PDMS (Sylgard 184; 10:1 base-catalyst ratio, by wt.; Dow Corning) embedded with vertically-aligned columns of \( D = 15 \mu m \) diameter Ag-coated Ni microparticles (SN15P30: 69.5\%Ni-30.5\%Ag, by wt; Potters Inc.). Optical images (InfiniteFocus, Alicona, Inc.) of the samples are presented in Figure. 4.5 for samples with 40\% (top and side views) and 20\% (side view only) by weight of Ag-Ni filler. The sample with 40\% \( (f = 0.4) \) concentration has a thickness of \( t = 256 \mu m \) and an estimated column-column spacing in the range of \( x = 50 \) to 75 \( \mu m \). Noting that Ag and Ni have \( \chi = 10 \) times the density of PDMS, the microparticle volume fraction is \( v = 1/[1 + \chi(f^{-1} - 1)] = 0.0625 \). Assuming hexagonal spacing, each column will contain between \( N \sim 20 \) and 44 microparticles, where \( N = 3\sqrt{3}tvx^2/\pi D^3 \). This corresponds to staggered columns that are approximately 2-4 particles wide, which is consistent with what is observed in the side view. Since the columns are seperated by polymer, no in-plane conductivity is measured. Instead, the capacitance of two co-planar capacitors sealed with z-film were measured to be 1.85 and 1.29 pF with Q factors of 24.7 and 11.4, respectively, indicating that the electrodes were well insulated.

An SEM image of the top of a column (Figure. 4.6a) shows protrusion through the surface and an EDS (energy-dispersive x-ray spectroscopy) scan (Figure. 4.6b,c)
Figure 4.5: Top down and side views of PDMS film embedded with magnetically-aligned columns of 15 μm diameter Ag-Ni microparticles (20 and 40%, by weight).

Figure 4.6: (a) An environmental SEM image of a particle at the surface of the z-film. (b,c) An environmental SEM image of the particles at the surface of z-film and the corresponding EDS image indicating the distribution of the elements.

indicates the abundance of Ag and Ni. Together, these results provide evidence for the presence of Ag-Ni microparticles for forming direct electrical contacts at the surface.
4.2.1 Conductivity

Figure 4.7a presents conductivity measurements for z-film composites with a $A = 9$ mm$^2$ area (3mm×3mm) and $t = 90$ μm thickness. As expected, the z-axis resistance $R$ and volumetric resistivity ($\rho = AR/L$) decrease with increasing mass fraction $f$. Likewise, for a sample with fixed mass fraction, resistance decreases with increasing area and decreasing thickness. This is demonstrated in Figure 4.7b for $f = 0.4$ and samples with dimensions of 1×1, 2×2, and 3mm×3mm. In the case of a 3mm×3mm sample with a thickness of 69 μm, the electrical resistance is 0.2 Ω, comparable to typical values for cm-scale wiring and contacts. Even for samples with a lower concentration of 20 wt%, the electrical resistance is low enough (0.6 Ω for $A = 9$ mm$^2$ and $t = 90$ μm) to support basic circuit and sensing functionality. The conductivity of z-film with strain and cyclic loading is also measured with the method mentioned in last section. The z-film tested had a Ag-Ni particle content of 20 wt% and thickness of approximately 180 μm. Five samples were tested. The resistance of each sample as it is stretched to 50% strain is shown in Figure 4.8. The resistance of the samples were also measured before and after being stretched to 50% strain for 100 cycles. Before loading, the five samples had an average resistance of 1.54 Ω with a standard deviation of 0.24 Ω. After the cyclical loading, their average resistance decreased to 1.44 Ω with a standard deviation of 0.21 Ω. Although the average was observed to decrease, it is within the sample error and so an electromechanical dependency is not established.

The reason for this is that conductivity is supported by the Ag/Ni-columns and their size and number remain invariant to the deformation of the surrounding elastomer.

Connecting LM circuits to external electronics through ribbon cables is challenging since they typically require mechanical adapters that clip on to the circuit terminals. For liquid-phase soft electronics, the clip will exert stress concentrations that may puncture the seal or otherwise damage the circuit. To avoid this, conductive paper
Figure 4.7: (a) Z-axis resistance of composites for $f = 0.1$, 0.2, and 0.4; $A = 9$ mm$^2$ (3mm×3mm), $t = 90$ μm thickness. (b) Results for z-film with 40% filler concentration; varying $A$ and $t$. 
Figure 4.8: The resistance of Z-film when stretched to 50% strain.
is used to connect the terminal ends of the liquid circuit to the cable adapter, as described in Figure. 4.2. The conductive paper is 60 μm thick and composed of Ni/Cu-coated non-woven polyester fiber (CN-3490; 3M). As with the other materials, it can be patterned with a CO₂ laser and readily bonds to both uncured PDMS and pressure sensitive polyacrylate adhesives (e.g. F-9473PC, VHB 4905, and ECATT 9703).

### 4.2.2 Mechanical Durability

The mechanical properties of z-film specimens (f = 0.2 and 0.4, three samples for each) and pure PDMS (f = 0, three samples) are tested using a materials testing system (5969 Instron). The PDMS and zPDMS films are made with the same spin coating parameters, 600 rpm for 10s, cured at 100 °C for 2 hours, and cut with CO₂ laser systems into straight specimens with planar dimensions of 45 × 5 mm (length × width). The dimensions are limited by the size of magnets used. The thickness of specimens with f = 0, 0.2, 0.4 are 0.175, 0.190, 0.170 mm, respectively. The length of the between-grips section of the specimens is 20 mm. Failures were observed to happen at different locations of the specimens, but mainly near the grips. Typical strain-stress curves of samples with f = 0.2 and 0.4 are shown in Figure. 4.9. Samples with 20 wt% content were stretched 3× to strains of 30%, 60%, and 90% strains in sequence. Samples with 40 wt% content were loaded 3× to strains of 20%, 40%, and 60%. After the nine loading cycles, each sample was stretched to mechanical failure. The average 10%-strain elastic moduli are 1.974 MPa (SD=0.109 MPa) and 2.596 MPa (SD=0.172 MPa) for f = 0.2 and 0.4, respectively, which are higher than that of unfilled PDMS cured at 100°C for 2 hours (1.758 MPa), as shown in Figure. 4.10a. For all samples, the maximum failure strain was ~94% (SD=5.1%) and ~80% (SD=1.7%) for f = 0.2 and 0.4, respectively, as shown in Figure. 4.10b.
Figure 4.9: Typical strain-stress curves of samples with $f = 0.2$ and 0.4 particle concentration.
Figure 4.10: The Young’s moduli (a) and failure strain (b) of samples with $f = 0$, 0.2, and 0.4 particle concentration.
To assess the durability of the electrical interface, circuits with surface mounted LED chips were fabricated and subject to tensile loading. Two samples were tested, both with a 130-μm-thick PDMS substrate and a 230-μm-thick z-film (f = 0.4) layer embedded with EGaIn traces. LED chips were then mounted above the z-film. One of the samples was coated with an extra encapsulating layer (60 μm) of PDMS to seal the LED contacts. The samples were then fixed on a vertical tensile tester and powered with 2.4V as they were stretched (Figure. 4.11). In the absence of the extra layer of PDMS, the LED chip remained fully functional until delamination occurred at approximately 30% strain. As delamination developed, the contact between LED and EGaIn was weakened and the circuit completely failed at about 67% strain. For the sample with the PDMS seal, although delamination was observed at a strain of approximately 60%, the circuit remained functional even after elastomer tore below the LED at a maximum strain of approximately 110%.

Figure 4.11: Tensile test of samples with a surface mounted LED.
Figure 4.12: (a) Wearable touchpad with z-film seal to allow for direct electrical contact with skin. (b) Schematic of voltage divider architecture for tactile sensing. (c) Voltage output on each electrode for user test with 5V input and 10MΩ resistor.
4.2.3 LM-based Tactile Sensor

Referring to Figures. 4.1d and 4.12a, a soft-matter tactile sensors is produced by sealing a laser-patterned film of EGaIn between layers of insulating PDMS and PDMS-based z-film \((f = 0.2)\). An image showing the quality of the line cut with a \(\text{CO}_2\) laser (VLS 3.50; Universal Laser Systems) is shown in Figure. 4.13. The peak-valley difference of the edge is approximately 40 \(\mu\text{m}\), which is 50 times less than the line width of the laser-patterned traces. The combination of low elastic modulus and <400 \(\mu\text{m}\) thickness allows the tactile sensor to conform to the skin without introducing mechanical constraints or causing discomfort. The sensor contains five leads that are connected to an external microcontroller (Arduino Uno) for recording fingertip-activated changes in voltage drop between the sensor electrodes. The leads interface through a ribbon cable and adapter. The sensor is composed of a central electrode that is connected to a 5V input and four outer electrodes that are electrically grounded through a \(R = 10\ \text{M}\Omega\) resistor (Figure. 4.12b). The 470 pF capacitor in the diagram is used to filter out high frequency noise. When the fingertip makes simultaneous contact with the central and outer electrode, the outer electrode charges and the voltage rapidly increases above 0V. Fingertip contact can also be detected by monitoring the electrical conductivity between the central and outer electrodes (Figure. 4.12b).

In the absence of fingertip contact, there is no measurable conductivity between the electrodes. When a finger is lightly pressed against a sensing node, a \(\sim1\ \text{M}\Omega\) resistance is measured. This resistance is dominated by the resistivity of the skin and the contact resistance between the skin and the z-film. Measurements performed with the tactile sensor are presented in Figure. 4.12c. As shown by the plots, the voltage output on each electrode rises when the electrode is connected to the input trace via the fingertip. With different placements of the finger, the electrodes can be activated either individually or simultaneously.
Figure 4.13: Microscopic image of the edge of laser-cut z-film coated EGaIn. Scale bar: 300 μm.
An example of a multi-layer tactile sensor is presented in Figure. 4.1e and a similar sensor is demonstrated in Figure. 4.14. In this implementation, rows and columns of EGaIn electrodes are embedded inside a z-film matrix (f = 0.4). There is a thin strip of insulating PDMS at the bottom of each electrode in the top layer that prevents direct electrical shorting. Each column and row pair becomes electrically connected to a voltage input by touching the surface with fingertip and forming electrical vias between the electrodes and the voltage input. As with the other sensor, the multi-layer sensor shows satisfactory voltage change in each channel as a response to the interaction with human skin.

Tactile sensing is also implemented with commercial acrylate elastomers. As shown in Figure. 4.15a, the EGaIn circuit is embedded within layers of adhesive acrylate elastomer (F-9473PC and 4905 VHB; 3M) and z-axis electrically conductive adhesive transfer tape (ECATT 9703; 3M). The arrowpad is used to operate a Tetris gaming script in Python (Figure. 4.15b,c). Referring to Figure. 4.16, the circuit is produced through a combination of CO$_2$ laser patterning and stencil lithography. In contrast to PDMS-based EGaIn circuits, fabrication with commercial acrylates eliminates the need for time consuming and labor intensive steps with elastomer preparation and curing.

4.2.4 Wrist Motion Sensor: Interfacing LM Circuit with External Wiring

Another example of robust electrical interfacing with a soft LM circuit element is the joint motion sensor presented in Figure. 4.17a. The sensor is composed of two overlapping films of EGaIn that function as capacitive electrodes insulated by a thin layer of PDMS. When placed on a wrist, the joint motion causes the electrodes and dielectric to deform and change capacitance. As in the tactile sensor, z-film and
Figure 4.14: (a) A multi-layer tactile sensor with EGaIn embedded as electrodes. (b) Schematic of voltage divider architecture for tactile sensing. (c) The voltage output for user test with 5V and 10 MΩ resistors. The sensor is tested with fingertip touching on top of the four nodes (intersections of electrodes). When a node is touched by a fingertip, voltage outputs from the corresponding upper and lower electrodes change.
Figure 4.15: (a) EGaIn circuit embedded within layers of adhesive acrylate elastomer (F-9473PC and 4905 VHB; 3M) and z-axis electrically conductive adhesive transfer tape (ECATT 9703; 3M). (b,c) The arrowpad is used to operate a game of Tetris coded in Python; the same sensing architecture is used as in Figure. 4.1d.
Figure 4.16: Rapid fabrication of acrylate-based tactile sensor with CO$_2$ laser engraving: (i) prepare a 4905 VHB film (0.5 mm thick) with a non-stick paper backing; (ii) pattern the backing with CO$_2$ laser; (iii) remove excess paper; (iv) cover with F-9473PC VHB film (0.254 mm) (including one side with backing); (v) laser cut F-9473PC film with the same pattern as the paper traces underneath; (vi) remove adhesive film and underlying patterned paper; (vii) apply a thin film of EGaIn to fill in the open channels of exposed acrylate adhesive; (viii) remove top lining of F-9473PC film; (ix) cover with ECATT 9703 z-film. Because this process does not require elastomer curing, prototypes can be rapidly produced in $\sim$5-20 minutes.
Figure 4.17: (a) Capacitive strain sensor for measuring joint motion. EGaIn electrodes embedded in PDMS are connected to a ribbon cable via z-film and CN-3490 conductive paper. Plots show sensor output for bending on a (b) mechanical joint and (c) wrist.
Conductive paper are used to connect the LM circuit element to off-board electronic hardware through a ribbon cable. An LCR meter is used to measure the relative change in capacitance $\Delta C/C_0$ in response to changes in bending angle $\theta$.

When undeformed, the circuit has a capacitance of $C_0 \sim 7 \text{ pF}$. When placed on a mechanical joint (Figure 4.17b) the capacitance changes linearly with $\theta$, with an $R^2$ value of 0.99 and bending-mode gauge factor $GF = \{\Delta C/C_0\}/\theta(\text{rad})$ of 0.077. When placed above the wrist (Figure. 4.17c), the response is nonlinear: $\{\Delta C/C_0\} = \alpha(\theta \pi/180)^2 \{\text{sgn}(\theta)\}$, where $\alpha = 9.57 \times 10^{-2}$ and $R^2 = 0.95$. Although the internal contact resistance does not factor into the capacitive measurements, the reliability of these results (smooth, monotonic, low hysteresis) demonstrates the ability of the interface to support bend sensing applications without electrical failure.

### 4.3 Summary

Z-axis anisotropic conductors (“z-film”) are a versatile material for electrically interfacing Ga-based liquid metal (LM) circuits with human skin and external electronics. Previous studies of z-film have focused on solid-state electronics, packaging, and other applications that take advantage of their optical transparency and bonding properties. Here, we show that z-film elastomers are also well suited as electric vias for LM-based circuits since they seal in liquid without introducing elastic rigidity or electrical shorts within the plane of the circuit. This work improves upon previous efforts with EGaIn-based microfluidics by eliminating the need for manual wire insertion. Wire insertion is not only labor intensive and prone to error, but also introduces mechanical mismatches, stress concentrations, and non-bonded interfaces that can result in elastomer rupture, wire slip/pull-out, and LM leakage. Alternative interfacing methods based on isotropically conductive elastomers[16] and rigidity graded materials[114] address these issues but are challenging to implement due to their de-
pendency on precise alignment and/or specialized materials and fabrication methods. In contrast, a single layer of z-film can be applied to the entire surface of the LM circuit without any additional alignment step.

Since the use of z-film in conventional electronics is already well-established, the purpose of this work is to demonstrate their reliability for forming interfaces with LM alloys. We have not investigated the principles of ferromagnetic microparticle alignment as this has already been addressed in the literature.\[121, 123\] In summary, the z-axis column formation is controlled by a magneto-rheological response to external magnetic field. Once the surrounding elastomer has cured, the particles are constrained to their relative configuration and the field can be removed. The length and spacing of the columns is controlled by the composition (i.e. wt % of microparticles) and field intensity. For example, as previously shown, we find that the length of the columns of ferromagnetic microparticles scales with the intensity of the magnetic field. If the field is too weak, the column length will be less than the film thickness and z-axis conductivity will not be achieved. If the field is too strong, the microparticles will break the surface tension of the uncured elastomer and the column length will exceed the thickness of the film and form dendrite-like structures.

While successfully implemented with the help of z-axis conductive film, the tactile sensing devices demonstrated in this work still need to be further studied to clarify their tolerances and resistance against environmental factors such as humidity, sweat, and oil, etc. User-based studies, by which devices are tested by human participants, should be performed in the future to assess input accuracy for prescribed data entry tasks\[124\]. Lastly, as with other LM circuits, we observe mechanical and electrical failures when a sample is stretched to beyond the strain limit of the elastomer sealing. In the case of surface mounted electronics, poor bonding between the rigid circuit and z-axis conductor will lead to delamination and eventual mechanical failure. This is
caused by stress concentrations that initiate at the edges of the interface due to elastic mismatch between the soft elastomer and rigid electronic component. As external loading increases, a crack forms and propagates towards the center of the interface. Further studies are required to characterize the bond strength between z-axis adhesives and rigid or semi-rigid circuit elements. Nonetheless, the current study shows that under tensile strains of up to 50%, the bonding is adequate for supporting electronic functionality.
Chapter 5

Rapid Prototyping of Soft-Matter Electronics with UV Laser Patterned Liquid Metal

CO₂ laser patterning, as introduced in previous chapter, has shown it can be used for rapid prototyping of both conductive polymer and liquid metal. This method, however, has two major disadvantages. Firstly, the CO₂ laser beam cannot be used to patterned LM traces if width less than 250 μm. This is due to the indirect method of patterning as well as the fundamental coupling between laser wavelength, focal diameter and focal depth. For a Gaussian beam (as shown in Figure. 5.1), the depth of focus $F=\frac{2\pi w_0^2}{\lambda}$, where $w_0$ is the waist size and $\lambda$ is the wavelength of laser. The waist diameter of CO₂ laser has to be relatively large for a practical focal depth (100 μm focal diameter for a focal depth of 1.47 mm ), which results in a relatively low patterning resolution. Secondly, when patterning liquid metal with CO₂ laser, a layer of polymer sacrificial substrate is needed, which limits the application of this technique.
Figure 5.1: The schematic of a Gaussian beam. Here, $F$ is the depth of focus and $w_0$ is the waist diameter. The depth of focus $F$ of a Gaussian beam is 2 times the Rayleigh distance (the distance between the waist and the point where beam diameter is $\sqrt{2}$ times the waist diameter).
UV laser ablation is another patterning technique for laser-based rapid prototyping of LM-based stretchable electronics. With a UV laser micromachining system (UVLM), I have successfully fabricated liquid metal traces of varying shape and dimensions, as shown in Figure 5.2. The traces shown in Figure 5.2a,b show that the EGaIn film on a zPDMS can be patterned into features with dimensions as small as 50 $\mu$m. Figure 5.2c shows the scalability of UV laser patterning. The widths of the traces are 500, 200, 100, and 50 $\mu$m, respectively. The diameters of the circles are 1000, 500, 200, and 100 $\mu$m, respectively. Figure 5.3 shows a simple application of UV laser-patterned EGaIn. This application is a circuit consisting of an LED chip, liquid metal, and polymer (PDMS and zPDMS) and shows significant flexibility.
Figure 5.3: A circuit composed of UV laser-patterned EGaIn trace, LED chip, zPDMS, and PDMS.
Figure 5.4: (a) The result of parametric patterning quality testing. Patterning is performed on a polyurethane substrate for better contrast. The substrate is coated with a layer of EGaIn of thickness of approximately 15 μm. The EGaIn is then pattern with a UV laser micromachining system to produce cross-like features. The size of each square is 5 by 5 mm. Five speeds (200, 400, 600, 800, 1000 mm/s) and five powers (0.3, 0.6, 0.9, 1.2, 1.5 W) are tested. The same feature and parameters are tested on PDMS and zPDMS. (b) A typical sample with EGaIn not completely ablated and (c) a sample with EGaIn completely ablated. Scale bars: 2 mm

5.1 Principles

Liquid metal circuits are produced by depositing thin films of EGaIn on substrates with a roller made of PDMS, and selectively ablating the liquid metal with a 355 nm wavelength Nd:YAG UVLM (Protolaser U3; LPKF). Unlike that of IR light, the UV reflecttance of metals are much lower (0.9-0.99(IR)/0.4-0.95(UV))[125], which means metal can be ablated by UV laser at a lower power. The photon energy of UV laser is firstly absorbed by the free electrons and then transferred to phonons by collisions[125]. During ablation, exposure to air causes the EGaIn traces to form a Ga2O3 skin that allows the patterned liquid to stay in place and maintain the circuit shape. The laser has a 200 kHz pulse rate, 15 μm waist diameter (i.e. spot
Figure 5.5: (a) The result of parametric patterning quality testing on PDMS. (b) A typical sample with EGaIn not completely ablated and (c) a sample with EGaIn completely ablated. Scale bars: 2 mm
Figure 5.6: (a) The result of parametric patterning quality testing on zPDMS. (b) The result of patterning quality testing on zPDMS with powers of 1 and 1.1 W and speeds of 100, 150, 200 mm/s. (c) A typical sample with EGaIn not completely ablated and (d) a sample with EGaIn completely ablated. Scale bars: 2 mm
size), and an adjustable power that ranges from 0.3 to 6 W. Samples are patterned with a variety of beam parameters on different substrates (PU, PDMS, zPDMS) to show the influence of powers (0.3-1.5 W) and marker speeds (200-1000 mm/s) on patterning quality, as reported in Figure. 5.4, 5.5, 5.6. These results are just for a rough understanding of the effect of power and markers speed on the patterning quality of GaIn. In actual patterning process, patterning quality is observed to be influenced by of factors such as the overall size of patterned area, the minimum feature size, etc. 1 W and 150 mm/s are selected for all the patterning presented in this chapter because of their effectiveness validated by LM circuits of different scales and designs. Compared with CO$_2$ patterning, which has a bigger spot size (25, 127 $\mu$m), UV laser patterning is capable of patterning much smaller features with higher resolution as shown above.

5.2 Results and Discussion

Most of the EGaIn patterning presented in this chapter is done on zPDMS substrate. zPDMS is used as the substrate material because a fabrication technique is proposed by me as the potential method for producing soft circuits with surface-mounted chips (shown in Figure. 5.7) and zPDMS plays a critical role in this process. Circuits made with this method can be found in Chapter 6.

The zPDMS used in this chapter is modified based on the zPDMS presented in Chapter 4. The magnetic particles used are silver-coated iron oxide particles (~40 $\mu$m, Potters, Inc.), and EGaIn is added to increase the adhesion between particles so the conductivity between them would not easily decrease because of the particle separation that may occur when zPDMS is deformed. This mechanism is shown in
Figure 5.7: The proposed fabrication technique for soft circuits with surface-mounted chips. (i) A layer of PDMS is applied on a substrate and covered with a colored tape. The PDMS film and tape is then patterned with laser, to create features with the sizes of corresponding chips and fiducials for the laser to align (black dots). (ii) The chips are coated with a layer of PVA solution, placed into the features, and cured. (iii) The chips are then sealed in PDMS. After curing, the sample is peeled from the substrate, along with the chips embedded inside. (iv) The sample is flipped over with the chips exposed on one side. The chips are then rinsed with deionized water to remove PVA, and dried. Subsequently, a layer of zPDMS is applied on top of the chips, and cured on magnet. (v) A EGaIn film is then deposited on the zPDMS film, and then (vi) patterned with the UV laser to create circuits. The alignment of laser is assisted by the fiducials made in step (i). The circuits are then sealed in PDMS.
Figure 5.8: The mechanism of increasing adhesion between magnetic particles. Magnetic particles without EGaIn coating (top) have weak adhesion with each other and this can potentially be improved by adding EGaIn as a coating of the particles (bottom)
Figure 5.9: (a) The 50-μm-wide EGaIn traces with defects (in circles). Scale bar: 0.5 mm. (b) A piece of typical unablated impurity in EGaIn that causes shorting. Scale bar: 250 μm.
Figure 5.8. The magnetic particles are first mixed with EGaIn with a mortar for 5 min and PDMS is subsequently added. The mixing ratio is 35 wt% magnetic particles, 15 wt% EGaIn, and 50 wt% PDMS. The curing condition is the same as the zPDMS presented in Chapter 4. EGaIn is deposited onto zPDMS substrate by using a PDMS roller as shown in Chapter 3.

On the zPDMS substrate, UV laser-patterned EGaIn traces with widths as small as 50 \( \mu m \) have been successfully patterned and exhibit stable electrical conductivity. When narrower than 50 \( \mu m \), however, the traces can be nonconductive because of the defects of EGaIn deposition (Figure. 5.9a). The statistical likelihood of being nonconductive increases with the length of trace. The minimum spacing between traces is 100 \( \mu m \), which enables the traces to be effectively insulated. With spacing smaller than that, the traces can be shorted due to the presence of EGaIn residue that contains impurities which prevent laser ablation as shown in Figure. 5.9b.

As mentioned above, the minimum trace size and spacing are limited by the defects in EGaIn. By using EGaIn with less impurity such as Ga\(_2\)O\(_3\) and better deposition quality, the patterning resolution can be further improved. The EGaIn spraying technique introduced by Jeong et al ([126]) has made a significantly improvement in EGaIn deposition by providing an LM film that is smoother and contains less impurity by spraying (with compressed nitrogen as a carrier) atomized eutectic Gallium based liquid alloy onto half cured PDMS substrate.

The thickness of the patterned EGaIn traces is measured by using a surface profilometer (VHX 5000, Keyence, Inc.). Since the surface of zPDMS is rough, EGaIn cannot form a uniformly distributed film on top. The surface profilometry results from multiple points show that the thickness of EGaIn varies from 10 to 20 \( \mu m \). For better accuracy, the measurements are taken at the points with no obvious surface extrusions due to the magnetic conductive particles. During laser patterning, a por-
tion of substrate material beneath the laser patterning area is ablated along with the liquid metal. The laser-patterned surface is examined with surface profilometry and the measurement results show that the thickness of zPDMS substrate removed during laser patterning is approximately 15 μm.

5.2.1 EDS and SEM Test

To show the effectiveness of liquid metal removal by UV laser patterning, zPDMS samples with and without laser patterned traces are examined with SEM and Energy Dispersive Spectroscopy (EDS). Figure 5.10 shows the SEM images of the zPDMS surfaces used for EDS testing. Figure 5.10a shows the zPDMS surface that is neither EGaIn-coated nor laser ablated. It is clear that magnetic particles are distributed uniformly in zPDMS. Figure 5.10b is the surface of EGaIn-coated zPDMS after laser ablation. The zPDMS surface ablated by laser obviously contains more surface features and texturing than unablated zPDMS.

EDS is a technique used to analyze the elemental composition of material. It works by using an electron beam to excite the electrons in inner electron shells of atoms. The electrons are then released from inner electron shells and leave electron holes behind. The electrons on outer shell fall into the holes and the energy difference between electron shells are released in form of X-rays. The characteristics of X-rays vary with elements. By analyzing the spectrum of x-rays, the elements in a sample can be determined qualitatively and quantitatively. EDS can be used to scan an area on a sample and analyze the elemental composition on its surface. Figure 5.11 shows the EDS results of surface scanning on zPDMS. It can be seen that elements are marked with different colors, and the X-ray spectrum shows the characteristics of different elements. EDS results include colormaps that show the distribution of different elements on the surface of sample. The colormaps of four different elements
(Ga, In, Ag, and Fe) on zPDMS and EGaIn-coated/laser-ablated zPDMS surfaces are shown in Figure. 5.12 and Figure. 5.13, respectively. The colormaps show that for zPDMS, the elements are highly concentrated on the magnetic particles. After EGaIn coating and laser ablation, however, the elements are much more distributed. The test results also show an increase in the content of Ga (from 2.21 wt% to 5.48 wt%) and In (from 0.51 wt% to 1.27 wt%) on the surface of samples where EGaIn was laser ablated compared with the samples of zPDMS which are neither coated nor ablated. This indicates that the EGaIn on surface is not completely removed.

SEM and EDS tests with higher magnification are also done to examine if the elemental contents converge on the surfaces of different zPDMS samples. The elemental colormaps are shown in Figure. 5.14 and Figure. 5.15 along with the contents of Ga and In of different samples. The test results show that the content of Ga and In increase from 3.08 wt% and 1.06 wt% to 5.99 wt% and 1.42 wt%, respectively. These results are roughly consistent with the measurement results from lower magnification. The zoomed-in SEM image and EDS colormaps of zPDMS (Figure. 5.14) confirm that most of the EGaIn is coated on the magnetic microparticles. The zoomed-in SEM image of EGaIn-coated/laser-ablated zPDMS (Figure. 5.15a) show that there are liquid metal droplets sized smaller than 5 μm that remain after UV laser patterning. This observation is consistent with elemental colormaps of Ga and In obtained by EDS (Figure. 5.15b,c). These particles are scattered on the laser-patterned surface and there is no conductivity between them.
Figure 5.10: The SEM image of (a) zPDMS and (b) EGaIn-coated/laser-ablated zPDMS. Scale bars: 500 μm

Figure 5.11: Overall elemental colormap of zPDMS obtained by EDS (left) and corresponding X-ray spectrum(right). Scale bar: 500 μm
Figure 5.12: The EDS elemental colormap of Ga, In, Ag, Fe on the surface of zPDMS, along with the content of each element, by weight. Scale bars: 500 μm
Figure 5.13: The EDS elemental colormap of Ga, In, Ag, Fe on the surface of EGaIn-coated/laser-ablated zPDMS, along with the content of each element, by weight. Scale bars: 500 μm
Figure 5.14: Zoomed-in SEM image(a) and EDS elemental colormaps of Ga(b) and In(c) for zPDMS surface.
Figure 5.15: Zoomed-in SEM image(a) and EDS elemental colormaps of Ga(b) and In(c) for EGaIn-coated/laser-ablated zPDMS surface. The circled areas in (a) show typical droplets of EGaIn remaining from laser ablation and this is confirmed by comparing with the circled areas in (b) and (c).
Figure 5.16: (a) The serpentine design of the samples for resistance test. The sample shown has a trace width of 200 μm. The resistance measurement is taken by connecting Electrode A and one of Electrodes B. (b) The interdigital design of samples for capacitance test. The sample shown has four pairs of 250-μm-wide electrodes with 150-μm-wide gaps and 7-mm-long electrode overlapping.
5.2.2 Resistance Test

Samples with different circuit designs are produced to examine the electrical properties of UV laser-patterned EGaIn traces. Samples with a serpentine circuit are fabricated to test the traces resistance with different length (Figure. 5.16a). EGaIn is deposited on a zPDMS film and patterned with UVLM. The traces have two different widths (100 and 200 μm, 4 samples for each trace width) and the measurements are taken between the terminal on the upper left (Electrode A) and one of the terminals on the right side (Electrodes B) by direct contact with the probes of a multimeter. The goal of this test is to examine the resistance change with different trace length (5, 14.6, 24.2, 33.8, 43.4, 53, 62.6 mm). The resistance-vs-length plots (Figure. 5.17) of the UV laser-patterned EGaIn traces shows high linearity with $R^2$ values greater than 0.96. The equations of the average resistances (in Ω) of 100-μm-wide and 200-μm-wide traces are $R_1=0.1615x+0.3713$ and $R_2=0.055x+0.2493$, respectively. Here, $x$ is the length of circuit, in mm. These equations indicate that the average resistance of 100-μm-wide traces (0.1615 Ω/mm) is 2.93 times that of 200-μm-wide traces (0.055 Ω/mm) with the contact/lead resistance excluded. The difference in resistance normalized by width is likely due to the uneven deposition of EGaIn. The thickness of the EGaIn traces is calculated from the resistance measurements using Ohm’s Law $R=\rho L/(wt)$, where $\rho$ is the electrical resistivity of EGaIn ($29.4 \times 10^{-8}$ Ω·m [35]), $L$, $w$, and $t$ are the length, width, and thickness of the conductor, respectively (all in m). The calculation results show that the thickness of the 100-μm and 200-μm traces are $\sim 18.4$ and 27.1 μm.
Figure 5.17: The plot of resistance vs. trace length for two different widths.
5.2.3 Capacitance Test

Capacitors with interdigital electrodes (Figure 5.16b) are made and tested to further examine the electrical properties of the UV laser-patterned traces and zPDMS. One group of the capacitors consist of four pairs of 250-μm-wide and 150-μm-spaced electrodes with different overlappings (5, 7, 8, 9 mm and nine samples for each electrode length). The electrodes are patterned on a 120-μm-thick zPDMS substrate. The capacitance is measured by touching the probes of an LCR meter with Electrode A and B. The capacitance-vs-length plot (Figure. 5.18) shows high linearity with an $R^2$ value greater than 0.9. With the slope of the fitted curve (0.2065 pF/mm), the relative permittivity of zPDMS can be estimated with the following model derived by Gevorgian et al.[127]:

\[ C = C_3 + C_n + C_{end}. \]  \hspace{1cm} (5.1)

\[ C_n = (n - 3)\varepsilon_0\varepsilon_{en} \frac{K(k_0)}{K(k'_0)} l \]  \hspace{1cm} (5.2)

\[ C_3 = 4\varepsilon_0\varepsilon_{e3} \frac{K(k_{03})}{K(k'_{03})} l \]  \hspace{1cm} (5.3)

Here, $C$ is the total capacitance of an interdigital capacitor. $C_n$ is the capacitance of the periodical section of the capacitor, and $C_3$ is the capacitance of the 3-finger section. Since the length of electrodes is much longer than its width, the capacitance of the end zone, $C_{end}$, is negligible. $l$ is the overlapping length of the electrode. $K$ is the complete elliptical integral of the first kind, where $k_0$ and $k_{03}$ are calculated based on the geometrical dimensions of the interdigital capacitor, and $k'_0=\sqrt{1-k_0^2}$, $k'_{03}=\sqrt{1-k_{03}^2}$. Based on this model, the effective relative permittivity of zPDMS is $\sim5.35$. The reason to use the term “effective” is that the conductive columns
Figure 5.18: The plot of capacitance vs. electrode length for samples with four pairs of 250-μm-wide electrodes with 150-μm-wide gaps. The least square fit assumes a linear dependency.

connected to EGaIn trace can actually form parallel capacitors, which is hard to model at this point.

5.2.4 Stretching Test

As shown in Figure 5.19, an EGaIn circuit sample was fabricated to show the trace’s resistance change when being stretched. This sample contains UV laser-patterned EGaIn trace and was interfaced with multimeter probes through zPDMS. The EGaIn
Figure 5.19: The sample of EGaIn circuit used for stretching test. The circuit is a 120-mm-long serpentine EGaIn trace that interfaces with external wires through zPDMS.
trace is 120 mm long and 0.5 mm wide. The sample is stretched to 60% from 0, with an increment of 5%. The total initial resistance of the circuit is measured to be 8.12 Ω. According to elastic deformation, total resistance of the circuit is

\[ R = R_1 + R_0 \lambda^2, \]

where \( R_1 \) is the resistance for the unstretched part of the circuit, \( R_0 \) is the initial resistance of the serpentine portion of the circuit which is stretched, and \( \lambda \) is the stretch. The resistance-vs-stretching plot is shown in Figure. 5.20 and the least-square fitting curve is based on the polynomial above. According to the fitting curve, the initial resistance of the serpentine circuit is approximately 4.32 Ω. This implies that the average thickness of this EGaIn circuit is approximately 16.3 μm, which is roughly consistent with previous calculation.

5.3 Summary

In this chapter I reported a novel method for rapid prototyping of liquid metal (EGaIn). UV laser ablation is used to break the metallic bonds between liquid metal atoms and as a result the liquid metal atoms are released as vapor into the system exhaust. With this method, thin films of EGaIn deposited on polymer surface can be UV laser-patterned into planar features with dimensions as small as 50 μm and the spacing between features can be as small as 100 μm. The experimental results reported above show that the UV laser-patterned EGaIn traces provide reliable conductivity. Combined with our z-axis conductive polymer (zPDMS), UV laser-patterning is feasible for the rapid prototyping of applications such as soft-matter circuits.

Although a rapid and reliable prototyping method for liquid-metal-based circuits, this method is not suitable for certain polymer surfaces such as pure PDMS. The UV transparency of PDMS is so high (\(~0.99, [128]\)) that a large portion of the UV energy
Figure 5.20: The plot of resistance vs. stretching for the sample shown in Figure 5.19.
will be absorbed by the substrate underneath. As a result, proper material selection is necessary before using UV laser for liquid metal patterning.
Chapter 6
Conclusion and Future Directions

Particle-filled elastomers and LM alloys are promising as conductive materials for soft-matter circuit. However, despite their growing popularity among academic researchers, limitations with fabrication and electronics integration remain a critical impediment to broader adoption. Further progress in this area depends on reliable techniques for rapid materials patterning and interfacing with packaged microelectronics and flex circuits.

Laser-based fabrication techniques, which has been used in the electronics industry for decades, is especially promising since it’s compatible with a very broad range of materials. Here, I have shown that these methods can be extended to liquid metal and elastomers to create soft-matter circuits that remain electrically functional when deformed. By introducing a series of novel fabrication methods with Ga-based LM alloy, I have addressed the following research objectives:

1. Demonstrate and examine the ability to pattern films of Ga-based liquid metal and conductive elastomers and produce soft-matter circuits using laser ablation.

Successful EGaIn and Galinstan patterning is accomplished with both a CO\textsubscript{2} laser engraver (VLS 3.50) and UV laser micromachining system (Protolaser U4; LPKF). The mechanism of ablation and pattern resolution is primarily dictated by the laser
wavelength, power, patterning speed, pulse duration, and optics. These factors, in turn, are dictated by the choice of laser system, which can range from an entry-level CO\textsubscript{2} laser engraver to an industrial-grade UV laser system used in microelectronics prototyping.

1a. Develop methods to produce LM circuits with both CO\textsubscript{2} and UV laser patterning.

With a CO\textsubscript{2} laser, the reflectance is too high for direct photophysical ablation of the liquid metal. Instead, a thin LM film should be deposited on a polymer substrate (e.g. PDMS, PU) that produces a recoil vapor that punctures through the film and induces patterning. This mechanism relies on the immediate oxidation of the liquid metal, which allows it to hold its pattern after the polymer vapor is released.

With UV laser micromachining, the photon absorption is high enough to induce photophysical ablation. Moreover, a tighter beam waist can be maintained over a longer cutting depth (e.g. 15 μm over ~1 mm). The Protolaser U3 is designed for industrial-level electronics prototyping and has native features (e.g. fiducial recognition, CAD software) for circuit patterning and multi-layer alignment.

1b. Validate rapid prototyping methods with representative circuits and sensors that remain electrically functional during elastic deformation.

Thanks to the rapid prototyping techniques, different types of applications are implemented and evaluated. These applications, including strain sensors and tactile sensors, have shown that they can maintain functionality during mechanical loading. The strain sensor (Chapter 4), which is essentially a parallel-plate capacitor, shows excellent performance in terms of repeatability and robustness. The tactile sensors (Chapter 3 & 4) demonstrate that laser-patterned conductive polymer composite and
liquid metal can be used for next-generation wearable electronics. These applications validate the overall effectiveness of the techniques reported in this work.

2. Explore methods to engineer a mechanically robust electrical interface between the terminals of the soft-matter circuit and the pins of rigid electronic components.

The interfacing technique developed provide a potentially effective solution to the implementation of stretchable integrated circuits. Circuit shorting has been a significant issue when interfacing liquid metal with solid-state components. With conventional methods, delamination may easily occur and, as a result, liquid metal seep through interfaces and cause shorting. Z-axis conductive material is developed to address this issue. Uniaxial (z-axis) conductivity is achieved by using conductive particles that are magnetically aligned to form discrete columns. With these z-axis conductive materials, liquid metal traces form electrical connections with surface-mounted components without any in-plane shorting. This interfacing technique has also been validated by application shown in Chapter 4.

6.1 Future Directions

Although the presented work has made significant improvement in soft-matter circuit fabrication, the methods can still be further improved. Below are several suggestions for potential follow-on research in LM rapid prototyping and interfacing.

6.1.1 Laser-Substrate Interaction

As mentioned in Chapter 5, a UV laser is not well-suited for LM patterning on PDMS because of its high UV transparency. Since UV laser can go through PDMS easily, the materials underneath the PDMS substrate can be ablated. The redisure
or vapor formed in this process may cause deformation of PDMS substrate that can significantly influence the effect of laser patterning. Additionally, it is noticed that the substrate can become partially damaged by the laser during patterning with a coating of EGaIn film. Neither of these effects are desired and further study of laser-elastomer interaction is required to better understand and mitigate these effects.

The addition of liquid metal and conductive particles to PDMS can greatly reduce its UV transparency. This suggests that the UV transparency of materials like PDMS can be potentially modified by using additives. However, such additives should not interfere with elasticity (i.e. elastic modulus, strain limit, rate-controlled hysteresis) or other desired properties like optically transparency and biocompatibility.

One candidate to be considered as an additive is Polyvinylbutyral (PVB). Compared to PDMS, which has a UV transmittance of nearly 100%, PVB has a much lower UV transmittance of approximately 26%[129]. PVB is also a transparent and nontoxic polymer, which make it suitable for common uses of PDMS. However, there are disadvantages for PVB as an additive in PDMS. Firstly, it is not as soft as PDMS. With a Young’s modulus of 0.97 GPa[130], PVB may significantly increase the stiffness of the composite. Secondly, PVB is far less stretchable than PDMS and fails when elongated to approximately 4.9% strain[130]. To use PVB as an UV-absorption-enhancing additive in PDMS, deterministic techniques are needed. For example, instead of mixing PVB directly with PDMS, it can be made into micro particles[131] and then mixed with PDMS to produce a transparent, PDMS-based composite with enhanced UV absorption. The rigidity of the composite will be larger than that of pure PDMS but can be adjusted by using different doses of PVB particles. This proposed method needs to be examined in detail to validate the mechanical and optical effectiveness.

As for the interaction between the laser and polymer substrate, an in-depth para-
metric study is needed to find out the optimal laser settings to minimize the substrate ablated during laser processing. According to Helvajian[132], laser material processing can be influenced by a variety of factors, namely wavelength, power fluence, dose (number of laser pulses delivered), processing beam character and a variety of spatial and temporal properties. These factors need to be studied along with the properties of the materials, including their thermal conductivities, molecular bondings, etc. For this type of systematic study, a highly customized laser ablation system is needed. In addition to the parametric study, another approach to obtaining precise LM laser patterning is to use ultrafast pulsed laser oscillators and amplifiers by multiphoton absorption[133].

6.1.2 Biocompatible Conductive Materials

Because they match the mechanical properties of natural biological tissue, soft-matter electronics have the potential to be used in biomedical/implantable devices. Safety is prioritized and this requires that the materials used should be biocompatible with tissues. In addition to this, the functional materials should be conductive enough for practical electronics applications. Polymers like PDMS have excellent biocompatibility and are popular as stretchable materials for implantable devices. A challenge is to introduce materials for electrical conductivity while preserving elasticity and biocompatibility.

Though nontoxic, Gallium-Indium-based liquid metal alloys are corrosive and therefore should not be in direct contact with tissue or organs. Alternatively, PEDOT:PSS is a biocompatible polymer that can provide conductivity as high as 4000 S/m[134]. However, it needs other chemicals added in order to be stretchable. Noh et al[135] have reported that a PDMS/PEDOT:PSS composite with a fracture strain of 75%. The conductivity of this material, however, is only 200 S/m. Some carbon
nanotube-filled polymer composites are biocompatible with conductivity as high as 5.7×10^5 S/m, but the conductivity drops dramatically when stretching[136]. Of the popular filled-elastomer composites, carbon black-PDMS (CPDMS) is biocompatible but the conductivity is very low and the biocompatibility of AgPDMS remains to be established [137].

Hydrogels are another potential direction for future research on biocompatible[31] and stretchable conductive materials. It has been reported that biocompatible hydrogels can be stretched to as much as 2100% strain (PAAm-alginate hydrogel[138]), which makes them suitable as base materials for a stretchable conductor. Hydrogels are attractive because they can be modified with other chemicals for certain properties. As for high conductivity, Sasaki et al [139] have fabricated a PEDOT/PU-hydrogel hybrid with a conductivity as high as 1.2×10^4 S/m at 100% strain. Thanks to their high stretchability and potentially high conductivity, it is reasonable to expect that hydrogels can be utilized as the conductor for stretchable biomedical devices.

6.1.3 Improving Z-axis Conductivity

We have noticed that zPDMS may have trouble providing reliable interfaces if the electrode pads of surface-mounted components are smaller than 0.5×0.35 mm. This is due to the relatively low density of conductive columns – in such small areas, the number of conductive columns is around 15. When the polymer is deformed, some of the columns will lose connection because the particles are separated. As a result, the through-thickness conductivity is further reduced. To solve this problem, smaller particles can be used to form more conductive columns in a relatively small area. In this work, I used magnetic particles of two sizes, namely 15 and 40 μm. Silver-coated magnetic micro/nanoparticles have been developed by multiple research groups[140][141][142], and shows that the dimension of magnetic particles can be
significantly reduced. Using these smaller particle sizes may allow for a higher density of columns and reduce the required dimensions of the via.

In addition to this, the overall thickness of zPDMS should be reduced to minimize the influence of polymer deformation and reduce resistance of each column. This, however, will require improvements on control of the laser ablation process. The thickness of zPDMS presented in this work is around 100 μm and each column is formed with about 3-5 particles, depending on the size of particles. The ideal thickness of zPDMS, or other uniaxially conductive materials, is the same as the diameter of particles. This single-particle column can hopefully eliminate the likelihood of conductivity loss due to separated particles.

As it may take extensive research to figure out optimal laser parameters for LM ablation, it may not be possible to accurately pattern anisotropically conductive materials that are as thin as the diameter of a single particle. Until then, methods for patterning multi-particle columns are still in need. This indicates that a method to enhance the contact between particles is important. In Chapter 5, I have shown a zPDMS with EGaIn-coated magnetic particles. EGaIn, as well as other LMs, can help connect particles when they are undergoing mild separation because of their high surface tension, fluidic integrity, and strong wetting to metal. This technique has yet to be extensively studied and should be a important topic for near-term research in LM stretchable electronics interfacing.

Lastly, because of the poor bonding between PDMS and surface-mounted components, delamination is often observed. This failure results in a loss of conductivity and device malfunction. The potential solution is to make anisotropically conductive composites with polymers that bond strongly to surface-mounted components. PVB, which is discussed previously, has great bonding ability to almost any surface. The drawback is that its large Young’s modulus and small fracture strain can
Figure 6.1: Subfigure(a) shows a stretchable and flexible circuit with six LED chips connected by LM interconnects patterned using the Protolaser U3. The LEDs and LM circuits are interfaced with a layer of zPDMS. As shown in subfigures (b) and (c), this circuit remains functional when stretched, twisted, and flexed.

lead to brittle fracture and cohesive failure (i.e. failure in the polymer instead of at the interface). Hopefully, these limitations can be addressed by addition of other polymers or chemicals in order to achieve an acceptable mechanical compliance and strain limit. Experimental data on modified PVB is currently insufficient, although researchers have successfully fabricated PVB-modified epoxy with improved mechanical properties[143].

6.1.4 Applications with Integrated Techniques

With the techniques presented in this thesis work (process shown in Chapter 5), several applications, including a Lilypad circuit consisting of LED chips (Figure. 6.1) and a heart-rate/blood-oxygen monitoring chip have been fabricated (Figure. 6.2). The yield rate of the fabrications is relatively low because the size of the contact pads is approaching the limit mentioned above in Sec. 6.1.3. With all the research topics mentioned above being studied, the techniques presented in this work can hopefully be improved and feasible for mass production of reliable and robust stretchable electron-
Figure 6.2: (a) Layout of a heart-rate/blood-oxygen sensing unit embedded in PDMS. The surface-mounted components are interfaced with LM through zPDMS. Scale bar: 2 mm. (b) Surface-mounted PulseOx chip remains functional during bending. (c) a close-in look of the unit.
ics. The potential areas in which these stretchable electronics can be applied include wearable health and fitness monitoring systems, personal electronics, and peripherals for augmented and virtual reality.

Unlike the existing wearable technologies, the next-generation of wearable devices should be soft and elastic enough to be integrated into clothing or adhere directly to the skin. The latter could be thought of as a “second skin” or “data skin” that monitors both physiological condition and external information such as UV intensity, humidity, toxicity level, etc. In addition to this, the second skin can be used as an input device for mobile computing. Technologies like virtual reality and augmented reality will benefit from these new systems since the user experience can be guided by more “natural” inputs like heart rate, muscle activity, and limb movement.

The application of stretchable electronics are currently limited by certain components such as stretchable displays and batteries. These represent additional open areas of research that could incorporate some of the materials and fabrication methods presented in this thesis. Breakthroughs in these areas will speed up the implementation of fully stretchable and wearable electronics.

6.2 Closing Remarks

In contrast to flex PCBs based on conventional electronic materials, stretchable circuits composed of conductive elastomers and liquid metal have the potential to be used in emerging applications that undergo large deformations. These include wearable computing devices that must match the stretchability of elastic textiles, inflatable and self-reconfiguring deployable structures, bio-inspired soft robotics, and “second skin” adhesives that bond to the skin and monitor physiological activity. Progress, however, depends on reliable and rapid techniques to produce these soft-matter circuits and populate them with solid-state electronic components.
As shown in this thesis, laser-based prototyping technique and particle-based anisotropically conductive materials are potential solutions for achieving these two objectives. The methods presented here lead to circuits that are mechanically robust and remain functional under stretching, bending, and twisting. With further improvements in materials and interface architectures, these approaches will eventually lead to stretchable electronics that can be integrated into wearables, soft robotics, and other emerging technologies.
Bibliography


