1. Introduction

Progress in soft functional material synthesis\cite{1,2} and manufacturing technology\cite{3,4} has enabled bioinspired and skin-like soft electronics for applications ranging from entertainment to healthcare.\cite{5,6} Unlike conventional solid-state electronics, soft electronics can be lightweight, stretchable, and reconfigurable, with biocompatible characteristics for skin-mountable and wearable sensing electronics.\cite{7,8} Thereby, flexible and stretchable characteristics are achieved by using either 2D or 3D compliant wave-like, solid metal patterns\cite{9,10} or elastic conductors based on conductive nanomaterials embedded in a polymer matrix.\cite{11,12} An alternative approach to realize all-soft microsystems is the use of intrinsically soft conductors, such as gallium-based liquid metal (eutectic gallium–indium alloy, EGaIn). EGaIn-based soft electronics benefits from its nontoxicity, mechanical stability (unlimited stretchability, but ultimately limited by the mechanical properties of the encasing material), thermal conductivity ($\kappa = 26.6 \text{ W m}^{-1} \text{K}^{-1}$), and electrical conductivity ($\sigma = 3.4 \times 10^6 \text{ S m}^{-1}$).\cite{13–15} The low melting temperature ($M_\ell < 15 \degree \text{C}$) and negligible vapor pressure of EGaIn facilitate room-temperature and ambient pressure manufacturing processes.\cite{13–15} Moreover, thanks to the formation of a thin oxide layer ($t = 1–3 \text{ nm}$) on the EGaIn surface under atmospheric oxygen level, EGaIn structures maintain their mechanical shapes,\cite{16,17} allowing 2D/3D EGaIn patterns on a soft elastomeric substrate, such as poly(dimethylsiloxane) (PDMS).

The moldable characteristic of EGaIn has enabled a broad range of patterning methods based on lithography-enabled stamping and stencil printing, injection, as well as additive and subtractive direct write/patterning processes,\cite{18–28} as summarized in Table S1 in the Supporting Information. Thereby, printing using lithography-defined stencils\cite{23–24} yields simple and high throughput EGaIn patterning on elastomeric substrates with small features of $w$ (width) $\approx 200 \ \mu \text{m}$, $t$ (thickness) $\approx 50 \ \mu \text{m}$ using metal stencil films,\cite{21} $w = 20 \ \mu \text{m}$, $t = 2 \ \mu \text{m}$ using microfabricated metal stencil films,\cite{22} and $w = 20 \ \mu \text{m}$, $t = 10 \ \mu \text{m}$ using polymer stencil films.\cite{23} Limitations of this approach are the relatively low resolution, rough EGaIn surface, and excessive EGaIn loss during the stencil lift-off process. Subtractive
direct patterning techniques using laser ablation\cite{25,26} or electrochemical reduction\cite{27,28} enable an inexpensive and facile approach to pattern fine EGaIn lines, but the serial process makes EGaIn removal slow in the case of patterning small EGaIn features on large substrates. The major technical challenge for both lithography-enabled stencil printing and subtractive direct patterning approaches is that creating thin and uniform EGaIn films is difficult due to the high surface tension of EGaIn ($\gamma = 624$ mN m$^{-1}$).\cite{18,19} Manually spreading EGaIn using a roller typically results in rough surfaces with holes in the EGaIn film.\cite{18} Additive direct write and injection approaches address this issue: 2D/3D direct writing techniques\cite{29-32} allow to deposit EGaIn patterns in desired locations only, but the resolution is limited to $w \approx 100$ $\mu$m and $l > 50$ $\mu$m because of the size limitation of the injection nozzles. Microfluidic injection\cite{33,34} and vacuum filling\cite{35,36} approaches provide better resolution with $w > 10$ $\mu$m, but the microchannels require relatively large thicknesses of $l > 50$ $\mu$m to reduce pressure drops and their practical use is limited when EGaIn film needs to be exposed to the surface for additional processing. Using a microtransfer deposition process based on soft lithography\cite{37} the smallest EGIn features so far with $w > 2$ $\mu$m were demonstrated, but the technology suffered from EGaIn residues formed outside of the channel areas during the molding process.

Soft lithography\cite{38,39} in combination with wetting/nonwetting surface modifications and 3D heterogeneous integration\cite{40,41} can potentially solve current EGaIn patterning challenges, by providing i) smooth and uniform deposition of thin films ii) multiscale patterning, and iii) high-density and functional integration. Soft lithography offers a simple, fast, and low-cost way to pattern multiscale EGaIn films. However, the surface oxide layer on EGaIn interferes with uniform wetting on soft elastomeric substrates, typically resulting in non-uniform thickness and rough surfaces, as mentioned before. Therefore, stamp lithography is often regarded as the lowest resolution and least reliable technique among the additive printing methods.\cite{18,19,42} By utilizing chemical and physical surface modification of the elastomeric substrates,\cite{43,44} the PDMS surfaces can be modified to have selective nonwetting or wetting properties for EGaIn: the nonwetting characteristics of chemically modified PDMS surfaces hinder formation of EGaIn residue, while the uniform wetting characteristics of physically modified PDMS surfaces assist to form thin and smooth EGaIn films. In combination, these wetting/nonwetting properties enable multiscale and uniform EGaIn thin-film patterning in simple and effective manners. Recently, we demonstrated high resolution, uniform, and residue-free EGaIn thin-line patterning ranging from single micrometer to millimeter scales by utilizing a chemical surface modification and residue transfer process in a reverse stamping approach to enable high-density, soft passive electronics.\cite{45} Building on our previous work,\cite{45-48} this paper presents multiscale and uniform EGaIn thin-film patterning by utilizing an additive stamping process for large-scale ($\text{mm-cm}$) soft electronics and the subtractive reverse stamping process for microscale ($\mu$m-$\text{mm}$) soft electronics. By combining these complementary patterning techniques using 3D heterogeneous integration, fabricated and optimized soft electronic components built with different patterning processes can be integrated to form high-density and multifunctional soft microsystems.

2. Results and Discussion

2.1. Multiscale and Uniform Liquid Metal Thin-Film Patterning Based on Soft Lithography

Figure 1 illustrates the complementary multiscale EGaIn thin-film patterning processes based on soft lithography: a) a subtractive reverse stamping approach relying on chemical surface modification for microscale and high-density EGaIn lines embedded in a PDMS microchannel and b) an additive stamping approach for centimeter-scale and large-area EGaIn patterning relying on physical surface modification of the PDMS substrate. Thereby, physical and chemical surface modifications provide proper wetting/nonwetting properties, which enable fine, uniform, and residue-free EGaIn thin films. The chemical surface modification using toluene increases the hydrophobicity of PDMS surfaces, which minimizes EGaIn residue formation and allows them to be readily transferred to a sacrificial PDMS layer.\cite{45} Using the reverse stamping process, molded EGaIn in predefined PDMS microchannels enables high-resolution, residue-free, and uniform lines. On the other hand, the physical surface modification creates a paper-textured PDMS surface,\cite{43,44} increasing the surface areas and adhesion forces, ultimately enhancing wettability to form uniform and smooth EGaIn thin films using the additive stamping process.

The underlying liquid metal patterning processes mainly consist of three steps: 1) microtransfer molding of EGaIn, 2) EGaIn transfer using subtractive reverse stamping or additive stamping, and 3) sealing and interconnection. The fabrication process starts with the selective chemical surface modification of the PDMS mold (for the subtractive approach) or the PDMS stamp (an optional step for the additive approach), which increases the surface hydrophobicity (Figure 1-i). The PDMS mold/stamp is then pressed onto a donor PDMS substrate coated with EGaIn and separated from it (Figure 1-ii). During the molding process, EGaIn adheres to all surfaces of the PDMS mold/stamp, including concave channels and protruding surfaces. In the next step, the PDMS mold/stamp wet with EGaIn can be reverse stamped to remove EGaIn residue on the protruding surfaces or additive stamped to deposit EGaIn film (Figure 1-iii,iv). Using a PDMS mold with 5 $\mu$m deep concave microchannels, unwanted EGaIn residue can be readily transferred to a sacrificial PDMS layer, as shown in Figure 1a. The reverse stamping process yields fine, uniform, and thin EGaIn lines inside of the PDMS channels without residues on the surrounding protruding surfaces, achieving a lateral line width resolution as small as $w = 2$ $\mu$m at film thicknesses of $l = 1.8$ $\mu$m (Figure S1, Supporting Information). The subtractive approach showed size scalability from 2 $\mu$m to 1--2 $\text{mm}$ in line width.\cite{45} However, for channel widths > 2 $\text{mm}$, EGaIn is not effectively transferred to the inside of the PDMS channels because of the sagging deformation of wide channels during the pressure-based molding process. Therefore, the pattern dimensions, in particular line widths achievable using the
subtractive reverse stamping approach are limited to the single \( \mu m \) to a couple of mm scale.

In contrast, the additive stamping approach provides large-area EGaIn thin-film patterning. In this process, a PDMS stamp wet with EGaIn is stamped onto a paper-textured PDMS substrate, as shown in Figure 1b. Thereby, the paper texture, which comprises randomly distributed micro cellulose fiber structures (Figure S2a, Supporting Information), is transferred to a PDMS surface using a conventional PDMS casting method (Experimental Section). The resulting paper-texture PDMS exhibits a surface microstructure which allows it to wet with EGaIn compared to a normal, smooth PDMS surface (Figure S2b–d, Supporting Information). As a result, continuous EGaIn films can be transferred to paper-textured PDMS with multiple stamping steps. The smallest feature size of stamped EGaIn was \( \mu m = 500 \mu m / t = 1.5 \mu m \) (with five times stamping). While this additive stamping process shows relatively poor lateral resolution, it is suitable for large-area patterning and thin-film deposition and thus can complement the high-resolution, subtractive reverse stamping process described above. To demonstrate the multiscale and large-area patterning capability of both processes, we fabricated EGaIn-based microscale and centimeter-scale Georgia Tech logos using the subtractive and additive approaches, respectively (Figure 1a,b). To this end, we prepared a microfabricated PDMS mold with a 3 \( \times \) 16 array of Georgia Tech logos with a critical dimension of 2 \( \mu m \) and a PDMS stamp of a Georgia Tech logo, which is 23\( \times \) wider in length and 529\( \times \) larger in area compared to the microfabricated PDMS mold, with critical dimensions of \( \approx 500 \mu m \). The resulting patterns using both additive and subtractive methods show fine, uniform, and smooth EGaIn films with sharp edges, which demonstrates the complementary nature of both fabrication processes (Figure S3a–c, Supporting Information). In the final step, the patterned EGaIn films were covered with an additional PDMS layer by either casting or spin-coating, and EGaIn-filled soft vias were used for vertical interconnection, as introduced in our previous work.\(^{[45]}\) Thanks to the self-healing characteristics of EGaIn, the soft vias facilitate 3D heterogeneous integrated soft microsystems for high-density integration and multifunctionality. The detailed fabrication processes for both the subtractive reverse stamping and the additive stamping are described in the Experimental Section.

Microscale EGaIn thin-line patterning using the subtractive reverse stamping technique was characterized in our previous work.\(^{[45]}\) This paper focuses on the large-scale EGaIn thin-film patterning to characterize resolution, scalability, smoothness, and uniformity of the additive stamping technique. Figure 2a,b shows the measured EGaIn width as a function of the designed.
stamp width and the resulting film thickness as a function of the patterned EGaIn width, respectively. PDMS stamps with >500 µm in width and 10 mm in length (Figure 1b) were prepared and stamped five times (alignment done under stereo microscope) on the paper-textured PDMS substrates. Thereby, stamping five times is the optimized number of stamping steps to achieve smooth and uniform films (Figure 2e). The tested smallest feature was 500 µm, and the patterned EGaIn lines showed reliable and uniform lines with on average <6.5% deviation from the designed PDMS width. This deviation can be explained by the deformation of PDMS elements during the stamping process as well as the misalignment during multiple stamping processes. The resulting film thickness after five times stamping was 1.56 ± 0.22 µm over the range of patterns tested. The uniform EGaIn thin film formation using the additive stamping approach is made possible thanks to the physical surface modification and the multiple stamping processes. To evaluate the impact of the physical surface modification on the EGaIn wettability, Figure 2c,d compares patterned EGaIn films on a normal, smooth PDMS surface, and a paper-textured PDMS surface. To this end, a square-shape PDMS stamp with 1 cm × 1 cm dimensions was prepared, and EGaIn was stamped on both the normal PDMS and the paper-textured PDMS substrate. As expected, patterned EGaIn on the normal,
smooth PDMS surface exhibits nonuniform films with EGaIn droplets and noncovered areas, as shown in Figure 2d-i. This nonuniform thickness resulting from poor surface wetting made stamping lithography the least reliable EGaIn patterning method, as highlighted in other literatures. In contrast, patterned EGaIn on the physical-surface-modified PDMS shows smooth and uniform thin films, as seen in Figure 2d-ii. 2d-iii shows a cross-sectional view of the stamped EGaIn thin film on the paper-textured PDMS after being covered with an additional PDMS layer, demonstrating the uniformly molded ≈1.45 µm thick EGaIn film patterned on the paper-textured PDMS substrate.

The uniformity and smoothness of the EGaIn thin film was then evaluated as a function of the number of stamping steps, as shown in Figure 2e. In the EGaIn stamping process, the PDMS surface roughness plays an important role in uniform wetting, which promotes EGaIn adhesion on a physically modified elastomeric surface. The paper-textured PDMS exhibited micrometer-sized surface structures with an average roughness $R_s$ of 1.22 ± 0.18 µm (Figure S2c,d, Supporting Information). The additive stamping process decreases the surface roughness $R_s$ with increasing number of stamping steps, which indicates that the stamped EGaIn film is getting smoother. The random, micrometer-sized features on the paper-textured PDMS surface were partially wetted during the initial one to two stamping steps, ultimately filling the grooved areas with EGaIn. After stamping five times, the surface structures of the paper-textured PDMS were fully covered with EGaIn, showing a uniform and smooth EGaIn thin film with low $R_s$ of 0.18 ± 0.01 µm. Interestingly, after stamping more than six times, excessive EGaIn partially formed small EGaIn edge beads along the edges of the patterned areas. To make films uniform again, these excessive edge beads can be effectively transferred to a sacrificial PDMS layer. The proposed additive EGaIn stamping enables simple patterning of uniform and smooth EGaIn films for large-scale electronics applications and shows competitive capabilities compared to other EGaIn patterning methods, as summarized in Table S1 in the Supporting Information. It is noted that the final EGaIn thickness (1.45 µm) needed to obtain smooth films is of the order of the initial surface roughness (1.22 µm). It would thus be interesting to see how paper textures with different surface roughness would affect these results.

### 2.2. Electrical and Mechanical Characteristics of Fabricated Soft Electronic Devices

Using the additive stamping process, soft passive components and circuits were fabricated, and their electrical and mechanical characteristics were investigated. Figure 3a shows fabricated soft passive components, namely a resistor, a planar spiral inductor, and an interdigitated capacitor, all having 1 mm line width and 1 mm line spacing, attached to a nonplanar object. One of the main requirements for soft Microsystems is to maintain their electronic functionality during mechanical deformation. Thus, we experimentally investigated the electronic functionality by applying bending and stretching forces. Figure 3b shows a simple circuit comprising a 5 × 5 array of light emitting diodes (LED). The commercial surface-mount LEDs were manually attached to EGaIn contacts using a tweezer along the patterned EGaIn lines under a stereo microscope for alignment and sealed using an additional PDMS casting. Applying a constant current to the soft circuit, the LED array was subjected to bending (bending radius, $r = 10$ mm) and stretching (strain, $\varepsilon \leq 50\%$) deformation. The soft circuit withstood these bending deformations and strain up to 50% while maintaining its electrical functionality, which also confirmed that the EGaIn thin film was uniformly patterned without voids.

The electrical and mechanical characteristics of the soft passive components were also investigated. Figure 4a shows the $I$–$V$ characteristic of the resistor (1 mm in width, 70 mm in length, and 1.35 µm in thickness), exhibiting an ohmic behavior with <7% deviation between the calculated and measured behavior. We then applied uniaxial strain up to 30% in both width and length directions to the resistor, the planar spiral inductor, and the interdigitated capacitor. Figure 4b–d shows the measured and calculated relative resistance, inductance, and capacitance changes, respectively, as a function of the applied uniaxial strain in both length ($\varepsilon_l$) and width ($\varepsilon_w$) directions. The calculated values are based on either analytical or numerical simulation using COMSOL (COMSOL Multiphysics v.5.3, COMSOL Inc.). For the resistor, the relative resistance change $\Delta R/R$ increases by stretching along the length direction and decreases by stretching along the width direction, as shown in Figure 4b. This behavior can be simply explained by the effect of geometry changes on
the resistance $R = \rho l/(wd)$, considering the Poisson’s ratio of the elastomeric materials. In the case of the planar spiral inductor ($d_{\text{out}} = 11 \text{ mm}$, $d_{\text{in}} = 5 \text{ mm}$, and $n$ (number of turns) = 2), the measured inductance was 43.7 nH, which agreed well with the simulated value <15% deviation (Figure S4a,b, Supporting Information). To evaluate its characteristics under mechanical loading, we used a modified Wheeler formula and calculated the relative inductance change $\Delta L/L$ as a function of the applied strain in both length and width directions.$^{[51,52]}$

Under the assumption of equibiaxial strain ($\varepsilon_{\text{wd}} = \varepsilon_{\text{w}} = \varepsilon_{\text{l}}$) and incompressible ($w_l + 1)(\varepsilon_{\text{l}} + 1) = 1$ conditions, the calculated $\Delta L/L$ can be simplified to $\varepsilon_{\text{s}} + 1)/2 + 1/(2(\varepsilon_{\text{w}} + 1)^{0.5}) - 1$ for strain in both width and length direction.$^{[52]}$

Overall, both calculated and measured $\Delta L/L$ increase by stretching in both width and length directions with similar behavior because of the orthotropic geometry, as shown in Figure 4c. The rather large deviation between analytically calculated and measured values at a strain of 30% may arise from the simplified 2D analytical model and the shape distortion of the planar spiral inductor while stretching.$^{[52,53]}$

In the case of the interdigitated capacitor, the measured capacitance ($l$ (length of the electrode) = 6 mm and $n$ (number of electrodes) = 6) was 1.48 pF, which agreed well with the simulated value with <5% deviation, as shown in Figure S5a,b in the Supporting Information. By adopting a conformal mapping technique,$^{[54]}$ which provides a closed-form expression for the capacitance by transforming the coplanar line geometry to a parallel-plate structure, the calculated capacitance $C$ can be simplified to $C_0(\varepsilon_{\text{l}} + 1)$ under the conditions of $l = l_0(\varepsilon_{\text{l}} + 1)$, $w = w_0(\varepsilon_{\text{w}} + 1)$, $s = s_0(\varepsilon_{\text{w}} + 1)$, and $(\varepsilon_{\text{w}} + 1)(\varepsilon_{\text{l}} + 1) = 1$, with $s$ being the electrode spacing. Therefore, the calculated relative capacitance change $\Delta C/C$ can be simplified to $1/(\varepsilon_{\text{w}} + 1)^{0.5} - 1$ under strain in width direction and $\varepsilon_{\text{l}}$ under strain in the length direction.$^{[42,52]}$

Both calculated and measured $\Delta C/C$ linearly increase when the interdigitated capacitor is stretched along the length direction, whereas they decrease when stretched along the width direction, as shown in Figure 4d. As discussed above for the planar spiral inductor case, the 2D nature of the analytical model and the shape distortion of the interdigitated capacitor likely cause the deviation between analytically calculated and measured values.$^{[52]}$

### 2.3. 3D Heterogeneous Integration for Skin-Mountable, Soft Functional Microsystems

By combining the proposed multiscale EGaIn thin-film patterning techniques with 3D heterogeneous integration, more complex soft microsystems can be realized. In this work, three different skin-mountable, soft functional microsystems based on 3D heterogeneous integration are demonstrated: i) a soft LC (inductor-capacitor) sensing platform with high areal capacitance, ii) a fingertip-mountable, soft biological sensing platform, and iii) soft heaters with localized and distributed heating capability. Figure 5 shows the 3D-integrated, skin-mountable LC resonator. The interdigitated capacitor ($w = 50 \mu m$, $s = 100 \mu m$, and $n = 34$) was fabricated using the subtractive reverse-stamping technique, and its area capacitance was 13.2 pF cm$^{-2}$, which is a roughly nine times higher density than what would be achievable with the (coarse) additive stamping technique. In contrast, the planar coil inductor ($d_{\text{out}} = 20 \text{ mm}$, $d_{\text{in}} = 11 \text{ mm}$, and $n$ (number of turns) = 2) of the LC resonator as well as the readout coil were fabricated using the additive stamping technique. After 3D integration of the LC components using liquid-metal filled vias, the soft microsystem was wirelessly interrogated using a readout coil to collect the resonance frequency (Figure S6, Supporting Information). The measured
resonance frequency was 276 MHz, which also agreed well with the calculated value with <5% deviation, and the Q factor was ≈20 (Figure 5). Using the LC resonance platform, battery-free and wireless sensors can be designed and fabricated for wearable chemical sensing applications.\(^{[51,55]}\)

To highlight the multiscale EGaIn patterning capabilities, a fingertip-mountable, soft, and 3D-integrated biological sensing platform, comprising a soft sensing layer with a commercial pulse oximeter and a soft interfacing circuit layer, is demonstrated for noninvasive and real-time heart rate (HR) and blood oxygen monitoring (Figure 6a). The soft sensor layer is fabricated using the subtractive reverse stamping technique for microscale EGaIn patterning to connect the integrated pulse oximeter (MAX30100, Maxim Integrated Products Inc.), while the additive stamping technique is utilized to fabricate the soft printed circuit board (sPCB) for the interfacing circuit (Figure 6b–d). These soft sensor and circuit layers are then vertically interconnected through soft vias to form a 3D-integrated soft system (Figure 6e). Figure 6f,g shows the measured photoplethysmogram (PPG) waveforms of IR LED and red LED and the extracted HR and saturation of peripheral oxygen (SpO\(_2\)) extracted using the PDMS/EGaIn-based soft sensing system (HR = 79 ± 3.6 bpm and SpO\(_2\) = 97%), demonstrating identical sensing performance compared to a PCB-based rigid sensing platform.

Figure 6. a) Demonstration of fingertip-mountable, soft, and 3D-integrated biological sensing platform using integrated pulse oximeter mounted on fingertip for heart rate and blood oxygen monitoring; b) circuit diagram; fabrication and electric component integration of c) soft sensor layer patterned using subtractive reverse stamping technique and d) soft circuit layer patterned using additive stamping technique; e) 3D-integrated biological microsystem mounted on fingertip; measured photoplethysmogram (PPG) waveforms of IR LED and red LED using f) PCB-based rigid sensing system (heart rate (HR) = 79.1 ± 3.1 bpm and saturation of peripheral oxygen (SpO\(_2\)) = 97%) and g) PDMS/EGaIn-based soft sensing system (HR = 79 ± 3.6 bpm and SpO\(_2\) = 97%).
system (MIKROE-2000, MikroElektronika Ltd., HR = 79.1 ± 3.1 bpm, and SpO₂ = 97%).

Finally, to highlight the EGaIn thin-film patterning capability, soft heaters based on EGaIn thin-film resistors are demonstrated for localized and distributed heating applications with <60 °C operating temperature because of the thermal degradation of PDMS at higher temperatures.[56] For the localized heating applications (Figure 7a), a 50 µm wide heating resistor fabricated using the subtractive reverse stamping technique is vertically integrated on a soft circuit. For large area heating applications (Figure 7b), a 1 mm wide serpentine heating resistor fabricated using the additive stamping technique is patterned along with a soft circuit. Figure 7c shows the measured and simulated hot-spot temperature as a function of the applied heating power. To reach a temperature increase of ΔT = 10 °C, the vertically integrated microheater requires 65 mW and heats a localized area, while the serpentine-shaped heater heats a larger area but requires 113 mW. Compared to other literatures,[57–59] the fabricated soft heaters using thin-film resistors showed 8x higher heating efficiency (0.158 °C mW⁻¹ for the localized heating approach and 0.093 °C mW⁻¹ for the distributed heating approach). In addition to efficient heating, the soft heater provides flexibility and stretchability for wearable and skin-mountable electronics applications (Figure S7, Supporting Information).

3. Conclusion

This paper reports multiscale and uniform EGaIn thin-film patterning based on soft lithography by utilizing a subtractive reverse stamping approach for microscale and high-density patterns and an additive stamping approach for centimeter-scale, large-area patterns. Considering the size scalability of the thin-film patterning and the possibility to heterogeneously integrate structures fabricated with either technique, soft electronic components can be fabricated and integrated to form high-density and multifunctional microsystems for physical, chemical, and biological sensing applications.

4. Experimental Section

Subtractive Reverse Stamping Technique Based on Soft Lithography[45]

For PDMS mold preparation, a microfabricated photore sist master on a silicon wafer with critical dimension of 2 µm was fabricated. To create the PDMS mold, liquid PDMS (10:1 ratio of PDMS prepolymer and curing agent, Sylgard 184, Dow Corning) was either drop casted or spin coated on the fabricated silicon master molds and cured at 60 °C for 8 h. A 500 µL droplet of toluene (Toluene, ACS grade, VWR International) was drop-coated on a glass substrate and subsequently dried at room temperature and under atmospheric pressure for 5–10 min. Then, the PDMS mold was placed on the glass substrate for chemical surface modification. In the microtransfer molding process, EGaIn (gallium–indium eutectic, >99.99% trace metal basis, Sigma-Aldrich) was...
dispensed and spread using a PDMS roller on a donor PDMS substrate. Afterward, the PDMS mold was gently pressed onto the EGaIn thin film and separated from it. Unwanted liquid metal residue on the outside of the channel was transferred to a sacrificial PDMS layer, and this transfer process was repeated several times (>15 times) until all residue is removed. The EGaIn-filled PDMS mold was finally bonded to an additional PDMS layer using either drop casting or spin coating.

Additive Stamping Technique Based on Soft Lithography: For the PDMS stamp preparation, an acrylic master with critical dimension of 500 μm was fabricated using a CO2 laser cutter, and liquid PDMS (10:1 ratio of PDMS prepolymer and curing agent, Sylgard 184, Dow Corning) was drop casted on the fabricated molds and cured at 60 °C for 8 h. For paper-textured PDMS preparation, a small piece of standard printing paper (Office Depot #348-037) was taped on a flat substrate, and liquid PDMS (10:1 ratio of PDMS prepolymer and curing agent, Sylgard 184, Dow Corning) was either drop casted or spin coated on the paper substrate. After curing at 60 °C for 8 h, the polymerized PDMS was gently peeled off from the paper substrate. With this process, the micro cellulose fiber structures can be effectively transferred to the PDMS surface, as shown in Figure S2 in the Supporting Information. EGaIn (gallium–indium eutectic, >99.99% trace metal basis, Sigma-Aldrich) was dispensed and spread using a PDMS roller on a donor PDMS substrate. The PDMS stamp was gently pressed onto the EGaIn film, and then the EGaIn film was stamped to the paper-textured PDMS substrate. The patterned EGaIn films on the paper-textured PDMS were then sealed by an additional PDMS layer using either drop casting or spin coating, and commercial copper tape was used for electrical contacts. All PDMS samples were polymerized at 60 °C for 8 h.

Electrical, Mechanical, Thermal, and Optical Characterizations: Electrical characterizations of the soft passive components, circuits, and LC resonator were performed using a multimeter (Hewlett Packard 34401A), a source meter (Keithley 2636A), and an LCR meter (Agilent 4284A). For mechanical stretching and bending characterizations, a linear motion stage and circular glass cylinders (radius: 10 mm) were prepared, respectively. Thermal characterizations of the soft heater were performed using a source meter (Keithley 2636A) to supply power and temperature increases were recorded using an IR camera (FLIR T640). Surface characterizations were performed using 3D laser confocal microscopy (Olympus, LEXT OLS4000) and scanning electron microscopy (SEM, Hitachi S-3700N Variable Pressure SEM). All characterizations were conducted at room temperature and under atmospheric pressure.

Participants: Informed consent was obtained from two participants (M.-g. Kim and C. Kim), who volunteered to perform these studies. All testing reported conformed to the ethical requirements of Georgia Institute of Technology.

Supporting Information
Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest
The authors declare no conflict of interest.

Keywords
all-soft electronics, gallium-based liquid metal (eutectic gallium–indium alloy, EGaIn), heterogeneous 3D integration, multiscale liquid metal thin-film patterning, soft lithography

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