SUPPLEMENTARY INFORMATION

Personal Electronics Printing via Tapping Mode Composite Liquid Metal Ink Delivery and Adhesion Mechanism

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SUPPLEMENTARY METHODS AND DISCUSSION

1. PRINCIPLES OF COMPOSITE PRINTING PROCESS OF THE LIQUID METAL PRINTER

The current tapping mode enabled print mechanism is particularly developed to tackle the extremely high surface tension of the liquid metal ink (such as 0.624N/m for $GaIn_{24.5}$) [1, 2]. And our performed series of theoretical and experimental works have fully demonstrated the capability of this printing principle. Basically, the printing of the liquid metal ink is mainly determined by the rolling enabled ink adhesion, transfer and then tapping to press process. Under automatic control of the computer software, the printing bead was driven to roll smoothly on the surface of the substrate (Fig. S1A). The liquid metal ink pre-loaded inside the small tubule of the printing head is then uniformly delivered to the tip slot due to gravity effect and adhered to the surface of the roller-bead (Fig. S1B). Subsequently, it is then transferred and finally deposited on the surface of the substrate. The strong force from the upside down tapping motion of the printing-head and the rolling of the printing bead contribute to the extremely tight adhesion of the ink to the target substrate. All these processes are automatically controlled by the computer without needing any assistance from the users. It thus significantly simplifies the difficulty in electronics making and therefore paved the way for large scale applications of the liquid metal printer. The following sections is dedicated to systematically disclose the basic mechanisms of the roller-bead based high quality printing of liquid metal electronics to compose various desired circuits.

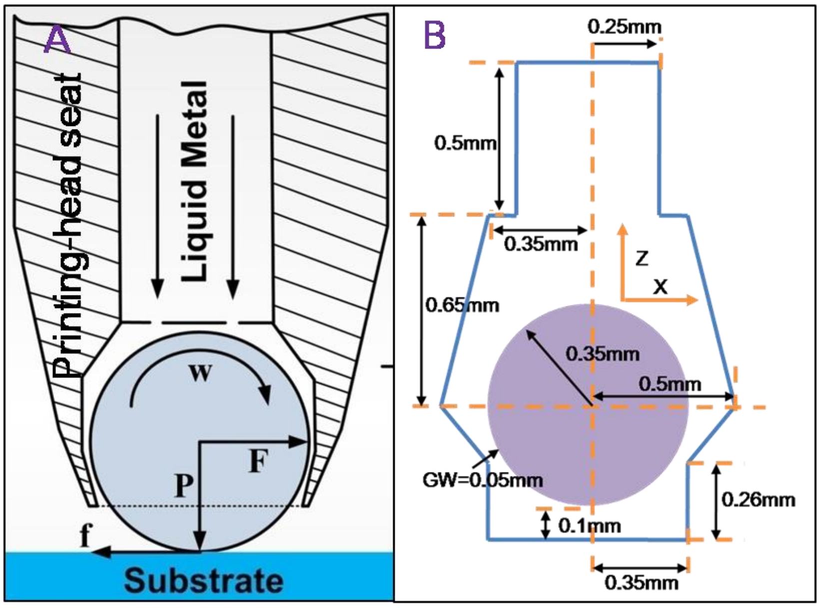

Figure S1. Schematic diagram of the roller-bead components to deliver and print liquid metal ink. (**A**) Illustration for liquid metal delivery through the small gap between roller-bead and its surrounding seat; (**B**) Flow domain of the computational model. (Jing Liu)

2. CONTACT MECHANICS BETWEEN ROLLER-BEAD AND ELASTIC SUBSTRATE

During the process for the printing bead to roll on the substrate, liquid metal is continuously transferred and deposited on the substrate surface. Our experiments indicate that a large sliding of the roller-bead would induce flow interruption of the liquid metal through the gap between roller-bead and its seat. Thus the contact mechanics between the roller-bead and the substrate is very important for guaranteeing the printing quality. Here we endeavored to clarify the mechanisms of the roller-bead sliding during the printing process.

Firstly, we consider a static rigid bead contacting with the elastic substrate surface (see insert illustration in Fig. S2) due to suffering from the external force *F*. A local coordinate for the contact surface position can be defined as $z = R^{-1}\left(x^2 + y^2\right) - h$, where *R* is the radius of the roller-bead, x and y are along the principal curvatures in the tangent plane (denoting here horizontal plane). When roller-bead approaches the substrate surface by a short distance *h*, the deformation displacement u_z of the substrate surface is given by

$$u_z\left(x, y\right) = R^{-1}\left(x^2 + y^2\right) - h \tag{S.1}$$

We denote by P_z the pressure at the contact region between the roller-bead and the deformed substrate. It is obvious that P_z =0 outside the contact region. P_z at the contact region is given by

$$\frac{1-\sigma^2}{\pi E}\int_S \frac{P_z\left(x, y\right)}{r} ds = h - R^{-1}\left(x^2 + y^2\right) \tag{S.2}$$

where, *E* denotes the Young's module and σ Poisson's ratios of the substrate. The solution of the above integral equation can be obtained through analogy with the potential theory [3]. Thus one has

$$P_z\left(x, y\right) = \frac{3\cos\left(\Phi\right)F}{2\pi a^2}\sqrt{1 - \frac{1}{R}\left(x^2 + y^2\right)} \tag{S.3}$$

where *F* is the amplitude of the external force, *Φ* the angle between the direction of *F* and z axis, *a* is the half-width of the indentation (shown in Fig. S2). Furthermore, we can derive the indentation width of the rigid bead and the radius of the contact region as follows:

$$\begin{cases} h = \left(\dfrac{3E\cos(\Phi)F}{4(1-\sigma^2)\sqrt{R}} \right)^{2/3} \\ a = \sqrt{Rh} \end{cases} \tag{S.4}$$

From Eq. (S4), it can be found that the contact area satisfies relation $A_c \sim \sqrt{F^3}$ ($\Phi \approx 0$ is set in our printing experiments), which indicates that the force F can be estimated from the contact surface measure. Large contact area A_c can promote the liquid metal to be easily deposited on the substrate when $\gamma_{sub-lm} < \gamma_{sub-sp}$, where γ_{sub-sp} is the surface tension of the liquid metal on roller-bead, and γ_{sub-lm} is its surface tension on substrate. In fact $\gamma_{sub-lm} < \gamma_{sub-sp}$ has to be enforced so as for the liquid metal to be easily dropped off from the roller-bead surface.

The tangential force for the static roller-bead is given as $\tau = \tan(\Phi) P_z$. No sliding exists for the static roller-bead when $\tau \le \mu P_z$, which means $\tan(\Phi) \le \mu$ (μ denoting friction coefficient). However, roller-bead experiences an elastic rotation (ω) in addition to the translational movement (V) during the printing process. A dimensionless quantity $s=1-R\omega/V$ is defined to characterize the sliding of the roller-bead. A detailed derivation of the expression s can be found in [4]:

$$s = \frac{-(4-3\sigma)}{4(1-\sigma)} \frac{\mu a}{R} \left[1 - \left(1 - \mu^{-1}\tan(\Theta)\right)^{1/3} \right] \tag{S.5}$$

We further define two dimensionless qualities as:

$$\begin{cases} Sr = -4(1-\sigma) sR / (4-3\sigma)\mu a \\ Fr = \mu^{-1}\tan(\Theta) \end{cases} \tag{S.6}$$

Then the relation between Sr and Fr based on Eq.(S5) and (S6) can be found in Fig. S2. It indicated that the external force plays an important key on the sliding of the roller-bead. In addition, the sliding ratio is also dependent on the contact area. From the above analytical interpretation, we have to control the sliding of the roller-bead, which leads to the instable shedding of the liquid metal from the roller-bead during printing process. It is noteworthy that increasing the contact area A_c helps liquid metal to well deposit on the substrate, which however also leads to the roller-sphere sliding. Thus, controlling force F is critical for printing the liquid metal ink, which can be fitly adjusted by monitoring the contact area A_c according to Eq. (S4).

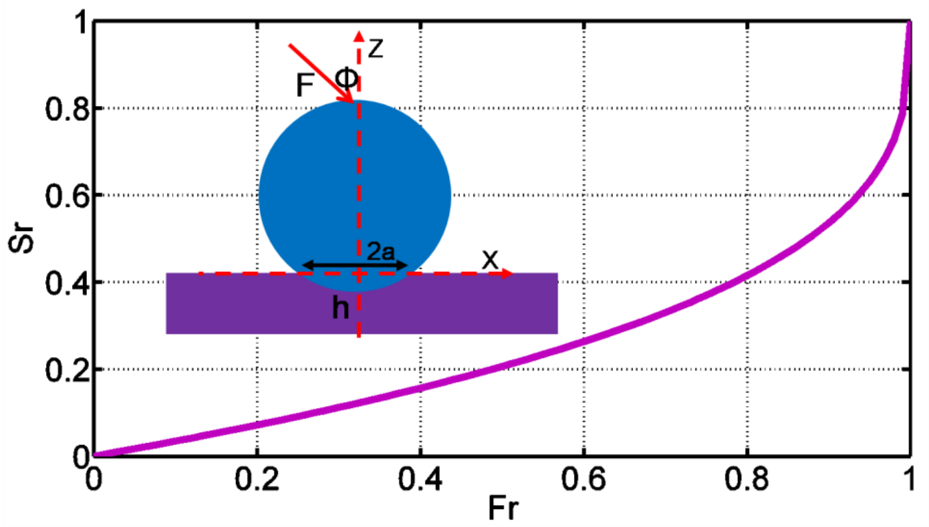

Figure S2. The relation between *Sr* and *Fr* (Jing Liu)

3. COMPOSITE FLUID DYNAMICS OF LIQUID METAL INK FOR RELIABLE PRINTING

The outflow of the liquid metal ink through the gap plays a key role in determining the final printing resolution and quality of the manufactured pattern. It is necessary to investigate the fluid dynamics of the liquid metal ink in the gap. A simplified geometrical model of the liquid metal flow is depicted as Fig. 1B. The liquid metal ink used here is $GaIn_{24.5}$ (its density: ρ=6.28×10^3 kg/m^3 and viscosity [1,2]: ν=2.7×10^{-7}m^2/s) which can be treated as Newton-fluid in small temperature variation case as studied in the present room temperature environment. The governing equation for the liquid metal ink adopts Navier-Stokes equation which reads as:

$$\begin{cases} \nabla \bullet \mathbf{u} = 0 \\ \left(\dfrac{\partial \mathbf{u}}{\partial t} + \mathbf{u} \bullet \nabla \mathbf{u} \right) = -\dfrac{\nabla p}{\rho} + \nu \nabla^2 \mathbf{u} + \mathbf{g} \end{cases} \tag{S.7}$$

where $\boldsymbol{g}$ denotes the gravity. The inlet and outlet of the computational domains are considered as gauge pressure boundary P_{in}=P and P_{out}=0, respectively. The reference pressure is set as the standard atmospheric pressure. The rotation velocity on the surface of the roller-bead is assumed as $\mathbf{V_R}=\boldsymbol{\omega}\times\boldsymbol{R}$. In order to characterize the translational motion of the printing head, the relative velocity $\boldsymbol{V}$ in x-y plane is given to the substrate surface. The boundary condition on the surface of the roller-bead seat is treated as non-slip. All simulations adopted Fluent 6.3 parallel model and run on the Dell PE2950 workstation with two quad-core CPUs (Intel Xeon x5365 @3.00Hz) and eight memories (1 GB each). In addition, we have performed mesh convergence tests to ensure adequate spatial resolution.

According to the theoretical prediction, the liquid metal ink can flow through the gap due to the pressure difference between inlet and outlet without roller-bead rolling (shown in Fig. S3A, B). When roller-bead rolls on the substrate, a composite flow state (Fig. S3C, D) of the liquid metal is initiated which is completely different from that driven by the single pressure. The flow velocity near the outlet of the gap is strongly enhanced by the rotation of the roller-bead, which promotes the liquid metal to be dropped off from the roller-bead surface. The fluid flux delivered to the substrate is determined by the flow state and the shortest width between the roller-bead and its seat (GW=0.05mm shown in Fig. S1B). In practice, the single pressure cannot drive the liquid metal continuously to flow through the gap due to pretty large surface tension of the liquid metal, which is hard to be considered in the simulation. Benefiting from the rotation of the roller-bead, the liquid metal is transferred rapidly to the substrate and ideally avoids the formation of large contacting surface and thus the high surface tension.

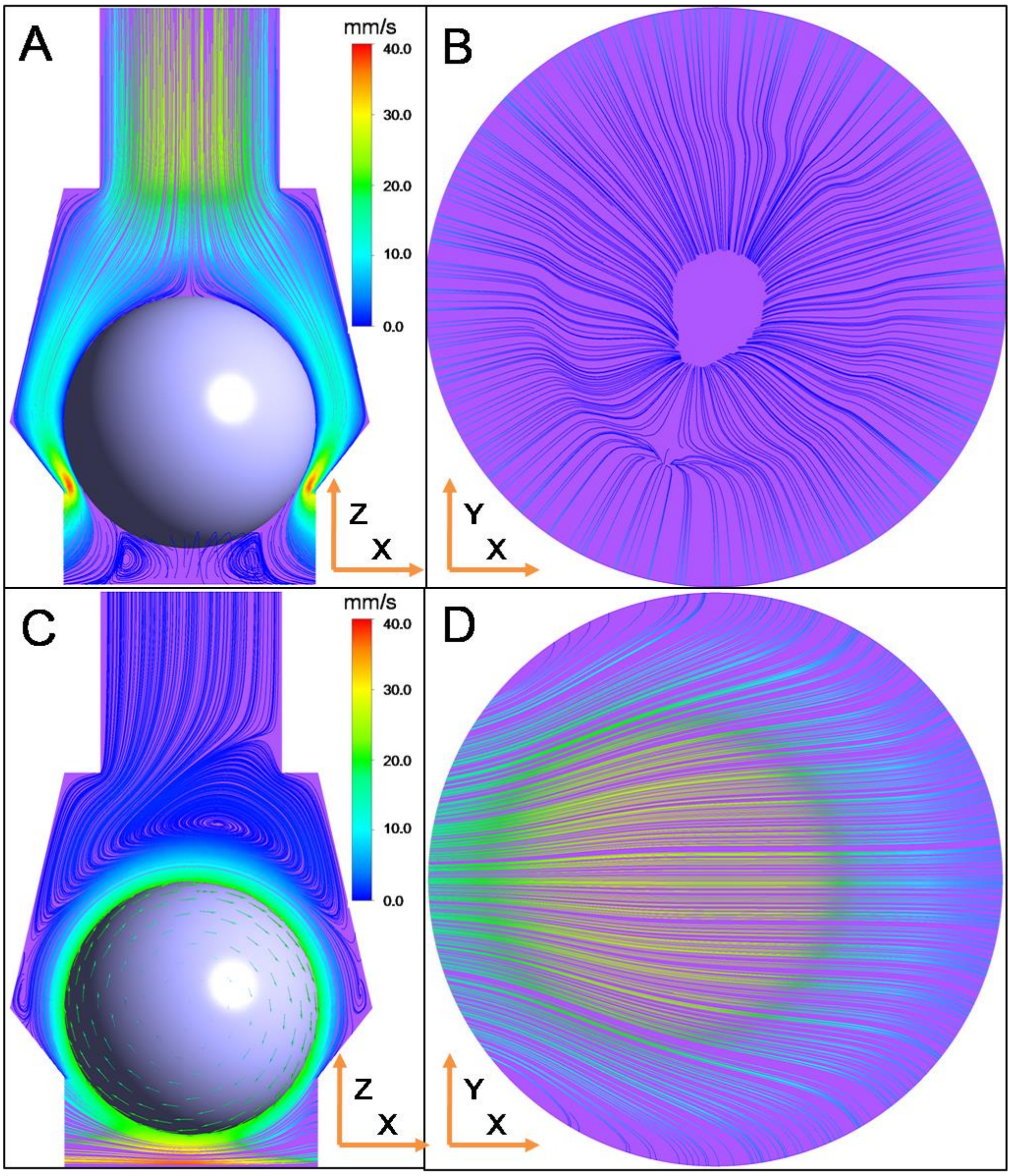

Figure S3. The flow streamline of the liquid metal ink in the gap. (**A**) 3D view and (**B**) 2D view at z=-1.5mm cross section with P_{in}= 1Pa, P_{out}=0, V=0 and ω=0; (**C**) 3D view and (**D**) 2D view at z=-1.5mm cross section with P_{in}=1, P_{out}=0, V=40mm/s and ω=70rad/s. (Jing Liu)

For printing a straight line, the rotation axis of the roller-bead is along y axis, which however may be frequently requested to change such initial direction for printing a nonlinear curve. This may lead to the discontinuous deposition of the liquid metal ink to compose target electronic-pattern. Here, an extreme case where roller-bead rotates the z axis and completely slides on the substrate, is particularly investigated, which may occur during turning around the printing head at the corner. The simulation results are shown in Fig. S4. Clearly, the rotation has no impact on the flow flux of the liquid metal delivering from the gap, which is determined by the pressure. This may make the printing at the turning point failed. Our experimental results also clearly disclosed such flow interruption of the liquid metal during turning at the corner, which was just induced by the rotation failure event.

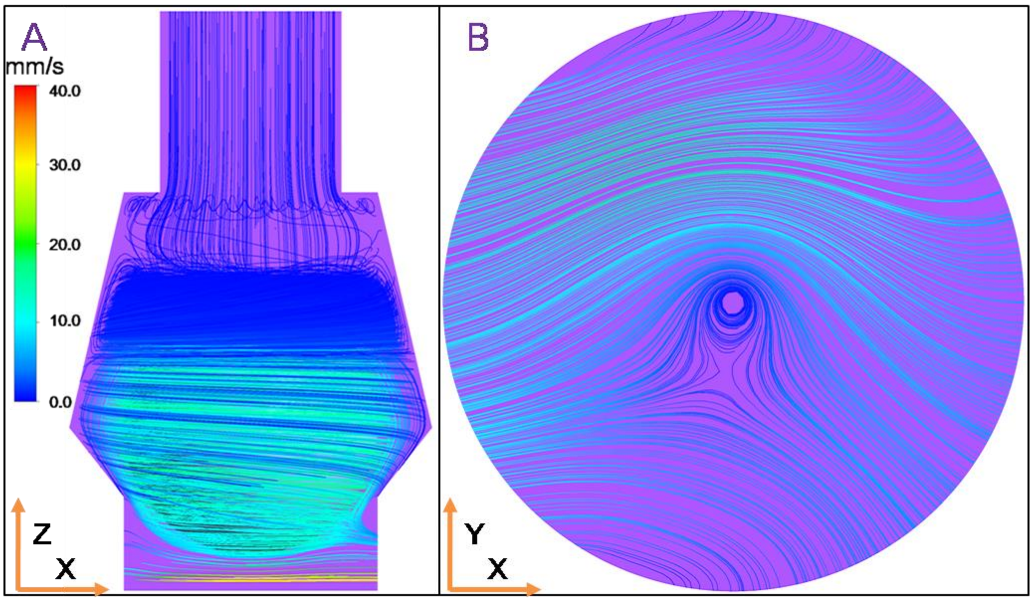

Figure S4. The flow streamline of the liquid metal ink in the gap. (**A**) 3D view and (**B**) 2D view at z=-1.5mm cross –section with P_{in}= 1Pa, P_{out}=0, V=40mm/s and ω=60rad/s. (Jing Liu)

4. ESTIMATING THE STABLE SHAPE OF LIQUID METAL LINE

After the liquid metal was deposited on the substrate through the roller-bead, the stability of the thin printed pattern is determined by the balance between the surface tension of the liquid metal and the surface energy of the substrate, which is often characterized by the contact angle of the liquid metal droplet on the substrate. The high contact angle (corresponding to less wetting) can easily lead to the instability of the printed pattern [5]. Here, we analyze the matching of the liquid metal surface tension with substrate surface energy to investigate the influencing factors of the unstable phenomenon of the printed pattern. The area of the line conductor cross-section can be approximated as $A=Q/V_s$, where Q is the flow flux of the liquid metal from the gap between roller-bead and its seat. The mechanical equilibrium between liquid metal and substrate can be described by Young's model, i.e.:

$$\gamma_{sub-air} = \gamma_{sub-lm} + \gamma_{lm-air} \cos(\theta) \tag{S.8}$$

where, θ is the contact angle of liquid metal with the substrate. According to thermodynamic equilibrium, we can derive the width of the stable line conductor L as

$$L = \frac{2\sin(\theta)}{\sqrt{\theta - \sin(\theta)\cos(\theta)}}\sqrt{\frac{Q}{V_s}} \tag{S.9}$$

This equation indicates that the width of line conductor is mainly determined by the contact angle, flow flux of liquid metal and translational speed of the printer, respectively.

For a typical printing process, where V=40mm/s, ω=60rad/s and P_{in}-P_{out}=1Pa, GW=0.05mm, the flow flux of liquid metal is about Q=0.0656mm3/s (from the above results of fluid dynamics simulation). For θ=40, the width of the stable liquid metal line is estimated about L=126μm based on Eq. (S9), which is consistent with the experimental results. Figure S5 presents the relation between the width of stable liquid metal line and the contact angle. It is noteworthy that the printed pattern would stay in liquid state at room temperature, which is easily affected by other factors, such as gravity and vibration. Thus packaging is very important for maintaining a stable electronic pattern for practical purpose.

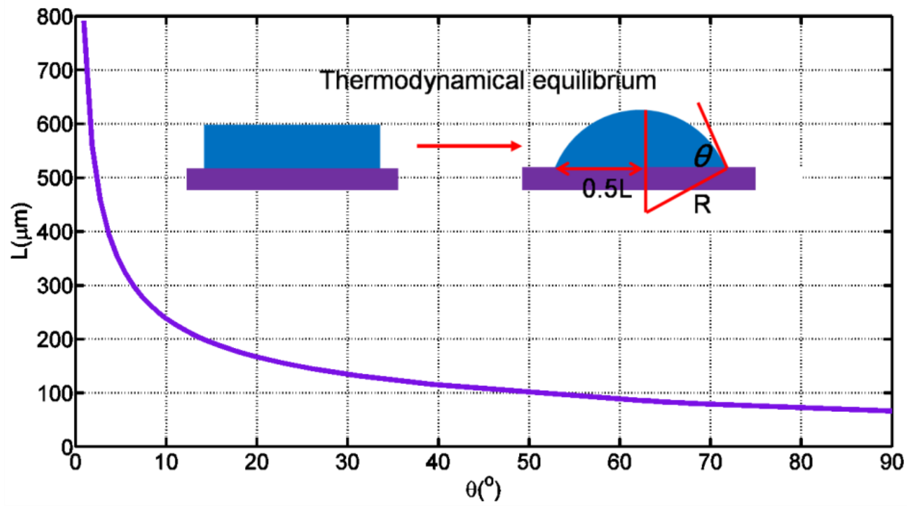

Figure S5. The relation between the width of stable liquid metal line and contact angle.

(Jing Liu)

5. DIVERSE CAPABILITIES OF THE LIQUID METAL PRINTER

To demonstrate the wide adaptability of the present composite ink printing method, a series of conceptual experiments were carried out to manufacture more representative electronic devices which all exhibit satisfactory outcomes on the well-matched substrate. It should be mentioned that all the original patterns of the currently printed electronics are either completely drawn or redesigned by the authors. Here, redesigning means that the basic elements of the printed patterns were partially derived from the public open webs for free-figures such as http://www.58pic.com or http://www.shutterstock.com, then synthesized, manually processed by using computer software, and finally transformed into vector graph by us. In addition, all these printed patterns, which have been modified with the size, shape and configurations, are printed out by the present liquid metal printer.

The present printed electronics way allows rather wide flexibility applications. In principle, various complex electrical patterns other than the pure line or curve can be easily printed out.

Figure S6 presents several regularly designed 3-dimensonal structures in effect. Here, due to dimension limitation of the current printer, only the projections (Fig. S6(A)) of 3-dimensional objects are printed. In the current manufacture, the printing head moves in X direction, while the substrate moves along Y direction. Therefore X-Y makes up Cartesian coordinate. The printer adopts various composite movements to draw specific target structures. Such printed patterns (Fig. S6(B)) further enrich the routing manufacturing styles.

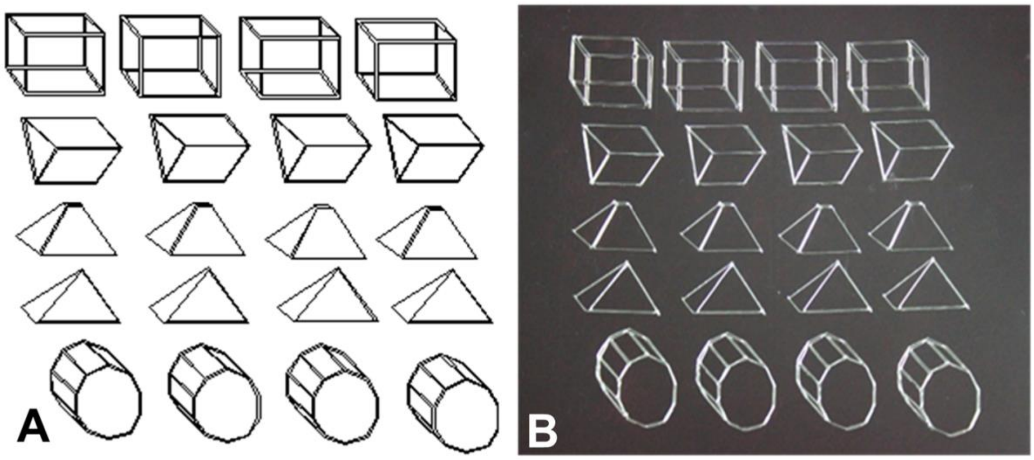

Figure S6. Regularly designed 3-dimensional structures. (**A**) Original structure and pattern in vector graph format; (**B**) Printed results according to the draft in (A). (Jing Liu)

6. PRINTING OF ELECTRONIC DECORATION ART

As shown in the above, the present liquid metal printer is capable of manufacturing various electrically conductive lines, curves or patterns which can then be used to compose more complex electrical targets. With such printer in hand, it would help incubate a group of interesting consumer electronics. One particular important area may come to the fabrication of electronic decoration drawings. To test the performance of the printer and its potential role for home, office or public use purpose, we choose to print a carefully designed decorative texture as Fig. S7A, B, respectively. Figure S8 presented more printed electronic decoration drawings. Various objects like eagle (Fig. S8A), flower (Fig. S8B) and dragon (Fig. S8C) etc. can be directly printed out in a few minutes. Clearly, with such complex metal pattern, some functional device such as LED as well as IC chip can be incorporated into the circuit. In this way, the IC chip can serve to control the lighting of LED according to the loaded program. Clearly, the combination of printable electronic and artistic field opens exciting new areas for the coming society. For brief, we will not present here the working of such functional electronics but leave the demonstration of the performance of the printed electronics to other circuits in later sections.

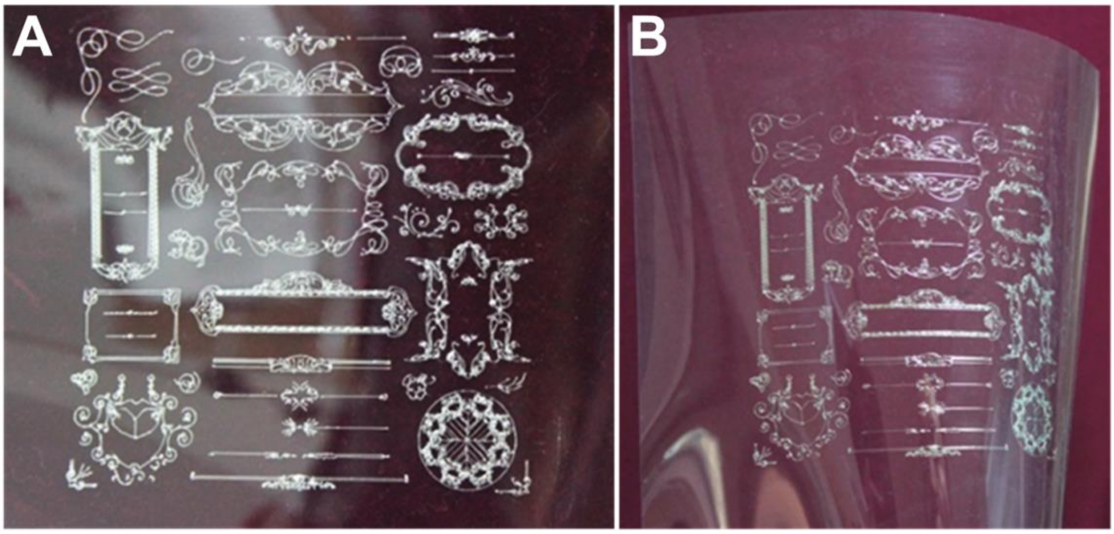

Figure S7. The electrical decorative textures directly printed out by the liquid metal printer. (**A**) Planar picture; (**B**) Folding state. Such electric drawing is rather appealing for pleasant home decoration since people may wish to design and print their own texture arts themselves. In a certain way, colorful LEDs can be connected with these instantly printed decorative pictures. In dark or weak light circumstance, these arts would look more beautiful when switching on the LEDs light, which makes it terrifically different from the traditional decorative arts. (Jing Liu)

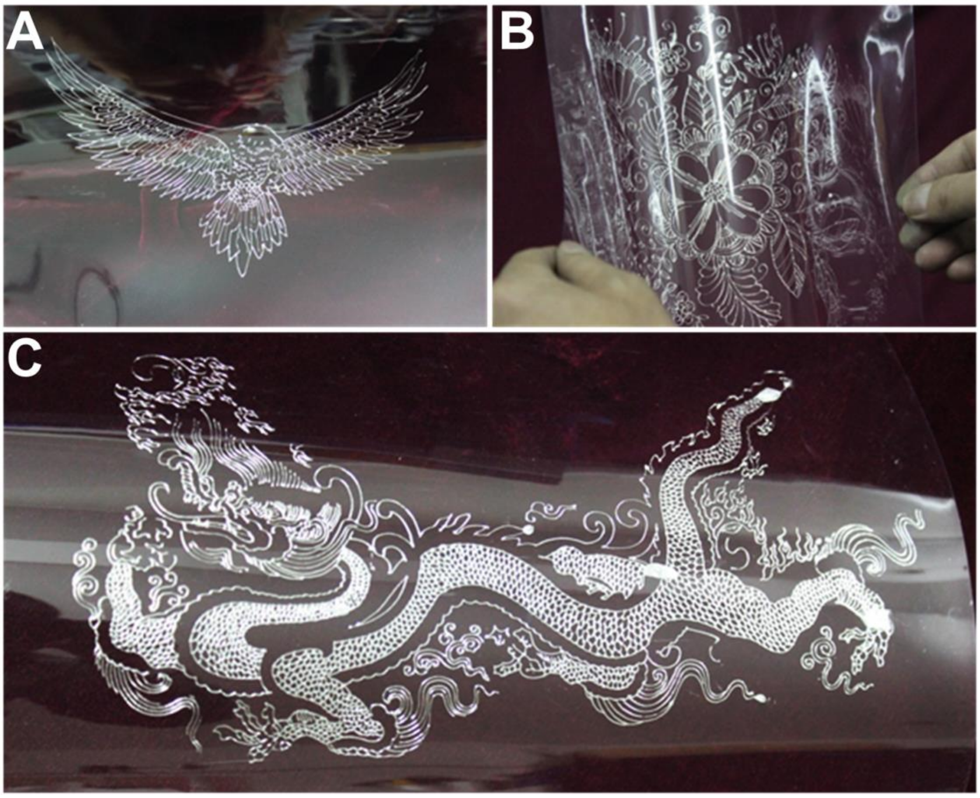

Figure S8. More printed electronically conductive decoration drawings. (**A**) Eagle; (**B**) Flower; and (**C**) Dragon. (Jing Liu)

7. PRINTING REGULARLY STRUCTURED PATTERN OF ELECTRICAL BUILDINGS

To move on, the present machine allows easily print out any desired electrically conductive building pictures. For example, Fig. S9 depicts one of such images of the electrical circuit to duplicate electrically a long historical Chinese building: The Temple of Heaven (Fig. S9A) as well as a famous US building: The White House (Fig. S9B). With electrical functions like controlled lighting in the drawing, the pattern could offer more electronic functions of the design. This electronics manufacture strategy would help incubate the new area of vivid electronic architecture design. Meanwhile, it is also important for future educational training purpose.

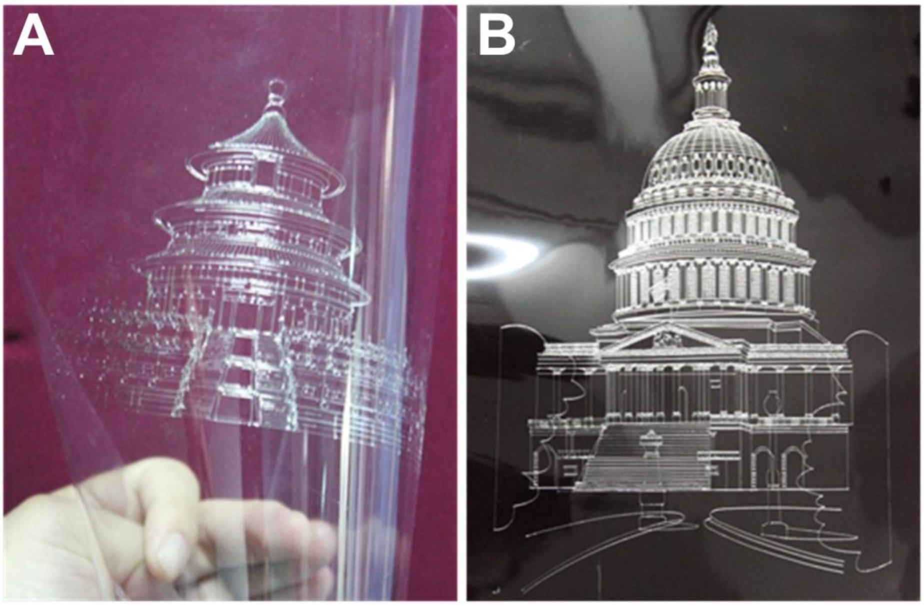

Figure S9. Printed electronic building pattern. (**A**) This printed building is called the Hall of Prayer for Good Harvests settling in Temple of Heaven as a principal building in Beijing, China. (**B**) Printed building of the White House, Washington D. C., USA. (Jing Liu)

8. PRINTING COMPLEX STRUCTURE AND PATTERN OF CLASSICAL PICTURE

With the big capability offered by the present technology, we were motived to print more complex electronic structures. One example is to print electronic picture of a famous Chinese painting: Along the River during the Qingming Festival. A total of the original picture is rather large and not necessary to cover. For illustration purpose, we chose to print only part of this drawing (see original in Fig. S10A). Before printing, it should be converted into a printable vector graph as shown in Fig. S10B. Then the final target pattern can be printed out. A result of such electronic picture is presented in Fig. S10C. For more illustration, what presented in Fig. S10D is another printed electronics pattern for the famous building: The Great Wall. Clearly, with electrical elements inside the picture, a vivid presentation of ancient arts can thus be obtained. This bridges well the arts and science which may shed light for future combinational research.

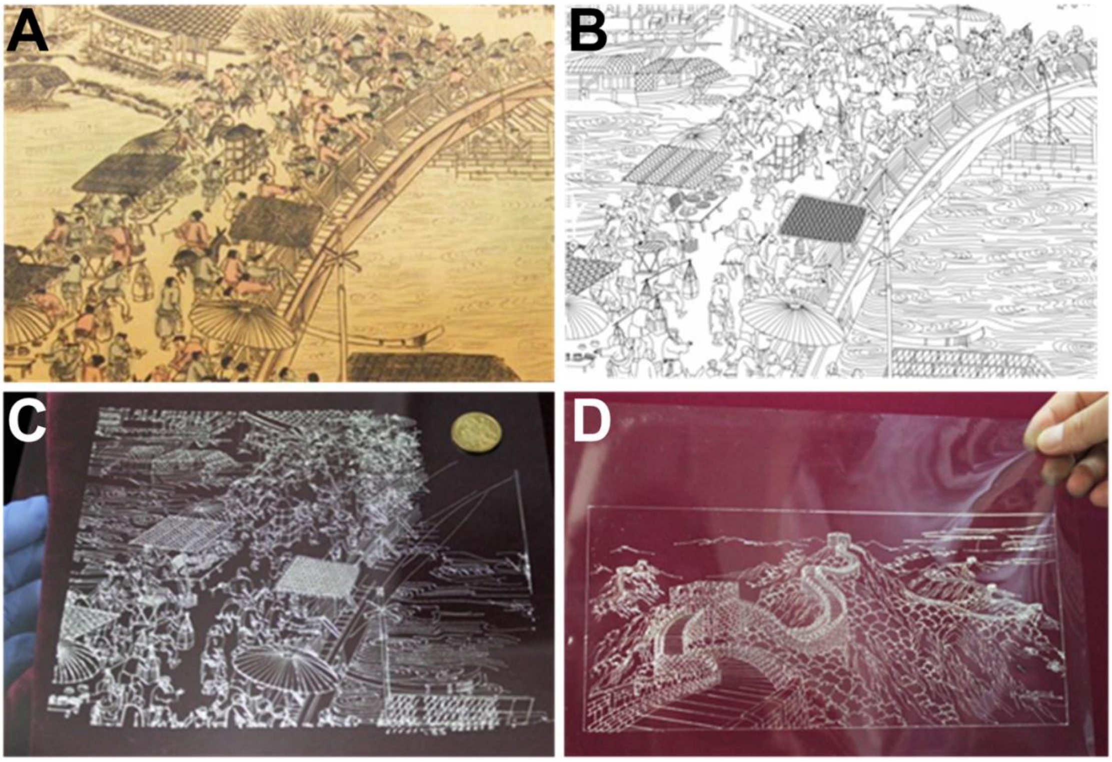

Figure S10. The printed electronics of Chinese famous drawing. (**A**) Original picture for Along the River During the Qingming Festival, which is one of the most famous ancient China painting created by Zeduan Zhang (a painter at China Song Dynasty) at around the year of 1205. (**B**) Vector graph for the original picture; (**C**) Printed metal drawing for Along the River During the Qingming Festival. (**D**) Printed metal drawing for China Great Wall. (Jing Liu)

9. PRINTING OF CONDUCTIVE HUMAN PORTRAITS

Figure S11 illustrates the printed portraits of four scientists which are physicists Issac Newton (Fig. S11A), Albert Einstein (Fig. S11B), and two China scientists Xuan Wang (Fig. S11C) (inventor of Chinese Characters Phototypesetting which significantly innovate the printing technology) and Sheng Bi (Fig. S11D) (inventor of ancient China typograph).

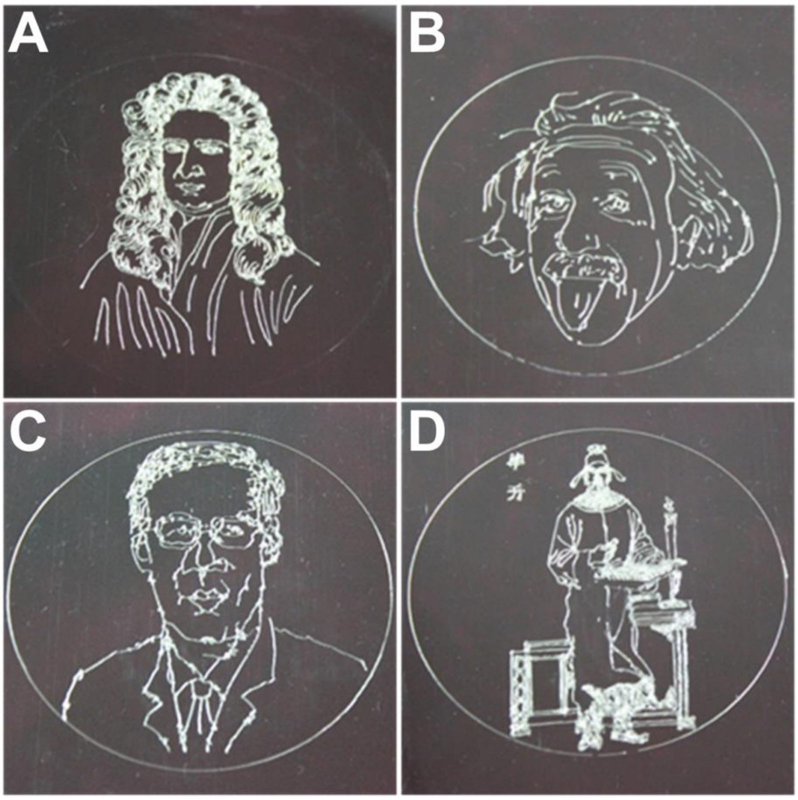

Figure S11. Printed electronic human portrait. (**A**) is a portrait of the great British scientist Isaac Newton. (**B**) is a portrait of Albert Einstein, who is one of the greatest scientists in 20th century. (**C**) is a portrait of Chinese scientist Xuan Wang, who is honored as the Father of Chinese Language Laser Typesetting. (**D**) is a portrait of an ancient scientist Sheng Bi in Song Dynasty China, who invented the world first known movable type printing press technology. (Jing Liu)

10. PRINTING OF PCB BOARD ON FLEXIBLE PLASTICS

The most core function of the present method is to freely manufacture a printed-circuit-on-board (PCB) in a short moment as desired. Figure S12 presented the design draft (Fig. S12A, D), printing process (Fig. S12B) and printed results of several PCB circuits (Fig. S12C, E), respectively. More discussion on such electronics can be found in the article. Overall, the present printed circuits are rather suitable for surface mounting electronic components. This significantly innovates the conventional PCB manufacturing technology.

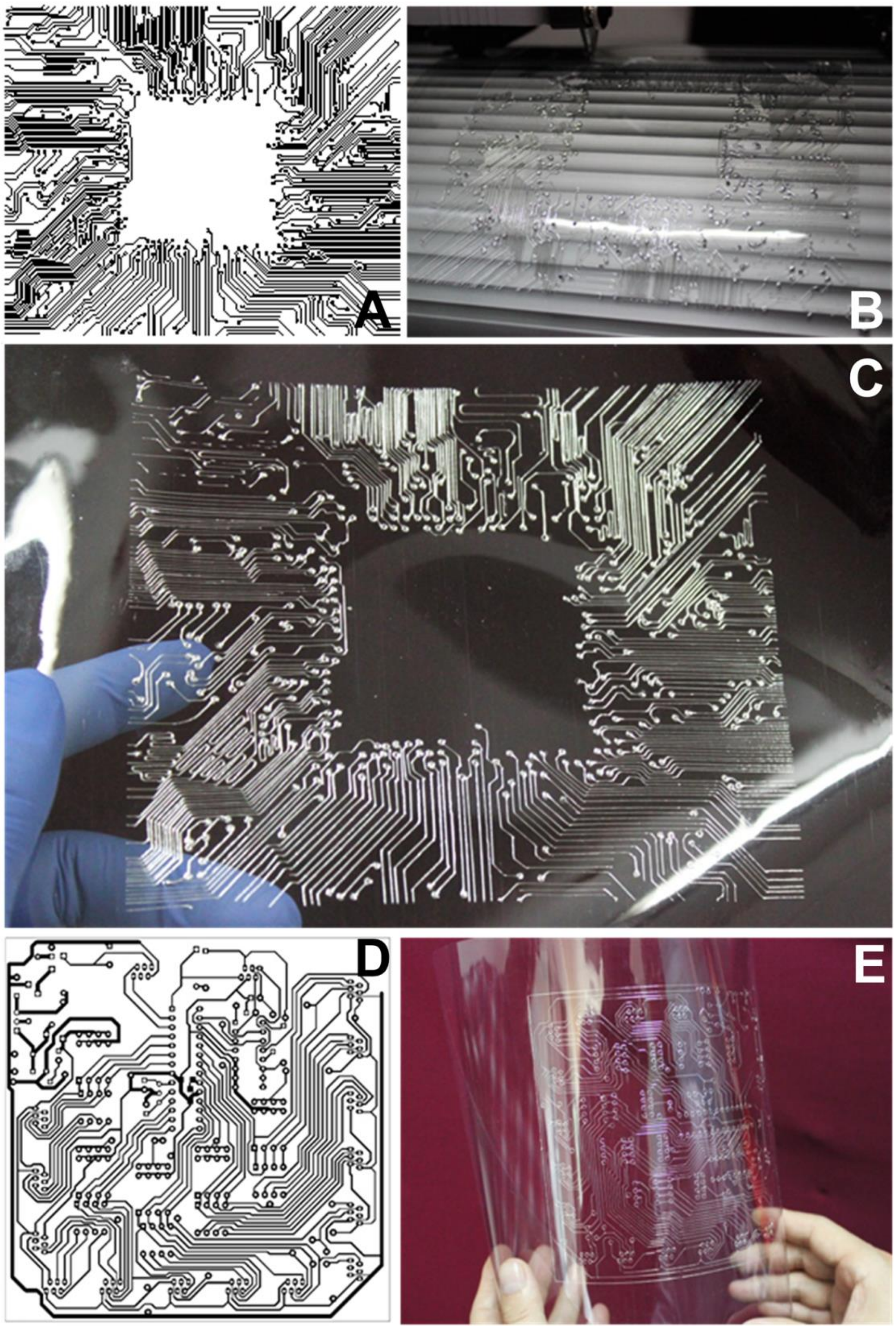

Figure S12. Printed PCB in a few minutes using the liquid metal printer. (**A**) Original PCB design draft; (**B**) PCB in printing on the machine; (**C**) Manufactured results of (A); (**D**) Additional PCB draft; (**E**) Printed PCB of (D). (Jing Liu)

11. DESIGN AND MANUFACTURE ELECTRONICS AS DESIRED

So far, there is a huge demand to develop personal electronics manufacture way which was even attributed as one important driving force for the coming "Third Industrial Revolution". The biggest barrier to impede the large scale practices of such endeavor lies in the serious shortage of a cost effective machine itself and the rather limited capability of the ink. The invention of the present liquid metal printer makes the dream towards electronics Do-It-Yourself a reality. What presented in Fig. S13 are just two of such typical working examples: electronic greeting card (Fig. S13A) and electronic decoration picture of the Great Wall (Fig. S13B). With lighting on, such drawings display more human's imaginations. Clearly, additional future practices along this direction are expected to stimulate increasing roles in a wide variety of coming consumer oriented electronics.

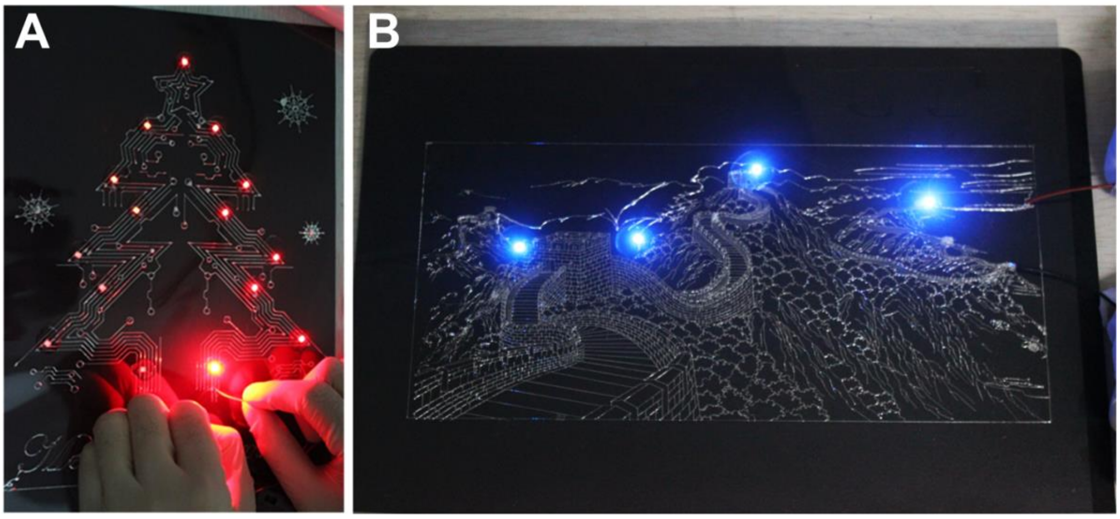

Figure S13. Printed personal consumer electronics. (**A**) Electronic greeting card with Christmas tree lighting on; (**B**) Electronic picture of China Great Wall with LED lighting on. (Jing Liu)

12. PACKAGE OF PRINTEED ELECTRONCS

If one wishes to completely ensure the environmental and mechanical stability of the printed electronic circuits, structures and patterns, a few commonly available materials such as PDMS or room temperature vulcanization (RTV) silicone rubber can be adopted to package the targets. Presented in Fig. 14 A and B are several such well packaged structures via RTV. The whole process in packaging each item just takes several minutes.

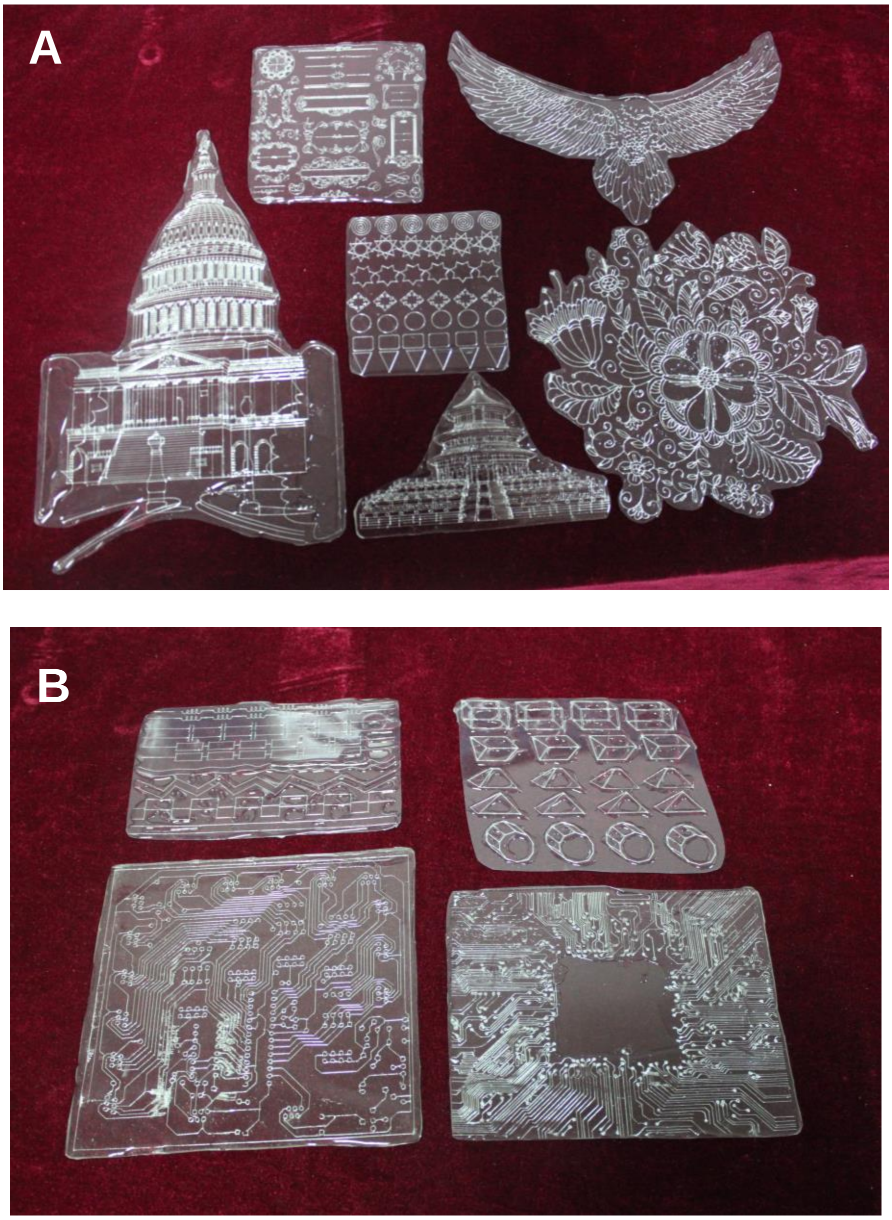

Figure S14. Packaged printed electronic patterns. (**A**) Printed electronic decoration arts. (**B**) Printed PCB or electronic elements. (Jing Liu)

In summary, we have demonstrated the diverse capability of the liquid metal printer in manufacturing various conductive structures and patterns spanning from a single line, curve to any desired complex paintings. This opens the way to directly and quickly fabricate flexible electronics as could as one can image. The total process looks just like printing a picture on the paper as it was often done in an office. The unique merit of the liquid metal printing technology lies in its entirely automatic controllability, extremely low cost, direct printing feature, and excellent adaptability. As is well known, conventional approaches of making a flexible circuit are generally complex, environment unfriendly, time and energy consuming, and thus expensive and hard to access by an ordinary user. With easy going feature, high resolution manufacture and extremely low cost merit, the present electronics printer makes the goal of printing personal electronics at home a reality. It is expected to be widely used over the world in the coming time.

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