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# Nimble native oxides: Printing circuits from the skin of liquid metal

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**Stretchable materials, such as liquid metals, promise to transform microelectronics hardware into soft, wearable devices. However, a recent report by Kong et al. shows that liquid metals are much more than stretchable wires; their metal-air interfaces offer surprising possibilities for synthesis and deposition of multifunctional two-dimensional (2D) transparent oxide nanomaterials.**

Stretchable electronic materials have promised to fundamentally change the paradigm of microelectronics hardware by making devices soft, wearable, and lightweight. One can imagine virtual reality systems and biomedical devices that seamlessly assist us in our daily lives. To realize this future of soft electronics, liquid metals melting at or near room temperature have become leading candidate materials. Recently, it has become clear that liquid metals are much more than stretchable wires, and that their interfaces offer surprising possibilities for synthesis and deposition of a wide range of multifunctional two-dimensional nanomaterials.

A recent report in Science[1] by Kong et al. entitled "Ambient printing of native oxides for ultrathin transparent flexible circuit boards," resolves two threads that run deep through the field of liquid metals and flexible electronics. The ultrathin (<4 nm) native oxide of liquid metals is known to be responsible for many of their fascinating properties, from their ability to assemble 3D structures[2] to their controllable electrochemical actuation and their ability to form stable nanoparticulate phases and colloids. Understanding the native oxide of eutectic Ga-In has been critical for overcoming challenges researchers face in printing stretchable circuits. Liquid metals' ability to wet surfaces, which is critical for printing liquid metals in high resolution patterns, depends on its ability to adhere its own native oxide to a surface. Without this native oxide, liquid metals remain spherical beads governed by their colossal surface tension (625 mN/m). The dynamics of liquid metal droplets also hinge on the motion of their contact lines,[3] demonstrating a large contact angle hysteresis associated with whether they adhere and leave their surface oxides behind. However, in the present paper by Kong et al., this residual oxide film, previously a nuisance, is the prize at the end of the meniscus.

Kong et al. control this phenomenon of liquid metal dewetting to print the native surface oxide in nanoscale (3–5 nm) thick two-dimensional layers. Their new mode of printing (Figures 1A and 1B) leverages a liquid-metal-filled slot to confine and control the translation of the liquid metal meniscus on a range of different surfaces ($SiO_2$, glass, PDMS, polyimide). They discover the regime of speeds and heights producing a uniformly receding meniscus, leaving behind an apparently perfect film of the surface oxide ($GaO_x$, $InO_x$, and $AlO_x$), as shown in Figure 1C. Their hypothesis reinforced by videos of dye-enhanced imaging is that the receding meniscus can "zipper" up, reclaiming residual liquid metal while leaving not one, but two continuous metal oxide films sandwiched together.

These printed 2D oxide films have surprising physical properties, a remarkable conductivity for $GaO_x$ despite its amorphous phase as well as the ability to strongly adhere nanoscale films of Au at thicknesses well below the traditional limits where Au films would remain discontinuous. These Au-decorated 2D oxide nanosheets are highly transparent and conductive while exhibiting high tolerance for bending as well as impressive adhesion, a critical feature for device durability in applications to flexible electronics. Using

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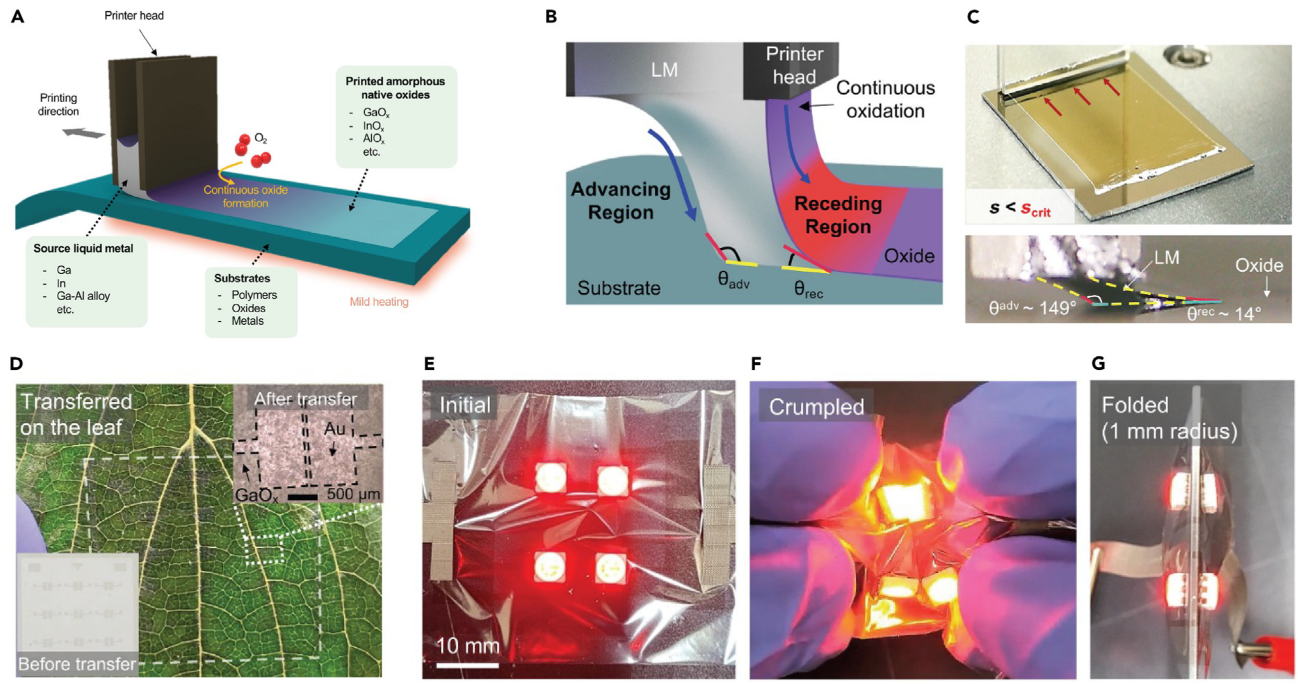


**Figure 1. Native oxide liquid metal printing and foldable transparent circuits**

(A) Schematic image of printing setup illustrating the continuous oxide printing process.

(B) Schematic of the dewetting-induced oxide printing method. An oxide-covered meniscus of liquid metal spans the gap between the printer head and substrate. The oxide at the leading edge continuously deposits onto the substrate.

(C) Photograph of oxide printing using dewetting liquid metal meniscus.

(D) Digital image showing the ultrathin transparent circuit line transferred to a leaf. Inset images show the circuit lines before (bottom left) and after (magnified, top right) the transfer.

(E-G) Digital images of LED lighting as a demonstration for initial, crumpled, and 1-mm-radius folded states. (Figures 1A–1G adapted with permission from[1]).

conductive grids formed by their Au/$GaO_x$ patterned by photolithography (Figure 1D), Kong et al. integrate LEDs into flexible and foldable circuits (Figures 1E–1G) and capture the unique ability of these materials to replace bulky conventional circuit boards with a thin, lightweight, and highly transparent alternative.

Kong's work addresses a central challenge in the field of 2D oxides derived from the skin of liquid metals. Since the demonstration of the van der Waals assisted transfer of two-dimensional oxides by Zavabeti et al. in 2017,[4] this method has been expanded across energy materials, sensors, and semiconductors,[5,6] generating a library of powerful functional materials. The unique features are its low temperature, thermodynamically favorable process of oxidation. The innovation by Kong et al. is in maintaining the uniformly shaped liquid metal meniscus and exfoliating the delicate 2D surface oxide in a controlled and large area fashion, which is difficult to accomplish while directly transferring 2D oxides through direct contact printing by rollers.[7]

What is clear from Kong's work and related work in the field is that liquid metals have room to grow as platform for synthesizing electronic materials. The technological and scientific impact of 2D oxides will depend on understanding of the physics of liquid metal's interfaces, e.g., what are the fundamental mechanisms by which we can control the kinetics and the chemistry of these reactions? The first opportunity here lies in the ability to manipulate the properties of the 2D liquid metal skin at the sub-nm scale. This level of electrostatic control could be critical to engineer advanced semiconductor devices composed of liquid metal derived 2D electrodes, semiconductors, and even dielectrics. Two other promising directions include transforming 2D oxides into high performance optoelectronic material systems, such as metal nitrides,[8] or using liquid metal interfaces to directly grow 2D transition metal dichalcogenide (TMDs) crystals.[9]

Future applications of flexible circuit technology could see liquid metal serving as both a literal and figurative bridge between hard and soft materials. For next generation wearable displays and imaging systems, for example, we might expect to see stretchable liquid metal interconnects as well as strain-tolerant 2D transparent conductors like those printed from liquid metals.[10] However, given how long 2D oxides have hidden in plain sight, we may have just scratched the surface of these native oxides' potential for fabricating transparent electronics with far greater capabilities.

## DECLARATION OF INTERESTS

The author declares no competing interests.

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