Room-Temperature Printing of Ultrathin Quasi-2D GaN Semiconductor via Liquid Metal Gallium Surface Confined Nitridation Reaction

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1. Introduction

Semiconductor technology has been one of the most fundamental cores in developing the integrated circuit industry. Since witness of the bottleneck of the first two generations of semiconductors, the third-generation high-temperature wide-bandgap semiconductor nanomaterials such as gallium nitride (GaN),[1] zinc oxide (ZnO), aluminum nitride (AlN),[2] silicon carbide (SiC),[3,4] diamond, etc., have become a focus in recent years. GaN nanomaterials are novel semiconductors owning many outstanding merits like high saturated electron mobility, radiation resistance, acid and alkali corrosion resistance, large thermal conductivity and strong breakdown field. In the GaN electronic devices, the inevitable electron transfer from the valence band to the conduction band can be suppressed through the wide energy bandgap. Accordingly, the devices can retain their electrical features in various scenarios. For such reason, the GaN was regarded as one of the most attractive semiconductor materials due to its idealistic efficiency and stability.[6,7] Tremendous efforts have therefore been devoted to developing and analyzing the GaN materials throughout the world.[8–10]

Compared with the bulk GaN materials, various nanoscale effects of the low-dimensional GaN materials can display better photoelectric, mechanical, thermal stability, as well as electrical[11,12] and chemical features.[13] Apart from the fundamental physicochemical features of GaN, they also own the surface, small-size, and quantum-confinement impacts as one of the most exciting areas for future growth of microelectronic devices. However, until now it is still not easy to construct a low-dimensional GaN. Only rather limited outstanding works are available. Syed et al. proposed a two-step process to synthesis 2D GaN nanosheets, the process includes obtaining 2D Ga2O3 by extrusion printing, and then converting Ga2O3 to GaN through ammonolysis in a tubular furnace.[14] Chen et al. reported the development of 2D GaN single crystals (~4.1 nm) attained using a surface-confined nitridation reaction (SCNR) through...
the chemical vapor deposition (CVD). In 2016, Balushi et al. employed the migration-enhanced encapsulated growth method to construct 2D GaN monolayer nanosheets. Unfortunately, the temperatures used in most of the processes are above 500 °C that is inconsistent with various electronic industry operations. This is because the long run time of the deposition process can increase the cost and feasibility. Clearly, the construction and research technology of low-dimensional GaN materials and devices should be developed and enhanced to satisfy the appeals from so many practical needs. In this side, a low temperature preparation of large-area, high-quality, and uniform GaN films will generate a significant impact on the high thermal stability 2D integrated circuit industry designed for power electronics. However, so far, there is still no report on the room temperature preparation of ultrathin or 2D GaN films.

Herein, we proposed and demonstrated for the first time to print the ultrathin quasi-2D GaN films on SiO2/Si substrates through introducing liquid metal-based synthesis and printing processes at room temperature. The basic principle relies on using nitrogen plasma to trigger nitriding of gallium droplets at room temperature, and then transferring them to the substrate through the proposed van der Waals (vdW) printing technology. The whole preparation process is rather easy going which is completely performed at room temperature and is consistent with the current electronics manufacturing processes. The proposed method can produce nanometer scaled thickness GaN film attained via individual or multiple prints. Moreover, we report excellent an electronic performance of the printed ultrathin quasi-2D GaN. For an illustration purpose, fully printed side-gated field-effect transistors (FETs) are fabricated, from which the quasi-2D GaN-FETs exhibit outstanding electronic performance with a large current on/off ratio (>105), high field-effect mobility (∼53 cm2 V−1 s−1), and tiny subthreshold slopes (∼98 mV dec−1) with a high degree of reproducibility. This study suggests a reliable and straightforward room temperature large-scale manufacturing technique for printing quasi-2D GaN with outstanding working features, which opens a significantly practical potential for wafer-scale processes. It also paves the way for the application of GaN semiconductor in a new generation of all printed electronic devices, integrated circuits, and more functional devices.

2. Results and Discussion

2.1. Liquid Metal-Based Ultrathin Quasi-2D GaN Printing Technology

Conventionally, quite a few processes have been available to prepare GaN films, such as metal–organic chemical vapor deposition (MOCVD) and atomic layer deposition (ALD), which are illustrated in Figures 1a and 1b. The high preparation temperature (>250 °C) and toxic material [Ga(CH3)3]4 are the main disadvantages of these processes to produce GaN at a large industrial scale. Recently, room temperature N2 plasma treatment as an emerging tool for green nitrogen fixation and surface modification of materials, have been tried in the field of ammonia synthesis, N doped in 2D materials plasma enhanced chemical vapor deposition (CVD) depositing thin film, nitride layer formation on rigid materials, etc. The N2 plasma technology owns merits of low cost, high efficiency, and pollution-free on nitriding treatment of material surfaces. With this in mind, here a simple and convenient process of GaN thin film prepared through N2 plasma irradiating clean liquid Ga surface is proposed.

The scheme in Figure 1c illustrates the mechanism of our introduced process of GaN formation. Such a plasma nitriding process employs an unusual glow discharge involving high current and charge densities. A potential difference is generated by applying a DC voltage between the discharger (anode) and the liquid Ga (cathode), which ionizes the nitrogen to produce the glow discharge. Generally, atomic nitrogen (Natom), excited nitrogen molecules (N2*), activated nitrogen molecule ions (N2+) and electrons (e−) are main contents of nitrogen plasma gas. In the plasma nitriding of gallium, the accelerated N2 atom, N2*, and N2+ can all hit the surface of the liquid Ga with high kinetic energy and react directly with Ga atom to form GaN molecule. In a word, we first define producing GaN from intimate reaction of nitrogen and liquid Ga in plasma as: N2 + 2Ga −→ Natom + N2* + N2+ + e−. Because the chemical reaction of nitrogen plasma is a thermodynamically stable excited state and ionic state, and the reaction activation energy is pretty low, it is much easier to generate GaN by the reaction of nitrogen plasma with liquid Ga than that through the traditional nitriding reaction (MOCVD) Ga([CH3]4) + NH3 −→ GaN + 3CH4.

The schematic in Figure 2a illustrates the preparation of ultrathin quasi-2D GaN on the surface of liquid Ga droplet using N2-plasma treatment technology at room temperature. The changes of fresh Ga droplet before and after nitrogen plasma treatment are shown in Figure S1, Supporting Information. The schematic setup of the system to transform the liquid Ga to GaN is elucidated in Figure 2b. The formation of GaN thin films was realized by nitrogen plasma triggering surface limited nitriding reaction. Gallium droplets with plasma treated surface were placed on SiO2/Si substrate and moved on the surface with the help of a scraper. During the scraping process of liquid Ga the surface nitride produced by plasma bombardment in nitrogen environment sticks to the substrate through vdW force and the clean Ga is exposed (Figure 2c) (detailed description of the process parameters will be given in the Experimental Section.). The nitridation reaction and printing of Ga both are conducted at room temperature that is consistent with the current electronic device production processes. The covered substrate area can be expanded by choosing a larger droplet diameter and a longer travel distance of the scraper. The thickness of the film can be increased via expanding the nitrogen plasma trigger time or frequent printing (Experimental Section and Figure S2, Supporting Information). Figure 2d shows a one touch print of the GaN film obtained from the surface of Ga droplets after being treated for 10 min by nitrogen plasma. It revealed a large and continuous ultrathin GaN film reaching lateral dimensions of more than many centimeters. Based on the atomic force microscopy (AFM), the thickness of the deposited GaN layer is measured as ≈4 nm that is moderately larger than a single GaN unit cell (Figure 2e). The printable GaN is slightly thicker than the single layer, indicating that the prepared GaN film is an ultrathin quasi-2D film. AFM also
indicates that the printed GaN film’s surface roughness is analogous to that of the SiO2 substrate, demonstrating that the GaN film has minimum cracks, holes, folds, or bubbles, reflecting conformal and homogeneous attachment.

It is noteworthy that the thickness of singly printed GaN film is below 1 nm (Figures S2 and S3a, Supporting Information), when the nitrogen plasma trigger time is lower than 1 min. The harvested GaN film can be classified as 2D materials. However, cracks and holes can also be seen on obtained 2D GaN. Actually, the uniformity and continuity of printed GaN films are not always good enough, when the nitrogen plasma trigger time is lower than 5 min. It might be because the insufficient nitrogen plasma treatment is not favorable for the perfect growth of GaN films on the surface of Ga droplets. Therefore, the 4 nm GaN films obtained after 10 min of treatment is employed in subsequent microstructure observation, optical analysis, electronic devices fabrication, etc.

The uniform thickness in broad regions and numerous samples supports the proposed Cabrera–Mott growth mechanism, where concurrent nitrides are formed among the whole metal interface, resulting in self-limiting growth to an accurate thickness. The mentioned approach is highly reproducible for

Figure 1. The schematic diagram of three GaN growth processes and temperatures. a) Illustration of MOCVD locus to fabricate GaN, at temperature above 950 °C. b) Illustration of ALD locus to prepare GaN, at temperature above 250 °C. c) Illustration of nitrogen plasma-liquid Ga reaction locus to harvest GaN, at room temperature.
growing large-area GaN sheets because the process was reproduced more than 50 times and always yielded identical, continuous, and laterally extensive thin GaN films with reproducible features. The GaN film was initially printed on the SiO$_2$/Si substrate, but further tests demonstrated that the formation of uniform centimeter-scale semiconductor films on various
substrates could be reproduced through the printing technology (Figure S3, Supporting Information), indicating that the presented construction approach is suitable to deposit GaN on several materials. Moreover, it should be pointed out that this method is also applicable to the fabrication of GaN heterostructures. In order to be compatible with silicon-based electronic technology, the ultrathin quasi-2D film printed on SiO$_2$/Si surface should be employed for further description and device construction.

It should be noted that, Figure S4, Supporting Information, presents the comparison of the temperatures and the thicknesses of GaN of our present printing technique with other classical techniques in the field of the preparation of ultrathin GaN in previous studies. Various processes have been introduced to the growth of GaN, such as MOCVD, metal–organic vapor-phase epitaxy (MOVPE), hydride vapor phase epitaxy (HVPE), atomic layer deposition (ALD), plasma-enhanced atomic layer deposition (PEALD), microwave plasma-assisted atomic layer deposition (MPALD), plasma-assisted atomic layer deposition (PA-ALD), plasma-assisted molecular beam epitaxy (PAMBE), laser molecular beam epitaxy (LMBE), flow modulation epitaxy (FME), pulsed direct current (DC) sputtering, RF magnetron sputtered, etc. The above processes basically require high temperature and vacuum conditions, which virtually increases the cost, and is not conducive to large-scale pervasive application, nor can it easily realize the fabrication of flexible devices, which seriously hinders the wide practices and research progress of the ultrathin GaN in flexible devices. Compared with the above technology, the room temperature GaN printing process based on liquid metal owns the advantages of simple and stable process, fast, large area, low cost, high efficiency, easy removal of excess metal, and the final sample surface is extremely clean.

2.2. Properties of the Ultrathin Quasi-2D GaN

Transmission electron microscopy (TEM) can be utilized to characterize the crystallographic features of the printed GaN. The GaN films were immediately moved to a TEM grid since it was printed. A high-resolution TEM micrograph (HRTEM) and selective area electron diffraction (SAED) pattern of the GaN are presented in Figures 3a and 3b, respectively, which confirm the crystallization of the printed GaN in the polymorph. The atomic spacing of 0.285 nm in the HRTEM image and the distance of the fringes at 0.248 nm correspond to the (100) and (101) planes of GaN. The collected lattice parameters show that the GaN sample is wurtzite structural.

The GaN film’s phonon modes also change compared to those of the bulk.$^{[20]}$ Based on the Raman spectra presented in Figure 3c, two peaks at 561.77 and 729.59 cm$^{-1}$ appear in printed GaN, respectively, corresponding to characteristic E$_2$ (high) located at 564.30 cm$^{-1}$ and A$_1$ (LO) located at 730.34 cm$^{-1}$ modes in the bulk phase. Moreover, the above distinction reflects the change in GaN’s phonons modes in quasi-2D limit. Obviously, the E$_2$ (high) and A$_1$ (LO) peaks of GaN film all show blue-shift compared to those of the bulk polycrystalline GaN. Generally speaking, the parameters influencing the Raman scattering include material size, order, internal stress, and structural defects. The tensile (compressive) strain in GaN films will result in a blue (red) shift in the Raman spectroscopy of the GaN film. Chen et al. reported the E$_2$ peak of 2D GaN is obviously blue-shifted compared with that of the bulk, due to the tensile-strain state in the 2D limit.$^{[23]}$ In the present work, the thickness of our printed GaN is 4 nm, which is close to the thickness of the reported 2D GaN.$^{[23]}$ The tensile-strain state may also exist in the prepared GaN film, as a consequence, the E$_2$ peak and A$_1$ peak all appear blue shift for the quasi-2D GaN film. The printed GaN film for Raman spectra test is on SiO$_2$/Si substrate, it is no doubt that the Raman spectra of GaN are influenced by that of the SiO$_2$/Si substrate. As a consequence, a large bulge exists in the Raman spectrum of the 2D GaN, while it is absent in the bulk GaN.

X-ray photoelectron spectroscopy (XPS) is utilized to attain the printed GaN’s chemical bonding states. Figures 3d and 3e present the spectra of Ga 2p and N 1s areas for the GaN, respectively. The doublet in the Ga 2p region corresponds to the 2p$_{3/2}$ and 2p$_{1/2}$ orbital of Ga, the characteristic gallium peak for Ga$_2$O$_3$ placed at $\approx$20.4 eV was not seen, indicating Ga’s quantitative transformation. The principal broad N 1s peak centered at $\approx$3976 eV corresponds to the N 1s region compatible with the desired N 1s area presented in GaN. Energy-dispersive X-ray spectroscopy (EDS) mapping study of the resulting film composition is shown in Figure S5, Supporting Information. The GaN films for scanning electron microscope (SEM) and EDS test was scraped from SiO$_2$/Si substrate and transferred to the surface of the transparent PET tap, in order to avoid the effect of abound Si on the results of GaN film’s EDS. The detected element C and O in EDS results come from PET tap, which is the content of element C, O and H. During the process of scraping GaN films, the uniformity of GaN film is destroyed and some films were wrinkled, so the distribution of N and Ga in observed GaN films is uneven. But the atomic ratio of Ga:N is approximate to 1:1, proving that the obtained material is GaN. The attained XPS data and the other characterization techniques aforementioned above provide the outcome that all of our printed GaN have good consistency and can be used for further empirical electronic device fabrications.

The obtained electronic band structure and density of states (DOS) of an individual unit cell of the printed ultrathin quasi-2D GaN are shown in Figure 4a. Vienna Ab initio Simulation Package (VASP, version: 5.4.4) combined with the projector augmented wave (PAW) approach were utilized to accomplish the first-principles computations.$^{[20–22]}$ The Perdew–Burke–Ernzerhof (PBE) functional integrated with the DFT-D3 correction was employed to treat the exchange-functional. The plane wave’s cut-off energy was adjusted at 520 eV.$^{[23]}$ The Brillouin zone integration was accomplished with $15 \times 15 \times 6$ Monkhorst–Pack point sampling to optimize the bulk GaN. The self-consistent computations can give a 10–4 eV convergence energy threshold. The optimal values of the equilibrium geometries and lattice constants were obtained with maximum stress on all atoms in 0.01 eV Å$^{-1}$. For the GaN (110) surface structure, we use the 7×7×1 K-points for structural optimization and self-consistent calculations. Because the PBE functional will underestimate the band gap of the semiconductor, we also use the hybrid functional method (HSE06)$^{[24]}$ to calculate the band gap and density of states (DOS). Our density PBE functional analysis indicates that the GaN has a 1.51 eV direct bandgap. According
to the HSE06 calculation, the printed GaN has a 3.32 eV direct bandgap. From Figure 4b, the measured band gap derived from the UV–Vis absorption is 3.3 eV, which agrees well with the value derived from HSE06 calculation. By comparison, the calculated bandgap using energy density PBE functional analysis is smaller than the measurement result. Figure 4c shows a periodic slab of GaN with a non-polar (110) surface sliced from the wurtzite bulk phase. Generally speaking, the bandgap of GaN can be modulated up to 5 eV down to monolayer limit. Notably, the measured band gap of the printable GaN film
(4 nm) is 3.3 eV, which is smaller than 3.5 eV of the reported 2D GaN film (1.3 nm). It is because the obtained ultrathin quasi-2D GaN is slightly thicker than 2D GaN. With the thickness increasing, the band gap of the ultrathin quasi-2D GaN decreases, which is close to band gap of the bulk GaN.

2.3. Application of Printed Ultrathin Quasi-2D GaN in Electronic Devices

For experimental assessment of the electron transport features of the printed GaN and to investigate the potential for electronic devices, field-effect transistors (FETs) were constructed to assess the GaN for electronic device applications. Figure 5a shows the structure diagram of the transistor based on the printed GaN. We have adopted an individual side-gate design for all of the devices fabricated in this study, and detailed description of the fabrication process will be presented in the Experimental Section. Figure 5b presents a scanning electron microscopy (SEM) image of the device. Ag was used as gate electrodes and source–drain metal contacts, and FET channels were patterned with a width of $W_{ch} = 1000 \mu$m and the length of $L_{ch} = 50 \mu$m. Electrical measurements were carried out for the printed side-gated GaN FET. Figure 5c presents the transfer (drain current, $I_{ds}$, with respect to the gate voltage, $V_{gs}$) features of a representative GaN FET. Figure 5d presents the $I_{ds}$ with respect to the drain–source voltage ($V_{ds}$) at various values of $V_{gs}$ applied to the device. As presented in Figure 5c,d, the printed GaN FET devices’ p-type switching feature with an on/off ratio is more than $10^5$. The sub-threshold swing (SS) for the FET was 98 mV per decade, near the desired action. The average value of the room-temperature field-effect mobility ($\mu$) was obtained as 53.1 cm$^2$ V$^{-1}$ s$^{-1}$, with a mobility of 57 cm$^2$ V$^{-1}$ s$^{-1}$ for the device with the best performance (the SS and mobility computations are presented in the Section S6, Supporting Information). A statistical analysis on the performance of many FET devices fabricated using the presently reported wafer-scale printing process was conducted. Electrical features of thirty printed GaN FETs were evaluated. The mean log ON/OFF current ratio, mobility, and SS were 5.31 ± 0.52, 53.1 ± 4.5 cm$^2$ V$^{-1}$ s$^{-1}$, and 97.6 ± 2.42 mV dec$^{-1}$ (Figure 5e–g), respectively. A desired output was seen between various devices, considering that the mentioned devices were constructed in an academic laboratory. This uniformity yields confidence in employing the mentioned devices and methods in numerous applications like integrated circuits and active-matrix back-planes for displays. Moreover, the GaN-based FETs were significantly stable, having a high cyclability with a stable on/off ratio and stable on currents for more than 100 switching cycles under ambient conditions (Figure S6, Supporting Information).

Notably, the observed field-effect mobility is greater compared to that of traditional high-efficiency broad-bandgap
GaN-based devices. However, it should be noted that most of the reported GaN FET devices adopted AlGaN/GaN hetero-junction as carrier transport layer, and the devices have complex structures. Because the 2D electron gas (2DEG) at the interface of AlGaN/GaN heterostructure has very high electron mobility, the performance of AlGaN/GaN heterostructure FET is better than that of the device composed of GaN film only, the two kinds of devices cannot be directly compared. Therefore, for further broader comparison, we compare the performance of GaN and other p-type and n-type FETs reported and the devices in this study. Figure 6 plots SS versus mobility for other FETs reported in the literature as well as the device from this study, the value of SS is comparable to that which appeared in some of the best p-type and n-type oxide semiconductors like indium oxide, this highlights the fact that our approach offers a high current mobility while maintaining a small SS, which is mainly attributed to the high-quality electron-level GaN semiconductor. Besides, no detectable performance degradation was observed in the devices while working under ambient situations without encapsulation. This highlights that the printed GaN is of exceptional quality while indicating that additional enhancements can be achieved via the enhanced device construction. Future studies should concentrate on constructing the devices that can integrate separately addressable transistors into more complicated circuits. Potential approaches can involve using vdW heterostructures with dielectrics like hexagonal boron nitride or Ga2O3 combined with top gates. The mentioned device configurations can be employed to determine essential parameters like the pinch-off voltage that can be informative for incorporating future devices into functional circuits.
3. Conclusion

Liquid metals have rather diverse unconventional functionalities. One of the essential values of such materials is that they can serve as a reactive and templating media simultaneously and thus to synthesize and print a wide range of 2D semiconductor materials with extensive practical values. In the present work, we demonstrated the successful room temperature printing of the high-quality ultrathin quasi-2D GaN films with a large scale via a plasma nitridation reaction and through transferring occurring in the liquid metal’s nitride skin constructed under this specific condition. Moreover, complete explanation and analysis have been performed on the electrical features of the printed GaN films with thickness spanning from 1 nanometer to several nanometers, giving essential advice for further evolution of liquid metal printed high-performance semiconductors and indicating their considerable potential for future nanoelectronics.

As an example, we effectively presented the printed FET devices using GaN for the first time, depicting significant field-effect mobility and relatively small values of SS, and demonstrating an extremely steep subthreshold voltage switching behavior. This high-efficiency, simple, large-size, and cheap production process provides a new way for advancing GaN devices for power electronics applications. Moreover, the 2D or quasi-2D semiconductor can be produced as the favored material to several nanometers, giving essential advice for further evolution of liquid metal printed high-performance semiconductors and indicating their considerable potential for future nanoelectronics.

4. Experimental Section

Materials: Gallium with a purity of >99.99% was bought from Sigma Aldrich and utilized without additional refinement. The remaining materials were bought from usual suppliers and employed without refinement.

Formation of GaN on Liquid Gallium by Nitridation Reaction: In order to obtain GaN films with high purity and low defects, the whole preparation and printing process was carried out in a glovebox in a pure N2 environment at a pressure of 1–3 atm. The O2 content in the glove box atmosphere was maintained below 3 ppm and the H2O content was controlled below 0.5 ppm. High purity Ga was melted and then placed in NaOH solution to remove the oxide scale on the surface. The clean liquid Ga was extracted to 10 mL by a syringe and placed on the surface of the low-voltage electrode stainless steel plate of the plasma trigger device, and the stainless steel plate was grounded. In the environment of high purity N2, it could be seen that the surface of liquid Ga was bright without any oxide film. The stainless steel disk wrapped in transparent quartz of plasma high-voltage electrode was suspended vertically above liquid Ga, and the distance between the disk and the surface of liquid Ga was 1 mm. A voltage of 50 kV was applied between the two electrodes of the plasma trigger device through the voltage regulating device. At this time, the electric field strength of N2 breakdown was 5 × 107 V m⁻¹, the output current was 16 A, and the uniform and dense GaN skin could be obtained on the liquid Ga surface after glow discharge treatment for a certain time.

Printing Process of Ultrathin Quasi-2D GaN Films: Wafers of 500 nm SiO2 on Si (SiO2/Si) were washed by the deionized (DI) water for 1 min, and then sonicated in acetone and isopropyl alcohol (IPA) for 10 min (25 °C) and blown dry with N2 gas. The 3 min of oxygen (O2) plasma (Emitech K-1050X) at 100 W was then applied to wafers under low vacuum (0.6 Torr). The nitrided Ga droplets were placed on the SiO2/Si substrate and the complete printing program was executed. The size of nitride film changed with the droplet diameter and the travel distance of the scraper. The nitride layer formed on the liquid Ga’s surface could be transformed and printed on the substrate through gently scraping the droplets from one end of the substrate to the other end with a scraper. An extra force within the extrusion phase could damage the nitride layer. By this extrusion printing method, high-quality GaN films with a transverse size greater than a few centimeters could be effectively printed on the substrate.

Mechanical and Chemical Cleaning Process: In order to remove all liquid metal parts that remained on the sample, a facile mechanical ethanol cleaning method was employed. At first, ≈100 mL of ethanol was taken in a beaker, and the beaker was heated on a heating plate to 100 °C. Then, the substrate with printed nitride film was immersed in hot ethanol with tweezers. In order to eliminate the metal residues, a swab tool was utilized for wiping the substrate immersed in ethanol. Due to a strong vdW adhesion between the nitride film and the bottom layer, the nitride film still stuck to the silicon oxide surface within the wiping proceeding. Besides, the weak adherence between the deposited nitride layer, the liquid metal, and the film could be quickly removed to maintain the film clean and intact. Moreover, a chemical process was employed to clean the samples for a complete elimination of the metal residue on the substrate. An iodine/triiodide (I⁻/I₃⁻) solution (100 mmol L⁻¹ LiI and 1 LiI).
and 5 mmol L⁻¹ I₂ was constructed in ethanol and then located on a hot plate to heat up to 50 °C. In order to eliminate metallic inclusions, a substrate printed with the GaN film was immersed in a heated 1⁻¹⁰⁻¹³ solution for a time interval. At last, the residual etchant was removed by cleaning the sample in deionized water. The liquid metal particles could be successfully eliminated using the mentioned two cleaning Processes.

**FET Fabrication:** In order to construct fully printed side-gated GaN FETs, first, part of the GaN/SiO₂ region was etched with HF solution processes. A substrate printed with the GaN film was immersed in a heated I− solution for a time interval. At last, the residual etchant was removed with concentration of 1 mol L⁻¹ for ≈10 s. Then, the etching area was cleaned with ethanol, the clean Si of a certain area was obtained on the substrate. The Ag ink (Ag40X, UT Dots, Inc.) involved 40 wt% Ag nanoparticles, with ~20 nm particle diameters, dispersed in a solvent mixture of xylene and terpineol (9:1 by volume). The constructed ink was printed on the GaN films and SiO₂/Si substrates. A scientific 3B mixture of xylene and terpineol (9:1 by volume). The constructed ink printer from Prtronic was adopted to verify the inkjet printing details. The obtained sample was located on the inkjet printer’s panel at room temperature. A target image file was applied to the computer, which could be converted into a printable file through the softw are. Under computer control, source/drain electrodes and side-gate electrodes were subsequently patterned by printing Ag on the substrate. Finally, the printed samples were then sedimented at 120 °C in the air for 30 min in a furnace (MDL 281, Fisher Scientific Co.) to improve the conductivity.

**Characterization:** The AFM images were employed by a Bruker Dimension Icon with “Scansyst-air” AFM tips. A JEOL 2100F TEM/STEM (2011) system working at a 200 kV acceleration voltage involving a bright-field Gatan OneView 4k charge-coupled device (CCD) camera was utilized for both the low-resolution HRTEM imaging and selective area electron diffraction (SAED). A laser micro-Raman spectrometer (Renishaw in Via, 532 nm excitation wavelength) was adopted to accomplish the Raman spectroscopy. Moreover, the EDS measurements were used to collect the elemental mapping of the as-prepared samples. A thermo Scientific K-alpha XPS spectrometer associated with monochromatic X-rays from an Al anode (hν = 1486.6 eV) was adopted to perform the XPS analysis. A UV–visible (UV–vis) absorbance spectrometer (Hitachi U3900 UV–vis spectrophotometer) was utilized to evaluate the film’s optical bandgaps. A Cascade Microtech spectrometer (Hitachi U3900 UV–vis spectrophotometer) was adopted to perform the XPS analysis. A UV–visible (UV–vis) absorbance spectrometer (Hitachi U3900 UV–vis spectrophotometer) was utilized for both the low-resolution HRTEM imaging and selective area electron diffraction (SAED). A laser micro-Raman spectrometer (Renishaw in Via, 532 nm excitation wavelength) was adopted to accomplish the Raman spectroscopy. Moreover, the EDS measurements were used to collect the elemental mapping of the as-prepared samples.

**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.

**Data Availability Statement**
The data that support the findings of this study are available from the corresponding author upon reasonable request.

**Keywords**
2D semiconductor, filed effect transistor, GaN, nitrogen plasma, printing strategy

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