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van der Waals integration of GaN light-emitting diode arrays on foreign graphene films using semiconductor/graphene heterostructures

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Abstract

We report the van der Waals integration of micropatterned GaN light-emitting diodes (LEDs) onto foreign graphene films. GaN micro-LEDs were selectively grown on a graphene substrate using a patterned SiO_2 mask, and then the whole device structure was laterally fixed by a polyimide insulator to form a united layer. After device fabrication, the LED/graphene heterostructure device was piled on the foreign graphene layers using a typical wet transfer technique of 2D crystals where the bottom graphene layer of the heterostructure was adhered to the foreign graphene only by van der Waals interactions. The transferred micro-LEDs showed well-aligned crystallographic orientations as well as reliable device performances, including strong light emissions, good rectifying behaviors of the current density–voltage curve, and good simultaneity between the electroluminescence intensity and the applied currents, ensuring reliable electrical connections and mechanical adhesions of the light-emitting layer to the foreign graphene films. Furthermore, the reliable adhesiveness allowed us to achieve device wearability, while the LEDs exhibited homogeneous light emissions under various bending conditions because of negligible external stress in the discrete micro-LEDs.

Introduction

Two-dimensional (2D) van der Waals heterostructures have gained significant importance in fabricating multifunctional devices because of their ability to integrate heterogeneous 2D materials with a compact form factor[1,2]. The van der Waals interaction among the 2D films enables the stacking of different kinds of 2D layers on top of each other, while various 2D layered materials have become available for utilization in electronic and optoelectronic device applications, such as light-emitting devices, field-effect transistors, photodetectors, photovoltaics, energy storage, and biosensors[3–5]. Furthermore, the fabrication process involves simple wet or dry transfer techniques[6,7]. Accordingly, the van der Waals integration

facilitates the combination of heterogeneous devices with minor compatibility issues.

Despite the promising features of van der Waals integration, however, there are still some limitations in the integration approach that need to be overcome. Since it is difficult to satisfy all the demanded devices only by 2D materials, the integration process can be restricted for specific device applications. In addition, the lack of chemical reactivity on the 2D film surface makes it difficult to grow sequential structures of different compositions of 2D layers. To fabricate a sophisticated device, such as light-emitting diodes (LEDs) comprising multiple stacks of quantum wells, barriers, and n-type and p-type doped layers, numerous and time-consuming transfer steps would be required to build the device structure. However, most of these issues can be resolved by fabricating semiconductor/2D-material heterostructures. Semiconductor heteroepitaxy ensures the monolithic fabrication of sophisticated device structures to achieve high device

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performance[8]. Various semiconductor nanostructures and microstructures have been successfully fabricated on 2D layered materials exhibiting their flexibility and wearability, similar to 2D devices[9,10]. In particular, the presence of a 2D layer in the semiconductor/2D-material heterostructure can lead to integrating such heterostructure devices on foreign 2D materials through van der Waals interactions.
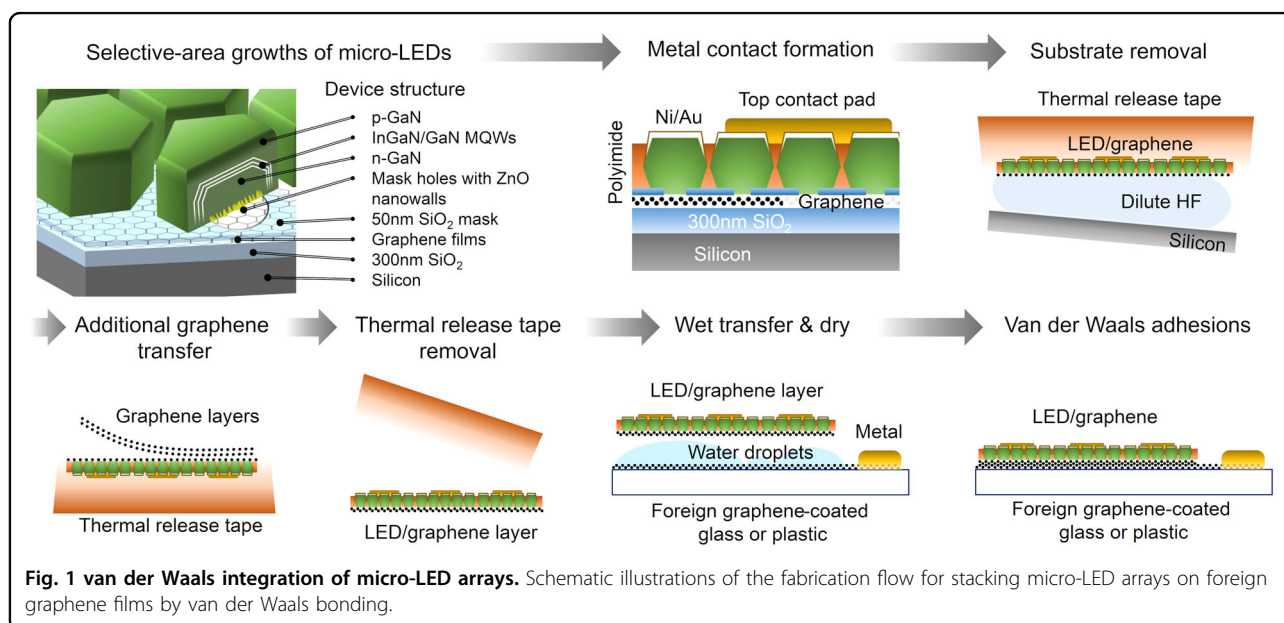
Here, we fabricated LED/graphene heterostructure devices by selectively growing micropatterned GaN LEDs on graphene films to demonstrate the properties of the heterostructure device in terms of van der Waals interactions for adhesiveness, the conductivity of the graphene contacts for current injections, and the device wearability at various shapes. By employing the LED/graphene heterostructure, the underlying graphene allowed us to mechanically and electrically link the LED arrays onto foreign graphene films without using any external adhesives. Moreover, because the discrete micro-LED arrays are rarely affected by external stress, the LED specimen exhibited reliable and consistent light emissions under various bending conditions.

The fabrication of semiconductor/2D-material heterostructures has been proposed as an effective method to realize transferable and flexible semiconductor devices[9–12]. Although organic devices have mostly been considered for flexible and wearable applications, inorganic semiconductors can provide superior device performance because of their excellent intrinsic properties, such as high radiative recombination rates, high carrier mobility, thermal and chemical stability, and long-term durability[13–15]. However, single-crystalline growth involves strong bonding between the semiconductor and the growth substrate,

which is restricted to meeting the flexibility demands. Nevertheless, by fabricating semiconductor/2D-material heterostructures, 2D layers can be placed between the semiconductor-substrate interface and/or semiconductor-semiconductor interface, resolving the semiconductor lift-off issue[11,16]. Furthermore, many notable advantages of using semiconductor/2D-material heterostructures have been successfully demonstrated, including fabrications of flexible and wearable devices, strain relaxations in the semiconductor epilayers, repeatable use of growth substrates, selective-area growths, and epitaxial lateral overgrowths on amorphous and/or metal substrates[17–25]. More promisingly, van der Waals integration has inspired the combination of various semiconductors and 2D materials for fabricating multifunctional devices[16,26]. Nonetheless, the sticking ability of semiconductor/2D-material heterostructures on foreign 2D films has rarely been studied, which is crucial to fabricate additive stacks of mixed-dimensional heterostructures.

Experimental section

To integrate semiconductor micro-LED arrays on foreign graphene films by van der Waals interactions, we fabricated a GaN/graphene heterostructure device where the top and bottom components of the heterostructure can work as an LED layer and an adhesive layer, respectively. Figure 1 illustrates our approach to assemble micro-LED arrays on foreign graphene films. First, GaN micro-LED arrays were selectively grown on continuous and full-coverage chemical vapor deposited (CVD) graphene films, which were prepared on 300-nm-thick SiO2-coated Si substrates. The thickness of the CVD graphene film was 10 nm with an estimate of 30 layers.

Fig. 1 van der Waals integration of micro-LED arrays. Schematic illustrations of the fabrication flow for stacking micro-LED arrays on foreign graphene films by van der Waals bonding.

For selective-area growth, a 50-nm-thick SiO_2 mask layer with a 5-μm-diameter hole pattern was formed on the graphene films prior to semiconductor growth. To avoid any damage to the graphene layer during patterning, we use a combination process of dry and wet etching to fabricate 50-nm SiO_2 hole arrays[27]. At first, reactive-ion etching was employed to etch the SiO_2 layer until an extremely thin SiO_2 layer remained. Then, the rest of the thin SiO_2 layer was removed using a buffered oxide etch.

All semiconductor layers were grown by metal-organic chemical vapor deposition (MOCVD). Two different MOCVD systems were employed for the growth of nitride films and ZnO nanostructures. First, using the ZnO MOCVD chamber, ZnO nanostructures were grown at 600 °C under 3.2 mTorr reactor pressure. High-purity diethylzinc (DEZn) (>99.9999%) and high-purity O_2 (>99.995%) were used as reactants, while high-purity Ar (>99.999%) was used as the carrier gas. During growth, the flow rates of DEZn and O_2 were both maintained at 40 sccm. We employed ZnO nanostructures because they can be grown on graphene films with high density[9], which provides growth selectivity between graphene hole openings and SiO_2 masks. Since GaN and ZnO are both wurtzite crystals with a small lattice mismatch within 2%, the ZnO nanostructures can also serve as a good seeding layer for GaN layer growth[20,27]. After preparing the ZnO nanostructures, the nitride layers of micro-LEDs were selectively grown on ZnO-coated graphene holes in another similar MOCVD. The LED structure is composed of sequential layers of ZnO nanostructure growth seeds, n-type GaN, three periods of $In_xGa_{1-x}N$/GaN multiple quantum wells (MQWs), p-type $Al_xGa_{1-x}N$, and p-type GaN. The micro-LEDs have sample-to-sample variations in emission wavelengths in the range of 490 to 520 nm, presumably because the growth conditions are not fully optimized yet.

After growth, a thermally annealed Ni(10 nm)/Au (10 nm) bilayer was formed on the micro-LEDs for the p-GaN ohmic contacts. Then, a 15-μm polyimide (PI) insulating layer was spin-coated at 1000 rpm for 40 s to fill the gaps between the discrete micro-LEDs where the PI laterally linked the micro-LEDs. Thereafter, the PI-coated LEDs were softly baked on a hot plate at 80 °C for 10 min and fully cured in a furnace under ambient N_2 in series at 80 °C for 30 min, 200 °C for 30 min, and 300 °C for 10 min. We hardly see peeling-off issues of the layer after full curing. Next, the PI layer was etched to 6 μm to expose the Ni/Au p-contact of micro-LEDs by O_2 plasma treatment. The PI layer, which was used in this study, measured transparency of 60%. We also deposited additional Ni(20 nm)/Au(20 nm) contact pads on the exposed micro-LED arrays for contact probing. The area of the contact pad was 100 μm × 100 μm. More details about the selective-area growths and the top contact formations of the micro-LED arrays are described elsewhere[27,28].

For the transfer process, the PI-surrounded micro-LED arrays were detached from the SiO_2/Si substrate by wet etching of the SiO_2 layer in buffered HF. A 1 cm² × 1 cm² micro-LED film takes ~12 h to completely detach from the substrate. We could peel off the as-grown micro-LED arrays from the substrate using a mechanical exfoliation method. However, the detached LED arrays become irregular (see Supplementary Fig. S1). Additionally, the use of PI layers prevents irregularities in micro-LED arrays but results in a relatively slow lift-off process. Nevertheless, we believe that the lift-off process can be further improved with better manufacturing scalability, as reported elsewhere by other research groups[12,25], since the as-grown micro-LED arrays showed mechanical lift-off feasibility.

Following rinsing with deionized water, we transferred a continuous CVD graphene film on the backside of the micro-LEDs, which helped the consistent current spread to the individual micro-LEDs. Thermal release tape was employed as a support to prevent curling problems of the PI-surrounded micro-LED arrays during the lift-off and graphene transfer processes. The PI-surrounded micro-LED arrays then became a free-standing layer by removing the thermal release tape using a 180 °C hot plate. Finally, the free-standing LED layers were piled on foreign graphene under wet conditions with a small amount of deionized water, where the foreign graphene films were prepared on a typical slide glass and a flexible PI sheet. The free-standing LED layer and the foreign graphene films adhered to each other after drying the sample on a hot plate at 60 °C. The wet transfer technique is widely used for the preparation of graphene layers on a supporting substrate[4-6]. Moreover, since the van der Waals force inherent on the graphene surface allows graphene to act as an adhesive on arbitrary substrates, we could attach LED/graphene heterostructure devices to glass and PI sheets using the same transfer method.

Prior to the adhesion process, we prepared a Ni(10 nm)/Au(10 nm)/Ag(500 nm) metal contact pad on foreign graphene films for reliable contact probing. To operate the LEDs, we injected currents by probing with metal needle tips at the top contact pad of the micro-LEDs and the metal electrode formed on the foreign graphene-coated substrates. The electroluminescence (EL) measurement system is composed of a probing stage, an optical microscope, and a spectrometer. Optical microscopy was used for contact probing, and the EL emissions were collected by an objective lens of the optical microscope. The EL emission was passed through the optical microscope and measured by the spectrometer. For the EL emission comparison under various bending conditions, we kept the same distance between the micro-LEDs and the objective lens by refocusing the micro-LED arrays after every bending. All the graphene films we used in this
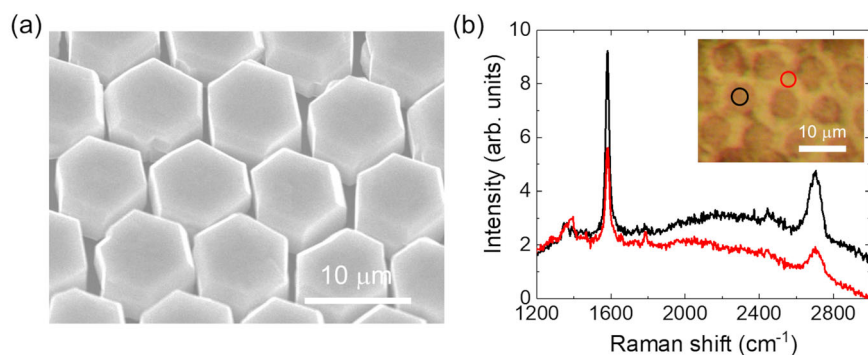
Fig. 2 Fabrication of stackable GaN micro-LED/graphene heterostructures. a FE-SEM image of the micro-LED arrays. **b** Optical image (inset) and Raman spectra of the flipped micro-LEDs. To obtain the Raman spectra, focused 532-nm laser beams with a spot size of 1 μm illuminated at the dark circle (black solid line) and the dark circle-free (red solid line) regions.

study were multilayer graphene, which was synthesized on Cu foil by chemical vapor deposition, showing 74% optical transparency in the visible range.

Results and discussion

The micro-LED/graphene heterostructures were designed such that the visible light-emitting device arrays can be transferable and stackable on external 2D layers by van der Waals forces. Figure 2a shows a field emission scanning electron microscope (FE-SEM) image of the as-grown micro-LEDs on SiO_2/Si substrates using the graphene intermediate layer. Each micro-LED showed a hexagonal crystal shape. As an array, the overall micro-LEDs exhibited consistent spacings, crystal sizes, and facets due to the selective-area growth on the graphene films. Since the electrical resistance depends on the device geometry, the nearly homogenous morphology of the micro-LEDs suggests uniform current injections to the individual micro-LEDs.

Additionally, after detaching the PI-surrounded micro-LED arrays from the original SiO_2/Si substrate, we investigated the presence of the underlying graphene films of the LED/graphene heterostructure using Raman spectroscopy with a 532-nm Nd:YAG laser. The optical microscope image of the flipped micro-LED/graphene heterostructure exhibited regular arrays of dark color circles (inset of Fig. 2b). The color contrast probably resulted from the selective-area growth using two-step temperatures where the low-temperature growths occurred only at the hole opening. The Raman spectra were obtained in both the dark circle region and dark circle-free region. As shown in Fig. 2b, a G-band peak at $1582 \, \text{cm}^{-1}$ and a 2D-band peak at $2705 \, \text{cm}^{-1}$ of graphene films were clearly observed regardless of the regions of the dark circles. This signifies that the graphene films were reliably preserved on the bottom surface of the micro-LEDs. For the as-grown LED sample, there was a 50-nm

SiO_2 mask between the micro-LED and the graphene films, which was etched out during the lift-off process using buffered HF. Nevertheless, the graphene films seemed to adhere to the PI-surrounded micro-LEDs after the wet etching process because of the very thin SiO_2 gap.

Furthermore, the stacking feature of the LED/graphene heterostructure layer on foreign graphene films was examined using X-ray diffraction (XRD). As shown in Fig. 3, we obtained rocking curves of the GaN(0002) XRD peak for the as-grown micro-LEDs on the original SiO_2/Si substrates (black dotted line) and the free-standing PI-surrounded micro-LEDs attached to the foreign graphene film-coated glass (red solid line). The full width at half maximum (FWHM) values of the rocking curves were measured to be 3.25° for the former and 3.35° for the latter. The comparable FWHM values suggest that the micro-LED arrays adhered to the foreign graphene films and maintained their original crystallographic alignments.

The physical and electrical connections between the micro-LED arrays and the foreign graphene films were further investigated by operating the LEDs. Figure 4 a shows a picture of the PI-surrounded micro-LED arrays uniformly attached to the graphene-coated glass. The good morphological bonding of the LED arrays agrees well with the XRD rocking curve result. The PI-surrounded micro-LED arrays were semitransparent, which enabled us to see alphabet letter displays underlying the glass. This confirms that no additional conducting adhesives, such as silver paste, were used to bond the micro-LEDs. We also observed strong blue light emissions from the micro-LED arrays. Moreover, the magnified light emission image showed homogeneous light emission distributions over the $100 \, \mu\text{m} \times 100 \, \mu\text{m}$ contact pad area, suggesting uniform current injections to the individual micro-LEDs (inset of Fig. 4a). Notably, the magnified light emission image was obtained from the backside through the glass substrate because they are metal-free, while the uniformity of the
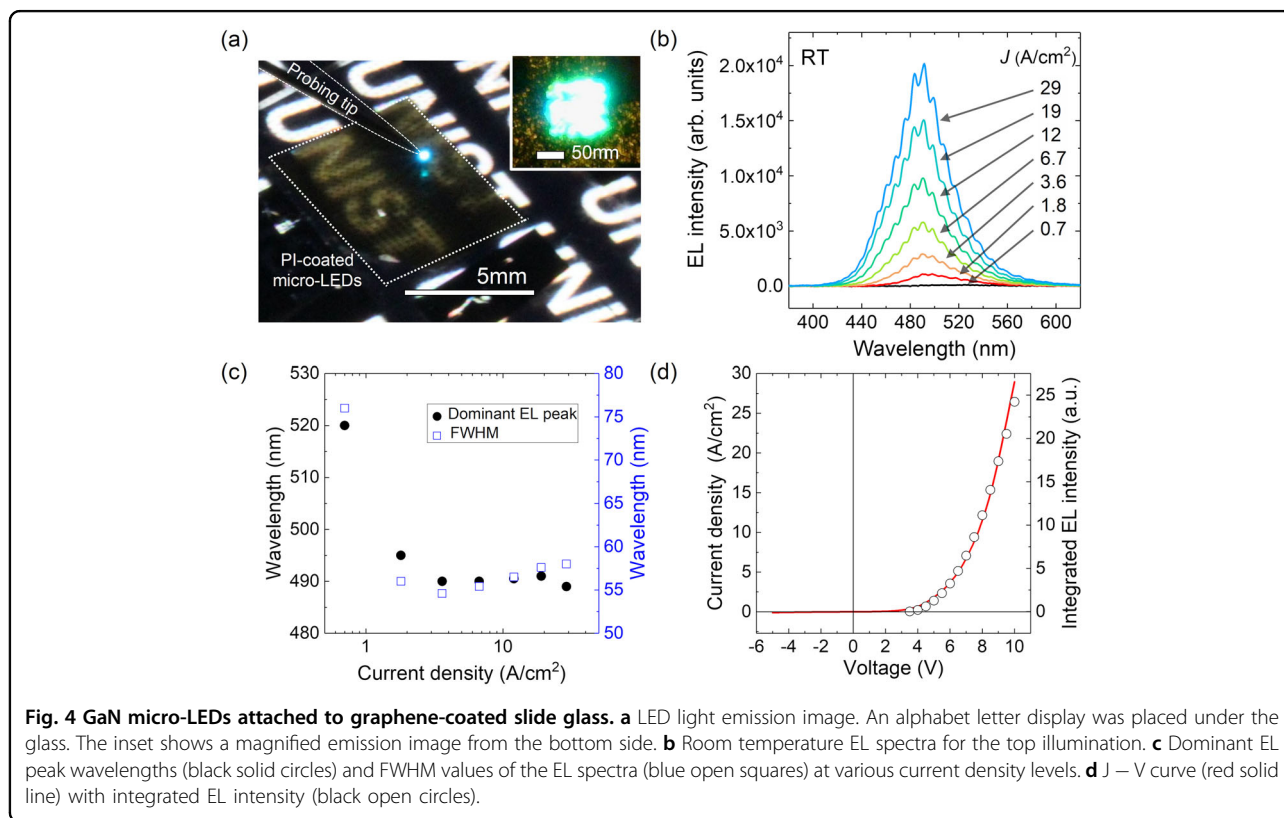
upper emissions can be disturbed by the relatively thick metal contact pad. However, there was little color difference between the top emissions and the bottom emissions such that the bottom emission color was green rather than blue. This was presumably because of the color mixing of the blue micro-LED emission with a weak fluorescent emission from the glass (see supplementary Fig. S2a). The
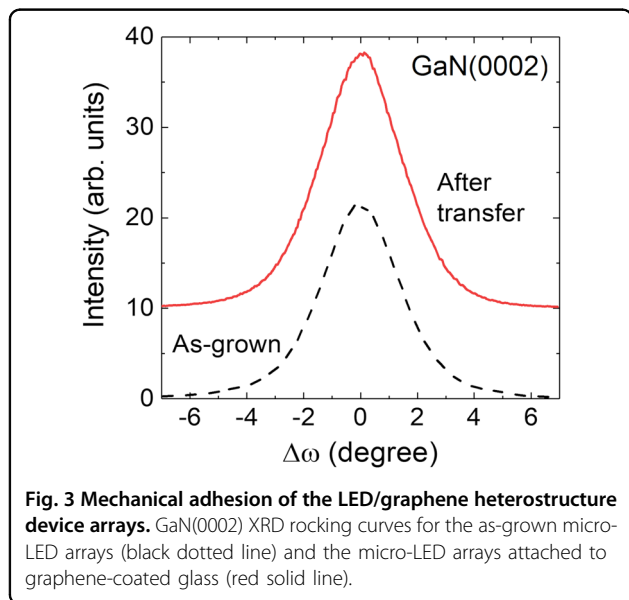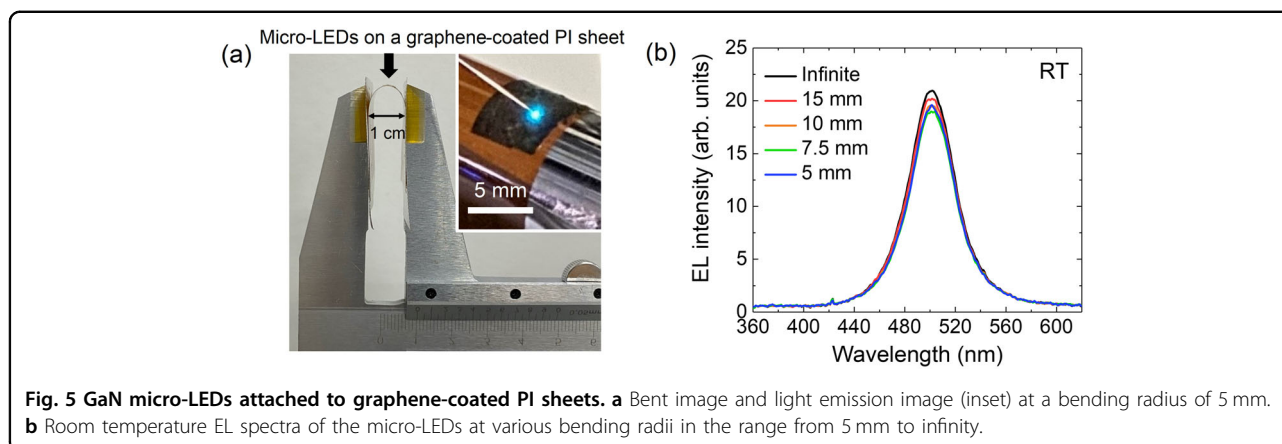
fluorescent emission spectrum exhibited a dominant peak at 512 nm with a shoulder peak at ~660 nm (see supplementary Fig. S2b).

The optical characteristics of the micro-LEDs were examined by measuring room temperature electroluminescence (EL) spectra for the top illumination, as shown in Fig. 4b, c. The dominant EL peak was observed at 490 nm, showing negligible peak shifts in the applied current density range above 3.6 A/cm^2. However, at a lower current density of $0.7-3.6 \text{ A/cm}^2$, we observed a blueshift of the EL spectra from 520 to 490 nm as the current density increased. Additionally, the FWHM values of the EL spectra decreased from 76 to 54 nm for the corresponding current density region. The EL peak shift with FWHM narrowing can be explained by screening quantum confined stark effects[29,30]. In the meantime, a shallow shoulder peak appeared at ~450 nm at high current densities above 6.7 A/cm^2 presumably because of carrier injections at different InGaN QW facets. Similar behaviors can be found commonly in multifaceted microstructure LEDs[31–34]. Although the shoulder peak yielded in the peak broadening, the FWHM value variations were within 4 nm.

Figure 4d shows the current density (red solid line) and integrated EL intensity (black open circles) of the micro-LEDs as a function of voltage. The current density–voltage (J–V) curve exhibited good rectifying behavior of a typical

Fig. 3 Mechanical adhesion of the LED/graphene heterostructure device arrays. GaN(0002) XRD rocking curves for the as-grown micro-LED arrays (black dotted line) and the micro-LED arrays attached to graphene-coated glass (red solid line).

Fig. 4 GaN micro-LEDs attached to graphene-coated slide glass. a LED light emission image. An alphabet letter display was placed under the glass. The inset shows a magnified emission image from the bottom side. **b** Room temperature EL spectra for the top illumination. **c** Dominant EL peak wavelengths (black solid circles) and FWHM values of the EL spectra (blue open squares) at various current density levels. **d** J − V curve (red solid line) with integrated EL intensity (black open circles).

Fig. 5 GaN micro-LEDs attached to graphene-coated PI sheets. a Bent image and light emission image (inset) at a bending radius of 5 mm. **b** Room temperature EL spectra of the micro-LEDs at various bending radii in the range from 5 mm to infinity.

p−n junction diode. The integrated EL intensity was on the same track as the J−V curve. These results strongly suggest that the micro-LEDs were electrically well connected to foreign graphene films using intermediate graphene channels. Taking advantage of the van der Waals integration with the good electrical connection of the graphene channels, we believe that it will become easier to expand the application of semiconductor device integrations with various other 2D materials. Graphene is widely used as an electrode in fabricating most 2D material-based devices. For example, a 2D-material heterostructure transistor consists of stacks of graphene/h-BN/MoS$_2$/h-BN/graphene[5], where one of the graphene electrodes can be considered a foreign graphene film similar to our approach and can be assembled with micro-LED/graphene heterostructure devices using van der Waals interactions. Hence, our demonstration of graphene makes the semiconductor a part of the van der Waals heterostructure system through graphene as an intermediate conducting and adhesive layer.

The PI-surrounded LED/graphene heterostructure device was also attached to a flexible graphene-coated PI sheet for the bending test. Since our device structure consists of a very thin graphene film, we use a flexible PI sheet as a supporting substrate in our demonstration. The bending radii ranged from infinity to 5 mm, where the 5-mm radius is smaller than the typical curvature of a contact lens. Figure 5a shows that the sample was bent as one united body without any observable peeling-off issues, suggesting good mechanical adhesions among the sample components. Furthermore, the curved micro-LED arrays were reliably operated under various bending conditions to generate strong light emissions, as displayed in the inset of Fig. 5a. More quantitatively, Fig. 5b shows nearly the same EL emissions of the micro-LED arrays regardless of the bending radii in terms of the dominant peak position of 502 nm, the FWHM value of 46 nm, and the light intensity. This result suggests that discrete micro-LEDs are beneficial for demonstrating flexible and

wearable devices because the mechanical stress and strain would be negligible under various bending conditions, while strain in the LEDs can yield emission intensity changes and wavelength shifts[35]. However, over numerous bending cycles, the micro-LED arrays along with the foreign graphene film partially peel off from the corners of the supporting substrate, which is presumably because of the strain due to the relatively thick PI layer. With further optimization of the PI coating conditions, we should be able to solve the peeling-off issue.

Conclusion

In summary, we fabricated micro-LED/graphene heterostructures to investigate the feasibility of using a semiconductor device as a component in van der Waals heterostructures. Similar to typical 2D materials, the micro-LED/graphene heterostructure device could be attached well to foreign 2D films of graphene layers using van der Waals interactions. The micro-LED arrays were fully operational under various bending conditions with good mechanical adhesion and electrical connections. In this study, we matched graphene layers with LEDs to fabricate semiconductor/2D-material heterostructures because the graphene films not only enabled the growth of epitaxial LED arrays but also served as a good bottom electrical channel for LED operations. We believe that this idea can be further extended to other combinations of semiconductor devices with 2D materials, such as hexagonal-BN and MoS$_2$, for fabricating multifunctional devices where the semiconductor/2D-material heterostructure will play an important role in obtaining an efficient and reliable device. More generally, our finding enlarges the advantages of van der Waals integration in 2D layered materials to the range of inorganic semiconductor devices.

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