nature portfolio

# Peer Review File

## Graphene-enabled laser lift-off for ultrathin displays

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## REVIEWER COMMENTS

**Reviewer #1 (Remarks to the Author):**

Laser lift-off of ultra-thin devices is a key technology for flexible electronics manufacturing. This paper proposes a graphene-enabled laser lift-off (GLLO) method, exhibiting improved processability and lift-off quality. The role of graphene is attributed to three factors: lateral heat diffusion, reduced adhesion, and enhanced UV absorption. I think the more important finding of this article is that the laser ablation effect can be converted from the vertical dimension (in the vertical direction, laser energy is compressed due to the enhanced absorption) to the horizontal dimension (due to the lateral heat conduction, and cracks can expand horizontally). This effect is equivalent to a horizontal redistribution of the laser energy, it may be of great significance to regulate the laser processing effect. This is a very interesting and high-quality paper, recommended for publication after in-depth revision. Some questions and suggestions are as follows
1- Could the author discuss any better methods for preparing graphene layers? Is CVD too expensive? Is it possible to achieve large-scale applications in the future?
2- The results of Fig 4b show that the adhesion strength of the PI-Gr-Glass structure is significantly reduced compared with that of PI-Glass and does not change with the number of Gr layers. It seems that the PI-Gr interface has the lowest adhesion strength. Why? Is it the van der Waals force? Is this adhesive strength strong enough for the device preparation?
3- The reusability of the carrier is important. After LLO, a very thin layer of new material (carbon?) on the surface will appear, will it affect the next use? Can the carbon layer be removed?
4- There are only four graphene layers at most in the experiment. What about more graphene layers? What is the programmable range of the LLO performance by controlling the number of integrated layers?
5- The characterization of the original laser spot is too little (the size and energy distribution of the laser spot), and the scanning pitch is best introduced in combination with the properties of the laser spot (to show the coverage degree). Besides, what are the differences and advantages of DPSS laser compared with excimer laser?
6- Even under high-energy laser irradiation, the Glass-Gr-PI structure is not damaged (so-called smooth effect), which is a very important and interesting phenomenon. The reason can be a more in-depth analysis. Is the lateral heat conduction to homogenize the Gaussian spot? Or does lateral heat conduction cause the edge region to peel, making the constraint relax when the film deforms? Or is there crack growth at the edge due to the gas ejection? To analyze this process, it is necessary to pay attention not only to the interface peeling mechanism but also to the deformation process of the film after peeling.
7- The results in Fig 2d show that the lift-off area is larger than the laser irradiation (spillover effect), but the spillover effect is not uniform (e.g., GLLO: 110.9 mJ/cm2). Why?
8- How to make readers better understand the so-called ‘narrow pyrolysis zone’ (in Fig. 3a)? Narrow is supposed to mean horizontal. The energy distribution is wider in the vertical direction, so the ablation degree per unit area is greater for Conv. LLO.
9- In Fig. 4e, is there a problem with the horizontal coordinate (nm scale?)?

10- For laser lift-off ultrathin devices, whether the interface Gr layer has the effect of protecting the device from high temperature since heat can be dissipated from the side of the light spot. Can the paper provide a simulation of this effect? How about thinner PI films (~1 um)?
11- Lastly and most importantly, what is the separation mechanism of the GLLO? The center area of the spot can be considered as a one-dimensional model, is the separation mechanism related to material ablation or gas-induced cavitation? The edge area of the spot needs to be considered as a two-dimensional model, is the separation mechanism related to thermal diffusion or gas impact-induced mechanical crack growth? Consider carefully observing the separation interface at different spot locations.

**Reviewer #2 (Remarks to the Author):**

In this paper, graphene is used to facilitate the laser lift off (LLO) of thin polyimide films from a glass carrier. Compared to conventional LLO, the graphene LLO process ("GLLO") changes the bubble formation between the polyimide (PI) film and the glass during laser ablation. With GLLO the bubbles are wider and flatter, producing less strain the PI film and thus less mechanical deformation. The authors attribute this to the enhanced absorption and lateral heat conduction in the graphene, and weaker adhesion of the PI to the graphene. The result is that GLLO is shown to have a wider process window than conventional LLO, and the resulting PI films tend to be less wrinkled. A demonstration of using GLLO to release fully-fabricated OLEDs on a thin PI film shows that – unlike the conventional LLO – the OLEDs remain operational, with little to no change in performance after GLLO. Overall, the use of graphene is an interesting idea that does provide some improvement to the LLO process, and the ability release the OLEDs without any noticeable change in performance is impressive. However, it is not clear to me that this work would have broad enough interest for publication in Nature Communications.

There are many points on which the paper could be made stronger:

1. The results would be more broadly applicable if this process was demonstrated to work with other substrate materials. It seems likely that (assuming the authors' hypotheses are correct) that this method would work with many polymer substrates.
2. The focus on ultra-thin (sub-5 micron) polyimide seems overly narrow, and not well-motivated. There are many substrates that may be a better choice if the goal is "conformal contact onto soft and curvilinear surfaces", because PI does not stretch. The title of the manuscript includes "for ultrathin displays", and the authors use "implantable and wearable information displays" as a motivation for such thin polyimide. What is an "implantable" display, and how can it be viewed by a user if it is implanted in the body? This use case needs more explanation to justify why the ultrathin polyimide is needed. On the other hand, there are many applications where thicker polymer substrates would be beneficial. Would this process work (and still provide any benefits over conventional LLO) if the substrate was NOT ultrathin PI? Is a thicker substrate more resistant to the wrinkling effect, and thus the GLLO is not really necessary or beneficial? The key point here is that the work is narrowly focused on ultrathin PI, the need for such an ultrathin substrate is not well

motivated, and the authors don't provide evidence that GLLO would provide benefits if the PI substrate were thicker.

3. The authors claim that the "graphene enables large-area integration" (pg 4). However, lift-off area demonstrated here is only 1.5 x 1.5 mm2 for all devices, which is quite small. Given that this process relies on graphene flakes that are transfer-printed with a stamp onto the glass substrate, it is hard to see how this could be reasonably scaled to be relevant for large-area displays.
4. The authors only include one brief paragraph summarizing other approaches to reduce laser-induced damage during LLO. If this is a problem with very broad interest (e.g., sufficiently broad to warrant publication in Nature Communications), I would suspect that there had been more previous efforts to solve this problem that should be discussed here. For example, there are at least two groups that have used a photonic lift-off process to delaminate flexible (opto)electronics on polyimide substrates from glass carriers. They also rely on an interfacial layer that absorbs the light and laterally spreads the heat at the interface. (See: Liu et al, "Photonic Lift-off Process to Fabricate Ultrathin Flexible Solar Cells", ACS Appl. Mater. Interfaces 2021, and Weidling et al, "Large-area photonic lift-off process for flexible thin-film transistors", npj Flexible Electronics 2022.)
5. Is the PI thickness of 2.9 um measured on substrates with and without the graphene layer to verify that the PI thickness is the same? If so, it is not stated or shown in the Supporting Info. The authors argue that the graphene surface and the interaction with the PI improves the lift-off; it would not be surprising if that difference in surface interaction also caused the spin-coated PI to wet and spread differently, resulting in films of different thicknesses on substrates with graphene vs without graphene. This should be clarified to ensure that the comparison is valid.
6. What is the criteria for "successful lift-off" (pg 7)? The authors categorize the lift-off results into 4 groups, but it is unclear what roughness/wrinkling/??? would be acceptable and categorized as "successful". This should be clarified and the substrates should be more thoroughly analyzed.
7. Figure 2d and the Supplementary Info show optical microscopy images of devices with different (G)LLO conditions. These images are not very informative without better description and/or more detailed analysis.
8. The simulation results shown in Figure 4 lead to many questions. Is a temperature of 1200 degC on the glass/PI interface actually feasible and reasonable?? Why is the glass in the simulation only 300 nm thick? What boundary conditions were applied at the back of the glass (distance = 0 nm) to keep the temperature there near room temperature? Is absorption in the glass included in the simulation (there is no absorption coefficient for glass included in Supp Table 1)? What fraction of the laser power is absorbed in the graphene vs the polyimide?
9. The authors mention several times that reusing the glass substrates would be beneficial and that re-use may be possible with the graphene-coated substrates: pg 3 "the deposited sacrificial layer cannot be reused", pg 6 "thick carbonaceous residues remaining after the process, impeding the recycling of the expensive glass carrier", pg 8 "facilitate the recycling of the graphene-integrated glass carrier", pg 18 "indicating the potential reusability of the expensive graphene-integrated glass carrier". However, they do not actually provide evidence that the graphene-coated substrates CAN be re-used, and thus these arguments are not well-founded and might mislead the reader.
10. The authors should specify the "controlled temperature and humidity" conditions that were used for the LLO experiments (pg 20).

**Reviewer #3 (Remarks to the Author):**

The authors reported Graphene-assisted laser lift-off (GLLO) to solve problems of conventional LLO technologies. The graphene can facilitate lateral heat diffusion, reduce adhesion between carrier glass and polyimide, and enhance UV absorption, resulting in wide process windows. Although this manuscript contains extensive data and their discussion, but we could find insufficient explanation. Moreover, this work is quite specific for publication in Nature Communications. I therefore do not feel that is suitable for publication in Nature Communications. I think the manuscript to be better suited for sister journals. Here are the points which need to be addressed before it can be considered for publication.

1) The author mentioned that the deposited sacrificial layer cannot be reused after the LLO process, leading to an increase in manufacturing costs. Does graphene can be reused after the LLO process? Are there any experimental results?

2) The author mentioned that "thick carbonaceous PI residues remaining after the process, impeding the recycling of the expensive glass carrier." The thickness of the PI residues can be thinner in the GLLO process, but there were still carbonaceous PI residues (~9.5 nm as in Fig. 2f) in the GLLO process. It seems difficult to reuse glass carriers in the GLLO process. Is it possible to reuse glass carriers in the GLLO process?

3) In figures 3e-3g, how do you measure the geometry of the blister? Is it experimentally determined or calculated? No explanation for this.

4) In Figure 5b, what materials is interconnection? The authors seem to use an Al cathode, but there is no description of interconnection (might be connected with an Al cathode).

We are submitting a **revised** manuscript entitled ***"Graphene-enabled laser lift-off for ultrathin displays"*** for publication in ***Nature Communications***.

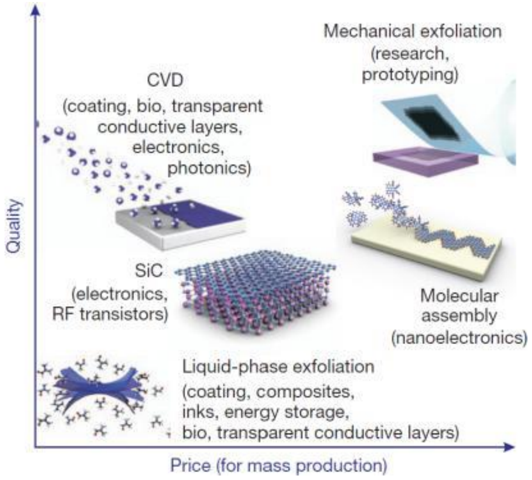
We greatly appreciate the valuable comments of the reviewers on our manuscript. We have responded in detail below to their comments and have revised the manuscript as noted. The changes are highlighted in a red color.

## The first reviewer's comments

*Laser lift-off of ultra-thin devices is a key technology for flexible electronics manufacturing. This paper proposes a graphene-enabled laser lift-off (GLLO) method, exhibiting improved processability and lift-off quality. The role of graphene is attributed to three factors: lateral heat diffusion, reduced adhesion, and enhanced UV absorption. I think the more important finding of this article is that the laser ablation effect can be converted from the vertical dimension (in the vertical direction, laser energy is compressed due to the enhanced absorption) to the horizontal dimension (due to the lateral heat conduction, and cracks can expand horizontally). This effect is equivalent to a horizontal redistribution of the laser energy, it may be of great significance to regulate the laser processing effect. This is a very interesting and high-quality paper, recommended for publication after in-depth revision. Some questions and suggestions are as follows*

**(#1)** *Could the author discuss any better methods for preparing graphene layers? Is CVD too expensive? Is it possible to achieve large-scale applications in the future?*

**Response to the first reviewer's comment (#1):** We appreciate the reviewer's valuable comment. There are several methods for the production of graphene, including CVD, liquid-phase exfoliation, synthesis on SiC, mechanical exfoliation, and molecular assembly (as shown in the figure below, [K. S. Novoselov. et al., *Nature* **490**, 192-200 (2012)]). The production method should be selected by considering the size, quality, and cost according to the applications. In this context, CVD-grown graphene has an affordable price because it has been recently commercialized by various mass production companies. Owing to its roll-to-roll manufacturing manner and reproducibility [S. Bae et al., *Nature Nanotechnology* **5**, 574-578 (2010)], we expect that large-scale applications can be achieved in the near future. Nevertheless, we recognize that the endeavor to find an advanced interlayer material is worthwhile to enhance the LLO performance and industrial applicability. Therefore, we plan to conduct follow-up studies not only to scale up the GLLO method but also to investigate other suitable materials, such as graphene oxide and transition metal dichalcogenide (TMD) monolayers. Additionally, we have added a brief discussion on the interlayer materials in the manuscript as follows.



The 1st paragraph of page 22 (Discussion section):

~ Several endeavors, such as optimization of laser irradiation conditions and beam profiles, and advances in interlayer materials, will be worthwhile for enhancing LLO performance, reusability, and industrial applicability.

**(#2)** *The results of Fig 4b show that the adhesion strength of the PI-Gr-Glass structure is significantly reduced compared with that of PI-Glass and does not change with the number of Gr layers. It seems that the PI-Gr interface has the lowest adhesion strength. Why? Is it the van der Waals force? Is this adhesive strength strong enough for the device preparation?*

**Response to the first reviewer's comment (#2):** We appreciate the reviewer's comment on the underlying mechanism of adhesion reduction due to the graphene layer. Polymeric materials on rigid substrates typically form interfacial adhesion through intermolecular forces [T. Miwa et al., *Polymer* **34**, 621-624 (1993)]. In this regard, it has been revealed that the atomic charge state at the interfacial region (coulombic interaction) is a primary factor determining the adhesion strength between the PI and silica glass [K. Min et al., *Polymer* **98**, 1-10 (2016)]. Therefore, we carefully anticipate that the inserted graphene layers hindered the coulombic interaction between the glass carrier and PI film, resulting in the PI film to interact with the graphene layer through weak dispersion forces. Although the graphene layer reduced the interfacial adhesion significantly, our results demonstrate no reliability or manufacturing issues in preparing OLED devices on PI substrates.

**(#3)** *The reusability of the carrier is important. After LLO, a very thin layer of new material (carbon?) on the surface will appear, will it affect the next use? Can the carbon layer be removed?*

**Response to the first reviewer's comment (#3):** We greatly appreciate the reviewer's comment on the reusability of the carrier. To validate the reusability, we performed additional experiments as shown in Fig. 6, and discussed the results as follows. Please refer to the revised manuscript (Pages 20 and 22).

The page 20:

Furthermore, the reusability of the graphene-integrated glass carrier was investigated by repeating the processes of spin coating the 2.9-μm-thick ultrathin PI substrate, OLED fabrication, and GLLO sequentially (**Fig. 6a**). The OLEDs fabricated in the first cycle were separated with a laser fluence of 63.4 mJ/cm$^2$. Consistent with the results in **Fig. 5**, the devices maintained their current density-voltage-luminance properties and current efficiency before and after the GLLO process (**Figs. 6b** and **6c**). The reusability of the graphene-integrated glass carrier was evaluated by considering the electrical performance variation of OLEDs fabricated on reused carriers. In this regard, delamination of the OLEDs in the second and third fabrication cycles required a slightly higher laser fluence of 79.2 mJ/cm$^2$ for complete lift-off. Moreover, the analysis results indicate that although the OLEDs remained operational after the GLLO process, a slight degradation in luminance was observed (**Fig. 6b**). We carefully anticipate that the higher laser fluence requirement and degraded luminance resulted from the thin carbonaceous PI residues represented in **Fig. 2f**. In other words, the thin residues covered the integrated graphene layer, hindering the reduction of interfacial adhesion of newly coated PI substrates and increasing the distance from the graphene layer. Therefore, more effort will be needed to completely remove the carbonaceous residues, ensuring the intact reusability of the graphene-integrated carrier.

**Fig. 6**:

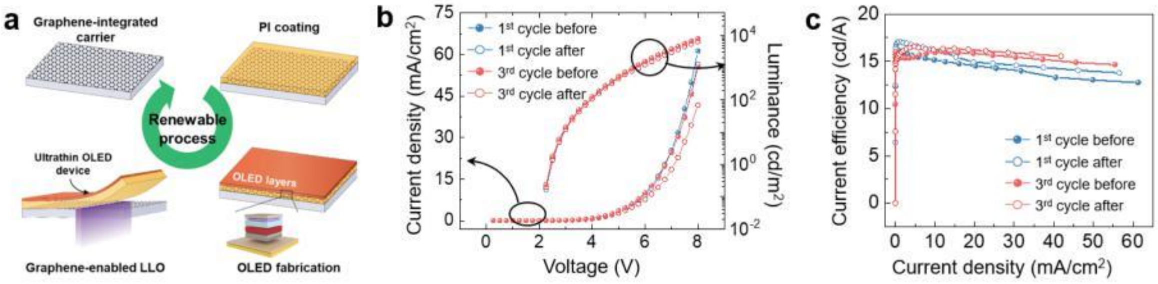


**Fig. 6 | Reusability of the graphene-integrated carrier. a,** Schematic illustration of the renewable process of the graphene-integrated carrier. **b,c,** Electrical performance variation of the OLEDs fabricated in the first and third cycles, before and after the GLLO process. (**b**) current density-voltage-luminance properties and corresponding (**c**) current efficiency-current density curve.

The 1st paragraph of page 22 (Discussion section):

~ Concerning the reusability of the graphene-integrated carrier, the separation of ultrathin OLED devices fabricated on reused carriers was achieved with a slightly higher laser fluence irradiation. Although the experiment demonstrated early-stage reusability, a slight degradation in OLED luminance was observed after the GLLO process. This degradation in LLO performance may be attributed to the remaining carbonaceous PI residues on the carrier which cover the graphene layers. It is anticipated that further study is required for the complete reusability of the graphene-integrated carrier, even though the presented GLLO method reduced the thickness of carbonaceous PI residues by approximately 92.8 % compared to the conventional LLO methods. Several endeavors, such as optimization of laser irradiation conditions and beam profiles, and advances in interlayer materials, will be worthwhile for enhancing LLO performance, reusability, and industrial applicability. Finally, we believe that this work will open up new possibilities for utilizing CVD-grown graphene in laser-based manufacturing applications, such as emerging displays, wafer-level packaging, and energy harvesting devices.

**(#4)** *There are only four graphene layers at most in the experiment. What about more graphene layers? What is the programmable range of the LLO performance by controlling the number of integrated layers?*

**Response to the first reviewer's comment (#4):** We are thankful for the reviewer's comment. The number of integrated layers is theoretically unlimited; tens or even hundreds of graphene layers can be integrated. However, in practice, the preparation time and cost increase proportionally with the number of integration layers because CVD-grown graphene is transferred layer-by-layer. For this reason, we decided to use 4 layers of graphene based on the blistering behavior observed in Figs 3e and 3f. According to the observed trends in blister height and diameter, we expect that integrating more graphene layers may lead to a further increase in blister diameter while maintaining similar blister height.

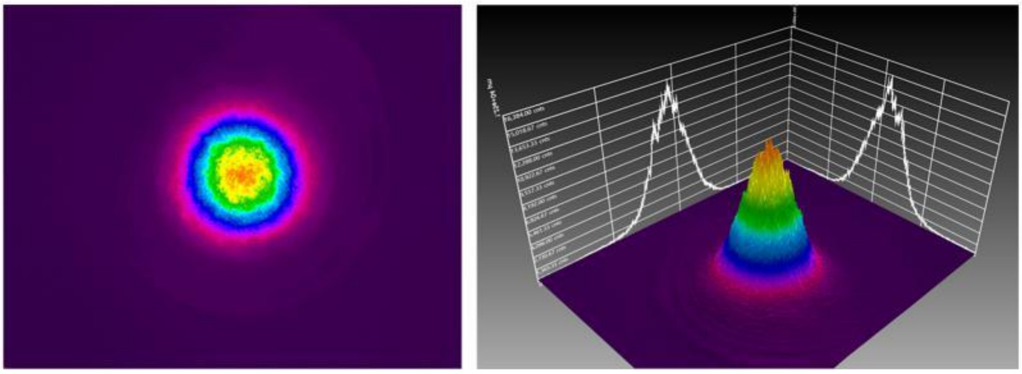
**(#5)** ***(Q5-1)*** *The characterization of the original laser spot is too little (the size and energy distribution of the laser spot), and the scanning pitch is best introduced in combination with the properties of the laser spot (to show the coverage degree).* ***(Q5-2)*** *Besides, what are the differences and advantages of DPSS laser compared with excimer laser?*

**Response to the first reviewer's comment (#5):** Thank you for the constructive comment. Here are our responses to each comment.

**(Q5-1)**: We agree with the reviewer's comment that the characterization of the laser spot is important because it is related to the scanning pitch conditions. To supplement the information, we have included a Gaussian beam profile in Supplementary Fig. 20, and provided a more detailed description of the control of the laser spot and scanning pitch in the Methods section as follows.

The 1st paragraph of page 24 (Methods section):

~ The pulse had a Gaussian profile (Supplementary Fig. 20), and its spot size was adjusted to 56.7 μm (full width at half maximum) using an optical system that contains power and polarization control. To focus the pulse on the plane of the specimen and control the scanning pitch, the Gaussian energy distribution laser pulse was inserted into a 2D laser scanner (hurrySCAN III 10, Scanlab) which includes a telecentric f-theta lens with a focal length of 160 mm. These LLO experiments were conducted in a cleanroom environment where the temperature and relative humidity were controlled at 22 °C and 40 %, respectively, under ambient air conditions.



**Supplementary Fig. 20** | Profile of Gaussian beam utilized in the laser lift-off processes.

**(Q5-2)**: As the reviewer is already aware, excimer lasers have typically been used in traditional LLO processes because they can generate a wide line beam with a uniform energy distribution due to their high energy. However, excimer lasers exhibit relatively low beam quality in precision manufacturing, and the systems are significantly expensive and difficult to maintain. To overcome these limitations, DPSS lasers have recently attracted attention in LLO technologies (Y. Kim et al., *Optics and Laser Technology* **142**, 107245 (2021)). Although the lower energy of the DPSS laser compared to the excimer laser limits beam size expansion, its high pulse repletion frequency enables fast and high-quality processing when integrated with a high-speed laser scanner. In addition, the DPSS laser system offers advantages in terms of cost and maintenance due to its low energy requirements. Therefore, the utilization of the DPSS laser system is suitable for high precision and economical LLO technologies.

For the sufficient understanding of general readers, we have included an explanation about the advantages of the DPSS laser compared to excimer laser systems in our manuscript as follows.

> The 1st paragraph of page 5:
>
> After specimen preparation, a 355 nm diode-pumped solid-state (DPSS) laser system was utilized for the laser lift-off process (Supplementary Fig. 4). The DPSS layer system offers advantages in terms of cost-competitiveness, high reliability, and outstanding beam quality for precision manufacturing compared to excimer laser systems.

**(#6)** *Even under high-energy laser irradiation, the Glass-Gr-PI structure is not damaged (so-called smooth effect), which is a very important and interesting phenomenon. The reason can be a more in-depth analysis. Is the lateral heat conduction to homogenize the Gaussian spot? Or does lateral heat conduction cause the edge region to peel, making the constraint relax when the film deforms? Or is there crack growth at the edge due to the gas ejection? To analyze this process, it is necessary to pay attention not only to the interface peeling mechanism but also to the deformation process of the film after peeling.*

**(#11)** *Lastly and most importantly, what is the separation mechanism of the GLLO? The center area of the spot can be considered as a one-dimensional model, is the separation mechanism related to material ablation or gas-induced cavitation? The edge area of the spot needs to be considered as a two-dimensional model, is the separation mechanism related to thermal diffusion or gas impact-induced mechanical crack growth? Consider carefully observing the separation interface at different spot locations.*

**Response to the first reviewer's comments (#6) and (#11):** We greatly appreciate the reviewer's valuable comments. Since the separation and deformation behaviors of the PI film are closely related, we are addressing them together in our response.

First, cross-sectional images of the PI blisters were observed using focused ion beam (FIB)-scanning electron microscopy (SEM) (Supplementary Fig. 12). These images provide evidence of the vertical expansion of the PI film in the conventional LLO process and excessive interfacial delamination between the graphene and PI films in the GLLO process.

To understand the in-depth mechanism, we analyzed the ablation diameter using FEA simulation as provided in Supplementary Fig. 17. The results show that the experimentally observed blister diameter of the GLLO method (Fig. 3f) is significantly larger than the simulation data. This means that, after the initial blister formation, gaseous products propagated into the graphene–PI interface, resulting in further expansion of the diameter by interfacial crack growth. In contrast, in the conventional LLO process, the strong adhesion of the PI–glass interface hindered the lateral crack growth, causing the gas products to expand the PI film vertically.

In addition, we considered the peel test results in Fig. 4b. The presence of only a single layer of graphene remarkably reduced the interfacial adhesion similar to multilayered graphene, but there is an obvious distinction in the blister height and diameter with the addition of graphene layers, as represented in Figs. 3e and 3f. This indicates that not only adhesion reduction but also the enhancement of interfacial UV absorption and lateral heat diffusion, which are related to the initial thermal decomposition of PI films, are important in explaining the blistering behavior.

Therefore, we revised the separation and deformation mechanisms by diving them into the initial formation of blister and gas products by the thermal decomposition of PI films, and the lateral and vertical expansion of the blister by the propagation of gas products. Based on this explanation, we have improved the manuscript as follows.

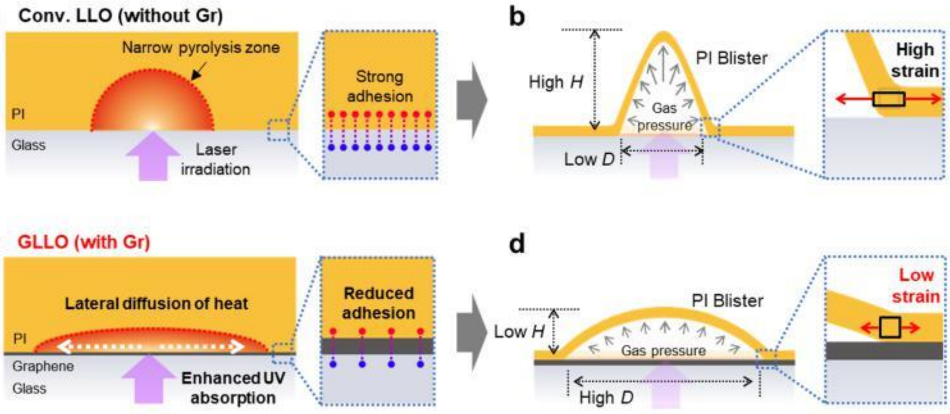
The 1st paragraph of page 11:

~ The ablation of PI is typically attributed to the photothermal decomposition of the material with a vertically distributed narrow pyrolysis zone[26]. Moreover, as a result of photothermal decomposition, gaseous products are produced inside the PI film, separating the PI films. In particular, the vertically distributed pyrolysis zone results in the initial formation of a sharp blister, and the gaseous products further expand the sharp blister vertically, resulting in high height and low diameter (**Fig. 3b**). At this point, the intrinsically strong interfacial adhesion[27,28] between the glass carrier and PI film hinders lateral crack growth into glass–PI interface. Consequently, the high mechanical strain associated with the expansion of the sharp blister causes plastic deformation of the PI film[29,30].

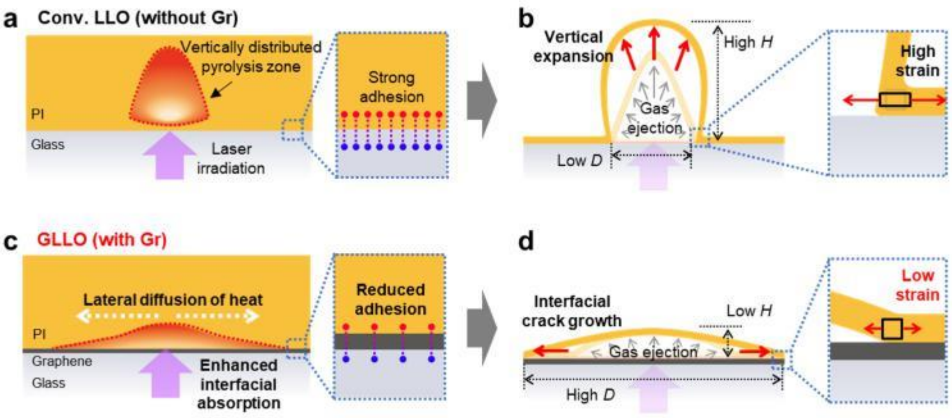
The 2nd paragraph of page 11:

~ First, the graphene layer enhances the absorption of photothermal energy at the interface, relocating the ablation sites from within the PI film to the graphene–PI interface. This relocation contributes to the reduction of the thickness of carbonaceous residue. Second, the outstanding in-plane thermal conductivity of the graphene layer effectively disperses the absorbed photothermal energy in the horizontal direction, thereby widening the narrow pyrolysis zone. These two factors, the enhancement of interfacial absorption and lateral heat diffusion, enable the smoothing of the initially generated blister by distributing the pyrolysis zone horizontally. Third, the graphene layer reduces the interfacial adhesion of the PI film by hindering molecular interactions between the glass carrier and the PI film. Owing to the low adhesion, the generated gaseous products are propagated to graphene–PI interface, leading to lateral interfacial crack growth and further increasing the blister diameter (**Fig. 3d**). The resulting smooth blister, with low height and high diameter, reduces the mechanical strain during the ablation process, allowing the lift-off of the ultrathin PI film without significant damage. More detailed separation and deformation processes of the PI film for the conventional LLO and GLLO methods are described in Supplementary Fig. 11, and cross-sectional images of the blisters observed via focused ion beam (FIB)-scanning electron microscopy (SEM) are provided in Supplementary Fig. 12.

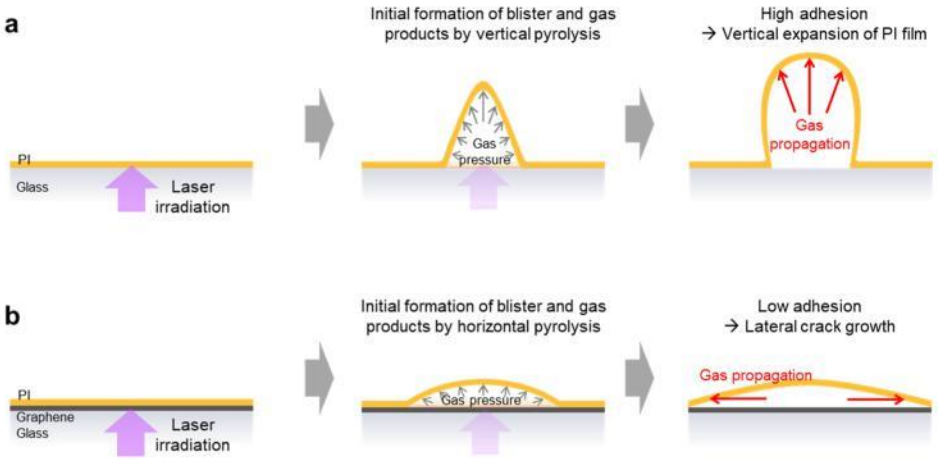
**Figs. 3a-d (Before)**:



**Figs. 3a-d (After)**:

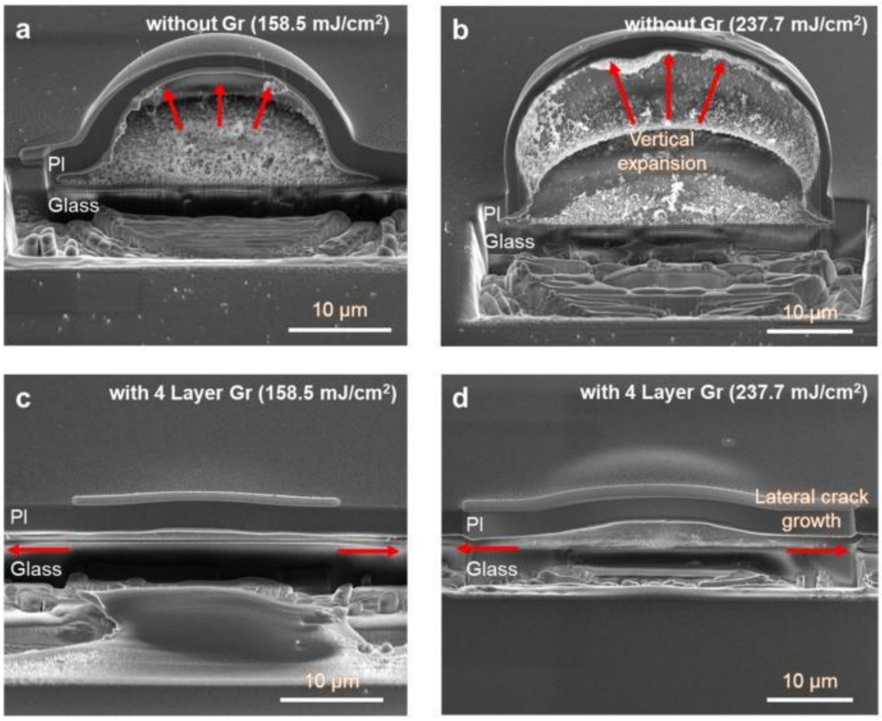


**Supplementary Fig. 11**:



**Supplementary Fig. 11** | Separation and deformation mechanisms of the PI film: **a**, conventional LLO process. **b**, GLLO process.

**Supplementary Fig. 12**:



**Supplementary Fig. 12** | Cross-sectional images of ultrathin PI blisters formed by laser irradiation. **a,b**, Specimens without graphene integration irradiated with a laser fluence of 158.5 and 237.7 mJ/cm$^2$, respectively. **c,d,** Specimens with 4-layer graphene integration irradiated with a laser fluence of 158.5 and 237.7 mJ/cm$^2$, respectively. The images were obtained using FEM-SEM. The FIB cuts were made along the central line of the blister, and SEM was exploited to capture the high-resolution images. A thin platinum layer was coated on the specimens to prevent deformation of the blister during the FIB milling process.

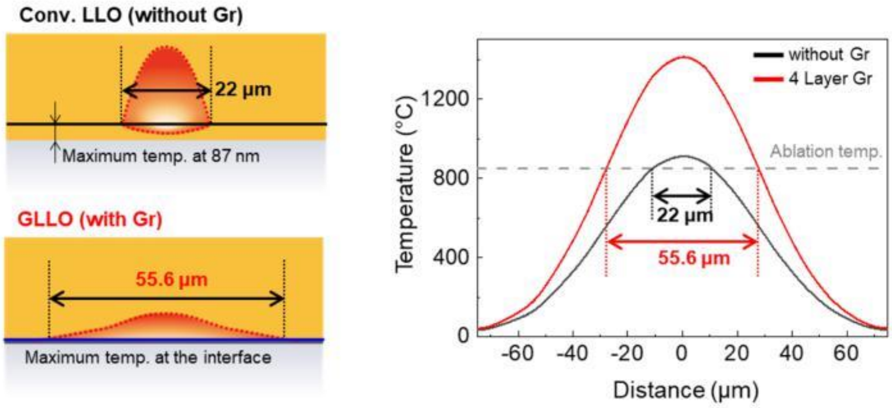
The 1st paragraph of page 15:

~ The peel test results in **Fig. 4b** also confirm that the graphene layer significantly reduced the interfacial adhesion by impeding molecular interactions between the glass carrier and the PI film. In particular, the presence of a single graphene layer remarkably reduced the interfacial adhesion similar to that of multilayered graphene. However, there was an obvious distinction in the blister height and diameter with the addition of the graphene layer as shown in **Figs. 3e** and **3f**. Accordingly, it can be concluded that the enhancement of UV absorption and lateral heat diffusion, which are related to the thermal decomposition of the PI film, are important in explaining the ablation behavior.

The 2nd paragraph of page 16:

In addition, taking into account the threshold ablation temperature of 850 °C for the PI films[31], it can be inferred that the ablation occurs at the interior region of the PI film with a diameter of 22 μm in a typical LLO process. In contrast, the multilayered-graphene relocates the ablation region to the interface with a diameter of 55.6 μm (Supplementary Fig. 17). The results can underpin the notion of lateral crack growth by considering that experimentally obtained blister diameters of the GLLO method presented in **Fig. 3f** are larger than the simulation-based ablation diameter. ~

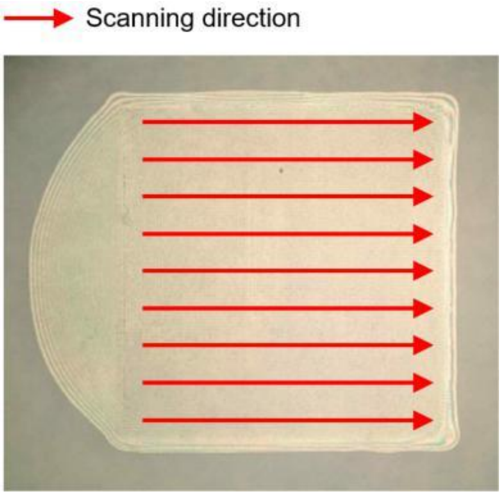
**Supplementary Fig. 17**:



**Supplementary Fig. 17** | Horizontal temperature distribution and ablation diameter at the height of maximum temperature for the conventional LLO (87 nm above the interface) and GLLO (at the interface). Ablation diameters were determined by considering the threshold temperature of 850 °C for the PI films

**(#7)** _The results in Fig 2d show that the lift-off area is larger than the laser irradiation (spillover effect), but the spillover effect is not uniform (e.g., GLLO: 110.9 mJ/cm2). Why?_

**Response to the first reviewer's comment (#7):** We appreciate the reviewer's comment on the spillover effect in the GLLO process. The spillover effect is attributed to the propagation of gas products into the low adhesion interface between graphene and the PI film. In this regard, laser ablation occurred sequentially, following the laser scanning direction as shown in the figure below. We anticipate that the sequential laser scanning resulted in anisotropic propagation of gas products during the GLLO process, causing non-uniform spillover.



**(#8)** *How to make readers better understand the so-called 'narrow pyrolysis zone' (in Fig. 3a)? Narrow is supposed to mean horizontal. The energy distribution is wider in the vertical direction, so the ablation degree per unit area is greater for Conv. LLO.*

**Response to the first reviewer's comment (#8):** Thank you for the reviewer's insightful comment. We agree that the expression 'narrow pyrolysis zone' requires clarification for better understanding. We anticipate that specifying the direction will resolve this issue. Therefore, we have revised the term 'narrow pyrolysis zone' to 'vertically distributed pyrolysis zone' in the manuscript and Fig. 3a as follows.

The 1st paragraph of page 11:

~ The ablation of PI is typically attributed to the photothermal decomposition of the material with a vertically distributed narrow pyrolysis zone[26]. Moreover, as a result of photothermal decomposition, gaseous products are produced inside the PI film, separating the PI films. In particular, the vertically distributed pyrolysis zone results in the initial formation of a sharp blister, and the gaseous products further expand the sharp blister vertically, resulting in high height and low diameter (**Fig. 3b**) ~

**Fig. 3a (Before):**

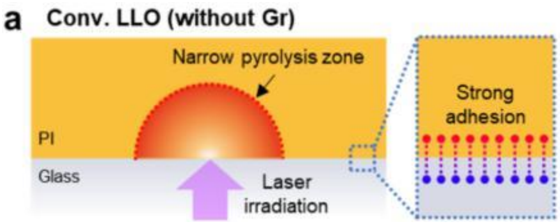
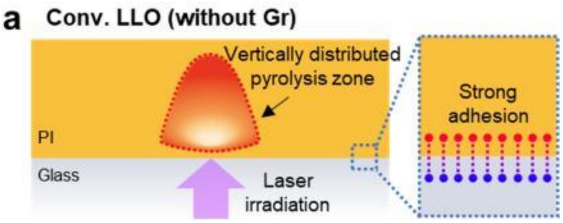


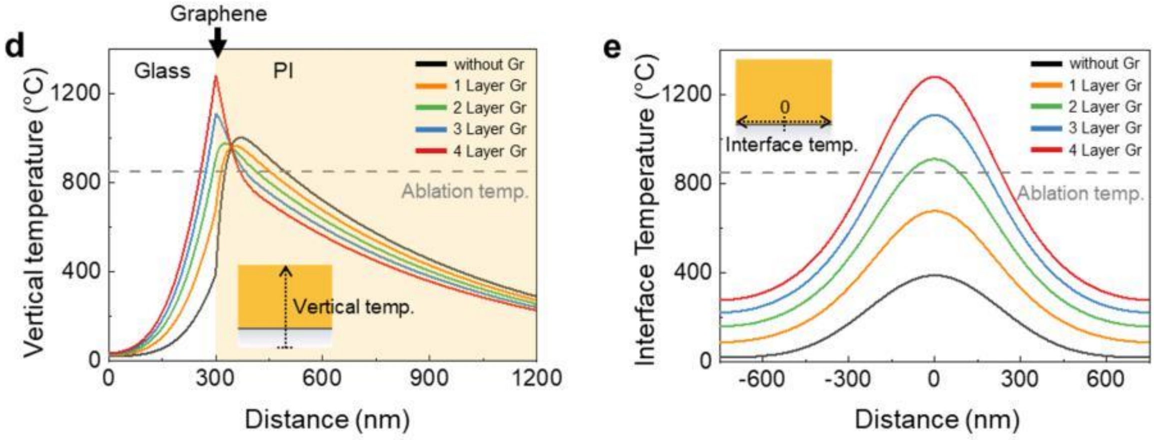
**Fig. 3a (After):**



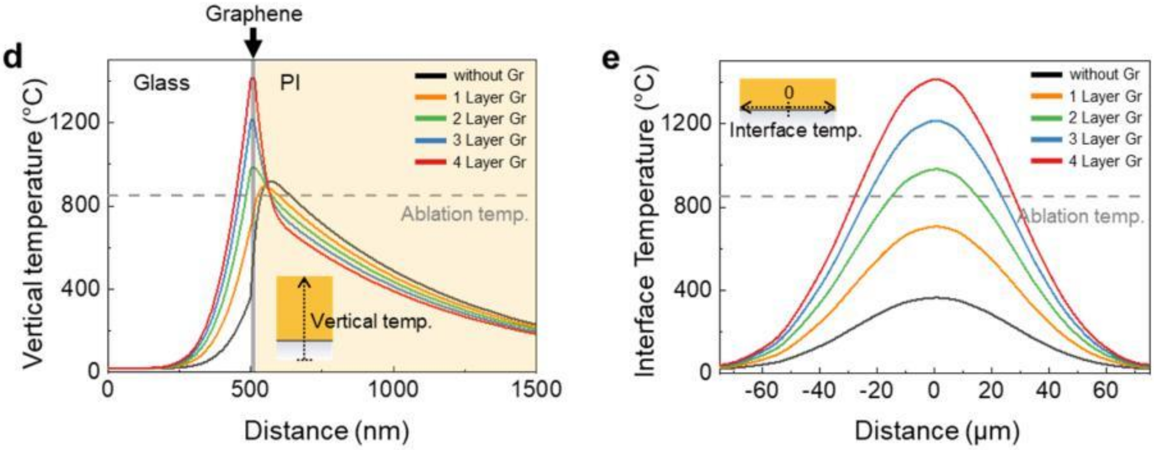
**(#9)** *In Fig. 4e, is there a problem with the horizontal coordinate (nm scale?)?*

**Response to the first reviewer's comment (#9):** We appreciate the reviewer's valuable comment on the simulation results. The simulation model in our original manuscript was designed to visualize the relative differences in temperature profiles during the ablation process, with respect to the number of graphene layers (**Fig. 4c**), so the simulation scale was reduced compared to the actual specimens. However, to provide more accurate temperature distributions in **Figs. 4d** and **4e**, we have revised the data to those obtained from a full-scale simulation model that reflects actual specimen structures (Supplementary Fig. 14). Additionally, we have included appropriate explanations for both the reduced and full-scale simulation models in the manuscript.

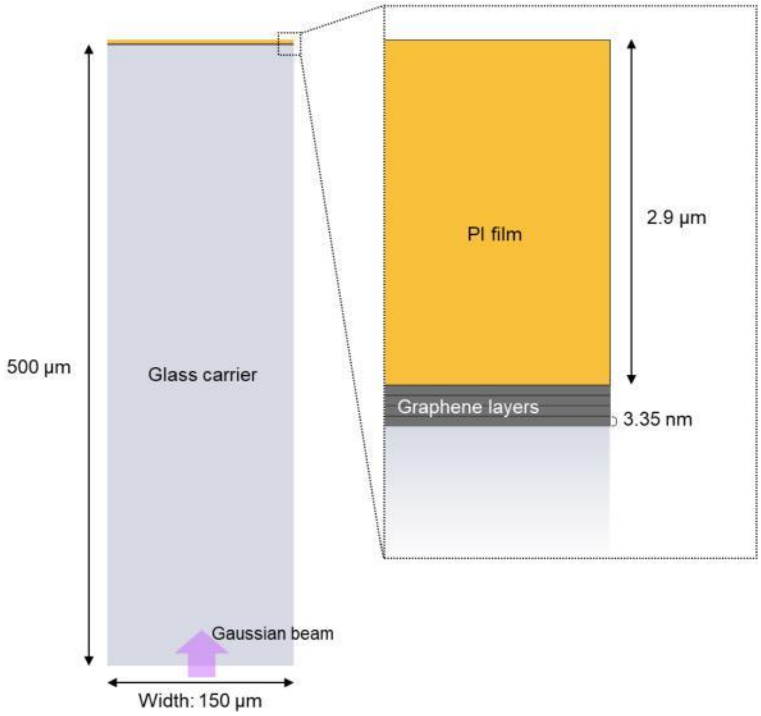
**Figs. 4d and 4e (Before – obtained from reduced model):**



**Figs. 4d and 4e (After – obtained from full-scale model):**



**Supplementary Fig. 14:**



**Supplementary Fig. 14** | Full-scale simulation model for accurate analysis of temperature distributions.

The 1st paragraph of page 15:

~ Moreover, the relative differences in temperature profiles during the ablation process, concerning the number of graphene layers, were visualized through finite element analysis (FEA) simulation (**Fig. 4c**). In this regard, due to the different thickness scales of the materials, the simulation model with a reduced scale was introduced for the visualization (Supplementary Fig. 13). ~

The 1st paragraph of page 16:

Further investigations were conducted using a full-scale simulation model (Supplementary Fig. 14), to examine the actual temperature distributions in both the vertical direction and at the interface (**Figs. 4d** and **4e**). ~

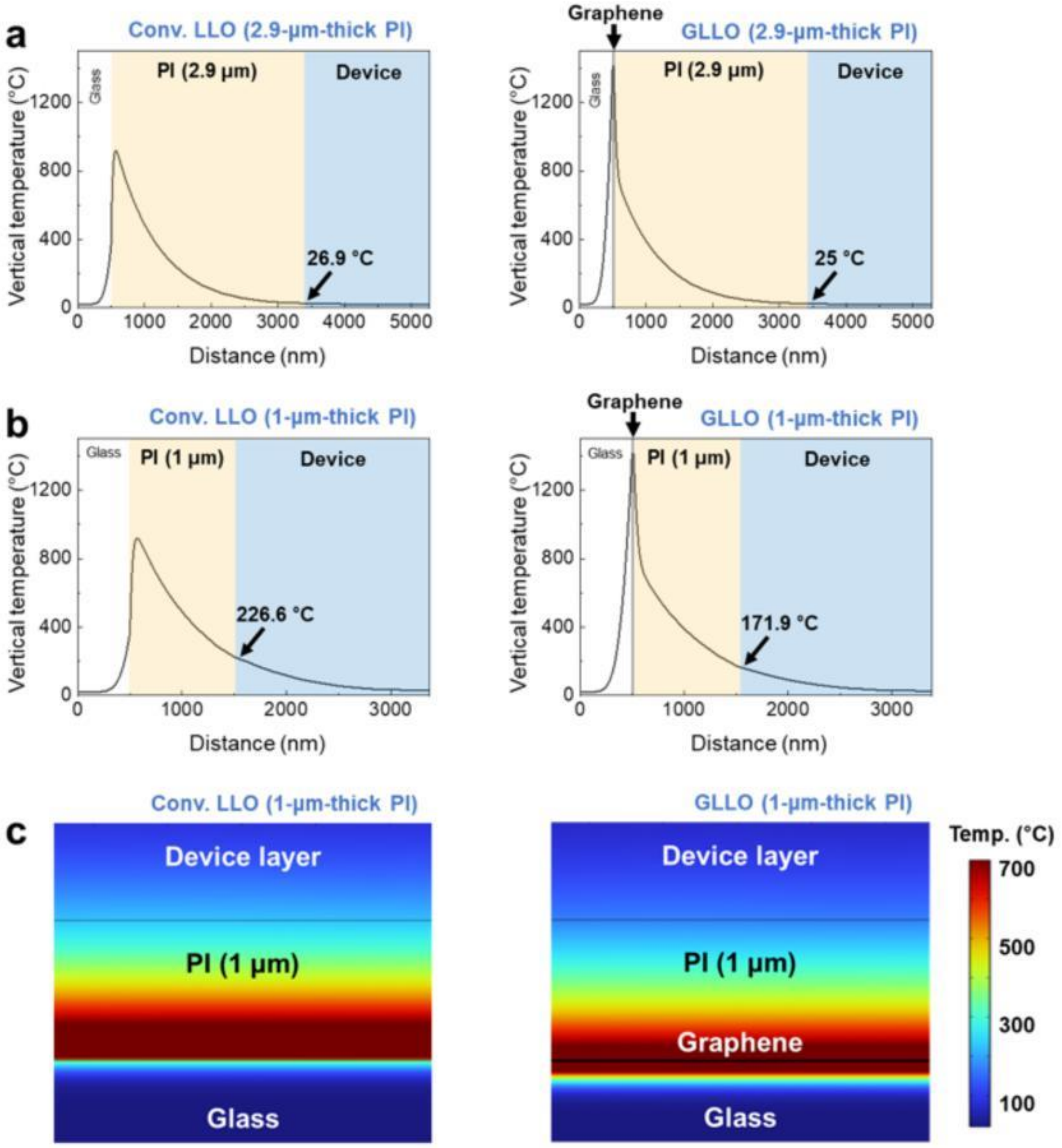
**(#10)** *For laser lift-off ultrathin devices, whether the interface Gr layer has the effect of protecting the device from high temperature since heat can be dissipated from the side of the light spot. Can the paper provide a simulation of this effect? How about thinner PI films (~1 um)?*

**Response to the first reviewer's comment (#10):** Thank you for the constructive suggestion. In compliance with the reviewer's comment, we further examined the effect of graphene on the thermal protection of a fabricated device layer during the lift-off process (Supplementary Fig. S18). The results indicate that the inserted graphene layer can contribute to reducing the maximum temperature of the device layer on thinner PI films (1 μm) due to the increasing UV light absorption at the interface and lateral diffusion of the absorbed heat during the GLLO process. We have included the corresponding results and discussion in our manuscript as follows.

The 2nd paragraph of page 16:

~ Furthermore, the effect of graphene on the thermal protection of a device layer during the lift-off process was investigated by analyzing the temperature distributions. The results demonstrate that the graphene layers can contribute to reducing the maximum temperature of the device layer on thinner PI films, by increasing UV light absorption at the interface and dispersing the heat in a lateral direction during the GLLO process (Supplementary Fig. 18)

**Supplementary Fig. 18**:



**Supplementary Fig. 18 │** The effect of graphene on the thermal protection of a device layer. **a**,**b**, Temperature distributions of a device layer integrated simulation model during the conventional and GLLO processes: (**a**) 2.9-μm-thick PI film, (**b**) 1-μm-thick PI film. **c**, Differences in temperature profile between the conventional and GLLO processes. The graphene layer reduced the device temperature, and the effect was more significant in thinner PI films.

## The second reviewer’s comments

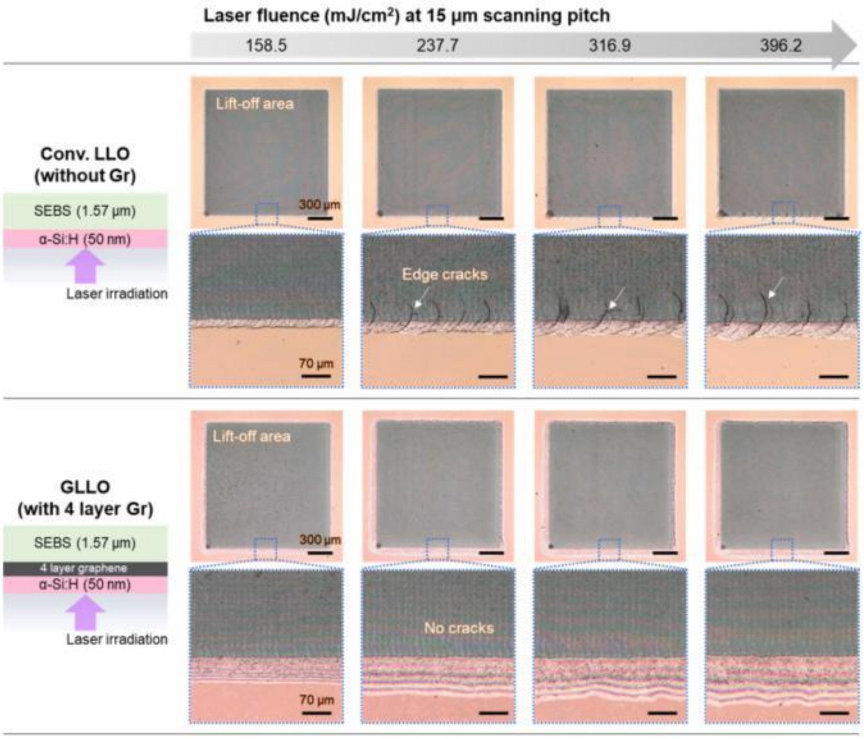
*In this paper, graphene is used to facilitate the laser lift off (LLO) of thin polyimide films from a glass carrier. Compared to conventional LLO, the graphene LLO process (“GLLO) ” changes the bubble formation between the polyimide (PI) film and the glass during laser ablation. With GLLO the bubbles are wider and flatter, producing less strain the PI film and thus less mechanical deformation. The authors attribute this to the enhanced absorption and lateral heat conduction in the graphene, and weaker adhesion of the PI to the graphene. The result is that GLLO is shown to have a wider process window than conventional LLO, and the resulting PI films tend to be less wrinkled. A demonstration of using GLLO to release fully-fabricated OLEDs on a thin PI film shows that – unlike the conventional LLO – the OLEDs remain operational, with little to no change in performance after GLLO. Overall, the use of graphene is an interesting idea that does provide some improvement to the LLO process, and the ability release the OLEDs without any noticeable change in performance is impressive. However, it is not clear to me that this work would have broad enough interest for publication in Nature Communications. There are many points on which the paper could be made stronger:*

**(#1)** *The results would be more broadly applicable if this process was demonstrated to work with other substrate materials. It seems likely that (assuming the authors’ hypotheses are correct) that this method would work with many polymer substrates.*

**Response to the second reviewer’s comment (#1):** We appreciate the reviewer’s valuable comment on the applicability to other substrate materials. Several materials can be used in the LLO process, including polyimide (PI), gallium nitride (GaN), lead ziconate titanate (PZT), hydrogenated amorphous silicon (α-Si:H), and amorphous gallium oxide (α-$GaO_x$). To validate the expandability of the GLLO method to other polymer substrates, we adopted an α-Si:H sacrificial layer and polystyrene-*b*-poly(ethylene-*co*-butylene)-*b*-polystyrene (SEBS) elastomer film, which is a representative thin polymer substrate for stretchable electronics. The results indicate that although the SEBS film exhibits superior elongation of approximately 800 % [M. A. Rahman et al., *Science Advances* **7**, eabk2451 (2021)], edge cracks were observed under high laser fluence conditions in the conventional LLO processes. In contrast, the graphene layer prevented cracking during the lift-off processes, implying the effectiveness of the GLLO method in a different material system. We have included the results and discussion in our manuscript as follows.

The 1st paragraph of page 8:

~ OM images of the other specimens after the conventional LLO and GLLO processes are shown in Supplementary Figs. 7 and 8, respectively, showing apparent differences between the lift-off results of GLLO and conventional LLO methods. Furthermore, the effectiveness of the GLLO method in reducing damage during the lift-off process was also confirmed in a different material system, indicating the method's expandability (Supplementary Fig. 9).



**Supplementary Fig. 9 |** Application of the GLLO method to a different material system composed of a 50-nm-thick hydrogenated amorphous silicon (α-Si:H) layer and a 1.57-μm-thick polystyrene-*b*-poly(ethylene-*co*-butylene)-*b*-polystyrene (SEBS) elastomer film. The α-Si:H was utilized as a sacrificial layer for the laser ablation process, and the graphene layer was inserted between the α-Si:H layer and SEBS film. The results indicate that edge cracks were observed under high laser fluence conditions in the conventional LLO processes, although the SEBS film exhibits superior elongation of approximately 800 %. In contrast, the graphene layer prevented cracking during the lift-off processes, demonstrating the effectiveness of the GLLO method in a different material system not only the PI film.

**(#2)** *The focus on ultra-thin (sub-5 micron) polyimide seems overly narrow, and not well-motivated. There are many substrates that may be a better choice if the goal is "conformal contact onto soft and curvilinear surfaces", because PI does not stretch. The title of the manuscript includes "for ultrathin displays", and the authors use "implantable and wearable information displays" as a motivation for such thin polyimide.* ***(Q2-1)*** *What is an "implantable" display, and how can it be viewed by a user if it is implanted in the body?* ***(Q2-2)*** *This use case needs more explanation to justify why the ultrathin polyimide is needed. On the other hand, there are many applications where thicker polymer substrates would be beneficial.* ***(Q2-3)*** *Would this process work (and still provide any benefits over conventional LLO) if the substrate was NOT ultrathin PI? Is a thicker substrate more resistant to the wrinkling effect, and thus the GLLO is not really necessary or beneficial? The key point here is that the work is narrowly focused on ultrathin PI, the need for such an ultrathin substrate is not well motivated, and the authors don't provide evidence that GLLO would provide benefits if the PI substrate were thicker.*

**Response to the second reviewer's comment (#2):** Thank you for the constructive comments. Here are our responses to each comment.

**(Q2-1) & (Q2-2)**: We agree with the reviewer's comments that the term "implantable display" is confusing and that more explanation is needed to justify why the ultrathin polyimide film is necessary. To support the motivation of this research, we emphasized that the mechanical reliability and stretchability of stretchable electronics are enhanced when thinner PI films are applied to the structures [added references 8, 9]. Moreover, we revised the expression 'implantable and wearable displays' to 'implantable and wearable photonic healthcare devices' [added references 10-12] as follows.

> The 1st paragraph of page 3:
>
> ~ Meanwhile, in stretchable display applications, the reduction of the thickness of PI films is advantageous for enhancing stretchability and mechanical reliability[8,9]. Moreover, next-generation applications, such as implantable and wearable photonic healthcare devices[10-12], require the use of ultrathin (sub-5 μm) substrate thickness to allow conformal contact onto soft and curvilinear surfaces based on their extreme flexibility[13]. ~

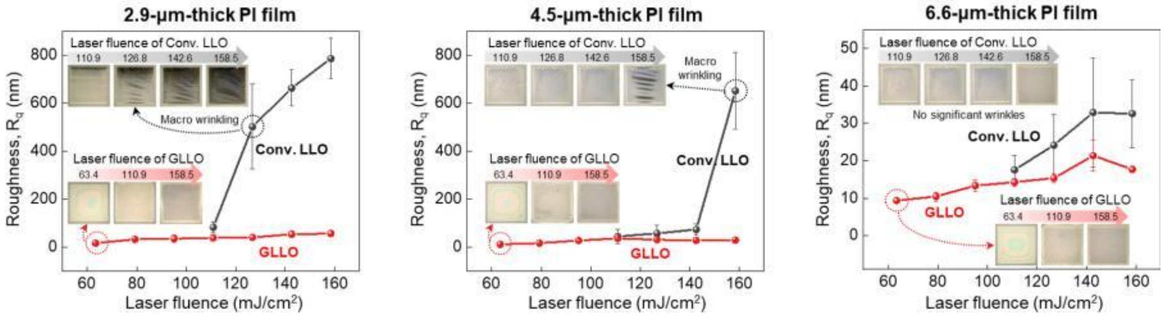
Added References:

8. Jang, H.-W. Kim, S. K & Yoon, S.-M. Impact of polyimide film thickness for improving the mechanical robustness of stretchable InGaZnO thin-film transistors prepared on wavy-dimensional elastomer substrates, *ACS Appl. Mater. Interfaces* **11**, 34076-34083 (2019).
9. Song, H. et al., Highly-integrated, miniaturized, stretchable electronic systems based on stacked multilayer network materials, *Sci. Adv.* **8**, eabm3785 (2022).
10. Lee, G.-H. et al., Multifunctional materials for implantable and wearable photonic healthcare devices, *Nat. Rev. Mater.* **5**, 149-165 (2020).
11. Cho, E. H. et al., Wearable and wavelength-tunable near-infrared organic light-emitting diodes for biomedical applications, *ACS Appl. Mater. Interfaces* **15**, 5741557426 (2023).
12. Kim H. et al., A flexible and wavelength-designable polymer light-emitting diode employing sandwich-encapsulation for wearable skin rejuvenation photomedicine, *Adv. Mater. Interfaces* **8**, 2100856 (2021).

**(Q2-3)**: In response to the reviewer's comment, we investigated the effectiveness of the GLLO method on thicker PI films, specifically 4.5-µm-thick and 6.6-µm-thick (Supplementary Fig. 10). It was observed that the plastic deformation was also reduced even in thicker PI films, although the differences were mitigated under low laser fluence conditions due to the third-power relationship between bending stiffness and film thickness. In this regard, higher laser fluence, such as those used in excimer lasers, is anticipated to enhance the effectiveness of the GLLO method in thicker PI films, not just ultrathin ones. Moreover, the graphene layer still enabled the lift-off process of the thicker PI films at a significantly lower laser fluence of 63.4 mJ/cm$^2$ compared to the conventional LLO method. We have added the experimental results and discussion in the manuscript to demonstrate the influence of PI film thickness.

The 2nd paragraph of page 8:

~ These results indicate that the laser-induced plastic deformation during the ablation process significantly reduced owing to the presence of graphene layers, and this reduction allows for the demonstration of damage-free separation of ultrathin devices. Additionally, the plastic deformation was also reduced even in thicker PI films by the GLLO method, although the differences were mitigated under lower fluence conditions due to the third-power relationship between bending stiffness and film thickness (Supplementary Fig. 10). Moreover, it was observed that the graphene layer still enabled the lift-off process of thicker PI films at a significantly lower laser fluence of 63.4 mJ/cm$^2$.



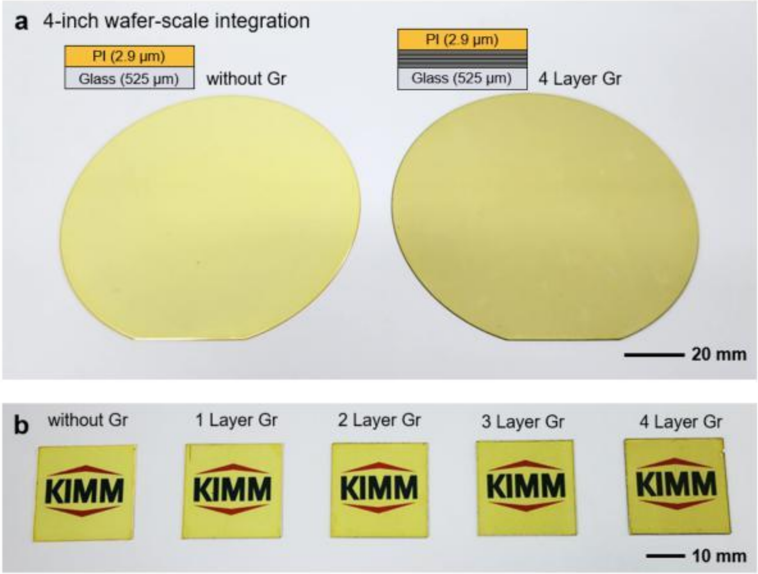
**Supplementary Fig. 10** | Surface roughness of PI films with thicknesses of 2.9, 4.5, and 6.6 μm separated by conventional LLO and GLLO methods. The surface roughness was measured by confocal microscopy, and the inset shows OM images of the lift-off area under each condition. The results indicate that the differences in surface roughness between the conventional LLO and GLLO methods were mitigated due to increased bending stiffness in thicker PI films. The integrated graphene layer enabled a lower laser fluence lift-off process.

**(#3)** *The authors claim that the "graphene enables large-area integration" (pg 4). However, liftoff area demonstrated here is only 1.5 x 1.5 mm2 for all devices, which is quite small. Given that this process relies on graphene flakes that are transfer-printed with a stamp onto the glass substrate, it is hard to see how this could be reasonably scaled to be relevant for large-area displays.*

**Response to the second reviewer's comment (#3):** We appreciate the reviewer's comment. The large-area processability is an important characteristic of CVD graphene. The small lift-off area of 1.5 x 1.5 mm$^2$ is used solely for investigating the LLO performance in this work. In addition, we lifted off 25 x 25 mm$^2$ ultrathin PI films to demonstrate the OLED applications shown in Figs. 5 and 6. Moreover, to provide more intuitive data on the large-area processability and programmability of the graphene layer, we have added photographs of our 4-inch wafer scale and 25 x 25 mm$^2$ square samples as a supplementary figure as follows.

The 1st paragraph of page 5:

The glass carrier–graphene–PI film structured specimen was prepared by transferring the graphene layer onto the carrier and spin coating the PI film. In this process, the graphene layer was transferred layer-by-layer using a roll-to-roll manner, controlling the number of layers (Supplementary Fig. 1).



**Supplementary Fig. 1 │** Prepared glass carrier–graphene–PI film structured specimens. **a**, 4-inch wafer-scale specimens without graphene and with the integration of 4-layered graphene. **b**, 25 x 25 mm$^2$ square specimens for various numbers of integrated layers.

**(#4)** *The authors only include one brief paragraph summarizing other approaches to reduce laser-induced damage during LLO. If this is a problem with very broad interest (e.g., sufficiently broad to warrant publication in Nature Communications), I would suspect that there had been more previous efforts to solve this problem that should be discussed here. For example, there are at least two groups that have used a photonic lift-off process to delaminate flexible (opto)electronics on polyimide substrates from glass carriers. They also rely on an interfacial layer that absorbs the light and laterally spreads the heat at the interface. (See: Liu et al, "Photonic Lift-off Process to Fabricate Ultrathin Flexible Solar Cells", ACS Appl. Mater. Interfaces 2021, and Weidling et al, "Large-area photonic lift-off process for flexible thin-film transistors", npj Flexible Electronics 2022.)*

**Response to the second reviewer's comment (#4):** We appreciate the reviewer's comment regarding previous studies, particularly those on photonic lift-off (PLO) processes. Although the PLO process utilizes a "light-absorbing layer (LAL) or light-to-heat conversion layer (LTHC)" which converts light energy into thermal energy, there are two significant differences between the GLLO and PLO methods.

First, the PLO method focuses on developing extremely high throughput lift-off processes using high-intensity light pulses from a flash lamp. However, the required energy density of the flash lamp is approximately 2.61 ~ 4.55 J/cm$^2$, which is 40 to 70 times higher than that of the GLLO method (63.4 mJ/cm$^2$), causing damage to sub-10-μm PI films. Consequently, the PLO method is unsuitable for lifting off the ultrathin PI films which is a primary target of this work. Second, the LALs in the PLO method consist of metal thin films for enhancing light absorption, such as Mo and W/Ti alloys, which have thicknesses of hundreds of nanometers range. In contrast, the synthesized CVD graphene used in our work has an atomic layer thickness, which not only facilitates the lateral spreading of absorbed photothermal energy but also reduces interfacial adhesion. These differences make possible the successful lift-off of sub-3-μm-thick PI films using the GLLO method.

To provide a comprehensive discussion on previous efforts, we have included a discussion in terms of high-throughput processes in our manuscript, with appropriate references [34-37] as follows.

The 2nd paragraph of page 21 (Discussion section):

In terms of performance, the GLLO method achieved successful lift-off of the significantly thinner PI films than the previous LLO studies[5-7,13-15,18,19], offering a wide process window and minimal plastic deformation. Moreover, the GLLO method requires only a single pass irradiation of a low fluence laser with a wide scanning pitch owing to the graphene layer, enabling higher throughput processes compared to the previously reported multiple irradiations of low fluence lasers. Meanwhile, the photonic lift-off (PLO) method, which employs high-intensity light pulses of flash lamps instead of lasers and relatively thick metallic light-absorbing layers such as Mo and W/Ti alloys, enables extremely high throughput processes[34-37]. However, the required light energy density of the PLO processes is much higher than that of the LLO processes, potentially causing damage to sub-10-μm PI films. Considering these characteristics, it can be concluded that the GLLO method allows for high throughput and precision lift-off of ultrathin PI films.

34. Liu, W., Turkani, V. S., Akhavan, V. & Korgel, B. A. Photonic lift-off process to fabricate ultrathin flexible solar cells. *ACS Appl. Mater. Interfaces* **13**, 44549–44555 (2021).

35. Weidling, A. M., Turkani, V. S., Akhavan, V., Schroder, K. A. & Swisher, S. L. Large area photonic lift-off process for flexible thin-film transistors. *npj Flex. Electron.* **6**, 14 (2022).

36. Lee, S. I. et al. Xenon flash lamp lift-off technology without laser for flexible electronics. *Micromachines* **11**, 953 (2020).

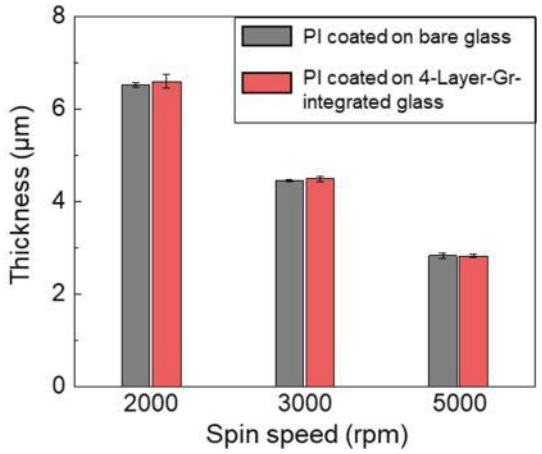
37. Jang, S. H. et al. Investigation of the chemical structure of ultra-thin polyimide substrate for the xenon flash lamp lift-off technology. *Polymers* **13**, 546 (2021).

**(#5)** *Is the PI thickness of 2.9 um measured on substrates with and without the graphene layer to verify that the PI thickness is the same? If so, it is not stated or shown in the Supporting Info. The authors argue that the graphene surface and the interaction with the PI improves the lift-off; it would not be surprising if that difference in surface interaction also caused the spin-coated PI to wet and spread differently, resulting in films of different thicknesses on substrates with graphene vs without graphene. This should be clarified to ensure that the comparison is valid.*

**Response to the second reviewer's comment (#5):** We appreciate the reviewer's valuable comment on the thickness of fabricated PI films. To address this point, we measured the PI thickness on the substrate with and without graphene layers under various spin coating conditions. The results indicate that the presence of graphene layers does not significantly influence the thickness of the spin-coated PI films. We have revised our manuscript as follows to reflect these results.

The 1nd paragraph of page 5:

~ The thickness of the spin-coated PI film, the target lift-off material, was fixed at 2.9 μm (Supplementary Fig. 2), which is significantly thinner than the reported thickness in previous LLO studies[5-7,13-15,18,19]. In this regard, the presence of graphene layers on the glass carrier does not significantly influence the thickness of the fabricated PI films (Supplementary Fig. 3). ~



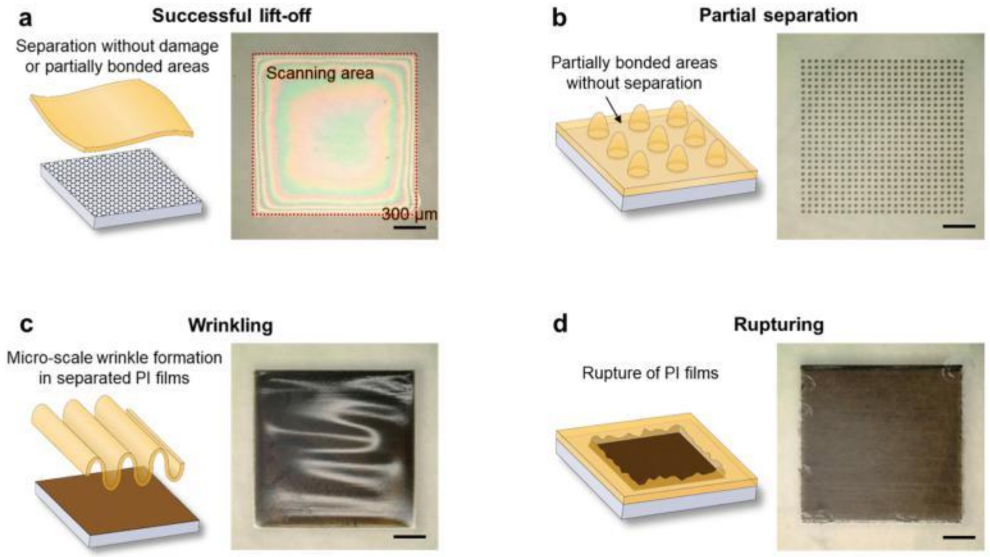
**Supplementary Fig. 3** | The measured thickness of the PI films spin-coated on bare glass carrier and 4-layer-graphene-integrated glass carrier.

**(#6)** *What is the criteria for “successful lift-off” (pg 7)? The authors categorize the lift-off results into 4 groups, but it is unclear what roughness/wrinkling/??? would be acceptable and categorized as “successful”. This should be clarified and the substrates should be more thoroughly analyzed.*

**Response to the second reviewer’s comment (#6):** We are thankful for the reviewer's comment. To address the unclear information on the classification of the lift-off results, we have provided detailed explanations in the manuscript as follows.

The 1st paragraph of page 7:

The results of each LLO process were categorized into partial separation, wrinkling, rupturing, and successful lift-off. The criterion for the successful lift-off is defined as the separation of the PI films without micro-scale wrinkles, ruptures, and partially bonded areas. Detailed explanations and representative images for each classification are provided in Supplementary Fig. 5.



**Supplementary Fig. 5 | ** Classification of the lift-off results and representative OM images. **a**, Successful lift-off is defined as the separation of the PI films without micro-scale wrinkles, ruptures, and partially bonded areas. **b**, Partial separation is defined as the presence of remaining bonded areas without separation. **c**, Wrinkling is defined as the presence of micro-scale wrinkles in the separated PI films. **d**, Rupturing is defined as the tearing of separated PI films due to mechanical deformation exceeding their fracture toughness.

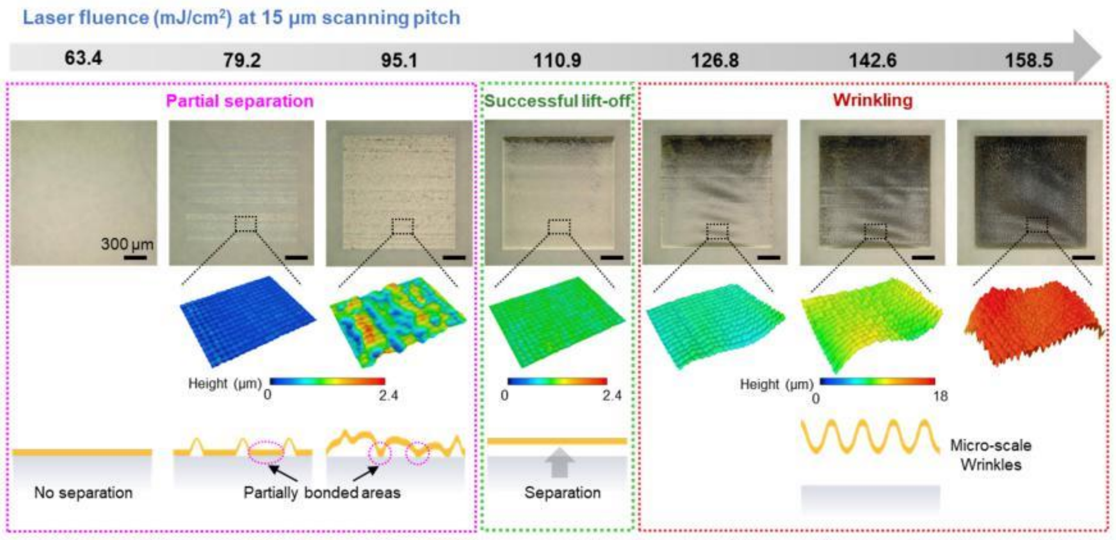
**(#7)** *Figure 2d and the Supplementary Info show optical microscopy images of devices with different (G)LLO conditions. These images are not very informative without better description and/or more detailed analysis.*

**Response to the second reviewer's comment (#6):** We appreciate the reviewer's valuable comment. In compliance with the reviewer's comment, we have revised the manuscript to provide a more detailed analysis and better description.

The 2nd and 3rd paragraphs of page 7:

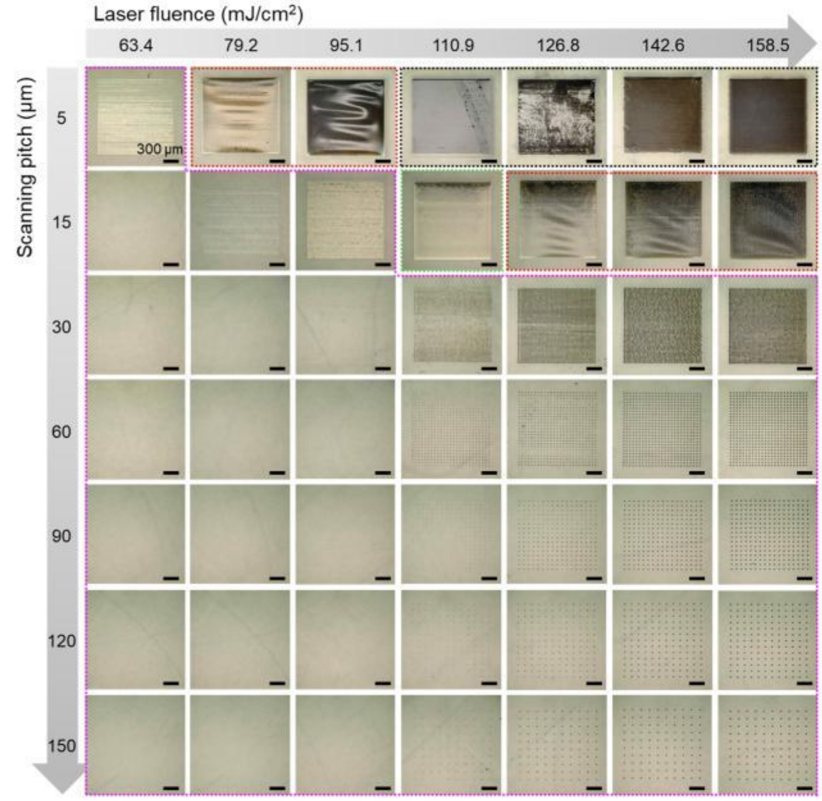
The optical microscopy (OM) images in the first row of **Fig. 2d** verify the aforementioned lift-off behavior. More detailed analysis on the lift-off results using the conventional LLO method is provided in Supplementary Fig. 6.

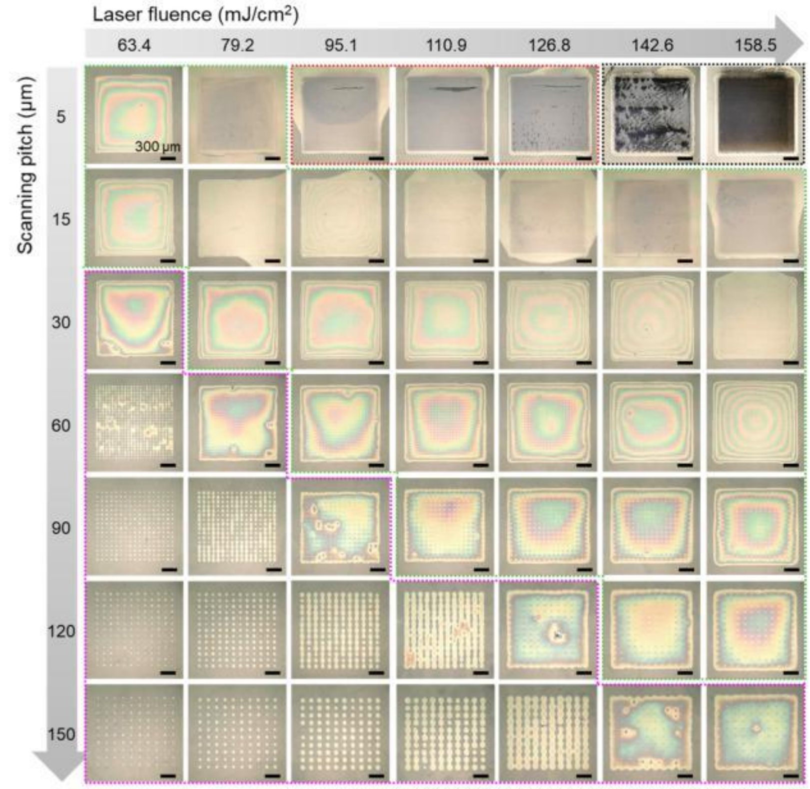
In contrast, the GLLO process exhibited a wide process window for successful lift-off (**Fig. 2c**). When considering the scanning pitch of 15 μm (indicated by the blue arrow in **Fig. 2c**), all laser fluences ranging from 63.4 to 158.5 mJ/cm$^2$ successfully separated the ultrathin PI film without significant damage. The OM images in the second row of Fig. 2d demonstrate the successful lift-off results, leading to moiré fringes under low laser fluence conditions[23].



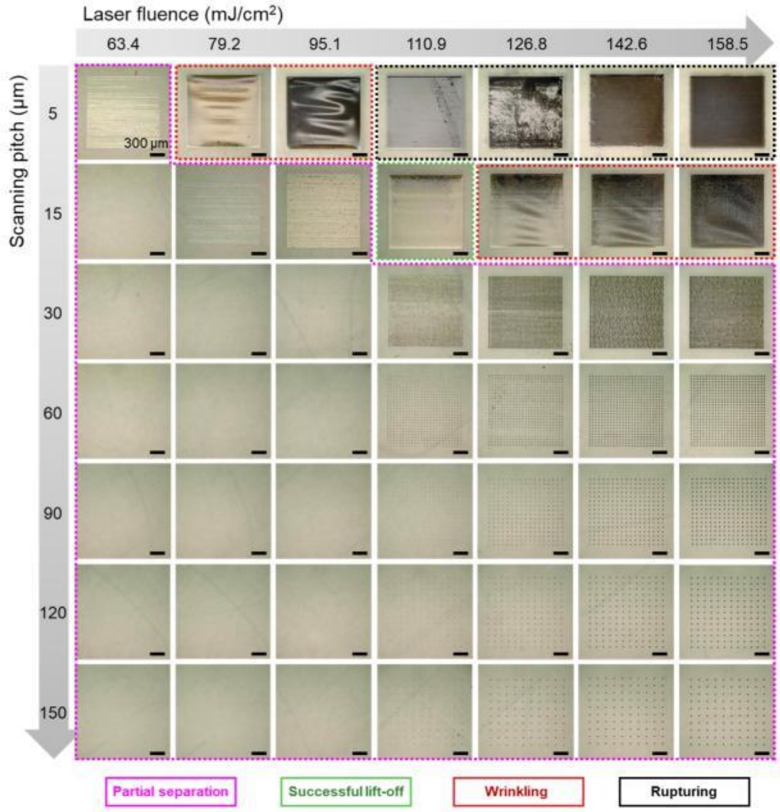
**Supplementary Fig. 6 |** Analysis of lift-off OM images obtained by the conventional LLO method. Confocal microscopy images (scale factor: 10) and corresponding schematics illustrate the lift-off behaviors of the PI film in detail under various laser fluence conditions.

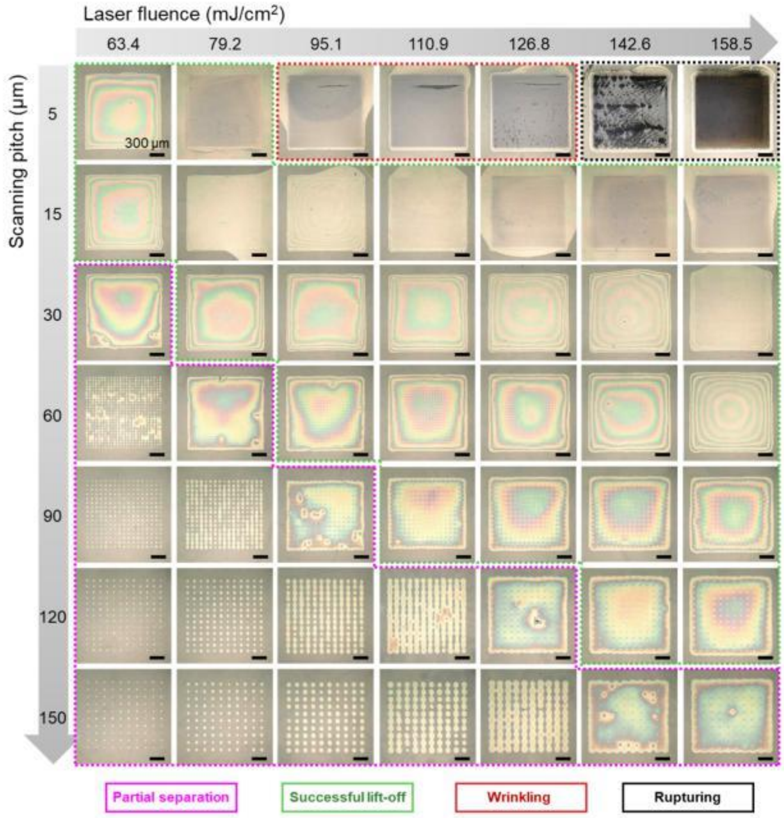
Supplementary Figs. 7 and 8 (Before):





Supplementary Figs. 7 and 8 (After):



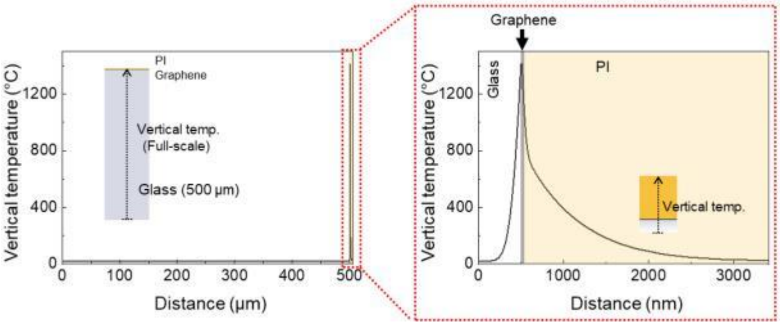


**(#8)** *The simulation results shown in Figure 4 lead to many questions.* ***(Q8-1)*** *Is a temperature of 1200 degC on the glass/PI interface actually feasible and reasonable??* ***(Q8-2)*** *Why is the glass in the simulation only 300 nm thick? What boundary conditions were applied at the back of the glass (distance = 0 nm) to keep the temperature there near room temperature? Is absorption in the glass included in the simulation (there is no absorption coefficient for glass included in Supp Table 1)?* ***(Q8-3)*** *What fraction of the laser power is absorbed in the graphene vs the polyimide?*

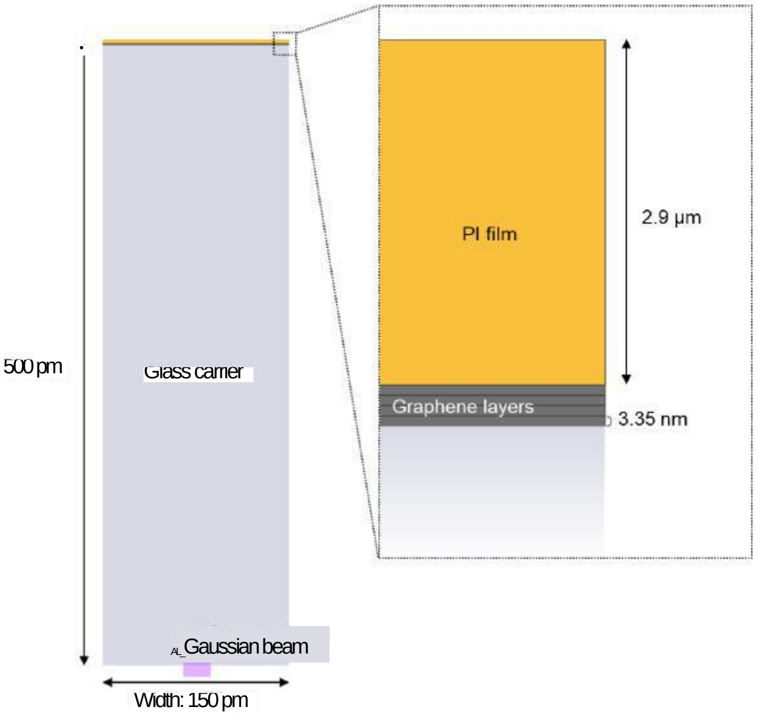
**Response to the second reviewer's comment (#8):** **(Q8-1)** We appreciate the reviewer's valuable comments on the simulation results. Several studies have reported similar temperature levels of 500 ~ 1500 °C during the laser ablation process of PI films [references 18, 19, 26], and these high temperatures typically persist for only a few nanoseconds. Therefore, the obtained results in this work are reasonable.

**(Q8-2)** The simulation model in our original manuscript was designed to visualize the relative differences in temperature profiles during the ablation process, with respect to the number of graphene layers (**Fig. 4c**). Consequently, the simulation scale was reduced compared to the actual specimens due to the significantly different thicknesses of the materials. Moreover, the glass carrier was assumed to be transparent (no absorption of the light energy), because the absorption coefficient of the glass ($2.107\ \mathrm{cm^{-1}}$) is significantly smaller than those of the PI and graphene which are $2\times10^{4}\ \mathrm{cm^{-1}}$ and $2.4\times10^{6}\ \mathrm{cm^{-1}}$, respectively. However, to provide more accurate temperature distributions, we have incorporated the actual specimen structures into a full-scale simulation model (Supplementary Fig. 14), and considered the absorbing characteristic of the glass carrier (Supplementary Table 1).

For the boundary conditions, an initial temperature of 20 °C was assigned to the entire structure. We found that there is no significant temperature variation at the back of the glass due to its low light absorption, and solely heat conduction of photothermal energy absorbed in graphene and PI films influences the temperature of glass (up to 500 nm). The figure below represents the full-scale distribution of temperature in the vertical direction.



## Supplementary Fig. 14:



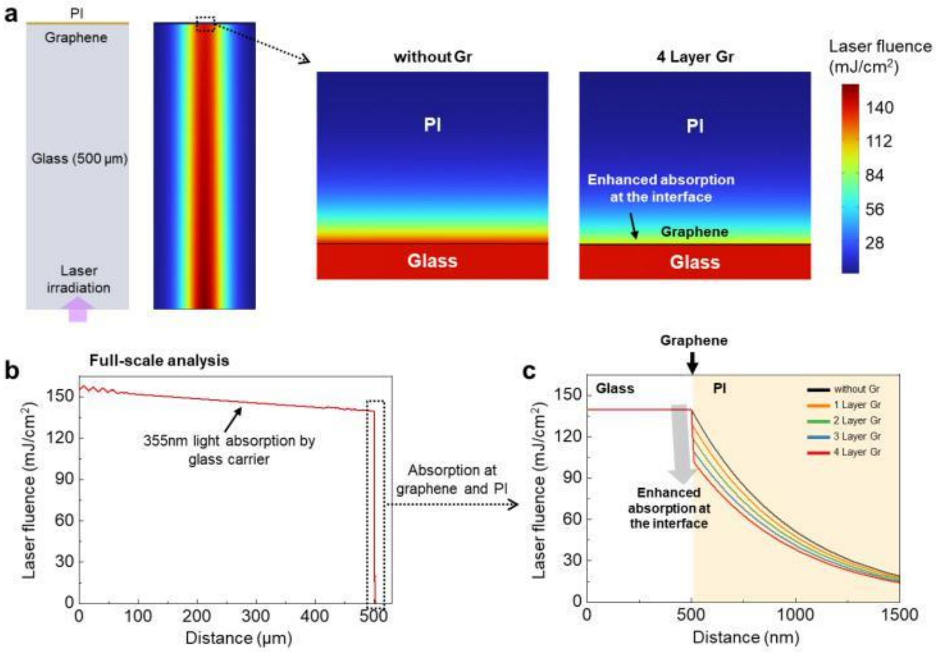
Supplementary Fig. 14 | Full-scale simulation model for accurate analysis of temperature distributions.

## Supplementary Table 1 (Glass absorption coefficient added):

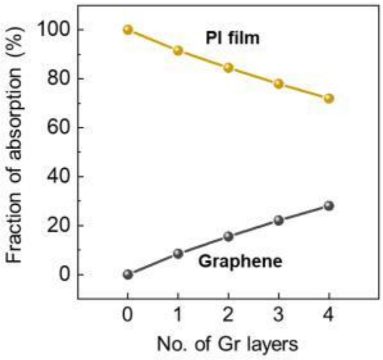
| Parameter | Value | Reference |
|---|---|---|
| Density of PI [g-cm$^{-3}$] | 1.42 | [26] |
| Density of graphene [g-cm$^{-3}$] | 2.21 | [38] |
| Density of glass [g-cm$^{-}$1 | 2.37 | [39] |
| Specific heat of PI [J-g$^{-'}$-K$^{-1}$] | 2.55 - 1.59 x exp[(T$_o$—T)/460] | [26] |
| Specific heat of graphene **[J-g$^{-'}$-K$^{-1}$]** | 0.709 | [38] |
| Specific heat of glass [..l-g$^{-1}$-K$^{-}$9 | 1.53 — 0.79 x exp[(T$_o$—T)/638] | [39] |
| Thermal conductivity of PI [W.cm$^{-1}$-K$^{-}$1 | 1.55 x 10$^{-,}$(171$^{-}_{0}$)$^{,2,}$ | [26] |
| Thermal conductivity of graphene [W.cm$^{-'}$K$^{-}$1 | 26.4 x (T/T$_0$)$^{-1}$-$^{38}$ | [40] |
| Thermal conductivity of glass [W-cm$^{-1}$.K$^{-}$9 | 7.9 x 10$^{-4}$T$^{,43}$ | [39] |
| Absorption coefficient of PI [cm$^{-1}$] | 0.2 x 10$^{5}$ | [41] |
| Absorption coefficient of graphene [cm$^{-1}$] | In(10) x 0.035 / (6 x 10$^{-7}$) | - |
| Absorption coefficient of glass [cm$^{-1}$] | 2.107 | Product datasheet provided by Corning |

**(Q8-3)** Thank you for the insightful comment. The variation in laser fluence was investigated using the FEA simulation to evaluate the fractions of laser power absorbed by the graphene layer and the PI film (Supplementary Figs. 15 and 16). The results indicate that approximately 28% of UV light was absorbed in the graphene layers, while the remaining 72% was absorbed in the PI film during the GLLO processes with 4-layered graphene.

**Supplementary Figs. 15 and 16**:



**Supplementary Fig. 15 │ a**, Laser fluence profiles obtained in the FEA simulation. The graphene layer enabled the enhancement of UV light absorption at the interface. **b**, Laser fluence distribution in the vertical direction of the full-scale model. **c**, Laser fluence distribution at the graphene and PI film.



**Supplementary Fig. 16 │** Fraction of UV light absorption by the graphene and PI film.

**(#9)** *The authors mention several times that reusing the glass substrates would be beneficial and that re-use may be possible with the graphene-coated substrates: pg 3 "the deposited sacrificial layer cannot be reused", pg 6 "thick carbonaceous residues remaining after the process, impeding the recycling of the expensive glass carrier", pg 8 "facilitate the recycling of the graphene-integrated glass carrier", pg 18 "indicating the potential reusability of the expensive graphene-integrated glass carrier". However, they do not actually provide evidence that the graphene-coated substrates CAN be re-used, and thus these arguments are not well-founded and might mislead the reader.*

**Response to the second reviewer's comment (#9):** We greatly appreciate the reviewer's valuable comment on the reusability. In compliance with the reviewer's comment, we performed additional experiments to investigate the reusability (Fig. 6) and discussed the results. Please refer to the revised manuscript (Pages 20 and 22).

The page 20:

Furthermore, the reusability of the graphene-integrated glass carrier was investigated by repeating the processes of spin coating the 2.9-μm-thick ultrathin PI substrate, OLED fabrication, and GLLO sequentially (**Fig. 6a**). The OLEDs fabricated in the first cycle were separated with a laser fluence of 63.4 mJ/cm$^2$. Consistent with the results in **Fig. 5**, the devices maintained their current density-voltage-luminance properties and current efficiency before and after the GLLO process (**Figs. 6b** and **6c**). The reusability of the graphene-integrated glass carrier was evaluated by considering the electrical performance variation of OLEDs fabricated on reused carriers. In this regard, delamination of the OLEDs in the second and third fabrication cycles required a slightly higher laser fluence of 79.2 mJ/cm$^2$ for complete lift-off. Moreover, the analysis results indicate that although the OLEDs remained operational after the GLLO process, a slight degradation in luminance was observed (**Fig. 6b**). We carefully anticipate that the higher laser fluence requirement and degraded luminance resulted from the thin carbonaceous PI residues represented in **Fig. 2f**. In other words, the thin residues covered the integrated graphene layer, hindering the reduction of interfacial adhesion of newly coated PI substrates and increasing the distance from the graphene layer. Therefore, more effort will be needed to completely remove the carbonaceous residues, ensuring the intact reusability of the graphene-integrated carrier.

**Fig. 6**:

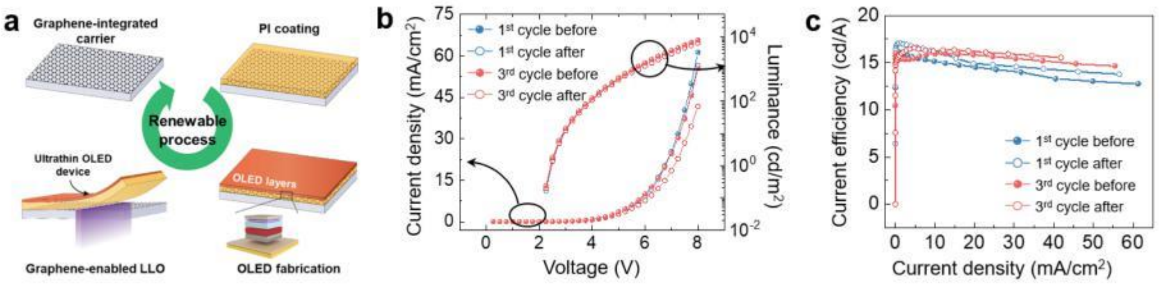


**Fig. 6 | Reusability of the graphene-integrated carrier. a,** Schematic illustration of the renewable process of the graphene-integrated carrier. **b,c,** Electrical performance variation of the OLEDs fabricated in the first and third cycles, before and after the GLLO process. (**b**) current density-voltage-luminance properties and corresponding (**c**) current efficiency-current density curve.

The 1st paragraph of page 22 (Discussion section):

~ Concerning the reusability of the graphene-integrated carrier, the separation of ultrathin OLED devices fabricated on reused carriers was achieved with a slightly higher laser fluence irradiation. Although the experiment demonstrated early-stage reusability, a slight degradation in OLED luminance was observed after the GLLO process. This degradation in LLO performance may be attributed to the remaining carbonaceous PI residues on the carrier which cover the graphene layers. It is anticipated that further study is required for the complete reusability of the graphene-integrated carrier, even though the presented GLLO method reduced the thickness of carbonaceous PI residues by approximately 92.8 % compared to the conventional LLO methods. Several endeavors, such as optimization of laser irradiation conditions and beam profiles, and advances in interlayer materials, will be worthwhile for enhancing LLO performance, reusability, and industrial applicability. Finally, we believe that this work will open up new possibilities for utilizing CVD-grown graphene in laser-based manufacturing applications, such as emerging displays, wafer-level packaging, and energy harvesting devices.

**(#10)** *The authors should specify the "controlled temperature and humidity" conditions that were used for the LLO experiments (pg 20).*

**Response to the second reviewer's comment (#10):** We appreciate the reviewer's comment. In compliance with the comment, we have specified the conditions of "controlled temperature and humidity" in the manuscript as follows.

The 1st paragraph of page 24 (Methods section):

~ These LLO experiments were conducted in a cleanroom environment where the temperature and relative humidity were controlled at 22 °C and 40 %, respectively, under ambient air conditions.

## The third reviewer’s comments

*The authors reported Graphene-assisted laser lift-off (GLLO) to solve problems of conventional LLO technologies. The graphene can facilitate lateral heat diffusion, reduce adhesion between carrier glass and polyimide, and enhance UV absorption, resulting in wide process windows. Although this manuscript contains extensive data and their discussion, but we could find insufficient explanation. Moreover, this work is quite specific for publication in Nature Communications. I therefore do not feel that is suitable for publication in Nature Communications. I think the manuscript to be better suited for sister journals. Here are the points which need to be addressed before it can be considered for publication.*

*(#1) The author mentioned that the deposited sacrificial layer cannot be reused after the LLO process, leading to an increase in manufacturing costs. Does graphene can be reused after the LLO process? Are there any experimental results?*

*(#2) The author mentioned that “thick carbonaceous PI residues remaining after the process, impeding the recycling of the expensive glass carrier.” The thickness of the PI residues can be thinner in the GLLO process, but there were still carbonaceous PI residues (~9.5 nm as in Fig. 2f) in the GLLO process. It seems difficult to reuse glass carriers in the GLLO process. Is it possible to reuse glass carriers in the GLLO process?*

**Response to the third reviewer’s comments (#1) and (#2):** We greatly appreciate the reviewer’s valuable comments on the reusability. **(#1)** To validate the reusability of the graphene-integrated carrier, we performed additional experiments as represented in Fig. 6. **(#2)** The results indicate that a slightly higher laser fluence of 79.2 mJ/cm$^2$ is required for the complete separation of the ultrathin OLED devices fabricated on the reused carrier, also a slight degradation in the luminance of OLEDs was observed after the GLLO process. As the reviewer mentioned, we carefully anticipate that the differences between the OLED devices fabricated on as-prepared and reused GLLO carriers can be attributed to the thin carbonaceous PI residues. The thin residues covered the integrated graphene layers, hindering the reduction of adhesion of newly coated PI substrates and increasing the distance from the graphene layer. Therefore, we are planning to conduct further research to address these issues with several approaches, such as optimizing laser irradiation conditions and beam profiles, and developing advanced interlayer materials. We have included these results and discussion in the manuscript (Pages 20 and 22) as follows.

The page 20:

Furthermore, the reusability of the graphene-integrated glass carrier was investigated by repeating the processes of spin coating the 2.9-μm-thick ultrathin PI substrate, OLED fabrication, and GLLO sequentially (**Fig. 6a**). The OLEDs fabricated in the first cycle were separated with a laser fluence of 63.4 mJ/cm$^2$. Consistent with the results in **Fig. 5**, the devices maintained their current density-voltage-luminance properties and current efficiency before and after the GLLO process (**Figs. 6b** and **6c**). The reusability of the graphene-integrated glass carrier was evaluated by considering the electrical performance variation of OLEDs fabricated on reused carriers. In this regard, delamination of the OLEDs in the second and third fabrication cycles required a slightly higher laser fluence of 79.2 mJ/cm$^2$ for complete lift-off. Moreover, the analysis results indicate that although the OLEDs remained operational after the GLLO process, a slight degradation in luminance was observed (**Fig. 6b**). We carefully anticipate that the higher laser fluence requirement and degraded luminance resulted from the thin carbonaceous PI residues represented in **Fig. 2f**. In other words, the thin residues covered the integrated graphene layer, hindering the reduction of interfacial adhesion of newly coated PI substrates and increasing the distance from the graphene layer. Therefore, more effort will be needed to completely remove the carbonaceous residues, ensuring the intact reusability of the graphene-integrated carrier.

**Fig. 6**:

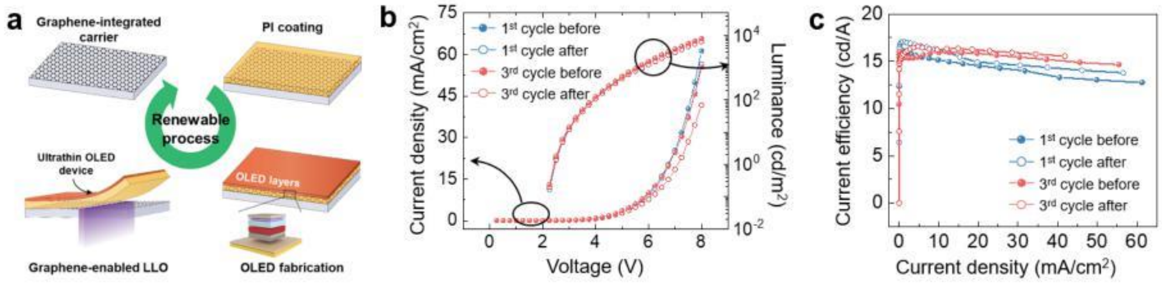


**Fig. 6 | Reusability of the graphene-integrated carrier. a,** Schematic illustration of the renewable process of the graphene-integrated carrier. **b,c,** Electrical performance variation of the OLEDs fabricated in the first and third cycles, before and after the GLLO process. (**b**) current density-voltage-luminance properties and corresponding (**c**) current efficiency-current density curve.

The 1st paragraph of page 22 (Discussion section):

~ Concerning the reusability of the graphene-integrated carrier, the separation of ultrathin OLED devices fabricated on reused carriers was achieved with a slightly higher laser fluence irradiation. Although the experiment demonstrated early-stage reusability, a slight degradation in OLED luminance was observed after the GLLO process. This degradation in LLO performance may be attributed to the remaining carbonaceous PI residues on the carrier which cover the graphene layers. It is anticipated that further study is required for the complete reusability of the graphene-integrated carrier, even though the presented GLLO method reduced the thickness of carbonaceous PI residues by approximately 92.8 % compared to the conventional LLO methods. Several endeavors, such as optimization of laser irradiation conditions and beam profiles, and advances in interlayer materials, will be worthwhile for enhancing LLO performance, reusability, and industrial applicability. Finally, we believe that this work will open up new possibilities for utilizing CVD-grown graphene in laser-based manufacturing applications, such as emerging displays, wafer-level packaging, and energy harvesting devices.

**(#3)** *In figures 3e-3g, how do you measure the geometry of the blister? Is it experimentally determined or calculated? No explanation for this.*

**Response to the third reviewer's comment (#3):** We appreciate the reviewer's comment on the method for measuring the blister geometry in Figs. 3e-3g. The results were obtained through experimentation using confocal microscopy. To provide more clear and exact information, we have revised the manuscript as follows.

The 2nd paragraph of page 12:

According to the explanations, the shape of the blister is the key difference between the conventional LLO and GLLO methods. Therefore, we investigated the blister shape by irradiating a single UV laser pulse to the specimens with different numbers of graphene layers, and experimentally measuring the blister geometry using confocal microscopy (**Figs. 3e** and **3f**).

The caption of **Fig. 3**:

**g,** The representatitve images of blisters at the laser fluence of 110.9 and 79.2 mJ/cm$^2$ (scale factor: 120). The shape and dimension of the blisters were experimentally measured using confocal microscopy.

**(#4)** *In Figure 5b, what materials is interconnection? The authors seem to use an Al cathode, but there is no description of interconnection (might be connected with an Al cathode).*

**Response to the third reviewer's comment (#4):** We are thankful for the reviewer's comment regarding the missing information. For the interconnections, we utilized a conductive tape composed of a Cu-Ni-coated fabric backing with a conductive acrylic adhesive (YCF50, Youngjin Co., LTD.). This tape has a total thickness of 50 µm and offers conductivity in the x-y-z directions. As correctly observed by the reviewer, we tailored pieces of this conductive tape and attached them to the OLED electrodes. The interconnections established by conductive tape provide reliable conductive pads for testing the OLEDs' optoelectronic performance. We have updated the manuscript to detail the interconnection material used as follows.

> The 1st paragraph of page 25 (Methods section):
>
> ~ A 50-µm-thick conductive tape (YCF50, Youngjin Co., LTD.), featuring a Cu-Ni-coated fabric backing and a conductive acrylic adhesive, was attached to the exposed active contact area as interconnection.

Again, we greatly appreciate the valuable comments and critical questions by the reviewers. We believe that we have fully responded to all of their comments and questions and revised the manuscript accordingly.

Thank you for handling our submission and your favorable decision would be greatly appreciated.

Sincerely,

Seungman Kim, Ph.D.
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Department of Ultra-Precision Machines and Systems
Korea Institute of Machinery and Materials (KIMM)
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E-mail: kimsm@kimm.re.kr

## REVIEWER COMMENTS

**Reviewer #1 (Remarks to the Author):**

All the issues that I concerned have been addressed very well. I recommend it to be accepted.

**Reviewer #2 (Remarks to the Author):**

I appreciate the authors' detailed responses and explanations. They have addressed most of the concerns, and have substantially improved their manuscript (including new data and expanded explanatations). In some cases, this new information raised additional questions. comments are below. I have included below the comments/concerns that were not fully addressed in this revision, NCOMMS-24-04978A. Overall, I think the revisions have strengthened the claims made by the authors in this manuscript, but the overall scope and motivation for the work are too narrow for Nature Communications. Perhaps this would be more appropriate in a sister journal with a more targeted audience.

1. My original comment (#1): The results would be more broadly applicable if this process was demonstrated to work with other substrate materials. It seems likely that (assuming the authors' hypotheses are correct) that this method would work with many polymer substrates.

Regarding the revision:
The additional data provided in Supplementary Fig. 9 is not very convincing here, and actually introduces new questions about GLLO. The authors suggest that graphene could potentially improve the lift-off process for a different material system, but the improvement seems to be minimal: all SEBS films were successfully lifted off without partial separation or wrinkling, and even at the highest laser fluence the edge cracks don't propagate very far into the film. More importantly, this additional data does not support the authors' hypothesis on the role of graphene in the GLLO process (lateral heat diffusion resulting in smoother blisters). The authors' statement in Supp Fig 9 caption, "although the SEBS film exhibits superior elongation of approximately 800 %", is unclear/misleading. I assume the 800% elongation is referring to the results published in Rahman et al (2021), and not their own work? If so the authors must include a citation.

2. My original comment (#2): The focus on ultra-thin (sub-5 micron) polyimide seems overly narrow, and not well-motivated. There are many substrates that may be a better choice if the goal is "conformal contact onto soft and curvilinear surfaces", because PI does not stretch. The title of the manuscript includes "for ultrathin displays", and the authors use "implantable and wearable information displays" as a motivation for such thin polyimide. What is an "implantable" display, and how can it be viewed by a user if it is implanted in the body? This use case needs more explanation

to justify why the ultrathin polyimide is needed. On the other hand, there are many applications where thicker polymer substrates would be beneficial. Would this process work (and still provide any benefits over conventional LLO) if the substrate was NOT ultrathin PI? Is a thicker substrate more resistant to the wrinkling effect, and thus the GLLO is not really necessary or beneficial? The key point here is that the work is narrowly focused on ultrathin PI, the need for such an ultrathin substrate is not well motivated, and the authors don't provide evidence that GLLO would provide benefits if the PI substrate were thicker.

Regarding the revision:
This comment has been somewhat addressed by the authors by adding a few references and verifying that the GLLO works with thicker (up to 6.6 micron) PI films. However, even that small increase in PI thickness reduced the wrinkling effect during LLO, thus diminishing the benefit of the GLLO method. I still find the focus of the paper to be a bit narrow, and the motivation for using PI <5 microns thick could have been made more strongly.

4. My original comment (#4): The authors only include one brief paragraph summarizing other approaches to reduce laser-induced damage during LLO. If this is a problem with very broad interest (e.g., sufficiently broad to warrant publication in Nature Communications), I would suspect that there had been more previous efforts to solve this problem that should be discussed here. For example, there are at least two groups that have used a photonic lift-off process to delaminate flexible (opto)electronics on polyimide substrates from glass carriers. They also rely on an interfacial layer that absorbs the light and laterally spreads the heat at the interface. (See: Liu et al, "Photonic Lift-off Process to Fabricate Ultrathin Flexible Solar Cells", ACS Appl. Mater. Interfaces 2021, and Weidling et al, "Large-area photonic lift-off process for flexible thin-film transistors", npj Flexible Electronics 2022.)

Regarding the revision:
The authors added some relevant points of comparison between GLLO and the photonic lift-off (PLO) method in the Discussion section near the end of the paper, which strengthens the paper. However, do they have any evidence that PLO damages ultra-thin PI films? That claim seems to be based solely on the incident energy density used for these two methods. If PLO uses light with a broad spectrum (the energy is distributed over many wavelengths), I'm not sure the energy density can be directly compared to a laser in that way. They should provide evidence or consider removing that suggestion.

[The rest of my original comments have been addressed by the authors.]

**Reviewer #3 (Remarks to the Author):**

I appreciate the author’s great efforts in revising the manuscript. The reviewers' comments have been well responded to, and I am satisfied with the changes made. I recommend the publication of this article in Nature Communications.

August 10, 2024

Dear Reviewers,

We greatly appreciate the valuable comments on the manuscript. We have responded in detail below to the comments and have revised the manuscript as noted. The changes are highlighted in red.

## The first and third reviewers’ comments

***First reviewer’s comment****: All the issues that I concerned have been addressed very well. I recommend it to be accepted.*

***Third reviewer’s comment****: I appreciate the author’s great efforts in revising the manuscript. The reviewers' comments have been well responded to, and I am satisfied with the changes made. I recommend the publication of this article in Nature Communications.*

**Response to the reviewers’ comments:** We greatly appreciate the favorable comments on our manuscript.

## The second reviewer’s comments

*I appreciate the authors’ detailed responses and explanations. They have addressed most of the concerns, and have substantially improved their manuscript (including new data and expanded explanatations). In some cases, this new information raised additional questions. comments are below. I have included below the comments/concerns that were not fully addressed in this revision, NCOMMS-24-04978A. Overall, I think the revisions have strengthened the claims made by the authors in this manuscript, but the overall scope and motivation for the work are too narrow for Nature Communications. Perhaps this would be more appropriate in a sister journal with a more targeted audience.*

*Original comment* **(#1)** *The results would be more broadly applicable if this process was demonstrated to work with other substrate materials. It seems likely that (assuming the authors′ hypotheses are correct) that this method would work with many polymer substrates.*

*Regarding the revision:*
*The additional data provided in Supplementary Fig. 9 is not very convincing here, and actually introduces new questions about GLLO. The authors suggest that graphene could potentially improve the lift-off process for a different material system, but the improvement seems to be minimal: all SEBS films were successfully lifted off without partial separation or wrinkling, and even at the highest laser fluence the edge cracks don’t propagate very far into the film. More importantly, this additional data does not support the authors’ hypothesis on the role of graphene in the GLLO process (lateral heat diffusion resulting in smoother blisters). The authors’ statement in Supp Fig 9 caption, “although the SEBS film exhibits superior elongation of approximately 800 %”, is unclear/misleading. I assume the 800% elongation is referring to the results published in Rahman et al (2021), and not their own work? If so the authors must include a citation.*

**Response to the second reviewer's comment (#1):** We appreciate the reviewer's valuable comment and have made the following revisions in response:

***Citation addition.*** We have added a citation related to the elongation of the SEBS material (Supplementary references, S2) to clarify our statement.

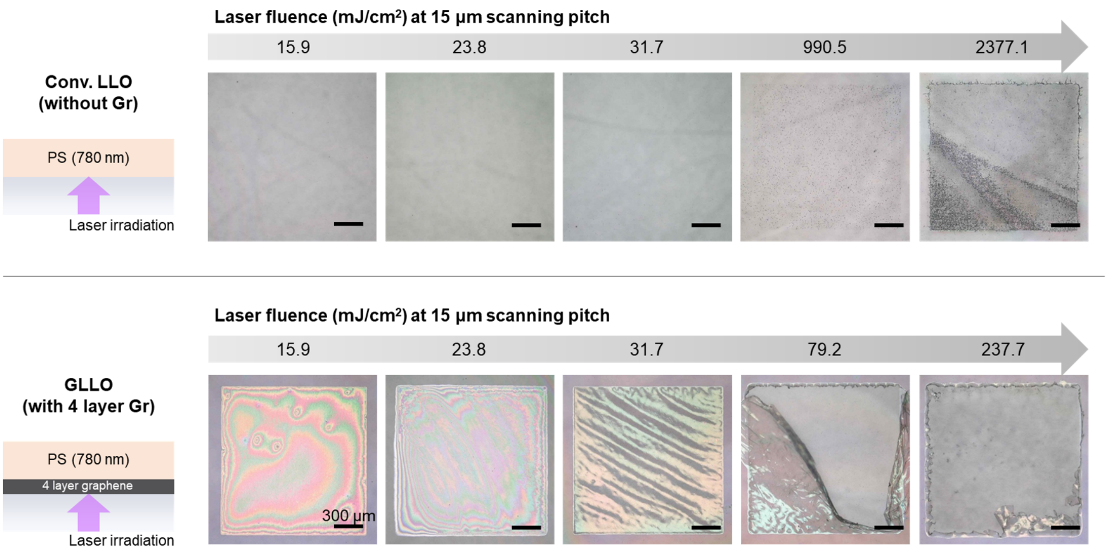
***Explanation for the reduced improvement.*** The relatively reduced improvement is attributed to the use of a sacrificial layer, hydrogenated amorphous silicon (α-Si:H). The sacrificial layer underlying the graphene was ablated by UV laser irradiation instead of the SEBS layer, forming blisters under the graphene. Therefore, the expected adhesion reduction and enhanced UV absorption effects might be minimized, although the lateral heat diffusion and protection effects by the graphene layer can influence the results.

***Reinforcement of the results.*** To address the issue related to the expandability of the method, we performed additional experiments using another material, a 780-nm-thick polystyrene (PS) film. The results indicate that the PS film is significantly difficult to separate by the conventional LLO method due to its low thermal stability. In contrast, the GLLO method enabled laser lift-off of the ultrathin PS film.

We have included the obtained results in the Supplementary Information and revised the manuscript.

The 1st paragraph of page 8:

~ Furthermore, the effectiveness of the GLLO method in reducing damage during the lift-off process was also confirmed in different material systems, indicating the method’s expandability (Supplementary Figs. 9 and 10).



**Supplementary Fig. 9 | ** Application of the GLLO method to a 780-nm-thick polystyrene (PS) thin film. For the conventional LLO process, the low thermal stability of the PS film resulted in no observable differences at low fluence conditions and the formation of an ablation trail without separation at high fluence conditions. In contrast, the GLLO process enabled the lift-off of the PS film under low laser fluence conditions. The separation mechanism of the PS film can be attributed to a photomechanical effect, which indicates that separation occurred at temperatures slightly above the glass transition temperature ($T_g$) due to the coefficient of thermal expansion (CTE) mismatch[S1]. It is anticipated that the graphene layer enlarged the CTE mismatch effect, including the enhancement of interfacial UV absorption, lateral heat diffusion, and adhesion reduction.

## Supplementary references

S1. Kappes, R. S. et al. A study of photothermal laser ablation of various polymers on microsecond time scales, *SpringerPlus* **3**, 489 (2014).

S2. Rahman, M. A. et al. Design of tough adhesive from commodity thermoplastics through dynamic crosslinking, *Sci. Adv.* **7**, eabk2451 (2021).

*Original comment* **(#2)** *The focus on ultra-thin (sub-5 micron) polyimide seems overly narrow, and not well-motivated. There are many substrates that may be a better choice if the goal is "conformal contact onto soft and curvilinear surfaces", because PI does not stretch. The title of the manuscript includes "for ultrathin displays", and the authors use "implantable and wearable information displays" as a motivation for such thin polyimide. What is an "implantable" display, and how can it be viewed by a user if it is implanted in the body? This use case needs more explanation to justify why the ultrathin polyimide is needed. On the other hand, there are many applications where thicker polymer substrates would be beneficial. Would this process work (and still provide any benefits over conventional LLO) if the substrate was NOT ultrathin PI? Is a thicker substrate more resistant to the wrinkling effect, and thus the GLLO is not really necessary or beneficial? The key point here is that the work is narrowly focused on ultrathin PI, the need for such an ultrathin substrate is not well motivated, and the authors don't provide evidence that GLLO would provide benefits if the PI substrate were thicker.*

*Regarding the revision:*
*This comment has been somewhat addressed by the authors by adding a few references and verifying that the GLLO works with thicker (up to 6.6 micron) PI films. However, even that small increase in PI thickness reduced the wrinkling effect during LLO, thus diminishing the benefit of the GLLO method. I still find the focus of the paper to be a bit narrow, and the motivation for using PI <5 microns thick could have been made more strongly.*

**Response to the second reviewer's comment (#2):** Thank you for the constructive comment regarding the motivation of our study. We wish to highlight the critical role of ultrathin PI films in the field of stretchable electronics applications (refs. 8 and 9 in the manuscript). Owing to their outstanding thermal stability, ultrathin polyimide films are utilized in various industries, including display technology, to develop stretchable electronics. However, as noted by the reviewer, the intrinsic stretchability of PI films is limited. Consequently, many researchers have explored several design strategies to enhance the stretchability of PI films such as network and serpentine structures (below **Fig. R1**). In this regard, references 8 and 9 report that the stretchability and reliability of the stretchable electronic are improved when thinner PI films are exploited in the designed device structures as shown in **Fig. R2**.

[REDACTED]

**Fig. R1**: Design strategy for stretchable electronics (ref. 9).

[REDACTED]

**Fig. R2**: Stretchability with respect to the thickness of metal and PI films (ref. 9).

Moreover, we further verified the role of the ultrathin PI film on stretchability by analyzing the stress-strain distribution of a metal electrode on the PI substrate using FEM simulation (**Fig. R3**). The PI substrate thickness was varied from 1 to 15 μm, and von Mises stresses and maximum principal strains were examined for the integrated 100-nm-thick Cu electrode. The results show that using a thinner PI substrate reduces the applied strain and stress in the electrode structure. This reduction is attributed to differences in buckling behaviors [Pan, T. et al, Adv. Funct. Mater. 27, 1702589 (2017)].

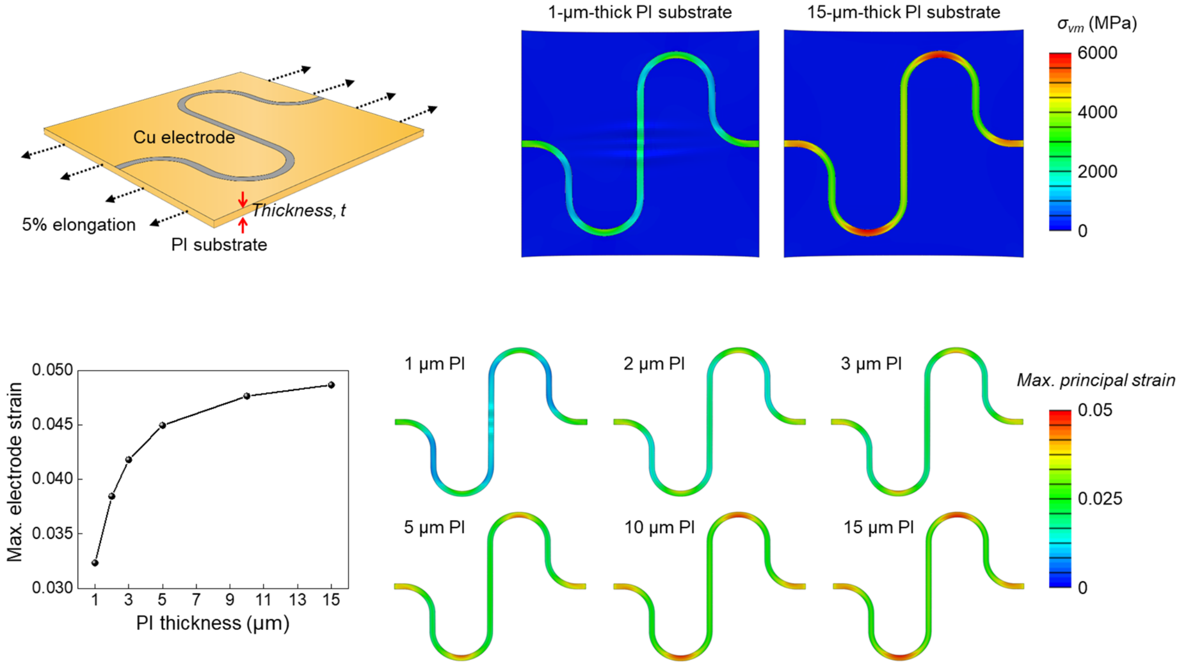


**Fig. R3**: Stress-strain analysis of the electrode for the thickness of the PI film (our work).

*Original comment* **(#4)** *The authors only include one brief paragraph summarizing other approaches to reduce laser-induced damage during LLO. If this is a problem with very broad interest (e.g., sufficiently broad to warrant publication in Nature Communications), I would suspect that there had been more previous efforts to solve this problem that should be discussed here. For example, there are at least two groups that have used a photonic lift-off process to delaminate flexible (opto)electronics on polyimide substrates from glass carriers. They also rely on an interfacial layer that absorbs the light and laterally spreads the heat at the interface. (See: Liu et al, "Photonic Lift-off Process to Fabricate Ultrathin Flexible Solar Cells", ACS Appl. Mater. Interfaces 2021, and Weidling et al, "Large-area photonic lift-off process for flexible thin-film transistors", npj Flexible Electronics 2022.)*

*Regarding the revision:*
*The authors added some relevant points of comparison between GLLO and the photonic lift-off (PLO) method in the Discussion section near the end of the paper, which strengthens the paper. However, do they have any evidence that PLO damages ultra-thin PI films? That claim seems to be based solely on the incident energy density used for these two methods. If PLO uses light with a broad spectrum (the energy is distributed over many wavelengths), I'm not sure the energy density can be directly compared to a laser in that way. They should provide evidence or consider removing that suggestion.*

**Response to the second reviewer's comment (#4):** We appreciate the reviewer's comment. Although we did not perform our own experiments on the PLO process, there is existing evidence of mechanical damage associated with PLO in the literature.

For example, reference 34 in the revised manuscript describes the performance of the PLO process on a 12-µm-thick PI film under various fluence conditions (0.85, 2.6, 4.2, and 5.8 J/cm$^2$). The results indicated that, in the absence of modifications to the molecular structures of the PI film, the PLO process led to substantial wrinkling and breakage of the 12-µm-thick PI film after separation (**Fig. R4**).

[*REDACTED*]

**Fig. R4**: Lift-off of 12-µm-thick PI film at fluences of 0.85, 2.6, 4.2, and 5.8 J/cm$^2$ (ref. 34).

Furthermore, in reference 35 (experiment condition: 20-µm-thick PI film and 4.55 J/cm$^2$ average pulse fluence), W. Liu et al. observed cracking and delamination of the brittle ITO layer integrated with the 20-µm-thick PI film (**Fig. R5**). In contrast, our study successfully achieved the lift-off of OLED devices fabricated on the 2.9-µm-thick PI film using the GLLO method.

[*REDACTED*]

**Fig. R5**: Cracking and delamination of ITO electrode during the PLO process (ref. 35).

Nevertheless, we acknowledge the reviewer’s concern regarding the comparison between PLO and GLLO methods. To address this issue, we have provided a more detailed statement in our discussion as follows.

The 2nd paragraph of page 21 (Discussion section):

~ Meanwhile, the photonic lift-off (PLO) method, which employs high-intensity light pulses of flash lamps instead of lasers and relatively thick metallic light-absorbing layers such as Mo and W/Ti alloys, enables extremely high throughput processes[34-37]. However, the required light energy density of the PLO processes is much higher than that of the LLO processes. The higher light energy density potentially causes damage to sub-10-µm PI films with typical molecular structures[34], and integrated devices composed of brittle electrodes and encapsulation layers[35]. Considering these characteristics, it can be concluded that the GLLO method allows for high throughput and precision lift-off of ultrathin PI films.

Again, we greatly appreciate the time and effort the reviewers have invested in providing constructive feedback. We hope that our responses and the modifications address all concerns satisfactorily.

Sincerely,

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## REVIEWERS' COMMENTS

**Reviewer #2 (Remarks to the Author):**

The authors have provided thorough and thoughtful responses to my comments. I appreciate their effort and the clear documentation of the changes made to the manuscript and supporting material. The additional references, experimental results, and simulations provide sufficient support for the claims made in the paper, so therefore I recommend publication.