

Facile electron-beam lithography technique for irregular and fragile substrates

Jiyoung Chang,^{1,2,3,a)} Qin Zhou,^{1,2,3,a)} and Alex Zettl^{1,2,3,b)}

¹Department of Physics, University of California at Berkeley, Berkeley, California 94720, USA ²Materials Sciences Division, Lawrence Berkeley National Laboratory, Berkeley, California 94720, USA ³Kavli Energy NanoSciences Institute at the University of California, Berkeley, California 94720, USA

(Received 22 September 2014; accepted 15 October 2014; published online 29 October 2014)

A facile technique is presented which enables high-resolution electron beam lithography on irregularly-shaped, non-planar or fragile substrates such as the edges of a silicon chip, thin and narrow suspended beams and bridges, or small cylindrical wires. The method involves a spin-free dry-transfer of pre-formed uniform-thickness polymethyl methacrylate, followed by conventional electron beam writing, metal deposition, and lift-off. High-resolution patterning is demonstrated for challenging target substrates. The technique should find broad application in micro- and nano-technology research arenas. © 2014 AIP Publishing LLC. [http://dx.doi.org/10.1063/1.4900505]

Electron-beam (e-beam) lithography, a powerful technique for sub-micrometer feature fabrication, is widely used in material characterization,¹ micro- or nano-electromechanical systems (MEMS or NEMS),² and in bioengineering.³ A critical requirement for reliable sub-micrometer e-beam lithography is uniform coverage of the e-beam resist (typically polymethyl methacrylate (PMMA)) on the surface. Although spin-coating PMMA is the most common method to achieve uniform coverage, it unfortunately fails for non-flat surfaces due to edge-bead effects.⁴ Furthermore, e-beam lithography on substrates with trenches or with highly curved surfaces (such as very thin cylindrical wires) remains highly problematic.

To help meet these challenges, alternative (but more complex) resist-deposition routes have been developed including spray coating,⁵ photo resist evaporation,⁶ use of dry-film photoresist,⁷ or even films of ice.⁸ The spray coating method is particularly suitable for sharp substrate features including deep trenches, but it requires special spray coating equipment and a complex optimized mixture of PMMA with acetate and ketone to facilitate vaporization during the coating process.⁵ Thermal evaporation of polystyrene (PR) has been recently used in ebeam lithography to achieve 15 nm resolution, but generally, PR has poor exposure sensitivity compared to PMMA and thermal evaporation equipment adds complexity and cost to the process. Ice resist has been applied to fragile and nonplanar substrates such as carbon nanotubes, but this method requires specially designed e-beam lithography and e-beam evaporation systems to maintain the temperature $< 120 \,\mathrm{K}$,⁸ thereby increasing cost and limiting the selection of target substrates. Focused Ion Beam (FIB) can offer similar benefit, but the cost is very high and deposition materials are quite limited.9 Most useful would be a facile, low-cost e-beam patterning method for fragile or non-planar substrates, ideally still retaining the many advantages of PMMA.

In this letter, a facile method is presented for the direct dry transfer of uniform-thickness PMMA films onto challenging substrates such as thin suspended beams, cylindrical wires, and chip edges. The method has distinct advantages over competing specialty techniques. First, a *pre-spun* PMMA film is directly dry-transferred onto the target substrate, making the process spin-free on the target substrate and allowing uniform-thickness PMMA on complex surfaces. Second, no special equipment or material is needed other than commercially available PMMA, a conventional photoresist spinner, a hot plate, Scotch[®] tape, and Kapton[®] tape. The direct and dry PMMA transfer method is simpler and cleaner than evaporated or spray-coating methods, yielding overall, a highly efficient e-beam lithography technique.

Fig. 1 schematically outlines key steps in the dry transfer PMMA process. First, clear Scotch[®] tape with adhesive on one side (bottom) is adhered to a glass slide (Fig. 1(a)). To insure a flat upper tape surface, any air bubbles are removed by driving them to tape edge with a razor blade. PMMA is then deposited onto the tape upper surface, and the slide is spun at 3000 rpm for 40 s. The slide is then baked at 150 °C for 3 min. The resulting PMMA film is approximately 200 nm thick. The cured PMMA film adheres only modestly to the upper surface of the Scotch[®] tape, and as described below can be easily peeled from it (this is not possible if the PMMA is directly spun on the glass). For this study, we employ PMMA 495PMMA A4 (microchem[®], PMMA A4) and developer 1:3 MIBK/IPA (microchem[®]). A proper thickness of the PMMA film is important for transfer. For instance, if PMMA A2 is used employing the same spin conditions, the PMMA film breaks while being peeled off. Based on our experiments, a PMMA film thickness over 200 nm is needed for reliable transfer of cm-scale films.

A transfer frame is prepared by attaching Kapton[®] tape (adhesive on one side only) on a different (bare) glass slide, followed by the cutting out of a circular or rectangular window from the tape. The Kapton[®] tape frame is then removed from the glass slide and pressed onto the glass slide supporting the spun PMMA. We find that before attaching the Kapton[®] frame on to the PMMA, it is beneficial to delineate via a razor blade the area of PMMA to be attached to the frame; this helps in the subsequent delamination of the

^{a)}J. Chang and Q. Zhou contributed equally to this work.

^{b)}Author to whom correspondence should be addressed. Electronic mail: azettl@berkeley.edu



1. Attach Scotch® tape on Glass slide

2. Spin PMMA(3000rpm) and bake the slide on hotplate (3minutes at 150°C)



5. Direct transfer to target substrate

FIG. 1. A process flow of spin-free dry transfer of PMMA film. (a) PMMA A4 is spun on clear Scotch[®] tape attached on the glass slide. Pre-baking at 150 °C for 3 min dries the film to be ready for peeling. (b) A Kapton[®] frame is pressed on the PMMA film and gently peeled-off, resulting in a free-standing PMMA film supported by the Kapton[®] frame (right). (c) Dry PMMA film transfer allows e-beam lithography on various irregular substrates, including trenches (left), small cylindrical wires (center), sharp edges and side faces (right).

PMMA dry film. After the frame is attached onto the selected region of PMMA, the Kapton® frame is gently peeled-off together with the PMMA film, as outlined in Fig. 1(b). The Scotch[®] tape is left behind on the glass slide. The right panel of Fig. 1(b) shows a 3 mm by 3 mm size PMMA dry film suspended over a Kapton[®] frame. The PMMA suspended in the frame is now ready to be transferred to the target substrate such as cylindrical wire, suspended beams or even sharp edge (Fig. 1(c)). Once the film is transferred, baking the substrate with its PMMA overlay on a hot plate at 150 °C for 1min allows the PMMA film to conform to the substrate topography and enhances its adhesion to the substrate. The transferrable size of PMMA film is dependent upon the intrinsic mechanical strength of the dry PMMA film and the adhesion between the PMMA film edge and the Kapton[®] tape frame. As noted earlier, dry films thinner than 200 nm do not reliably peel off. We consistently and easily transfer films 1 cm by 1 cm using 200 nm thick PMMA A4 film.

We demonstrate three specific embodiments of the dry transfer PMMA technique. The first is fine-feature metal patterning around the sharp corner, i.e., continuously on the top and the cleaved side edge (i.e., side face), of a silicon chip. In general, the side face of the chip is not an easy location to perform e-beam lithography due to aforementioned edgebead effects (indeed, spin-coating fails miserably here). However, the thin cleaved side face of a silicon chip, which can be atomically flat when properly prepared, may be an attractive patterning area. For example, the read/write magnetic head slider in hard disk drives is fabricated with multiple surface micromachining processes followed by dicing into strips and lithography on the edge of the strips.¹⁰ A viable PMMA transfer method could allow higher resolution lithography on such narrow surfaces not achievable with conventional methods.

Fig. 2 illustrates metal patterning over a cleaved chip edge region using the dry transfer PMMA method. When the suspended PMMA film is brought into contact with the sharp edge region of the chip, the film adheres to the substrate even over the edge (after hot plate baking), allowing continuous e-beam lithography on both top and side. Fig. 2(a) shows schematically the desired test pattern for the top and side region, and Fig. 2(b) shows a scanning electron microscope (SEM) image (after lift-off) of applied Ti/Au metal features to such a chip region; the metal electrodes here faithfully follow the sharp contours of the silicon substrate, connecting top and side chip faces across the edge (the patterning can even continue, if needed, to the bottom face of the chip). If necessary, variable tilt angles can be used during evaporation to insure uniform metal coverage. For the edge sample used in Fig. 2(b), only one shallow evaporation angle was used, and the slightly spotty deposition on the remote region of the side face is due to this non-optimal angle. If the substrate were tilted at 45°, making the edge or corner face upward, deposition on regions both at the corner and far from the corner would be more uniform.

The second application concerns extremely fragile substrates, such as thin suspended bridges or cantilevers as might be found in MEMS or NEMS mechanical resonators,^{11,12} chemical sensors,¹³ or thermal characterization instrumentation.¹⁴ Of interest is the ability to add, via e-beam lithography, features to the bridge structure following initial processing, or perhaps even to repair nanoscale defects in, for example, electrodes originally written onto the bridges.

We here consider post-production metal patterning on thin (250 nm thick) Si₃N₄ bridges, 50 μ m in length and 8 μ m in width, suspended over trenches 35 μ m in width, 70 μ m in length and 25 μ m deep in 3.5 mm × 3.5 mm silicon chips. Figs. 3(a)–3(c) show the process of adding the letters "CAL"



FIG. 2. (a) A schematic showing the metal patterning on the edge of the chip. Both top and side area can be e-beam patterned due to uniform PMMA film coverage over the edge contour. (b) Angled SEM image to show electrodes defined on the edge via lift-off Ti (5 nm)/Au (40 nm).



FIG. 3. High resolution e-beam lithography process applied to fragile suspended silicon nitride beam of 250 nm thickness. (a) SEM image of a beam before PMMA dry transfer. (b) Optical image shows beam after dry transfer and e-beam writing and developing. The duplicated words "CAL" are barely visible (arrows). (c) Zoomed in SEM image of the desired raised Ti/Au pattern. (d)–(f) show processing on a different silicon nitride suspended beam patching a gap between two electrodes.

on top of a 1 μ m-wide metal line on the suspended bridge. Fig. 3(a) shows an SEM image of the bridge structure prior to lettering. For such a fragile structure, spinning PMMA over the bridge would be disastrous, resulting only in bumpy and clogged PMMA making precise e-beam lithography impossible. Fig. 3(b) (optical image) shows instead the bridge after dry transfer of a pre-spun PMMA film and ebeam patterning and resist processing (the duplicate words CAL can just be made out on the lower trace). Following Ti (3 nm)/Au (17 nm) deposition and lift-off in acetone, a clean raised lettering results, as shown in the zoom-in SEM image of Fig. 3(c). Of interest is the fact that during e-beam writing (Fig. 3(b)), the suspended PMMA film over the entirety of the trench not only stabilizes the suspended bridge but also prevents debris or evaporated metals from entering the trench region; this results in crisply defined features and ultra-clean processing, particularly during lift-off.

Figs. 3(d)-3(f) show analogous post-production e-beam patterning and processing using the dry transfer PMMA method for a similarly suspended Si₃N₄ bridge carrying two electrodes with a narrow gap between them. Here, e-beam lithography is used to deposit a metallic patch or jumper between the electrodes. Of course, the deposited square patch need not be an inert metal but could be chosen to have added functionality, such as comprising a sensor material.

The final application demonstrates the utility of the dry PMMA transfer method for target substrates that are highly curved, such as those with cylindrical or even spherical



FIG. 4. E-beam writing onto a 1-mil ($d = 25\mu m$) gold wire. (a) PMMA film adheres not only to the wire but also to the substrate around the wire, providing immobilization of wire for controlled manipulation. The desired pattern to be written on the wire is a series of tower symbols and words "UCB." (b) Optical image of following lithography showing Ti writing onto the wire surface.

geometry. Patterning on cylindrical structures is an active concern for nanoimprint lithography¹⁵ or lab-on a fiber¹⁶ technology, yet it remains highly challenging due to difficulty in uniform PMMA film thickness control. A dry-transferred PMMA film here conforms and adheres closely to the contour of the non-planar surface after post-transfer bake, insuring uniform coverage of PMMA. E-beam lithography on highly curved surfaces then becomes no more difficult that than on flat surfaces.

Fig. 4 illustrates writing tower-shaped symbols and the letters "UCB" onto the surface of a 1-mil (25 μ m diameter) gold wire. Fig 4(a) shows schematically a cross-sectional view of the wire with added PMMA film and a tilted-perspective depiction of the desired pattern. We note that the PMMA film aids in immobilizing the wire on the substrate, an added benefit. It not only provides natural anchoring of the target without mechanical clamping or liquid-based paste for the e-beam writing process but also continues to give advantage during resist development and metal deposition, making the entire patterning process cleaner and simpler.

For the cylindrical wire demonstration, a 5 mm length of gold wire is laid flat on a silicon substrate. A 3 mm by 7 mm size Kapton[®] tape frame with suspended PMMA is placed over the wire and baked on hot plate at 150 °C for 1 min. The tower pattern and UCB letters are e-beam written at an area dose of $360 \,\mu\text{C/cm}^2$, the same dose we typically use for flat surface e-beam writing (and for the two other demonstrations above). After development, 20 nm of Ti are evaporated using an e-beam evaporator followed by acetone dip for lift-off. Figure 4(b) shows an optical image of the resulting stenciled pattern on the gold wire.

In summary, the facile spin-free PMMA film transfer method presented here overcomes many of the lithographic challenges found in multiple research fields and applications such as optics, nanoscience, NEMS/MEMS, and material characterization.

This research was supported in part by the Director, Office of Energy Research, Office of Basic Energy Sciences, Materials Sciences and Engineering Division, of the U.S. Department of Energy under Contract No. DE-AC02-05CH11231 under the sp2–bonded Materials Program, which provided for postdoctoral support, design and execution of the experiment, and e-beam writing and characterization instrumentation; and by the National Science Foundation under Grant DMR-1206512 which was provided for postdoctoral support and bridge test devices. We thank the Berkeley Marvell Nanofabrication Laboratory for providing additional specialized equipment for device fabrication.

- ¹S. Dubey, V. Singh, A. K. Bhat, P. Parikh, S. Grover, R. Sensarma, V. Tripathi, K. Sengupta, and M. M. Deshmukh, Nano Lett. **13**, 3990 (2013).
 ²H. G. Craighead, Science **290**, 1532 (2000).
- ³G.-J. Zhang, T. Tanii, T. Zako, T. Hosaka, T. Miyake, Y. Kanari, T. Funatsu, and I. Ohdomari, Small 1, 833 (2005).
- ⁴P. S. Kelkar, J. Beauvais, E. Lavallée, D. Drouin, M. Cloutier, D. Turcotte, P. Yang, L. K. Mun, R. Legario, Y. Awad, and V. Aimez, J. Vac. Sci.
- Technol. A 22, 743 (2004). ⁵J. Linden, C. Thanner, B. Schaaf, S. Wolff, B. Lägel, and E.
- Oesterschulze, Microelectron. Eng. **88**, 2030 (2011). ⁶J. Zhang, C. Con, and B. Cui, ACS Nano **8**, 3483 (2014).

Appl. Phys. Lett. 105, 173109 (2014)

- ⁷P. W. Leech, N. Wu, and Y. Zhu, J. Micromech. Microeng. **19**, 065019 (2009).
- ⁸A. Han, A. Kuan, J. Golovchenko, and D. Branton, Nano Lett. **12**, 1018 (2012).
- ⁹T. Tao, J. Vac. Sci. Technol. B 9, 162 (1991).
- ¹⁰TCS of Japan, Advanced Ceramic Technologies & Products (Springer Science and Business Media, 2012), p. 585.
- ¹¹J. S. Bunch, A. M. van der Zande, S. S. Verbridge, I. W. Frank, D. M. Tanenbaum, J. M. Parpia, H. G. Craighead, and P. L. McEuen, Science **315**, 490 (2007).
- ¹²A. K. Pandey and R. Pratap, J. Micromech. Microeng. 17, 2475 (2007).
- ¹³T. Giesler and J.-U. Meyer, Sens. Actuators B 18, 103 (1994).
- ¹⁴C. W. Chang, D. Okawa, A. Majumdar, and A. Zettl, Science **314**, 1121 (2006).
- ¹⁵Z. Li, Y. Gu, L. Wang, H. Ge, W. Wu, Q. Xia, C. Yuan, Y. Chen, B. Cui, and R. S. Williams, Nano letters 9, 2306 (2009).
- ¹⁶M. Consales, A. Ricciardi, A. Crescitelli, E. Esposito, A. Cutolo, and A. Cusano, ACS Nano 6, 3163 (2012).