Direct writing immersion laser lithography on graphene monolayers using 2-photon absorption

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Ever since, direct writing laser lithography is attractive because direct writing techniques require no mask, hence the design flexibility is large minimizing extra cost, i.e., for new exposure masks. Two-photon absorption direct laser writing (2PA-DLW) is one particular laser direct writing technique for very small features due to non-linear optical effects. Operating this technique in immersion, i.e., filling the air gap between lens and photoresist or probe reduces feature size and improves resolution [1]. In previous studies, immersion oil is usually applied [2]. In contrast, our experiments showed that while the immersion time is too long, oil would probably interact with the photoresist and create new and difficult-to-remove compounds with the photoresist. The changed photoresist can neither be exposed nor developed.

We proved this permanent resist modification by comparison of the exposure and development result on the resist after being contacted with oil (Immersol 518F) for different time intervals. Two spots were exposed on resist, one before oil contacting, one after oil washing by methoxyperfluorobutane (MPFB). Reference sample is without oil contact, the resist surface is clean after washing with MPFB and both exposed spots are clearly developed (Fig. 1a-c). For 1 h oil contact, although exposure spots are still successfully developed, circle like residues are observed and the resist color became lighter (Fig.1 d-f). For longer than 12 h oil contact, the resist at center in contact with oil changed completely its color, and is full of visible residues. The development of the exposure spots in center area failed completely (Fig.1 g-i). The step height between the center and outside is close to the initial resist thickness. Based on these results, we suspect that the interaction of oil and resist creates some new compounds. It starts near the resist surface and goes deeper along time. The longer oil contact time is, the more residues would be left behind after washing. When the contact is too long, the total resist changes to the new compounds. Hence, the long-time writing could be risky due to this resist change.

We applied 2PA-DLW with oil to pattern graphene. Fig.2a shows clear residues after graphene patterning by O_2 plasma and after resist stripping by acetone. We tried to apply glycerol as alternative. Although glycerol has lower refractive index, it is still better than water. Its high absorption at the wavelength of 193 nm is not a problem. Fig. 2b shows a sample successfully fabricated by 2PA-DLW with glycerol. No residues are observed on this sample surface.

[1] Sanders, D. P. (2010). Advances in patterning materials for 193 nm immersion lithography. Chemical Reviews, 110(1), 321–360.

[2] Jakkinapalli, A., etc. (2020). Scanning digital oil immersion lithography providing high-speed large area patterning with diffraction limited sub-micron resolution. Journal of Micromechanics and Microengineering, 30(12).

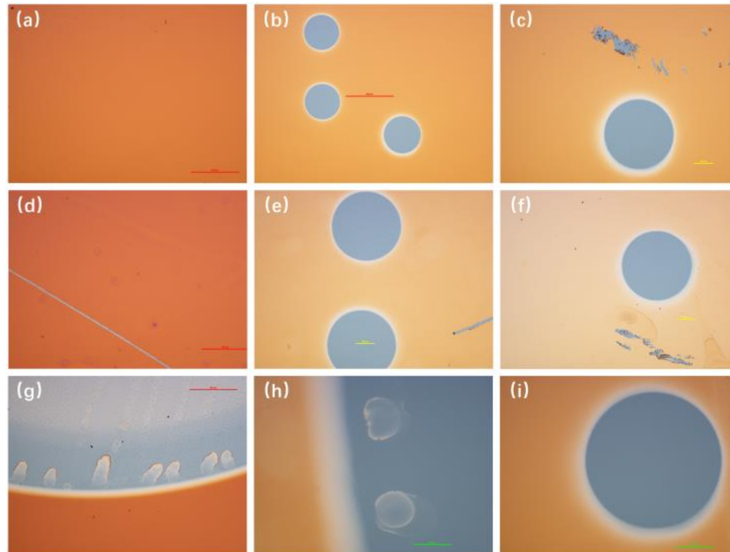

Figure 1. After methoxyperfluorobutane washing [a] surface of reference sample without oil. [b] Successful development of 3 spots at center without oil. [c] Successful development at edge without oil [d] surface of sample for 1 hh oil contact [e] Successful development at center (in contact with oil) for 1 h oil contact [f] Successful development at edge for 1 h oil contact [g] surface of sample (boundary of oil contact area at center) >12 h oil contact [h] Failed development at center (in contact with oil) >12 hours oil contact [i] Successful development at edge >12 hours

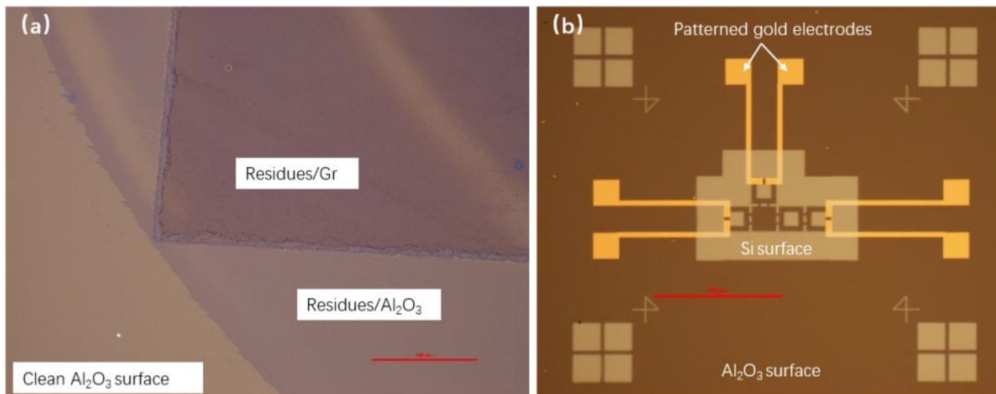

Figure 2. [a] Graphene patterning after 2PA-DLW resist pattering and subsequent O_2 plasma etching and resist removing by acetone. [b] Part of the sample successfully fabricated by apply 2 photon absorption direct writing (longer than 10 hours) with glycerol and metal pattern lift-off using the 2PA-DLW resist mask.