Electronic Supplementary Material (ESI) for Nanoscale.

Electronic Supplementary Information

Scalable Fabrication of High-Quality, Ultra-Thin Single Crystal Diamond Membrane Windows

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Electrochemical etching process

A saturated boric acid electrolyte solution was prepared in deionized water with equal proportions. The diamond sample was transferred to the glass petri dish. The sample to be etched was bonded to a base of a glass Petri dish via suitable adhesive (Command™ double sided tape), and then immersed in electrolytic solution. There was sufficient solution in the petri dish to cover the sample. Two Platinum electrodes were fixed as close to the sample as possible from both sides to maximize the electric field. A DC power supply was used to apply 300 V and approximately 30 mA across the sample.

The graphite like carbon layer was etched and the thin sheet of diamond was lifted off. It is to be noted that during the experiment the etching direction was reversed by either switching the power supply connections or rotating the diamond substrate resulting in proportional etching rates from all four sides of sample. The etching rate depends on the quality of graphite (directly proportional to annealing temperature), area of the etchable layer and a distance between electrodes (the electric field generated across substrate). The typical etching rate was 0.25mm^2/hr. A schematic diagram is shown in figure S1.

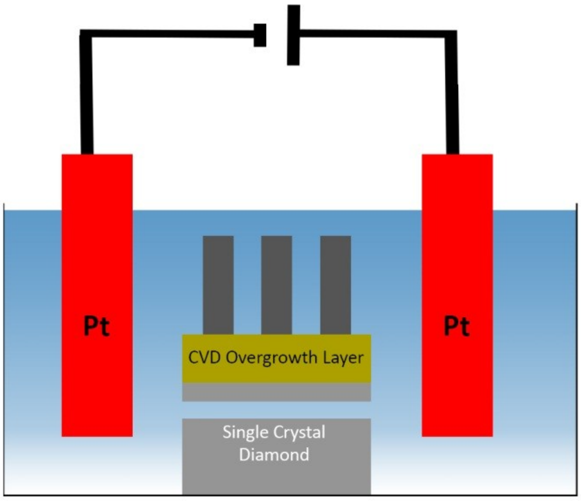

Figure S1 Schematic of electrochemical etching process

Photoluminescence (PL) setup configuration

The PL measurement was collected using a home built confocal microscope using a 532 nm continuous wave (CW) laser through a 100x and a 1.4 numerical aperture objective lens. A diamond sample was mounted on the glass substrate and placed on a piezo XYZ stage under an oil immersion objective to collect the fluorescence from the individual colour centers. The laser power used for the measurement was in order of 0.5 mW for the excitation at the microscope objective. The spatial resolution of the microscope was ~ 0.4 µm and a depth of field was ~ 0.8 µm.

Thickness measurement

As a general rule, this is how we estimated and measured the thickness of membranes.

- CVD growth rate was 20nm/min for the recipe used in this work.
- RIE thinning rate of diamond membrane was 8nm/min.

Equation#1

Plate thickness (after lift-off) = 1.7 µm (ion damaged layer) + CVD layer (overgrown)

- The above equation gives the Plate thickness (after lift-off) which can be estimated from the CVD growth rate and RIE thinning rate; and measured using white light interferometry. Total thickness of CVD (overgrown) agrees with the thickness calculated using equation#1.

Equation#2

Final membrane thickness (after RIE thinning) = CVD layer (overgrown)

- We know from the RIE thinning rate of 8nm/min, that the 1.7 µm (ion damaged layer) was etched away after calculated time as confirmed with Raman spectrum shown in manuscript (figure 3).

In a similar manner we can estimate the thickness of all other membranes. However, the technical limitation of white light interferometery prevents us measuring the final membrane thicknesses since they are below 1µm. The membrane (see figures 2b) was cut for the cross sectional SEM and its thickness was measured as ~300nm as shown in figure 2e. The membrane shown in figure 2c was processed to achieve the final thickness of ~1µm and measured using white light reflectometery as 1µm thick (shown in figure S2). Both SEM and reflectometery measurements were in good agreement with the calculated thickness using equation 1&2.

Optical constants; refractive index (real part) and extinction coefficient (imaginary part) of the Index of Refraction (ň = n - ik) were measured as shown in figure S3 (a,b). The value of n = 2.414 and k = 0.005 at wavelength of 637nm. This measurement was taken on the membrane shown in figure 2c. The values of n and k are in good agreement with pristine diamond [1,2,3].

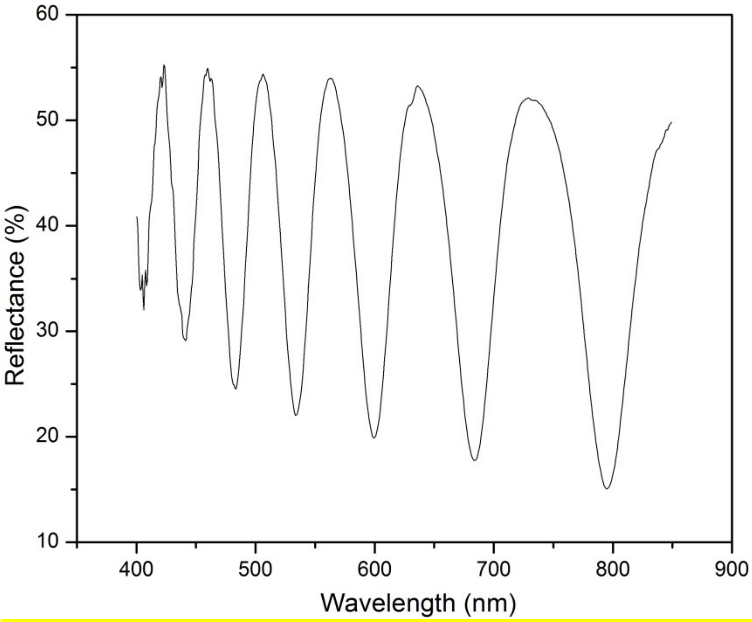

Figure S2 Membrane thickness measured from the reflectometer after fitting to the physical model based on number of layers. The model used for fitting was Air/diamond film/Air. Thickness obtained after best fit was 1 µm which was in agreement to the estimated value. This measurement was taken from the membrane shown in figure 2c.

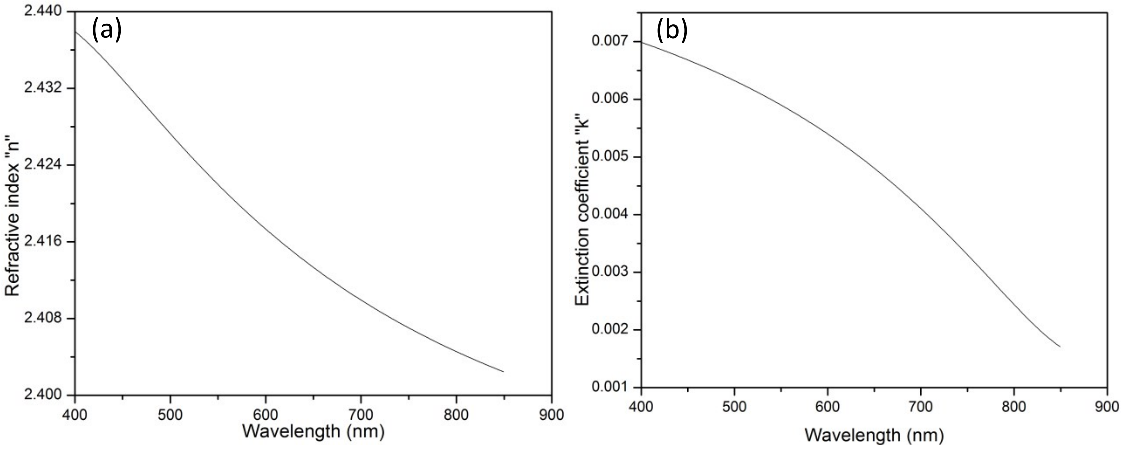

Figure S3 Optical constants n,k (a) Refractive index (real part), and (b) Extinction coefficient (imaginary part) of the Index of Refraction (ň = n - ik). The value of n = 2.414 and k = 0.005 at wavelength of 637nm. This measurement was taken on the membrane shown in figure 2c.

[1] M. A. Draganski, E. Finkman, B. C. Gibson, B. A. Fairchild, K. Ganesan, N. Nabatova-Gabain, S. Tomljenovic-Hanic, A. D. Greentree, and S. Prawer, *Opt. Mater. Express*. 2012, **2**, 644-649.

[2] M. A. Draganski, B.A. Fairchild, A. Alves, E. Finkman, J. Orwa, S. Rubanov, B. Gibson, K. Ganesan, P. Spizziri, A. D. Greentree, P. N. Johnston, D. N. Jamieson and S. Prawer, *Australian Institute of Physics, 18th National Congress,* December 2008, Adelaide, Australia.

[3] A. M. Zaitsev, *Optical Properties of a Diamond, Springer, Berlin* 2001.