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Highly Conductive and Long-term Stable Films from Liquid-Phase Exfoliated Platinum Diselenide

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We would like to note that during the course of this work, dozens of devices were fabricated and tested. While all of the devices with relatively pinhole free networks showed good sensor performance, we found that film homogeneity was key for a stable and reproducible performance. We realised that the reproducibility of the film production (and thus device performance) is greatly improved when dispersions with narrowed nanosheet size distributions are used. Therefore, for the main part of the work in the main manuscript, a $PtSe_2$ dispersion was used where larger/thicker nanosheets were removed as sediment after centrifugation at 3000 *g* and small/thin nanosheets were removed as supernatant after centrifugation at 5000 *g* (see methods). For details on the characterisation of the nanosheets produced this way (including size/thickness distributions), the reader is referred to our previous report.[1] The disadvantage of this approach is that only a fraction of the nanosheets produced by sonication is selected which means that the mass is relatively low. While we attempted spraying such as size-selected dispersion, the mass was not sufficient to produce a film with close coverage. Therefore, to compare LB-tiled and sprayed networks with respect to conductivity and film thickness, a different set of dispersion had to be used with a much broader size/thickness distribution. Specifically, the centrifugation boundaries were set to 400 *g* and 30,000 *g*. This greatly affects the film morphology (homogeneity and thickness) in the tiled network as shown by the visual comparison in Fig. S1. The films produced from dispersions with narrower nanosheet size distribution (Fig. S1a-c) show greater homogeneity, lower roughness, better edge to edge contact of the nanosheets and a reduced overall thickness compared to the dispersion with the broader distribution (Fig. S1d-f).

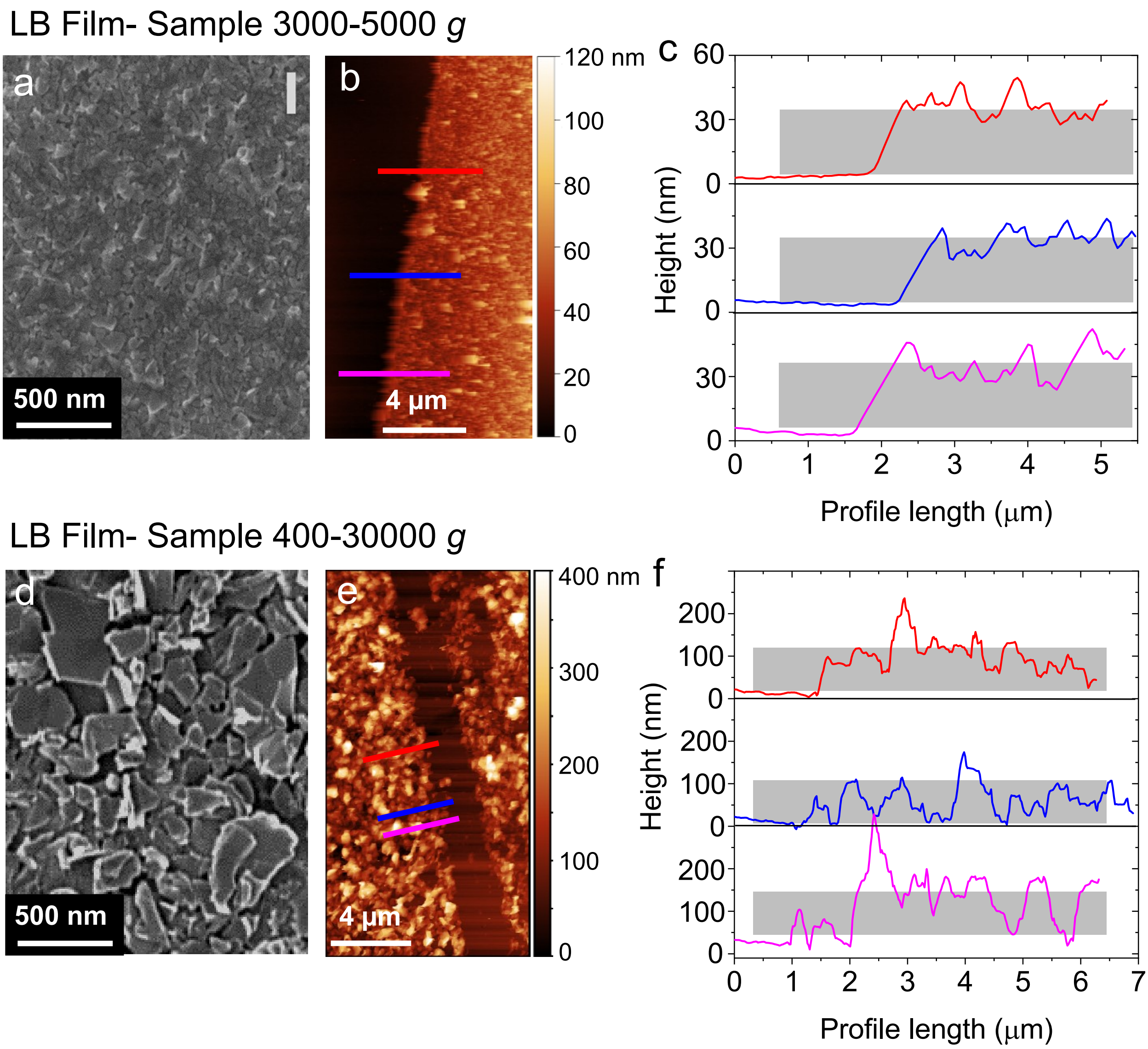

Fig. S1 Comparison of LB-type nanosheet networks produced from LPE-$PtSe_2$ using different size/thickness distributions through modification of the centrifugation conditions. (a-c) Data from the dispersion with narrower size distribution used for the gas sensing experiments. (a) SEM image, (b) AFM image of a scratched film. (c) Profiles of the lines indicated in (b). The gray box has a height of 30 nm and serves as guide for the eye to indicate the film thickness. (d-f) Same data from a dispersion with broader size distribution used to compare the conductivity of LB type and sprayed networks. (d) SEM image, (e) AFM image of a scratched film. (f) Profiles of the lines indicated in (e). The gray box in (f) has a height of 100 nm.

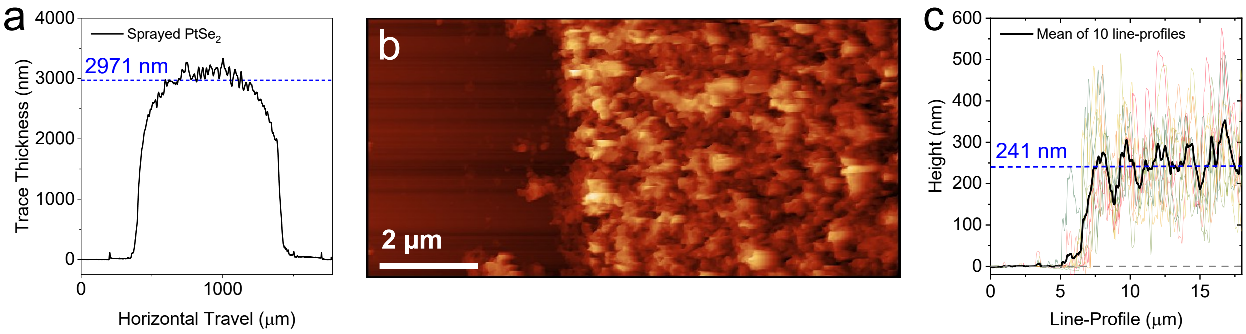

Fig. S2 (a) Profilometry curve indicates ~3 µm of film thickness produced by conventional spray deposition. (b) AFM image of an LPE-$PtSe_2$ film produced by the modified LB method using two subsequent deposition cycles of the 400-30000 g sample and (c) ~240 nm film thickness is estimated from the mean of 10 height profiles.

References

1 B. M. Szydłowska, O. Hartwig, B. Tywoniuk, T. Hartman, T. Stimpel-Lindner, Z. Sofer, N. McEvoy, G. S. Duesberg and C. Backes, *2D Mater.*, 2020, **7**, 045027.