Journal Name

ARTICLE TYPE

Evolution of inter-layer coupling strength in twisted atomically thin van der Waals heterostructures - Electronic Supplementary Information (ESI)

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S1 Effect of annealing the devices in ambient on their optical properties

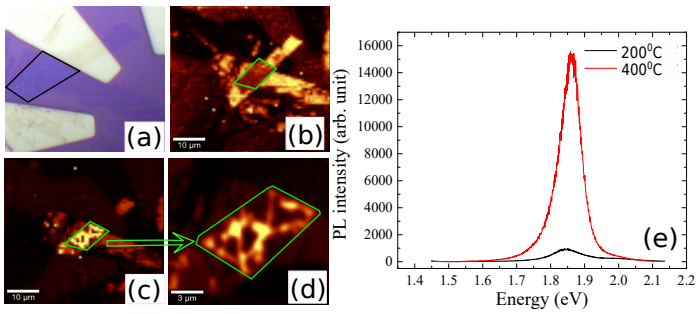

Fig. S1 (a) Optical micrograph of an ABL where we have identified stack area with solid black line. (b) Photoluminescence map of the device at 200°C. PL quenching on the stack region has been identified with green solid line. (c) Photoluminescence map of the device at 400°C marked with green solid line. (d) Small area scan of the stack region showing the appearance of the physical cracks and strong PL intensity in other areas of the stack. (e) PL intensity as a function of energy for sample heated at 200°C (black line) and 400°C (red line) in ambient. After annealing at 400°C, the sample showed a much higher PL count as compared to the 200°C PL quenched state.

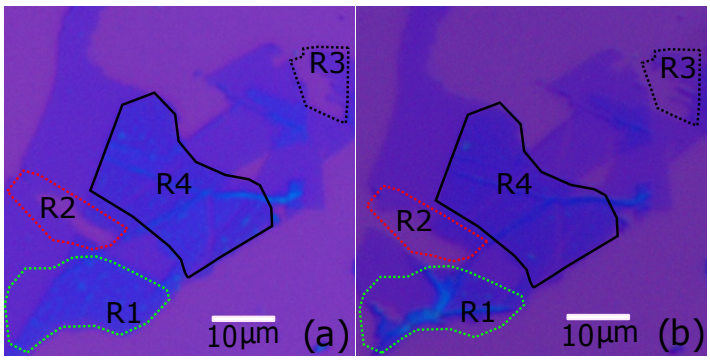

Fig. S2 (a) Optical image of the pre-annealed as prepared ABL sample with its constituent layers. Different regions have been marked - R1 is mostly ABL (marked with green dotted line), R2 is mostly SL (marked with red dotted line), R3 is another SL (marked with black dotted line) and R4 is ABL region (marked with solid black line). (b) Optical image of the same sample post-annealing at 400°C inside a vacuum chamber at a pressure 10^{-6} mbar.

We have shown in the main letter that quenching of photoluminescence in the artificial stack region indicates an increase of interlayer coupling. To identify the optimal temperature for cleaning the interface, we undertook a detailed study of quenching of PL with temperature. In Fig.S1 we show the results of annealing the artificial stack in ambient on a hot plate at different temperatures. The presence of O_2 in the environment can partially oxidize the MoS_2 and turn it into MoO_3. The amount of MoO_3 formation increases with temperature and causes a large increase in the PL signal in the ABL region[1](Fig. S1(e)). Once the sample is in this high PL count condition due to formation of MoO_3, it remains in this PL enhanced state which suggests a permanent physical change in the sample. Annealing at high temperatures also caused physical damage to the sample (Fig. S1(c) and (d)).

To avoid oxidation of the sample, we carried out the annealing process in a high vacuum (10^{-6} mbar) chamber. We find that annealing at 400^0C in vacuum cleanses the interface but still physically damages the sample (Fig. S2).

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S2 Results obtained on other heterostructures

This cleaning procedure and evolution of interlayer coupling is not only restricted to artificially stacked MoS_2 layers but is quite general to most heterostructures like graphene-MoS_2, graphene-graphene, hBN-graphene or hBN-MoS_2. In Fig.S3 we show an example of a graphene-MoS_2 heterostructure. In the as-prepared sample the interface of graphene and MoS_2 has a large PL signal (Fig.S3(a)). After annealing at 200°C, the PL gets quenched as was the case for ABL on SiO_2. Annealing at higher temperatures led to physical damage of the sample. We have observed in all cases that once the stacks are optimally annealed, they remain in the PL quenched state over months which confirms the stability of the sample for long time studies and practical applications.

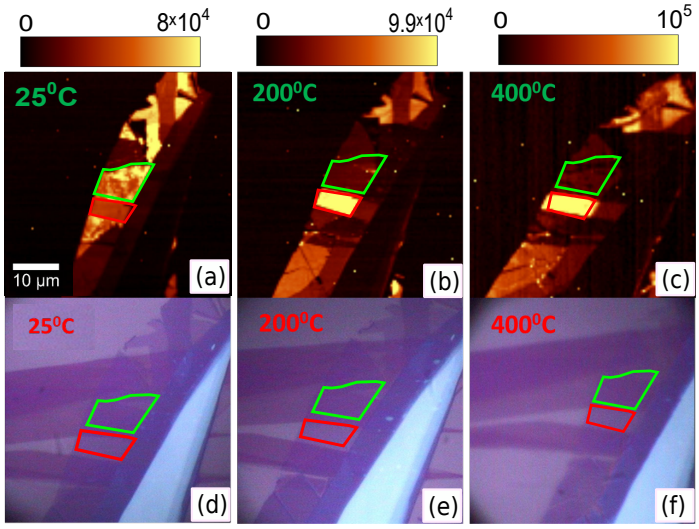

Fig. S3 Integrated PL intensity map (over 1.5-21. eV) of MoS_2-graphene heterostructure (a) at room temperature, (b) post-annealing at 200°C and (c) post-annealing at 400°C respectively. Colour bar on top shows the integrated PL intensity of the PL map. The SL MoS_2 on SiO_2 is outlined by solid red line and SL MoS_2 on graphene is outlined by solid green line. The corresponding optical images are shown in (d), (e) and (f) respectively.

S3 Long-term stability of annealed samples

The MoS_2 stacks, after optimal annealing, were found to remain optically and electrically stable for months. An example is shown in Fig. S4 where we show the Raman and PL emission spectra measured immediately after the annealing step sample and after a time-interval of three months. The optical features in both Raman and PL emission remain essentially unchanged attesting to the high stability of our devices. Example of the electrical stability of our MoS_2 devices over a time-period of one year has been discussed in a separate publication[2].

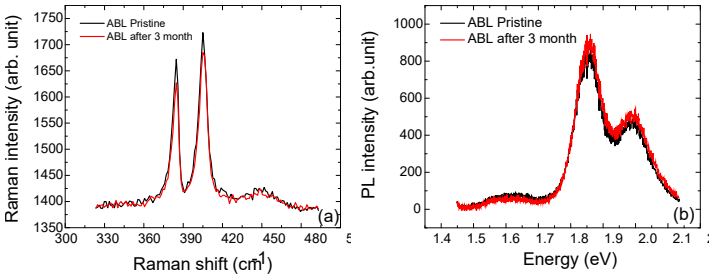

Fig. S4 (a) Raman spectra taken on the as-prepared device immediately after the annealing process (black line) and after three-months (red-line). (b) Photoluminescence taken on the as-prepared device immediately after the annealing process (black line) and after three-months (red-line). In both Raman and PL emission, the optical features are found to remain essentially unchanged.

Notes and references

1 H. Nan, Z. Wang, W. Wang, Z. Liang, Y. Lu, Q. Chen, D. He, P. Tan, F. Miao and X. Wang, *ACS nano*, 2014, **8**, 5738–5745.

2 S. Sarkar, A. Bid, K. L. Ganapathi and S. Mohan, *Phys. Rev. B*, 2019, **99**, 245419.