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Supporting information:

Automated mechanical exfoliation technique: a spin pumping study in YIG/TMD heterostructures

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Reproducibility of the method

To demonstrate the reproducibility of our technique, we have prepared five samples labeled R1 to R5, employing identical parameters as those used for the MoS_2/YIG samples discussed in the main article. The presented results pertain exclusively to MoS_2 thin film. However, it is important to emphasize that our technique applies equally to obtaining thin films of any Van der Waals (vdW) materials. The MoS_2 layers in these samples were controlled with a consistent six-layer configuration. We utilized Raman spectroscopy to determine the number of layers of MoS_2 accurately, and the corresponding data is shown in Fig S1.

Notably, all five samples exhibited characteristic peak shifts in the MoS_2 spectrum compared to the bulk material measurement. Following well-established literature methods [1], we accurately evaluated the number of layers of MoS2 of these samples. The analysis shows that each of the deposited films consists of 6 MoS_2 layers, as evidenced by the difference in positions between the E'_{2g} and A'_{1g} modes, which amounts to approximately 25 cm^{-1}. These compelling results confirm the excellent thickness reproducibility of our technique, thereby showcasing the remarkable consistency in the number of layers within the R sample series.

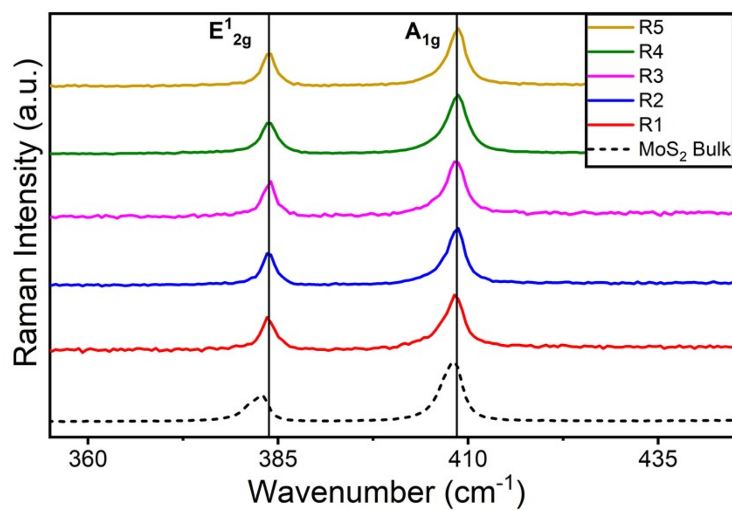


Fig. S1. Raman spectroscopy of the reproducibility test series. The Raman spectrum of MoS2 bulk is shown as a black dashed line, while solid colored lines represent the MoS2 few-layer films.

Homogeneity

One of the key parameters in our automated mechanical exfoliation method is the pressure applied by the deposition pad to the substrate during the deposition, which is controlled by the integrated piezoelectric system. As the main article details, this automated control offers a significant advantage over manual methods (traditional micro-mechanical exfoliation via original scotch-tape), enabling precise control of large-scale sample roughness.

To demonstrate the control of our method's roughness and homogeneity, we produced a series of four samples deposited on a glass substrate with all parameters fixed, except for the pressure, which was gradually increased from sample P1 to sample P4. The surface roughness was evaluated by the root means square (RMS) using atomic force microscopy (AFM) over a 5×5 μm^2 area, and the roughness dependency on the pressure parameter was shown in Fig. S2 (a). The dependency of roughness on the pressure parameter is presented in Fig. S2 (a). The roughness displayed an exponential asymptotic behavior, decreasing with increasing pressure. Moreover, a representative AFM measurement is displayed in Fig. S2 (b).

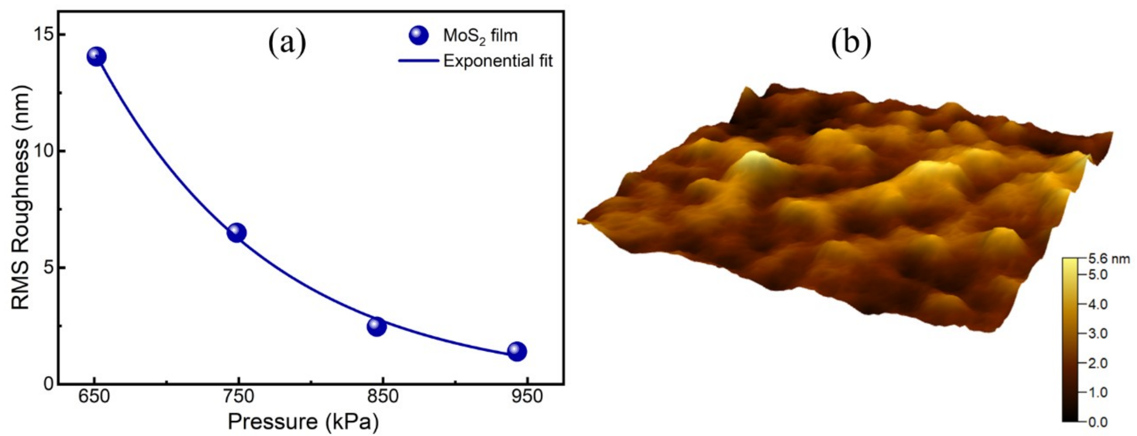


Fig. S2. (a) Roughness dependence as a function of the applied pressure. (b) Representative AFM map illustrating the surface topography with 1 μm^2 area.

Exfoliation

In our method, the sample thickness is primarily controlled by the number of exfoliations. To demonstrate the controllability of thickness, we prepared a series of MoS2 samples (labeled E1 to E4) on a glass substrate, incrementally increasing the number of exfoliations while maintaining all other parameters constant. To measure the thickness, we employed both atomic force microscopy (AFM) and focused ion beam scanning electron microscopy (FIB-SEM), as illustrated in Figure S1, where open red squares represent the data obtained from AFM measurements, while the blue triangles correspond to the FIB-SEM results. Notably, the FIB-SEM analysis indicated that the E1 sample was too thin for evaluation using this method.

As shown in Fig. S3 (a), the thickness has a quadratic behavior with the number of exfoliations, and the data obtained by the two methods are equal within the range of experimental error observed. It is crucial to highlight that the FIB-SEM method requires a metallic layer to prevent charge accumulation on the sample's surface, making the process destructive. Consequently, AFM and FIB-SEM measurements were performed on distinct samples, though utilizing identical disposition parameters, thereby substantiating the reproducibility of the methodology. The panel in Fig. S3 (b) shows representative images of FIB-SEM, while Fig. S3 (c) shows a representative AFM image of the thickness measurement.

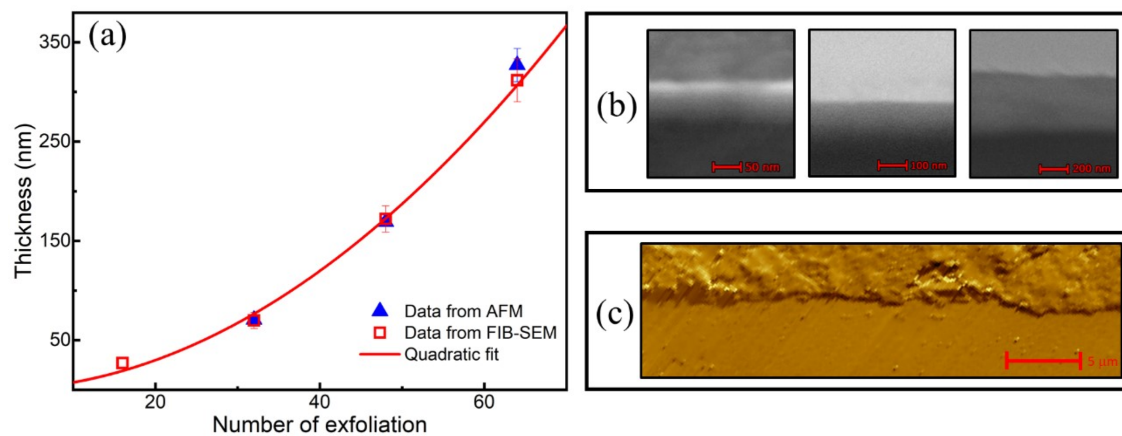


Fig. S3. (a) Thickness dependence as a function of the number of exfoliations. (b) Representative FIB-SEM measurement. (c) Representative AFM measurement.

Growth direction

To ensure the alignment of all samples along the same direction, with layers stacked through Van der Waals bonding, X-ray diffraction (XRD) and Raman spectroscopy was performed on all samples from series E and P. As shown in S4 (a) and (b), the XRD patterns exhibit only the peak family (001), confirming the organization along the c-axis of the hexagonal structure. The Raman spectroscopy in Fig S4 (c) and (d) also reveals only the modes E'_{2g} and A'_{1g}, further supporting the consistent quality of the samples.

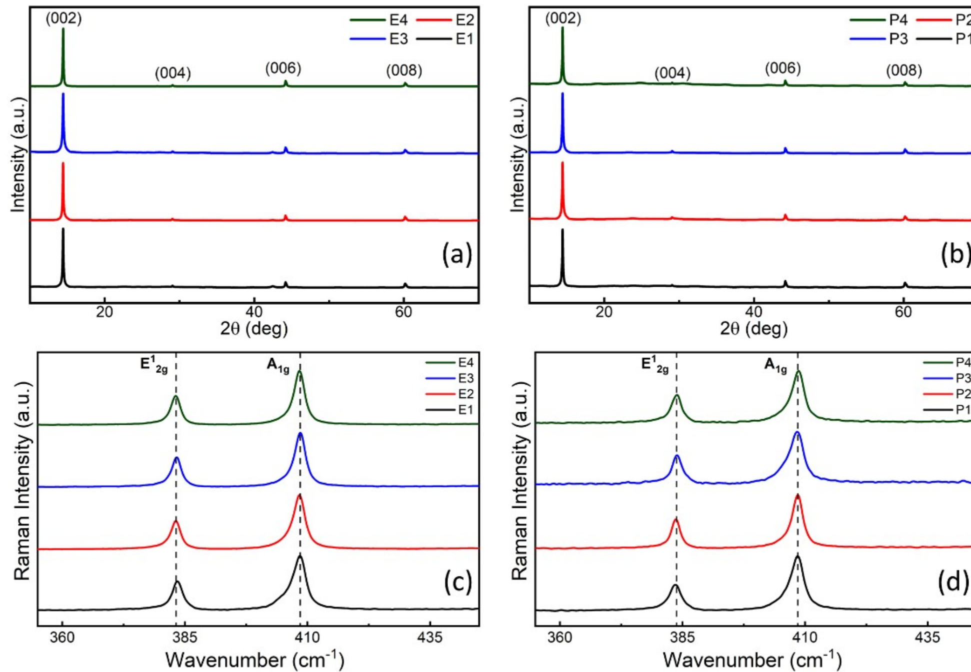
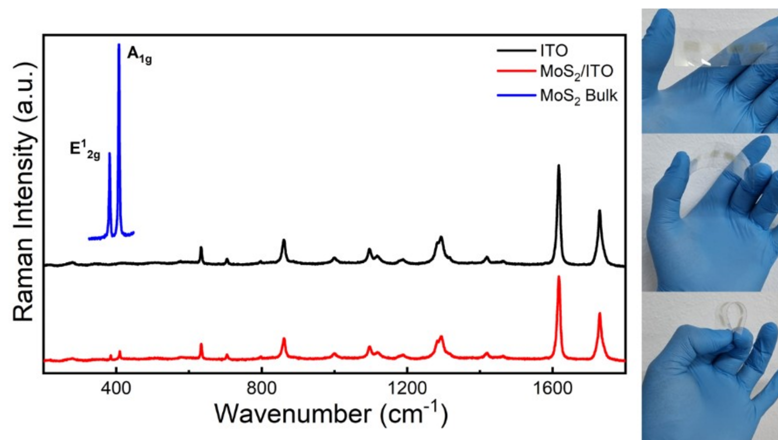


Fig. S4. XRD patterns of series E and P are shown in (a) and (b), respectively. Raman spectroscopy results for series E and P are displayed in (c) and (d).

Flexible substrate and large area deposition

A key advantage of our method is its compatibility with flexible substrates and its versatility in accommodating deposition areas ranging from small to large scales. In this regard, we prepared samples varying from 1 mm^2 to a few cm^2. Furthermore, we successfully produced TMDs samples on a flexible and conductor indium-tin-oxide (ITO) substrate (Indium tin oxide coated PET, surface resistivity 60 ω/sq, L × W × thickness 1 ft × 1 ft × 5 mil, sheet, from sigma aldrich), as depicted in Fig. S5. The integrity of the



sample was confirmed through Raman spectroscopy, as presented in Fig. S5.

Fig. S5. (Left) Raman spectroscopy of MoS2/ITO, ITO, and bulk MoS2. (Right) Photographs of the flexible ITO/MoS2 samples.

Interface quality of nanometer-thick YIG/TMDs

As discussed in the main text, spin pumping (SP) is highly sensitive to interfacial quality. To verify the interfacial quality of our deposition for films up to several nm thick, we conducted SP measurements on a YIG/MoS2 sample with a 27 nm thick MoS2, which is an order of magnitude thicker than the sample in the main text. Fig. S6 shows an increase in the Gilbert damping (α_{sp} = 6.2 ×10^{-4}) due to spin pumping, falling within the range of experimental error observed in the YIG/MoS2 sample from the main text. This result confirms the excellent interface quality, even for thicker samples.

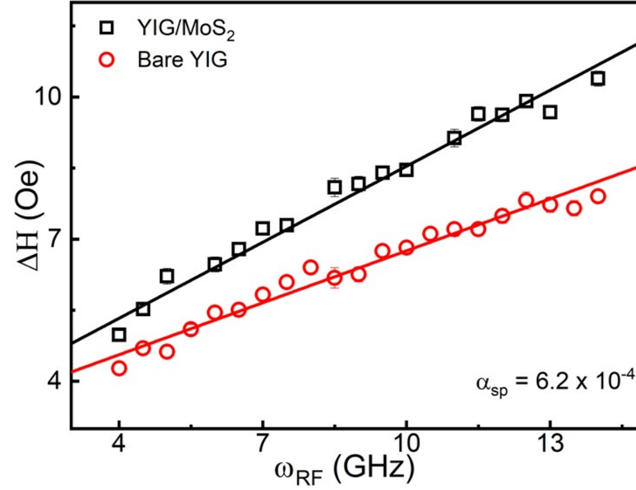


Fig. S6. FMR measurements of bare YIG in red and YIG/MoS2 with a thickness of 27 nm in black.

Automated mechanical exfoliation technique

Henceforth we aim to elucidate the difference between the exfoliation technique developed in this work, referred to as the non-conventional exfoliation technique, and the conventional exfoliation technique. Conventional mechanical exfoliation, also known as micro-mechanical exfoliation, involves the application of small mechanical forces during the exfoliation process. In the case of 2D transition metal dichalcogenides (TMDs), this technique breaks Van der Waals forces between adjacent layers by applying sufficient mechanical forces when the tape's adhesion force surpasses the newly produced surface energy. The conventional method has some inherent negative points; most of the time, a monolayer or set of layers is usually linked to the bulk TMDs crystal piece. Furthermore, the exfoliated flakes obtained are extremely small, which significantly limits their applicability for most practical uses.

The technique developed in this study exhibits notable distinctions from the conventional exfoliation approach in multiple aspects. One fundamental distinction lies in using the van der Waals (vdW) material powder as the initial material, in contrast to the crystal used in the conventional method. Another difference is that the material is exfoliated through friction between the deposition pad with the vdW powder and the substrate. Instead of using scotch tape for exfoliation, we use a soft polymer with a controlled surface to make the deposition pad. Finally, we have successfully developed an automated process capable of producing thin films across large areas, offering the advantage of easy scalability. This automated approach enables the efficient fabrication of thin films, making it a promising technology for various practical applications.

REFERENCES:

1. X. Zhang, X.-F. Qiao, W. Shi, J.-B. Wu, D.-S. Jiang, and P.-H. Tan, “Phonon and raman scattering of two-dimensional transition metal dichalcogenides from monolayer, multilayer to bulk material,” Chem. Soc. Rev. 44, 2757–2785 (2015).