Optoelectronic Properties of MoS_2/Graphene Heterostructures Prepared by Dry Transfer Method for Light-induced Energy Harvesting Applications

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Optoelectronic properties of atomic thin van der Waals heterostructures (vdWHs) comprising transition metal dichalcogenides that harvest light energy are of paramount interest. In this work, the effects of underlying single and bi-layer graphene (Gr) layers on structural and physical properties of MoS_2/Gr vertical heterostructures *i.e.,* (1-2L) MoS_2/(1-2L) Gr, besides additional interfaces including MoS_2 folds/edges [MoS_2(1L+1L)) /Gr(1L)] and MoS_2(1-2L) /Au, are investigated to unravel the excitonic properties. By employing correlative scanning probe microscopy combined with micro-spectroscopy, we observed multiple effects related to excitons (*i.e.,* redshifted neutral exciton, ratio of charged exciton or trion to neutral exciton population, and long-tailed trions) and surface electronic properties (*i.e.,* reduced work function suggesting electron transfer) in addition to significantly enhanced near-field Raman spectra, apparent n-p type current rectification behavior and increase in photo-generated carriers. These experimental findings are attributed to interlayer electronic interactions while minimizing Fermi level pinning at MoS_2/Au interface, commonly observed in 2D semiconductor−3D metal junction, and corroborated with theoretical DFT calculations, which deepened our understanding of dissimilar 2D materials junctions. Integrating MoS_2 with optimal number of graphene layers as 'nanospacer' signified substrate engineering that are versatile for key optoelectronic and photovoltaic applications [1, 2, 3, 4].

[1] Gupta S., Johnston A., Khondaker, A., Optoelectronic properties of MoS_2/graphene heterostructures prepared by dry transfer method for light-induced energy applications, *J. Electron. Mater.* 51 (2022) 4257.

[2] Gupta, S., Johnston, A., Khondaker, S., Correlated KPFM and TERS imaging to elucidate defect-induced inhomogeneities in oxygen plasma treated 2D MoS_2 nanosheets, *J. Appl. Phys.* 131 (2022) 164303.

[3] Gupta S., Dimakis N., First-Principles Calculations Integrated with Experimental Optical and Electronic Properties for MoS_2/Au and MoS_2/Graphene/Au Heterostructures, Appl. Surf. Sci. 623 (2023) 156948.

[2] Gupta *et al.,* Appl. Surf. Sci. **623**, 156948 (2023).

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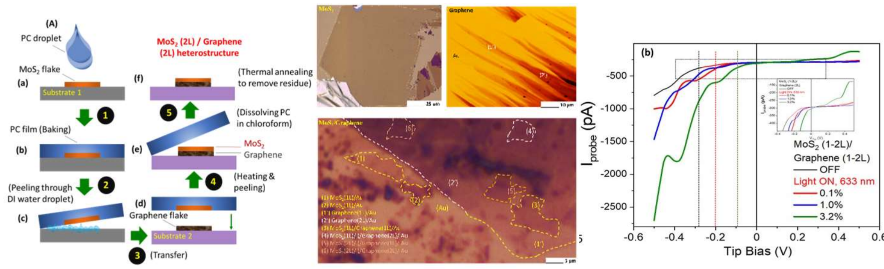

Figure 1. (Clockwise) Scheme of dry transfer technique to produce MoS_2/graphene vertical heterostructures, optical image of (a) metal exfoliated MoS_2, mechanical exfoliated graphene, and MoS_2/graphene heterostructures showing various heterointerfaces numbered from 1 to 6 and Local probe current measured using C-AFM as I−V curves. (a) MoS_2/Au heterointerface and (b) MoS_2/graphene vertical heterostructures, with and without light showing n-type behavior for MoS_2/Au and rectification behavior for MoS_2(n)/ graphene(p) heterointerfaces. The inset in (b) shows the zoomed area in dotted rectangle.

Supplementary information:

Optoelectronic properties of atomic thin van der Waals heterostructures (vdWHs) comprising transition metal dichalcogenides and graphene is continued to be of research interest for nanophotonics, nanoelectronics and energy harvesting applications. The structural and physical properties of MoS_2/Gr vertical heterostructures i.e., (1-2L) MoS_2/(1-2L) Gr, besides additional interfaces including MoS_2 folds/edges [MoS_2(1L+1L)) /Gr(1L)] and MoS_2(1-2L) /Au, are fabricated using dry transfer technique (Fig. 1) and investigated to unravel the excitonic and electronic properties. The use of multipronged approach such as optical and vibrational spectroscopy, Kelvin probe force microscopy and conductive-atomic force microscopy techniques, we established the interaction between MoS_2 and graphene layers, enhanced edge emissions, long-tailed excitons, charge transfer and n-p semiconducting junction like or rectification behavior useful for photodetectors. We corroborated our experimental findings with theoretical DFT calculations.

The effects of underlying single and bi-layer graphene on structural and physical properties of MoS_2/Gr vertical heterostructures i.e., (1-2L) MoS_2/(1-2L) Gr/Au/SiO_2/Si are investigated to unravel the intriguing excitonic and nanophotonic properties. We observed multiple effects related to excitons (i.e., redshifted neutral exciton, ratio of charged exciton or trion to neutral exciton population, and long-tailed trions) and surface electronic properties (i.e., reduced work function suggestive of electron transfer) in addition to significantly enhanced near-field Raman spectra at the folds and edges, apparent n-p type current rectification behavior and increase in photo-generated carriers. We attributed these observations to interlayer electronic interactions while minimizing Fermi level pinning at MoS_2/Au interface, commonly observed in 2D semiconductor−3D metal junction, and corroborated with theoretical DFT calculations, which deepened our understanding of dissimilar 2D materials junctions. Integrating MoS_2 with optimal number of graphene layers as 'nanospacer' signified substrate engineering that are versatile for key optoelectronic and photovoltaics [1-4].