Ultra-High Raman Enhancement on monolayer MoS_2

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Surface-enhanced Raman scattering (SERS) is usually associated with noble metal substrates. However, over the years modest Raman enhancements ($<10^4$) have been observed in semiconductor substrates[1]. This enhancement stems predominantly from the excitonic resonance of the semiconductors. Reducing the dimensionality using nanostructures was found to be an attractive pathway to further enhance this effect. Here we report for the first time > $3x10^5$ enhancement in SERS signal from an organic molecule (4-Mercaptopyridine) placed in the near field of a monolayer two-dimensional semiconductor (MoS_2). This large enhancement in the SERS signal is attributed to the charge transfer (CT) state formed between the two systems and is found to occur when the excitation source is in resonance with the CT state. We also show that exciting off resonance gives negligible enhancements. This approach of using the CT state resonances formed at the interface of 2D semiconductors and organic molecules provides a new strategy for carrying out SERS experiments on molecules with very weak Raman signatures without the need for nanopatterning.

1. Lombardi, J. R. & Birke, R. L. Theory of surface-enhanced raman scattering in semiconductors. *Journal of Physical Chemistry* **118**, 11120-11130 (2014).

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