

# Tuning the Fermi Level of Graphene by Two-Dimensional Metals for Raman Detection of Molecules

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Graphene-enhanced Raman scattering (GERS) offers great opportunities to achieve optical sensing with high uniformity and superior molecular selectivity.<sup>1</sup> The GERS mechanism relies on the charge transfer between molecules and graphene, which is difficult to manipulate by varying the band alignment between graphene and the molecules.<sup>2</sup> In this work, we synthesized a few atomic layers of metal termed two-dimensional (2D) metal to precisely and deterministically modify the graphene Fermi level. Using copper phthalocyanine (CuPc) as a representative molecule, we demonstrated that the Fermi level tuning can significantly improve the signal enhancement and molecular selectivity of GERS. Specifically, aligning the Fermi level of graphene closer to the highest occupied molecular orbital (HOMO) of CuPc results in a more pronounced Raman

enhancement. Density functional theory (DFT) calculations of the charge density distribution reproduce the enhanced charge transfer between CuPc molecules and graphene with a modulated Fermi level. Extending our investigation to other molecules such as Rhodamine 6G, Rhodamine B, crystal violet, and F<sub>16</sub>CuPc, we showed that 2D metals enabled Fermi level tuning, thus improving GERS detection for molecules, and contributing to an enhanced molecular selectivity. This underscores the potential of utilizing 2D metals for precise control and optimization of GERS applications, which will benefit the development of highly sensitive, specific, and reliable sensors.

#### Reference:

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