Coating-Induced Enhancement of Surface-Enhanced Raman Scattering: Multi-Scale Computational Insights

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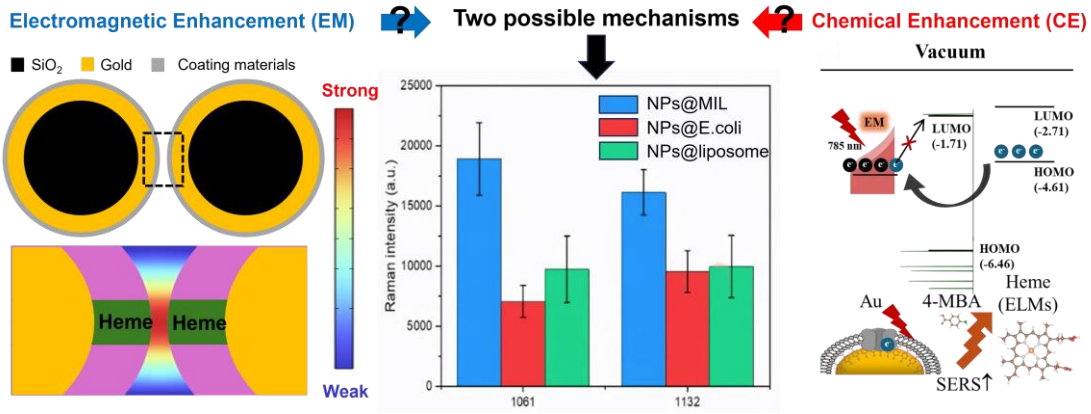
Surface-Enhanced Raman Scattering (SERS) is widely used to analyze the chemical composition and molecular structures of biomolecules. Experimental studies have shown that nanoparticles coated with different substrates exhibit varying SERS intensities under the same analyte conditions. Notably, MIL, a biomolecular membrane containing Heme, leads to higher SERS intensity compared to coatings with E. coli or liposomes. This intensity variation may result from two mechanisms: electromagnetic (EM) enhancement due to surface plasmon resonance on metal nanoparticles, and chemical enhancement (CE) caused by charge transfer between the analyte and nanoparticles. Identifying the dominant mechanism through experiments alone is challenging. This study uses multi-scale simulations to address this: EM effects are analyzed through numerical simulations, and CE is explored using Density Functional Theory (DFT) calculations. Our simulations show that the EM enhancement from different coatings is similar, with the Enhancement Factor (EF) for MIL being 8.56×10^6, slightly higher than for E. coli (8.54×10^6) and liposome (8.23×10^6). DFT calculations reveal that the HOMO of Heme in MIL is at -4.61 eV, enabling electron transfer to the gold nanoparticle's Fermi level at -5.2 eV. This interaction enhances charge transfer and boosts the SERS intensity. This approach helps clarify the underlying mechanisms in SERS studies and can guide the optimization of nanoparticle designs.



References

1. Wang, L.C., **Kuo, Y.C.**, Hung, M. Y., **Tian, H. K.***, Yeh, C. S.* **,** ***Advanced Materials*****, 2024, in revision.**