

Surface-enhanced Raman Scattering for Molecules with Interactions

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Surface-enhanced Raman scattering (SERS) has been considered as a powerful trace-level analytical technique for a diversity of fields. However, a molecule in real world inevitably interacts with other molecules or external stimuli, which often results in obvious changes of its SERS spectrum, e.g., relative intensity, broadening/narrowing, shifting, splitting/merging of the peaks, emergence of new peaks and/or disappearance of existed peaks, etc. These bring fundamental challenges to SERS in solving real-world problems through recognition of molecule's fingerprints, which has long been an open question in the field.

We proposed here a virtual molecules (VMs) and virtual spectra (VS) method to deal with molecule's interactions, and reformulated SERS with two concepts, i.e. cross-section function and relative scattering ability of molecules, which are valid for molecular systems with/without interactions. We established an approach to experimentally obtain the cross-section function and the relative scattering ability of VMs, by which the SERS behavior of molecular systems can be explicitly described by our formulation, with no need to recognize molecules' fingerprints.

We demonstrated the success of the VMs and VS method and the SERS formulation in several binary and complex molecular systems and the responses of molecule to external stimuli. Our study deepens the understanding of SERS and consolidates the foundation of SERS as a powerful trace-level analytical tool, pushing forward SERS applications in a diversity of fields to solve real-world problems.

Keywords: Surface enhanced Raman scattering; Molecules with interactions; Virtual molecules; Virtual spectrum;

References

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