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System-bath approach to electronic effect in Surface Enhanced Raman Scattering[1] SEMION SAIKIN, ROBERTO OLIVARES-AMAYA, CESAR RODRIGUEZ-ROSARIO, Department of Chemistry and Chemical Biology, Harvard University, MICHAEL STOPA, Center for Nanoscale Systems, Harvard University, ALAN ASPURU-GUZIK, Department of Chemistry and Chemical Biology, Harvard University, SEC TEAM — Raman scattering from molecules is greatly enhanced in proximity of a metal nanoparticle or a rough metal surface. The strong interest in this effect is driven by applications to selective detection of toxic chemicals, warfare agents, etc. The scattering enhancement has two distinct contributions. The electromagnetic effect originates in the field concentration by surface plasmons excited in the metal. The second, electronic or chemical contribution, which is important for molecules in direct contact with the surface, is more controversial. It is controlled by the charge transfer between a molecule and a metal with nanoscale roughness. We develop an open quantum system approach to the formation of charge-transferred states and apply it to describe electronic effect in SERS using specific examples of organic molecules adsorbed on a surface of a silver nanoparticle.

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