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Unified treatment of fluorescence and Raman scattering processes near metal surfaces PETER JOHANSSON, University of Orebro, Sweden, HONGXING XU, University of Lund, Sweden, MIKAEL KALL, Chalmers University, Goteborg, Sweden — We performed a general model study [1] of surface-enhanced resonant Raman scattering and fluorescence, focusing on the interplay between electromagnetic (EM) effects and the molecular dynamics as treated by a density matrix calculation. The model molecule has two electronic levels, is affected by radiative and non-radiative damping mechanisms, and a Franck-Condon mechanism yields electron-vibration coupling. Placing the molecule near two Ag nanoparticles leads to enhanced coupling to the EM field, and the Raman scattering can for realistic parameter values increase by some 10 orders of magnitude (to $\sim 10^{-14}$ cm²) compared with the free-space case. Also the fluorescence cross section grows with increasing EM enhancement, however, at a slower rate, and this increase eventually stalls when non-radiative decay processes become important. Finally, we find that anti-Stokes Raman scattering is possible with strong incident laser intensities, ~ 1 mW/ μ m.

[1]. H. Xu, X.-H. Wang, M. P. Persson, H. Q. Xu, M. Käll, and P. Johansson, to appear in Phys. Rev. Lett.

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