Quantum Interference between Energy Absorption Processes of Molecular Exciton and Interface Plasmons on Luminescence Induced by Scanning Tunneling Microscopy

KUNIYUKI MIWA, HIROSHI IMADA, RIKEN, MAMORU SAKAUE, HIDEAKI KASAI, Graduate School of Engineering, Osaka University, YOUSOO KIM, RIKEN — Luminescence induced by the tunneling current of a scanning tunneling microscope (STM) from molecule-covered metal surfaces is attributed to radiative decays of molecules and interface plasmons localized near the tip-substrate gap region. Since the dynamics of molecule and interface plasmons strongly influence each other, the interplay between these dynamics gives rise to peculiar phenomena originating from quantum many-body effects. In this study, we develop the effective model of the system and investigate the luminescence properties using the nonequilibrium Green’s function method. The results show that, in addition to the dynamics of molecule, energy reabsorption by interface plasmons have a critical role in determining the luminescence spectral profile of interface plasmons. The additional peak structure arises owing to the interference between these energy absorption processes. Origin of prominent peak and dip structures observed in recent experiments are identified by the developed theory. The details of the interference effects on the luminescence properties will be discussed.

1This work was supported by JSPS KAKENHI Grant Number 26886013.