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Quantum many-body effects in light emission from molecular exciton and plasmon induced by scanning tunneling microscopy

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Luminescence from the systems consisting of metal nanostructures (NSs) and adsorbed molecules can be strongly influenced by quantum many-body effects which arise from the interplay between dielectric response of metal NSs and intra-molecular electronic/vibrational excitations. In light emission induced by the tunneling current of a scanning tunneling microscope (STM) from molecule-covered metal surfaces, interface plasmons localized near the tip-substrate gap region play important roles in electronic excitations and radiative decays of the molecule. Recent experimental results have also suggested that the dynamics of molecules (e.g., luminescence and energy absorption) have an influence on the luminescence-spectral profiles of interface plasmons. Since the dynamics of molecules and interface plasmons have influence on each other, quantum many-body effects resulting from interplay between these dynamics are expected to occur. To unveil these effects from a microscopic point of view, there is a need to investigate the dynamics of the molecule and interface plasmons within the framework of quantum many-body theory. In this study, the effective model of the system was developed and investigated the effects of coupling between molecular exciton and interface plasmon (exciton-plasmon coupling) on the luminescence properties using the non-equilibrium Green's function method. It was found that in addition to the dynamics of the molecule, the dynamics of interface plasmons plays an essential role in determining the luminescence spectral profiles of interface plasmons. Prominent peak and dip structure observed in recent experiments are interpreted by the developed theory. The details of exciton-plasmon coupling on the luminescence properties will be discussed.

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