

Molecular Emission near Metal Interfaces: The Polaritonic Regime

Yuen-Zhou J., Saikin S., Menon V.

Kazan Federal University, 420008, Kremlevskaya 18, Kazan, Russia

Abstract

Copyright © 2018 American Chemical Society. The strong coupling of a dense layer of molecular excitons with surface-plasmon modes in a metal gives rise to polaritons (hybrid light-matter states) called plexcitons. Surface plasmons cannot directly emit into (or be excited by) free-space photons due to the fact that energy and momentum conservation cannot be simultaneously satisfied in photoluminescence. Most plexcitons are also formally nonemissive, even though they can radiate via molecules upon localization due to disorder and decoherence. However, a fraction of them are bright even in the presence of such deleterious processes. In this Letter, we theoretically discuss the superradiant emission properties of these bright plexcitons, which belong to the upper energy branch and reveal huge photoluminescence enhancements compared to bare excitons, due to near-divergences in the density of photonic modes available to them. Our study generalizes the well-known problem of molecular emission next to a metal interface to the polaritonic regime.

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