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Shaping anomalous molecular electroluminescence by resonant nanocavity plasmons¹

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Exciton formation and decay of molecules near metallic nanostructures is important for the control of energy conversion at the nanoscale and plasmonic devices. While extensive research has been carried out to such end using photon-excited techniques, complementary insights into the optical transitions and plasmon-exciton coupling can be obtained through electron excitations by a scanning tunneling microscope [1]. In this talk, we shall describe such reversed process of light-to-electricity conversion through single-molecule electroluminescence that demonstrates the critical role of nanocavity plasmons in the generation and decay of molecular excitons. By tuning the resonant interplay between molecular excitons and plasmons in nearby metallic nanostructures, we demonstrate that plasmons can do much more beyond intensity enhancement [2]: the emission band of molecular fluorescence can be effectively tuned by resonant nanocavity plasmons. New optoelectronic effects including resonant hot-electroluminescence and upconversion electroluminescence from higher vibronic levels of singlet excited states have been realized for porphyrin molecules near metals, which breaks the Kasha's rule and conventional Franck-Condon distribution. The highly confined nanocavity plasmons can behave like a strong coherent optical source with tunable frequency, and can be used to actively control the radiative channels of molecular emitters near metals over a wide spectral range. We shall also discuss critical factors that are responsible for the realization of single-molecule electroluminescence [3].

[1] Z.C. Dong, et al., Phys. Rev. Lett. 92, 086801 (2004).

[2] Z.C. Dong, et al., Nat. Photonics 4, 50 (2010).

[3] Y. Zhang, et al., Phys. Rev. Lett. (submitted).

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