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Exciton Diffusion and Annihilation in Nanophotonic Purcell Landscapes

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Abstract: Conventional nanophotonic emission enhancement neglects excitonic phenomena of diffusion and annihilation. We go beyond the localized Purcell effect and identify the enhancement mechanisms to turn their detrimental impact into additional emission.

Excitonic emitters in semiconductors such as perovskites, transition metal dichalcogenides and organic crystals disperse through diffusion and interact through exciton-exciton annihilation. [1–5] Nanophotonic resonators can improve light emission by enhancing excitation and radiative rates, and beaming the emission. [6–10] The conventional Purcell effect treats emitters like molecules and quantum dots as immobile and non-interacting. Total enhancement thus benefits from the product of excitation and emission at a point, which guides the design of nanoresonators and metamaterials made of metals and dielectrics. However, this approach is inadequate for excitonic emitters since it neglects diffusion over lengths comparable to nanophotonic scales (10 – 500 nm) and strong annihilation, which degrades the performance of light-emitting devices at high powers. [11]

Here, we go beyond the localized Purcell effect to exploit exciton dynamics for improving emission. We analyze the interplay of diffusion, annihilation, and nanophotonic enhancement for thin films of excitonic emitters in nanostructured landscapes (Figure 1). We provide analytical results for photoluminescence enhancement in the limiting regimes of high and low diffusion and annihilation. As excitons diffuse through optical hotspots and interact with each other, the balance of excitonic properties and nanophotonic geometry leads to either enhanced or suppressed photoluminescence. We present the conditions to turn the usually detrimental impact of diffusion on emission into additional enhancement and to overcome the adverse effects of annihilation at high exciton densities. Diffusion can increase photoluminescence by taking excitons to highly radiative locations when excitation and emission are spatially decoupled. Radiative rate enhancement can ameliorate the loss of efficiency arising from annihilation, while the interplay between annihilation and diffusion can improve performance by redistributing the local exciton density. Careful design of the relative strengths and spatial overlap of excitation and emission enhancements by tailoring the resonances and illumination conditions to the excitonic properties is thus key to efficient excitonic-nanophotonic systems. Our guidelines for customizing nanophotonic structures to diffusing and annihilating excitons will aid the design of efficient light-emitting devices based on excitonic nanomaterials.

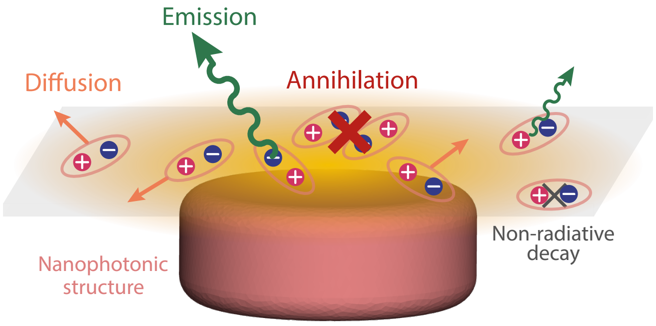

Fig. 1. Photoluminescence from a film of excitonic emitters near a nanophotonic structure. Excitons form in the near field (yellow), diffuse through the film (orange arrows), and annihilate by interacting with other excitons (red) before decaying radiatively with enhancement from the Purcell effect (green) or non-radiatively (grey).

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