Ultra-Efficient Förster-Type Nonradiative Energy Transfer via Tuning the Permittivity of Complex Dielectric Medium

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Förster-type nonradiative energy transfer (FRET) is a process of transferring excitons from the donor particles to the acceptor particles by nonradiative dipole−dipole interactions. FRET finds use in numerous applications important to color tuning, biosensing, light-harvesting, and light-generation. The FRET rate strongly depends on (1) the center-to-center separation between the donor and acceptor pair and (2) the Förster radius. Unfortunately, efficient FRET is only achievable for a limited length scale (approximately 10 nm) due to the strong distance dependence of the dipole−dipole interaction. This strong distance dependency can be alleviated by changing the nanocrystal geometry, for example, from a quantum dot to a quantum well [P. L. Hernández-Martínez, A. O. Govorov, and H. V. Demir, *J. Phys. Chem. C, 117 (19), 10203−10212 (2013)*]. This change in geometry consequently increases the FRET rate. However, this improvement remains limited. To further enhance the FRET rate, one can increase the strength of the donor-acceptor coupling by confining the surrounding electric field, in principle, using localized surface plasmon near metal nanoparticles. However, due to the lossy properties of such metal nanoparticles, placing metal nanoparticles can even decrease the FRET rate if not carefully designed. An alternative way of improving the FRET rate and efficiency is increasing the Förster radius. To do so, here we show that artificially engineered materials of complex dielectric medium with carefully tuned permittivity at a shifted complex point near zero, given a specific FRET pair, effectively confine and guide the electromagnetic energy within them owing to their low wavenumber and very large wavelength in the medium. Therefore, the proposed tailored complex medium can introduce the proximity effect in the long-range interactions and significantly enlarge the Forster radius, which can lead to a dramatic increase in FRET rate and consequently enable ultra-high FRET efficiency. In our work, we systematically studied the FRET mechanism by tuning the background medium's complex permittivity. In particular, the FRET rates of donor-acceptor pairs consisting of point-like, quasi-0-dimensional quantum dot (QD), and quasi-2-dimensional nanoplatelet nanostructures are analytically derived. The derived expressions are used to characterize the change of FRET rates with respect to the relative permittivity of the background medium. The analysis via the derived expressions reveals that the FRET rate becomes singular when the permittivity approaches zero and a fixed shifted non-zero value for the point-like and all other nanostructures, respectively. By setting the medium's relative permittivity to a realistic value near the singular point, an ultra-high FRET rate is achievable. For example, when cadmium selenide QDs are embedded in a medium with the relative permittivity of $-2+i$, the FRET rate is enhanced 53 times compared to that in the vacuum. Our numerical tests demonstrate the importance of carefully tunning the background medium permittivity for archiving an ultra-high FRET rate, which will potentially benefit and may find large-scale use in numerous FRET-based applications.