Enhancing Foerster-Type Nonradiative Energy Transfer by Tuning the Complex Dielectric Medium Permittivity

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Abstract: We systematically studied the FRET mechanism by tuning the background medium's complex permittivity. The FRET rates of donor-acceptor pairs for point-like, quantum dot, and nanoplatelet nanostructures were derived. The change in FRET rates with respect to the relative permittivity of the background medium was characterized. The analysis 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.

Förster-type nonradiative energy transfer (FRET) is a process where the nonradiative dipole−dipole interactions transfer excitons from the donor to the acceptor. FRET has numerous applications such as color tuning, biosensing, light-harvesting, and light-generation. The FRET rates strongly depend on (1) the center-to-center separation between the donor and acceptor pair and (2) the Förster radius, which limits its uses to a length scale of approximately 10 nm. To alleviate the strong distance dependency, one can change the nanocrystal geometry, for example, from a quantum dot to a quantum well [1], which decreases the distance dependency from d^{-6} to d^{-4} and consequently increases the FRET rate. However, this improvement remains limited. Another way to enhance the FRET is by strengthening the donor-acceptor coupling via a strong electromagnetic field using localized surface plasmon near metal nanoparticles. Nonetheless, due to the lossy properties of such metal nanoparticles, placing metal nanoparticles can even decrease the FRET rate if not carefully designed.

Here, we proposed an alternative way for improving the FRET rate and efficiency by using artificially engineered materials of complex dielectric medium with carefully tuned permittivity at a shifted complex point near zero, given a specific FRET pair, which 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 donor-acceptor interaction, leading to a dramatic increase in FRET rate and consequently enabling ultra-high FRET efficiency between them. For example, when cadmium selenide QDs donor-acceptor pair is embedded in a medium with the relative permittivity of $-2+i$, the FRET rate is enhanced 53 times compared to that in the vacuum (Figure 1) [2]. Our numerical results show the importance of carefully tunning the background medium permittivity for archiving an ultra-high FRET rate, potentially benefiting and finding large-scale use in numerous FRET-based applications.

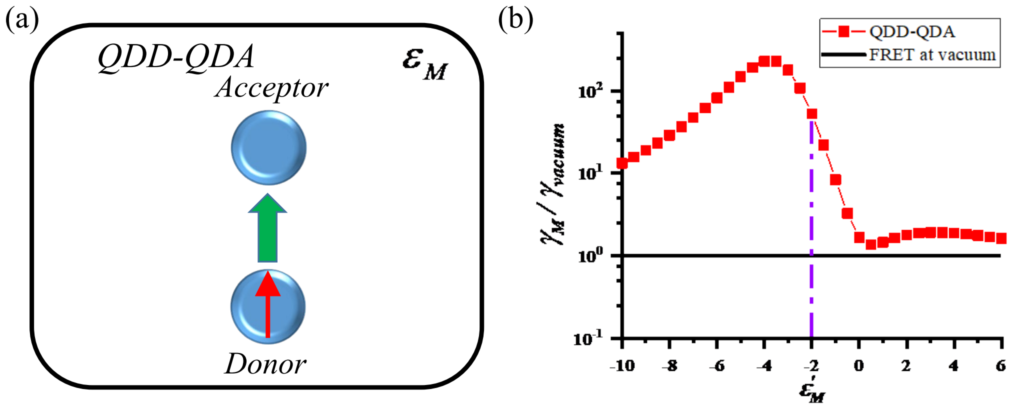

Figure 1. FRET enhancement factor, (γ_M/γ_{vacuum}), as a function of the real part of the permittivity of the dielectric medium for the FRET pairs of QDD-QDA.

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References

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