

Search for Optimal Arrangements of Perylene Pairs in Singlet Fission

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A multiple exciton generation, such as singlet fission (SF), in which one singlet exciton splits into two triplets could be utilized in organic photovoltaic materials to surpass the Shockley-Queisser limit.[1] The rate of SF depends not only on the excited states of the molecules but also their packing arrangements. Time-resolved spectroscopy has been carried out to investigate the SF rate for the crystal packing of perylene diimide (PDI).[2] Understanding the relationship between the packing arrangements and SF process plays an important role in tuning the favorable molecular pairs. A simple model described by the diabatic frontier orbitals is applied in searching for the local maximum rate of SF.[3] First, we examine the value of the square of the electronic matrix element in six-dimensional space. Then, the SF rates in the local maxima of molecular pairs were obtained with the approximation of the Marcus theory. A few molecular pairs of PDI and terylene diimide (TDI) have been found with fast SF rates to compare with their parental pairs in this simple model. These optimized geometries would provide a guideline for crystal engineering or covalent dimer synthesis in SF materials.

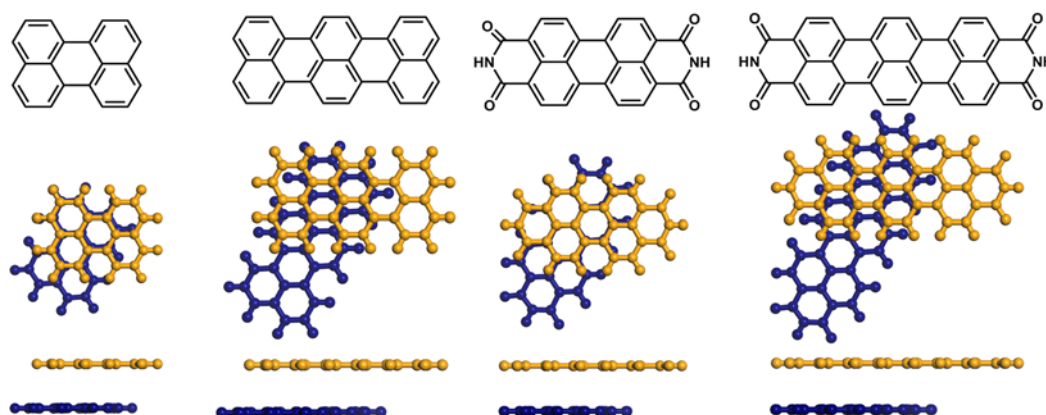


Figure 1: Optimal arrangements of molecular pairs for singlet fission.

References

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