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Molecular doping of nucleic acids into light emitting crystals driven by multisite-intermolecular interaction

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We reveal the fundamental understanding of molecular doping of DNAs into organic semiconducting tris (8-hydroxyquinoline) aluminum (Alq_3) crystals by varying types and numbers of purines and pyrimidines constituting DNA. Electrostatic, hydrogen bonding, and π-π stacking interactions between Alq_3 and DNAs are the major factors affecting the molecular doping. Longer DNAs induce a higher degree of doping due to electrostatic interactions between phosphate backbone and Alq_3. Among four bases, single thymine bases induce the multisite interactions of π-π stacking and hydrogen bonding with single Alq_3, occurring within a probability of 4.37%. In contrast, single adenine bases form multisite interactions, within lower probability (1.93%), with two-neighboring Alq_3. These multisite interactions facilitate the molecular doping into Alq_3 particles compared to cytosines or guanines only forming π-π stacking. Thus, photoluminescence and optical waveguide phenomena of crystals were successfully tailored. This discovery should deepen our fundamental understanding of incorporating DNAs into organic semiconducting crystals.

Since their inception as a typical genetic information carrier, nucleic acids have become a member of the material field and are widely used[1–4]. The unique physical and chemical properties make nucleic acid-associated materials the focus of numerous studies. For example, a nucleic acid molecule is generally complexed with π-conjugated organic semiconductors and serves as (i) an efficient receptor element for recognizing biological/chemical targets[5–7], (ii) a template for the assembly and polymerization of organic semiconductors[8–10], (iii) a walking component in a light-driven artificial nanomachine[11,12], (iv) a wide-bandgap material in organic light-emitting diodes enhancing their luminescence efficiency[2,13,14], (v) a molecular gadget for tuning organic semiconductor crystals bio-active when properly hybridized[15], and (vi) a biological moiety of organic hybrid crystals for remote sensing via optical waveguide effects[16].

Hybrid assemblies have become important in the field of self-assembly[17,18]. Binary or ternary hybrid assemblies have been prepared through molecular doping between organic semiconducting components[19], involving noncovalent intermolecular interactions, such as van der Waals force, π-π stacking, and hydrogen bonding[20–22]. The forms of hybrid assemblies can be classified into hetero structures[23,24] and uniform-[25,26] or gradient-doped[27] structures. However, deoxyribonucleic acids (DNAs) doped into light-emitting organic crystals exhibit distinctly different structures of molecular doping that has been unseen in conventional hybrid assemblies[3,15]. To date, studies have focused on the application of DNA-hybrid assemblies; however, little attention has been paid to how these nucleic acids interface with organic components at the molecular level. A fundamental understanding of the intermolecular interactions between nucleic acids and

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