FRET-mediated amplified spontaneous emission in biopolymer complexes

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Abstract

Förster resonance energy transfer (FRET), a long-range dipole-dipole interaction through which energy can be transferred between two chromophores separated by distances in the nanometer range, is becoming increasingly relevant in the field of photonics. Recently it has been shown how one can profit from such transfer mechanism in order to i.e. increase the efficiency of organic LEDs [1] or lasers [2].

Due to the nanometer sensitivity of FRET, materials presenting an intrinsic nanoscale structuration are commonly used as hosts to impose a chromophore distribution which can favor the efficiency of the FRET process. Some examples are zeolites, polymer blends or DNA. The latter, a biopolymer rapidly gaining attention in the field of photonics,[3] has been long studied in relation with FRET in both directions: using FRET as a means to detect structural changes in DNA and profiting from its natural scaffold to improve or control the transfer process. Recently DNA-lipid complexes [4,5] have been proposed as an environment where efficient FRET can be achieved by controlling the donor-acceptor concentration ratio [6]. These complexes are extremely attractive from the point of view of applications in photonics as they are soluble in organic solvents, represent a cost-effective approach and can be cast in the form of a polymer allowing the preparation of thin films, fibers, etc. To date, the feasibility of triggering two or even three-step FRET process [7] has been demonstrated in this polymeric matrices but a precise understanding of the transfer process is lacking. Further, its potential as FRET-based gain media has not been explored so far. Crucial to improving current and developing new applications for this system is a detailed knowledge of the FRET mechanism.

In this work we unveil the statistics of FRET in the above mentioned solid state biopolymer matrices. Tracking the dynamics of a population of donor molecules D (Coumarin 480) as acceptor ones A (DMASMPI) are added we find that rather than a single transfer process, a distribution of transfer processes (see Figure 1) has to be considered and an effective efficiency introduced. Additionally, the occurrence of amplified spontaneous emission (ASE) is demonstrated and the possibility of controlling it with the FRET efficiency is assessed (see Figure 2) enabling DNA-lipid complex lasers.

References

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Figures

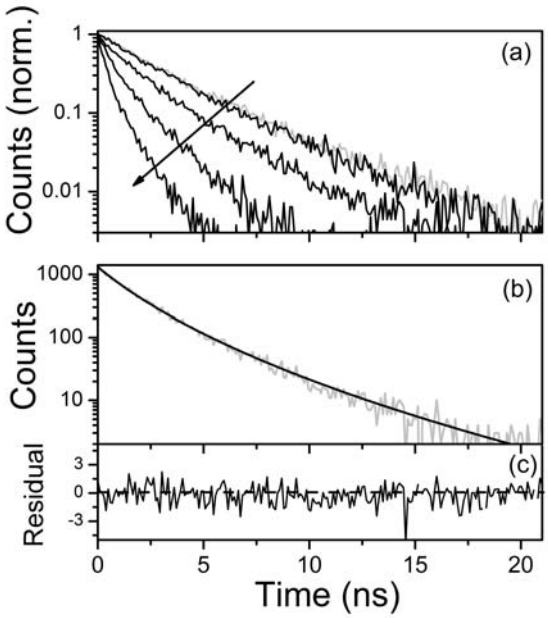

Figure 1. (a) PL decay curves for DNA-CTMA films with different ratios of D and A: 100:1 (grey curve) and 50:1, 25:1, 10:1 and 5:1 (black lines, arrow indicates decreasing values). (b) Decay curve for a sample having a 25:1 ratio (grey line) and a fit with a lognormal distribution of decay rates (black line). (c) Residual of the above fit.

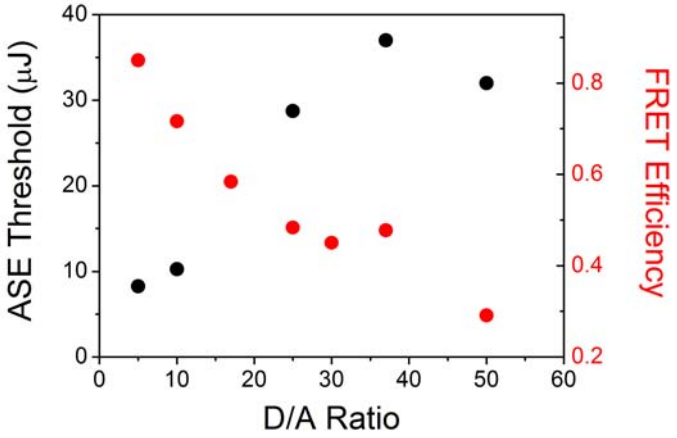

Figure 2. ASE threshold for DMASMPI emission (solid circles) and FRET efficiency (open circles) as a function of D/A concentration ratio.