Observation of efficient transfer from Mott-Wannier to Frenkel excitons in a hybrid semiconductor quantum dot/polymer composite at room temperature

Sedat Nizamoglu, Xiao Wei Sun and Hilmi Volkan Demir

Department of Electrical and Electronics Engineering, Department of Physics, UNAM - Nanotechnology Research Center and Institute of Material Science and Nanotechnology, Bilkent University, Ankara 06800, Turkey

Microelectronics Division, School of Electrical and Electronics Engineering, Nanyang Technological University, Nanyang Avenue, Singapore 639798, Singapore

Physics and Applied Physics Division, School of Physical and Mathematical Sciences, Nanyang Technological University, Singapore 639798, Singapore

Supplementary Information

For efficient NRET, the energy transfer is expected to be comparable to or faster than the recombination lifetime of the donor. If this condition is satisfied, then a significant fraction of the excitation energy can possibly be transferred to the acceptor.[4] Thus, it is desired to select an inorganic material having Mott–Wannier-type excitons as the donor and organics with Frenkel-type excitons as the acceptor. Since inorganic semiconductor quantum dot nanocrystals (NCs) show recombination lifetimes of tens of nanoseconds, they are suitable candidates as the donors for organic substances. Furthermore, the interface between the organic and inorganic materials is required to have a high purity and electronic perfection; if this condition is not satisfied, then the excitons can be trapped in defect states leading to

nonradiative recombination. For strong energy transfer, NCs and polymers are favorable candidates with their advantageous properties.[5–14]

Fig. S1 shows the photoluminescence (PL) spectrum of CdSe/ZnS core/shell heteronanocrystals (NCs) and the absorption and PL spectra of poly[2-methoxy-5-(3,7-dimethyl-octyloxy)-1,4-phenylenevinylene] MDMO-PPV homopolymer. The heteronanocrystals (Evident Technologies) are carefully chosen to match their emission peak with the absorption peak of the homopolymer (American Dyes Source Inc).

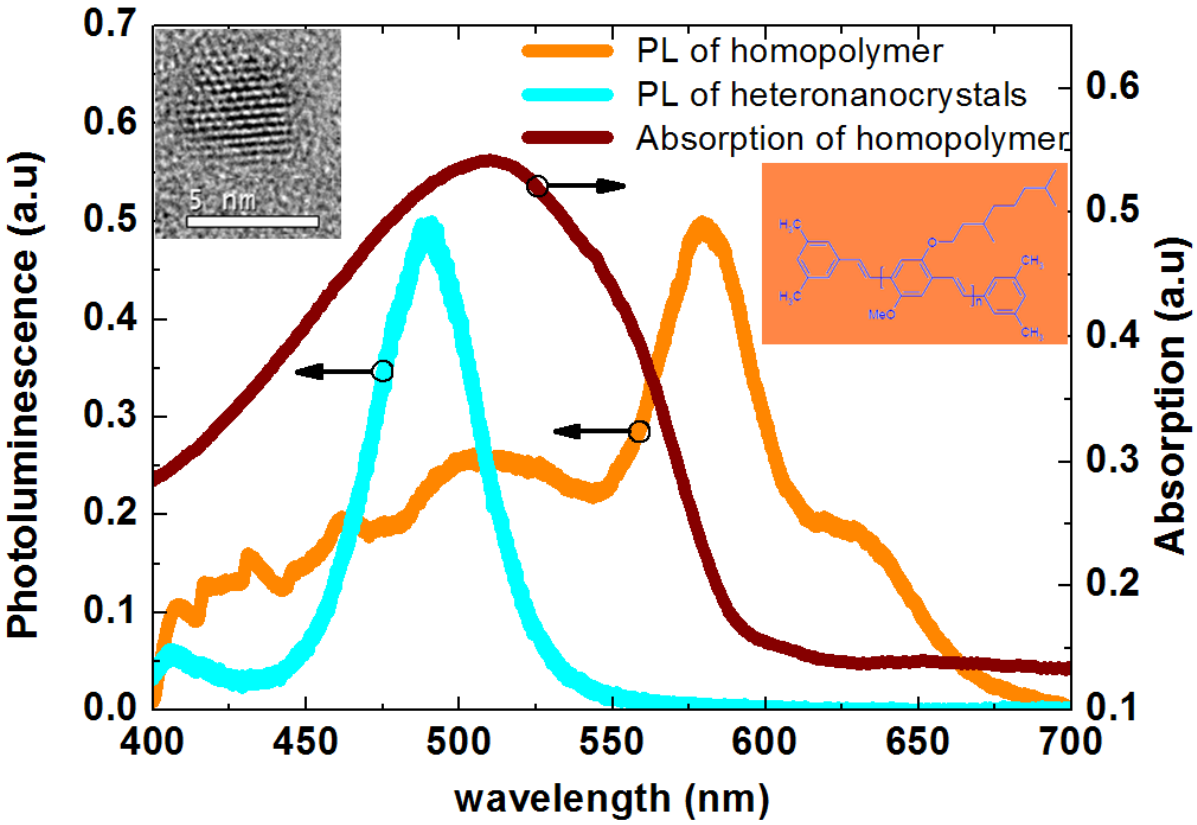

Fig. S1 Absorption and photoluminescence (PL) spectra of MDMO-PPV homopolymer along with the PL spectrum of CdSe/ZnS core/shell heteronanocrystals. Inset: TEM (transmission electron microscopy) image of the heteronanocrystal and chemical structure of the homopolymer.

To study the exciton migration, we utilized time-resolved photoluminescence measurements using FluoTime 200 spectrometer from PicoQuant with a time-correlated single photon counting (TCSPC) system of PicoHarp 300. As the excitation source, we used a pulsed diode laser operating at a wavelength of 375 nm with a pulse duration of 70 ps, and for signal detection, we employed a photon multiplier tube PMT. The response of the laser on the PMT detector is shown in the inset of Fig. 1. The resolution of our experimental apparatus can be adjusted from 4 to 512 ps. For heteronanocrystals, we use a resolution of 32 ps, and for polymers, we use a resolution of 4 ps (because of their relatively fast decay).

For the data analysis, the photoluminescence decays were fitted using FluoFit equipped with the FluoTime 200 spectrometer. In the fitting, a multiexponential least square error model was adopted, which is convoluted with the laser diode response (instrument response function - IRF) as given in Eq. (S1).

$$I(t) = \int_{-\infty}^{t} IRF(t') \sum_{i=1}^{n} A_i e^{-\frac{(t-t')}{\tau_i}} dt' \tag{S1}$$

Here n represents the number of lifetime components; A_i, the fitting amplitude; and τ_i, the lifetime.

For the reference NC-only sample, Eq. (S1) becomes

$$I_{ref}(t) = \int_{-\infty}^{t} IRF(t') \{ A_1 e^{-\frac{(t-t')}{\tau_{nc}}} \} dt' \tag{S2}$$

where A_1 is the fitting amplitude of single lifetime component for τ_{nc}.

In Table SI, the fitting parameters to the hybrid composite system decays are summarized.

Table SI. The multiexponential fitting parameters of only heteronanocrystal (only NCs) as the reference group and the hybrid composite systems consisting of both heteronanocrystals-

homopolymers (Samples 1-4) at nanocrystal emission peak wavelength (λ=495 nm), presented along with their associated Förster resonance energy transfer efficiency (η) and χ^2.

	A_1	τ_1 (ns)	A_2	τ_2 (ns)	η	χ^2
Only NCs	1162.40	16.16	-	-	-	1.006
Sample 1	736.70	16.16	406.4	7.00	0.566	1.003
Sample 2	465.20	16.16	642.7	5.87	0.636	1.482
Sample 3	165.75	16.16	844.8	4.15	0.743	1.276
Sample 4	114.42	16.16	947.6	3.08	0.809	1.124