Ultrafast energy and electron transfer in donor-acceptor molecules for photovoltaics


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We have investigated the excitation transfer in a novel polymer/dye system consisting of the blue-light-emitting conjugated polymer poly(6,12,18-tetra-2-ethylhexyl-2,8-indenofluorene) (PIFTEH) as a host and red-light-emitting perylene molecules covalently bonded to the PIFTEH chain ends as a guest. The spectral overlap between the host emission and the guest absorption leads to an efficient Förster excitation transfer, such that photo-excitation of the host results in emission originating predominantly from the guest (see Fig. 1). Using femto-second time-resolved photoluminescence (PL) spectroscopy, we find that the transfer of excitations from isolated PIFTEH chains to perylene molecules is completed within the first 30–40 ps after excitation, and we extract a Förster radius of (1.8 ± 0.3) nm from fits to the PL transients (Fig. 2).

We have modelled the polarization anisotropy for a host–guest system host to Förster interactions using a Monte Carlo simulation and find that the emission from the guest becomes unpolarized at sufficiently high guest concentrations, even if the host emission displays a significant polarization anisotropy due to excitation with linearly polarized light. These results allow the separation of two overlapping contributions to the film emission in the red spectral region: the luminescence from perylene molecules and from sites where the PIFTEH chains aggregate. We find that while spectral overlap calculations predict a large transfer of excitation to perylene molecules from sites where the PIFTEH chains aggregate, no transfer is observed experimentally. We attribute this to chain packing effects within the film: at sites where aggregation effects are dominant, the PIFTEH chains will be closely packed and oriented to some extent, prohibiting sufficiently close contact between the host (PIFTEH) and the guest (perylene). The influence of interchain interactions on the excitation transfer probability is an important factor in the design of highly efficient light-emitting diodes (LEDs) based on conjugated polymers.

Our novel approach, to bond covalently the dye guest molecules to the host polymer has distinct advantages over dye doping, since it prevents the phase separation of the dye molecules and therefore reduces the probability of concentration quenching for the dye emission. By incorporation of various dyes with suitable emission wavelengths, full-colour displays based on plastics are submitted to Advanced Materials.

References

duced. energy transfer to the fullerene moiety by
motion of the charge-separated state by the polar
medium. Moreover, at the same time an increase of
the PA $\lambda_{op}$ band of C$_{60}$, which has a much
lower cross section. In toluene this PA $\lambda_{op}$ band
has a long lifetime as inferred from fig. 2b. How-
ever, in odcb subsequent to this ultrafast pho-
tonic-induced energy transfer, a fast intramolecular
electron transfer is observed due to the stabiliza-
tion of the charge-separated state by the polar
medium.

A few ps after photoexcitation of C$_{60}$-OPV$_4$ in
odcb a rising signal at 490 nm is observed due to
the PA $\lambda_{op}$ band of C$_{60}$, which has a much
lower cross section. In toluene this PA $\lambda_{op}$ band
has a long lifetime as inferred from fig. 2b. How-
ever, in odcb subsequent to this ultrafast pho-
tonic-induced energy transfer, a fast intramolecular
electron transfer is observed due to the stabiliza-
tion of the charge-separated state by the polar
medium.

The phase-encoded sampling technique used in
the 505-MS/s photonic ADC reported here pro-
vides high linearity$^9$ and large suppression (50-
dB) of laser amplitude noise.$^9$

2. System description

Figure 1 illustrates the photonic ADC architec-
ture. The short-pulse optical source is a model-
locked sigma fiber laser that is driven with 130-ps
electrical pulses produced by a resonant tunnel-
ing diode (RTD) oscillator. The laser generates
25-ps pulses at the 505-MHz sampling rate.
Phase-encoded optical sampling is performed via
a dual-output LiNbO$_3$ Mach-Zehnder modulator
($V_m = 2.9$ V, BW = 3 GHz). A pair of 1-to-8 opti-
cal time-division demultiplexers is used to dis-
tribute the modulator's complementary output
pulse streams to an array of photonic integrate-
and-reset (PHIR) circuits followed by 14-bit
electronic quantizers (Analog Devices AD6644)
operating at 1/8 the sampling rate (63 MS/s).
Fiber delay lines are used to time-align the pulses
at the PHIR circuit inputs to simplify PHIR and
quantizer timing. The quantized samples are cal-
ibrated, phase-decoded, and re-interleaved to
produce a digital estimate of the electrical input
signal.

The Ti-indiffused LiNbO$_3$ 1-to-8 optical de-
multiplexers employ a high-extinction design
(Fig. 2) to minimize crosstalk between parallel
channels. Each of the seven demux switch stages
consists of a 1 x 2 switch element with an extra
extinction modulator at each output arm. The
stages are driven using phased sinusoids and the
three elements of a given stage are driven by a
common signal. The extinction for a single stage
ranges between 36 and 46 dB. The bandwidth of
a stage is 600 MHz. The half-wave voltages ($V_{\text{th}}$)
range from 6.5 to 7.4 V with <2.5% variation
within a single stage. The total insertion loss for a
channel ranges between 6.8 and 8.4 dB.

3. Photonic ADC performance

Initial characterization of the photonic ADC was
performed by sampling a 220-MHz, 0.32-V$_p$ signal
(Fig. 3). The linearity of the system can be es-
timated from the amplitude of the third harmonic
which is 83 dB below the main tone. This

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