## Ultrafast Dynamics of Charge Transfer in Fullerene-Free Polymer Solar Cells with over 11% Efficiency

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Figure 1: Transient absorption spectra showing hole transfer: Investigation of a polymer:small-molecule blend, used as the active layer in a solar cell. The hole donor molecules in the blend are selectively excited. The initial spectrum resembles the spectrum of the pure hole donor film. At later times the spectrum contains features of both the pure hole donor film and the pure hole acceptor film. GSB: Ground State Bleach, PIA: Photoinduced Absorption

Solution processed polymer solar cells (PSC) are promising candidates for next-generation solar energy conversion. Large-area processibility, low cost, flexibility, and low weight make them a potential replacement for current silicon based technologies. However, low photoconversion efficiencies and poor device stabilities currently prevent commercial applications.

Typical PSCs are based on blends of polymers with fullerene derivatives such as  $PC_{71}BM$ . In this configuration, photoexcitation of the polymer is followed by electron transfer to the fullerene derivatives. These derivatives have desirable properties such as high charge mobilities and form advantageous morphologies via aggregation. Yet, their efficiency is limited by a weak absorption in the wavelength region where the solar spectrum is most intense. Additionally, fullerene diffusion leads to a further decrease of performance over time.<sup>[1]</sup> Consequently, research into non-fullerene electron acceptor materials has recently attracted much attention. These materials can be designed to have a strong absorbance in the relevant spectral region, allowing photoexcitation of the electron acceptor as well. If this is followed by hole transfer into the polymer, it opens a new channel for photocurrent generation, thus increasing efficiency. Furthermore, good tunablity of energy levels allows the design of optimal donor-acceptor combinations with complementary absorption spectra.<sup>[2]</sup>

Here, we present a study on the ultrafast photophysics of a fullerene-free polymer solar cell with an efficiency exceeding 11%,<sup>[3]</sup> among the highest values reported in the field. In addition, this blend of a conjugated polymer with a small molecule shows an excellent thermal stability.<sup>[3]</sup> We investigate the dynamics of exciton decay, charge transfer, and charge separation with transient absorption spectroscopy, achieving a time-resolution of below 15 fs. The use of extremely short pulses enables us to measure the characteristic vibrational modes of the molecules and study how they evolve during charge transfer. In combination with other time-resolved and steady state optical measurements, we can identify the processes leading to the high efficiency and provide guidelines for the synthesis of new materials.

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