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Ultrafast and Nonlinear Optical Spectroscopies of Excited States in Pristine and Doped π-Conjugated polymers[1]

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A variety of ultrafast and optical nonlinear spectroscopies were applied to pristine and doped π-conjugated polymers, for elucidating the excited states energy levels and primary photoexcitation species in these materials. These spectroscopies include fs pump-probe photomodulation (PM), two-photon absorption (TPA), photoluminescence up-conversion (PL(t)), and electroabsorption (EA); as well as THz time domain spectroscopy (THz-TDS). The π-conjugated polymers include derivatives of PFO, PPV and PT, as well as t-$(CH)_x$; doping includes fullerene molecules, as well as heavily doping with strong acceptors. The results have been analyzed in terms of the exciton picture advanced by Mazumdar et al. The primary photoexcitations are singlet excitons of which PM spectrum is composed of two strong photoinduced absorption bands in the mid and near ir spectral range, that are correlated with a stimulated emission band and PL(t). These bands are in agreements with transitions from the lowest exciton with odd symmetry into higher lying excitons with even symmetry, as revealed by TPA and EA spectroscopies. Polaron excitations are also formed and are characterized by two PA bands in the mid-ir range, and correlated ir-active vibrations. Surprisingly t-$(CH)_x$ is not different from many other π-conjugated polymers, except that the primary polarons recombine at a later time to form charged solitons. In fullerene-doped polymers the primary singlet excitons are trapped and undergo ultrafast nonradiative decay in doping-related defects, and this explains in part the weak cw PL in these compounds. In heavily doped polymers with strong acceptors the ground state no longer is neutral, but rather contains substantial amount of free charge carriers characterized by the Drude free carrier response in the THz to mid ir spectral ranges.

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