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Frank Isakson Prize Talk: Optical Probes of π-Conjugated Polymers[1]

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We review several *optical probes* that have been applied to π-conjugated polymers over a time period of $\sim$ 30 years. These include linear and nonlinear optical spectroscopies, resonant Raman scattering, transient and steady state photomodulation, photoluminescence and laser action, and optically detected magnetic resonance spectroscopy. The application of these techniques has revealed a myriad of important information on the interaction that govern the optical, electrical and magnetic properties of these materials; including electron-phonon interaction, electron-electron (e-e) and electron-hole (e-h) interactions, interchain coupling, spin-lattice and spin-orbit coupling. These properties are very important for various *optoelectronic applications*, in which the polymers serve as active layers. The following picture of the excited state properties of these polymers has emerged. (i) The e-e and e-h interactions are substantial, and as important as the electron phonon interaction. This leads to relatively large intrachain exciton binding energy of $\sim$0.5 eV, and exchange energy between the singlet and triplet lowest states of $\sim$0.7 eV. (ii) There are few important excited states with odd and even parity symmetry that govern the nonlinear optical spectra of these materials. (iii) The primary photoexcitations are intrachain excitons in isolated chains, and both excitons and polaron pairs in chains coupled by interchain interaction. (iv) The most strongly coupled phonons are amplitude modes of which frequencies and oscillator strengths are very sensitive to the existence of excess charges on the chains. (v) Excess charges are accommodated on the chains in the form of polarons with relatively large relaxation energy ranging from 0.1 to 0.5 eV. (vi) The spin orbit coupling is very weak in these materials, but can be tuned by involving heavy atoms in the polymer building blocks. (vii) The spin relaxation time for spin $1/2$ polarons is relatively long of $\sim$ 1 microsecond. The two latter properties may lead to new applications in the field of Organic Spintronics.

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