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Two-dimensional Fourier transform spectroscopy of primary excitations, in conjugated polymers KENAN GUNDOGDU, CONG MAI, ANDREW BARRETTE, ROBERT YOUNTS, TERRY MCAFEE, HARALD ADE, NC State University — Conjugated polymers have tremendous potential for use in cheap, flexible, lightweight, energy efficient opto-electronic applications, Despite years of work, critical fundamental aspects about their optical and electronic properties are still poorly understood. Photo absorption in pure semi-conducting polymer thin films eventually results in both free charges and bound excitons with varying branching ratios. However the identification of the nature of early excitations and charge generation is an unresolved problem. There has been no direct observation of initial excitons or free electron-hole pairs, and competing views persist. Here we use 2D Fourier transform spectroscopy methods to separate the spectral signatures of various processes in the photoabsorption process in a homopolymer and show that initial excitation results in an intrachain electronic coherence that persists more than 200 fs. As these coherences evolve they collapse to transient population states i.e excitons, polarons and bipolarons.

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