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Temperature dependent interplay between emitting species in highly ordered P3HT single crystals

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In this study, the temperature dependent photoluminescence (PL) spectra measurement has provided us a feasible means to elucidate the nature of the emissive species and the melt transitions in P3HT single crystals. This has been achieved by performing *in situ* temperature dependent photoluminescence measurements followed by detailed spectral analysis. Spectrally resolved PL lineshapes through multipeak Gaussian functions simulating 0-0, 0-1, 0-n peaks have revealed multiple vibrational replicas yielding two and three emitting species (states) between 30°C–220°C and 240°C–290°C, respectively. Highly ordered crystalline domains with melt transition at ~302°C are suggested to form the structural and electronic origin of the species 1 emission while the less ordered domains with melt transition at ~240°C are suggested to form the origin of species 2 emission. At high temperatures, disordered domains whose PL lineshapes resembles that of P3HT solution emerged between 240°C and 310°C and is seen to be responsible for species 3 emission (single chromospheres emissions). This observation has further been corroborated by the temperature dependent birefringence measurements. We suggest that the temperature dependent vibronic progressions arise from different electronic origins i.e. different species (fluorophores) due to multiple crystalline polymorphs within the crystal with varied coupling of the excited states. We conclude that it is not sufficient to invoke only the intramolecular interactions to explain the nature of both the absorption and PL spectra of highly ordered P3HT single crystals which are widely dominated by both interchain and intrachain interactions.

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