MAR05-2004-010178

Abstract for an Invited Paper
for the MAR05 Meeting of
the American Physical Society

Understanding the Intra- and Interchain Electronic Structure of Conjugated Polymers by Encapsulation in Mesoporous Silica

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It is becoming increasingly clear that the morphology of a conjugated polymer sample – that is, the conformation of the individual polymer chains and the way they pack together – plays a direct role in the electronic properties conjugated polymer films. In this talk, we describe the results of work that provides a new method for controlling conjugated polymer morphology and hence electronic properties: encapsulation of the polymer chains into the channels of aligned mesoporous silicas. We find, for example, that in small-pore silica samples where only a single polymer chain can fit into each pore, essentially no polarons are formed upon excitation and phenomena such as exciton-exciton annihilation that are prevalent in bulk films do not occur. We also find that energy transfer (exciton migration) occurs much more slowly along the polymer backbone than between polymer chains, suggesting that the predominant mechanism for exciton diffusion in bulk films is interchain Forster energy transfer. In medium-sized pores that contain just a few polymer chains, we find that the chains are still aligned by the encapsulation, and that polarons can be formed upon directly upon excitation. We also see that exciton-exciton annihilation is now possible, but that it occurs predominantly at kinks or defect sites. In still larger pores, the polymer chains exhibit more bulk film-like properties, including a high degree of photogeneration of polarons and exciton-exciton annihilation. We also show that in the intermediate pore size regime, the alignment of the polymer chromophores leads to significantly lower lasing thresholds than bulk films of comparable optical density, even though the physical density of the chromophores encapsulated into the porous silica is much lower than that in bulk films. Taken together, the results allow a new picture of how interactions between polymer chains control the electronic properties of conjugated polymer films.