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Free exciton emission and vibrations in pentacene monolayers[1]

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Pentacene is a benchmark organic semiconductor material because of its potential applications in electronic and optoelectronic devices. Recently we demonstrated that optical and vibrational characterizations of pentacene films can be carried out down to the sub-monolayer limit. These milestones were achieved in highly uniform pentacene films that were grown on a compliant polymeric substrate. Films with thickness ranging from sub- monolayer to tens of monolayers were studied at low temperatures. The intensity of the free exciton (FE) luminescence band increases quadratically with the number of layers N when N is small. This quadratic dependence is explained as arising from the linear dependence of the intensity of absorption and the probability of emission on the number of layers N. Large enhancements of Raman scattering intensities at the FE resonance enable the first observations of low-lying lattice modes in the monolayers. The measured low- lying modes (in the 20 to 100cm^{-1} range) display characteristic changes when going from a single monolayer to two layers. The Raman intensities by high frequency intra-molecular vibrations display resonance enhancement double-peaks when incident or scattered photon energies overlap the FE optical emission. The double resonances are about the same strength which suggests that Franck-Condon overlap integrals for the respective vibronic transitions have the same magnitude. The interference between scattering amplitudes in the Raman resonance reveals quantum coherence of the symmetry-split states (Davydov doublet) of the lowest intrinsic singlet exciton. These results demonstrate novel venues for ultra-thin film characterization and studies of fundamental physics in organic semiconductor structures.

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