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Beyond the two-dimensional field-effect charge transport view in molecular thin film transistors

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Organic field-effect transistors (OFETs) are considered almost purely interfacial devices with charge current mainly confined in the first two seminconducting layers in contact with the dielectric with no active role of the film thickness exceeding 6-8 monolayers. The compact 2D layered film structure with the minimum possible thickness is targeted as an ideal condition to achieve the best device performance in terms of charge carrier mobility and source-drain current intensity. By a combined electronic, morphological, structural and theoretical investigation we demonstrate that in 2,3-thienoimide endcapped oligothiophene (NT4N) organic transistors the lateral field-effect charge transport between source and drain correlates with the vertical molecular packing and crystalline order in the direction perpendicular to the substrate.
OFETs based on polycrystalline films with thickness as high as 75 nm and 3D growth modality provide the best electrical and optoelectronic characteristics, highlighting that the molecular orientational order is the key-enabling factor to be maximized in organic systems other than the 2D monolayer crystalline structure. The cross-correlation of extensive structural analysis and two-step multiscale simulation provides a deeper insight into (i) the thickness-dependent molecular packing that allows electron mobility as high as 0,75 cm^2/Vs and (ii) the vertical charge distribution across the films thickness. These results call for a broader view of the fundamental mechanisms that govern field-effect charge transport in OFETs beyond the two-dimensional one, and demonstrate that the molecular packing motif and out-of-plane orientational order in polycrystalline films must be considered to design organic transistors with optimum electronic and optoelectronic properties.