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Orientation of conjugated polymers: epitaxy versus mechanical rubbing[1]

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Orientation of conjugated polymers like regioregular poly(3-alkylthiophene)s (P3AT) is of high importance as it can be used to exploit their high intrinsic charge transport anisotropy in the elaboration of devices e.g. OFETs and OLEDs. Orientation has been achieved by two different approaches: i) epitaxy and ii) mechanical rubbing. Herein we describe and compare these two orientation methods. On the one hand side, a large palet of structures, nanomorphologies and orientations can be achieved by controlled epitaxial crystallization of polymers like P3HT, PBTTT or polyfluorenes. Not only can epitaxy afford highly oriented and crystalline films of P3ATs, but also highly oriented fibers with a characteristic shish-kebab morphology. The application of Electron Diffraction analysis (rotation-tilt) on highly oriented polymer layers is an original an powerfull method to unravel the crystal packing of conjugated polymers. On the other hand side, mechanical rubbing of conjugated polymers, especially P3HT can also lead to highly oriented films without the use of an orienting substrate. The mechanism of thin film orientation has been analyzed in detail using Transmission Electron Microscopy, Grazing-Incidence X-ray diffraction and optical spectroscopy. It is demonstrated that the molecular weight Mw of the polymer impacts the maximum orientation achieved by rubbing. The Mw-dependence of orientation is explained in terms of chain folding and entanglements that prevent the reorientation and reorganization of the pi-stacked chains, especially for Mw$\geq$50kDa. Electron diffraction and HR-TEM show that epitaxied and rubbed films differ in terms of *intra-lamellar* order within layers of pi-stacked chains. Whereas the epitaxied P3HT films show a semi-crystalline structure with crystalline domains bearing 3D order, the rubbed P3HT films exhibit rather a 2D nematic-like order.

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