Supporting Information.

Spotlight on Excitonic Coupling in Polymorphic and Textured Anilino Squaraine Thin Films

Frank Balzer,[†] Heiko Kollmann,[‡] Matthias Schulz,[¶] Gregor Schnakenburg,[§] Arne Lützen,[¶] Marc Schmidtmann,[∥] Christoph Lienau,[‡] Martin Silies,[‡] and Manuela Schiek[*,⊥]

[†]*Mads Clausen Institute, University of Southern Denmark, Alsion 2, DK-6400 Sønderborg, Denmark*

[‡]*Ultrafast Nanooptics, Institute of Physics, University of Oldenburg, D-26111 Oldenburg, Germany*

[¶]*Kekulé Insitute of Organic Chemistry and Biochemistry, Rheinische-Friedrich-Wilhelms-University of Bonn, Gerhard-Domagk-Str. 1, D-53121 Bonn, Germany*

[§]*Institute of Inorganic Chemistry, Rheinische-Friedrich-Wilhelms-University of Bonn, Gerhard-Domagk-Str. 1, D-53121 Bonn, Germany*

[∥]*Department of Chemistry, University of Oldenburg, D-26111 Oldenburg, Germany*

[⊥]*Energy and Semiconductor Research Laboratory, Institute of Physics, University of Oldenburg, D-26111 Oldenburg, Germany*

E-mail: manuela.schiek@uni-oldenburg.de

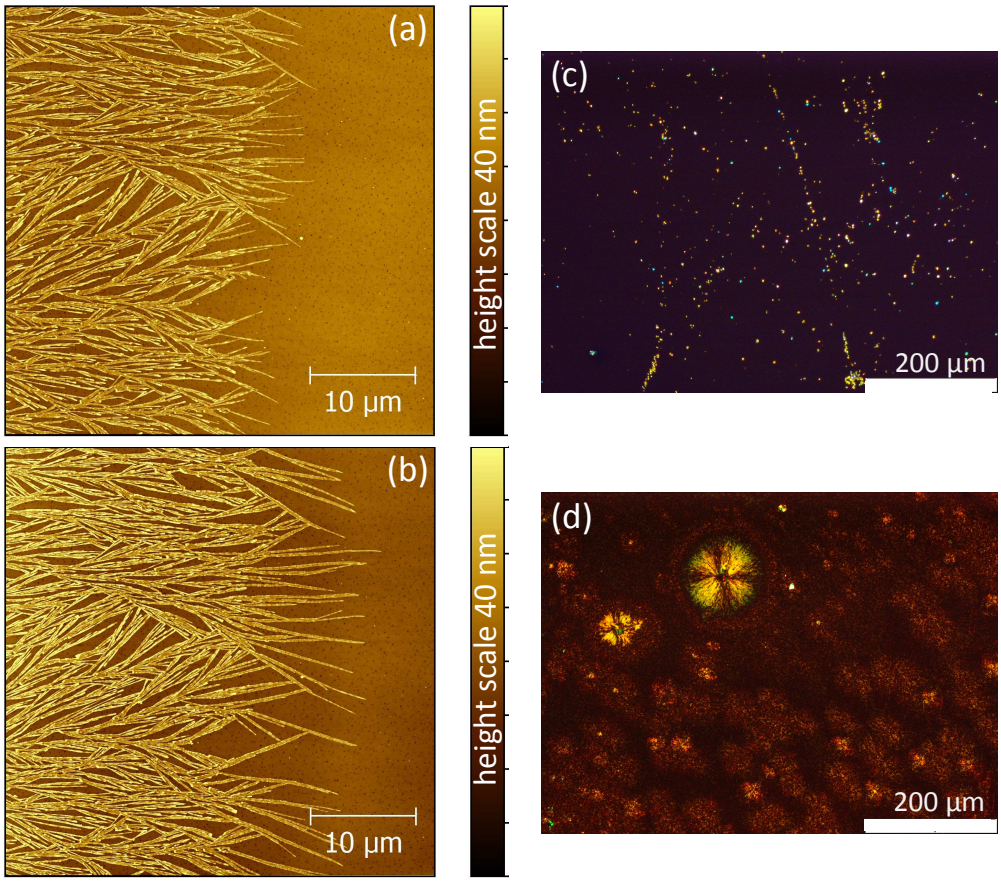

Figure S1: AFM images monitoring the growth of a sunflower in an amorphous SQIB film, time between (a) and (b) are approx. 42 hours. Optical microscopy images between crossed polarizers of (c) a fresh SQIB thin film annealed at 60 °C and (d) the same sample after two months storage.

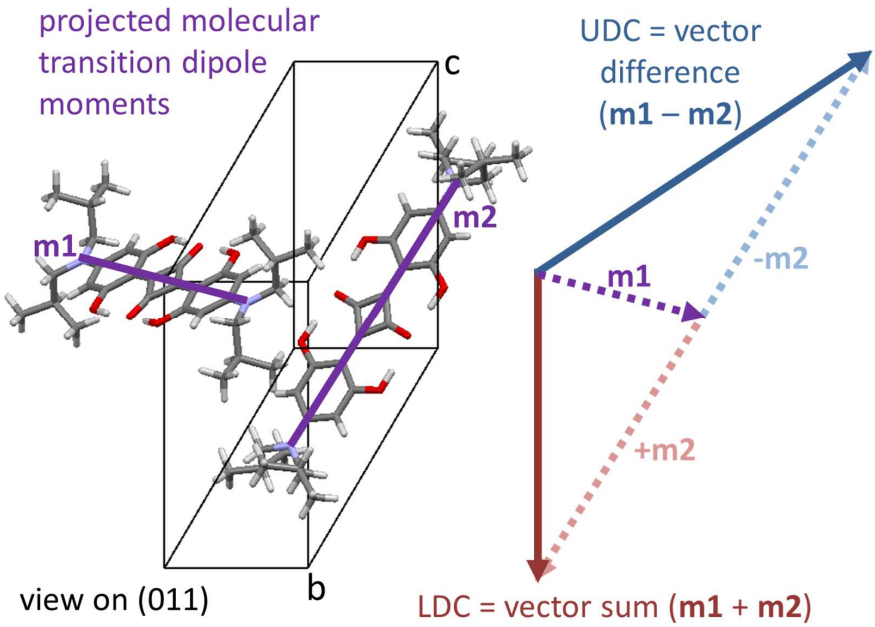

Figure S2: Graphical vector addition (red arrow, LDC) and subtraction (blue arrow, UDC) of the molecular transition dipole moments (violet color) for the monoclinic polymorph (sunflowers).

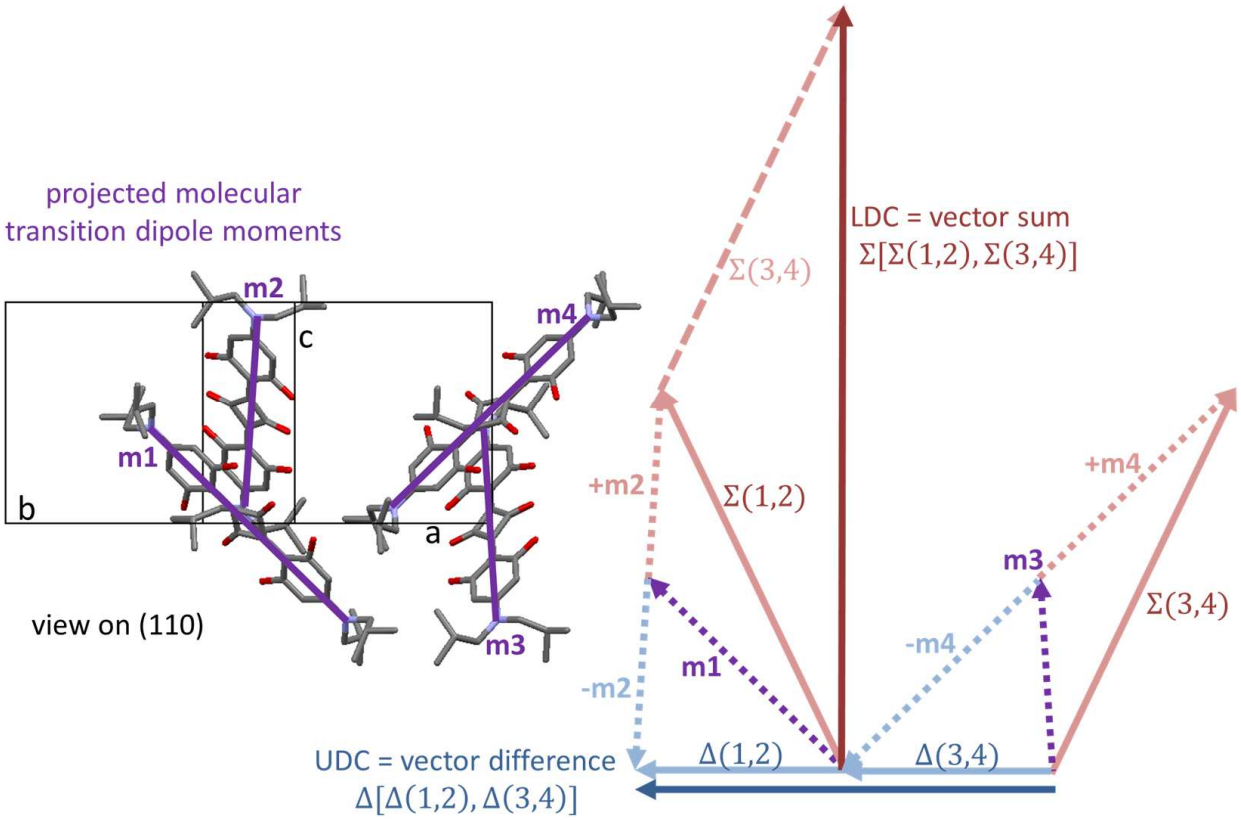

Figure S3: Graphical vector addition (red arrow, LDC) and subtraction (blue arrow, UDC) of the molecular transition dipole moments (violet color) for the orthorhombic polymorph (platelets). At first, the vector sums Σ [differences Δ] of the two pairs of stacked molecules are calculated: $\Sigma(1,2) = m1 + m2$ [$\Delta(1,2) = m1 - m2$] and $\Sigma(3,4) = m3 + m4$ [$\Delta(3,4) = m3 - m4$]. Secondly, the vector sum [difference] of the stacked pairs are graphically added [subtracted]: $\Sigma(1,2) + \Sigma(3,4) = \text{LDC}$ [$\Delta(1,2) - \Delta(3,4) = \text{UDC}$], resulting in the overall Davydov components.

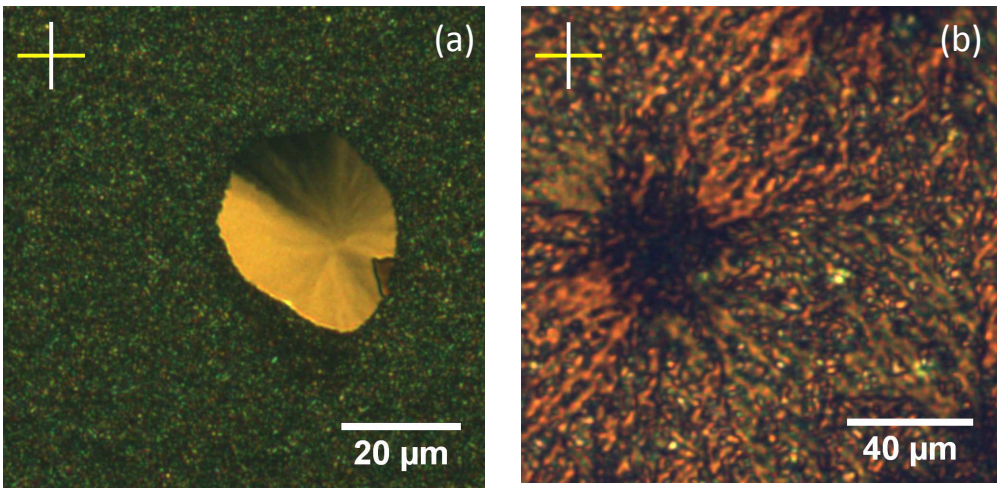

Figure S4: Birefringence of a single SQIB platelet (a) and a sunflower (b). The cross indicates the direction of polarizer (white) and analyzer (orange). Screenshots of the movie files.

In Figure S4, screenshots of two movies are displayed. In (a) a golden platelet is rotated between two crossed polarizers under a polarized light microscope. The images are observed in transmission. The polarizer and analyzer directions are depicted by a white and orange line, respectively, in the upper left. The same is shown in (b) for a sunflower. The movies are available as downloads.

A total of $N = 73$ images are recorded over a rotation angle ϕ of 360°. From the intensity variation $I_n^{x,y}$ $(n = 0 \ldots N-1)$ of each pixel at image position (x, y), the discrete Fourier coefficients $\tilde{I}_\gamma(x, y)$ are determined using

$$\tilde{I}_\gamma(x, y) = \frac{1}{N} \sum_{n=0}^{N-1} I_n^{x,y}\, e^{-i2\pi\gamma\, n/N} \quad . \tag{1}$$

From this, the extinction angle ϕ_{ext}, i.e. the angle for which the intensity is smallest, is extracted via

$$\phi_{\text{ext}}(x, y) = \frac{1}{2} \arg\left(\tilde{I}^*_{\gamma=4}(x, y) + i \left|\tilde{I}_{\gamma=4}(x, y)\right|\right) + \frac{\pi}{8} \quad . \tag{2}$$

Eventually, a value of $\pi/2$ is added to select the tangential extinction angle over the radial one.

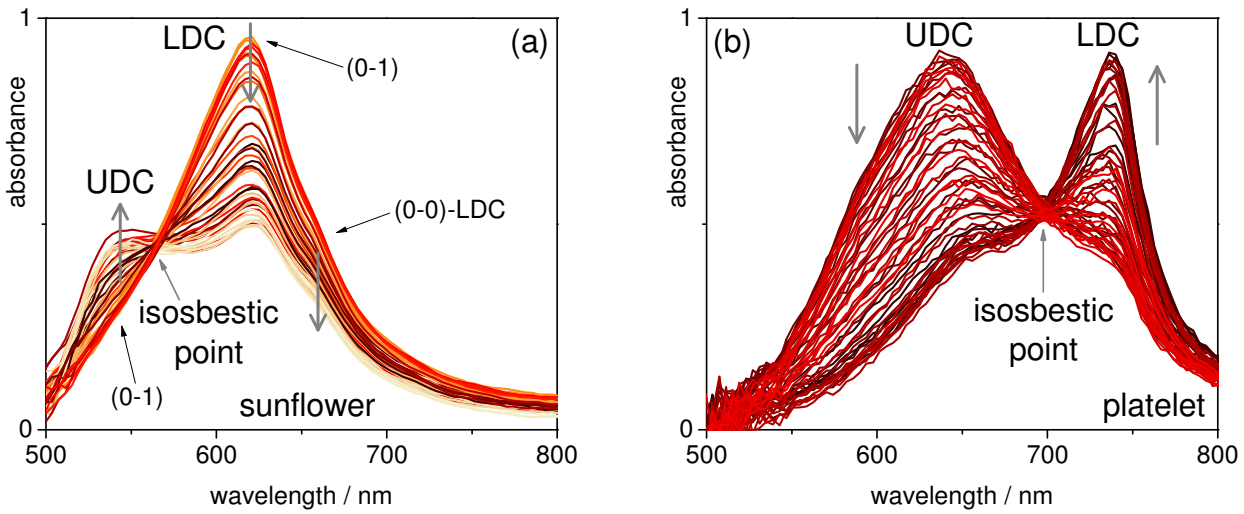

Figure S5: Absorption spectra recorded with a single polarizer dependent on the rotation angle of the polarizer (a) within a typical sunflower, and (b) on a typical platelet area. UDC = Upper Davydov Component; LDC = Lower Davydov Component. Note that for the sunflower in (a) the oscillator strength of the (0-1)-LDC might appear stronger than it actually is due to spectral overlap with the not resolved (0-0)-UDC.