Tuning and Tracking of Coherent Shear Waves in Molecular Films

Henrik Till. Lemke,[1,§*] Dag Werner Breiby,[2] Tine Ejdrup,[1] Peter Hammershøj,[1,¶] Marco Cammarata,[3] Dmitry Khakhulin,[3,£] Nerijus Rusteika,[4] Shin-Ichi Adachi,[5,¥] Shinya Koshihara,[5] Thomas Scheby Kuhlman,[1,‡] Simon Oddsson Mariager,[1] Thomas Nørskov Nielsen,[1] Michael Wulff,[6] Theis Ivan Sølling,[4,*] Niels Harrit,[3] Robert Feidenhans'l,[1] and Martin Meedom Nielsen[7]

[1]*Nano-Science Center, University of Copenhagen, Universitetsparken 5, Copenhagen, Denmark*
[2]*Deparment of Physics, Norwegian University of Science and Technology, Højskoleringen 5, Trondheim, Norway*
[3]*UFR Sciences et propriétés de la matière, Université de Rennes 1, Boulevard du général Leclerc, 35042 Rennes, France*
[4]*Department of Chemistry, University of Copenhagen, Universitetsparken 5, Copenhagen, Denmark.*
[5]*Tokyo Institute of Technology, 2-12-1-H61 Oh-okayama, Meguro-ku, Tokyo 152-8551, Japan.*
[6]*ESRF - The European Synchrotron, CS40220, 38043 Grenoble Cedex 9, France*
[7]*Technical University of Denmark, Risø, Frederiksborgvej 399, 4000 Roskilde, Denmark.*

Supplementary material

[§]*Present address: SwissFEL Bernina, Paul Scherrer Institute, WBBA/022, 5232 Villigen PSI , Switzerland*
[¶]*Present address: Ferrosan Medical, Sydmarken 5, 2860 Søborg, Danmark*
[£]*Present address: European XFEL GmbH, Holzkoppel 4, 22869 Schenefeld, Germany*
[¥]*Present address: Photon Factory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK) 1-1 Oho, Tsukuba, Ibaraki, 305-0801, Japan*
[‡]*Present address: Novozymes, Krogshøjvej 36, 2880 Bagsværd*

Transient Optical Reflectivity measurements

The transient change of optical reflectivity was measured using the third harmonic (266 nm) of an Ti:Sapphire system as pump and the second harmonic (400 nm) as probe. The intensity changes of the light reflected from the α-perylene thin film surface was measured after passing a grating monochromator in order to separate light from the probe pulse. The signal to noise ration was optimized using a lock-in amplifier and a boxcar integrator.

The transient change of optical reflectivity mainly represents the interference signal from light reflected at the film surface and that reflected from a laser-excited strain pulse. Two different strain discontinuities, propagating at different speeds through the
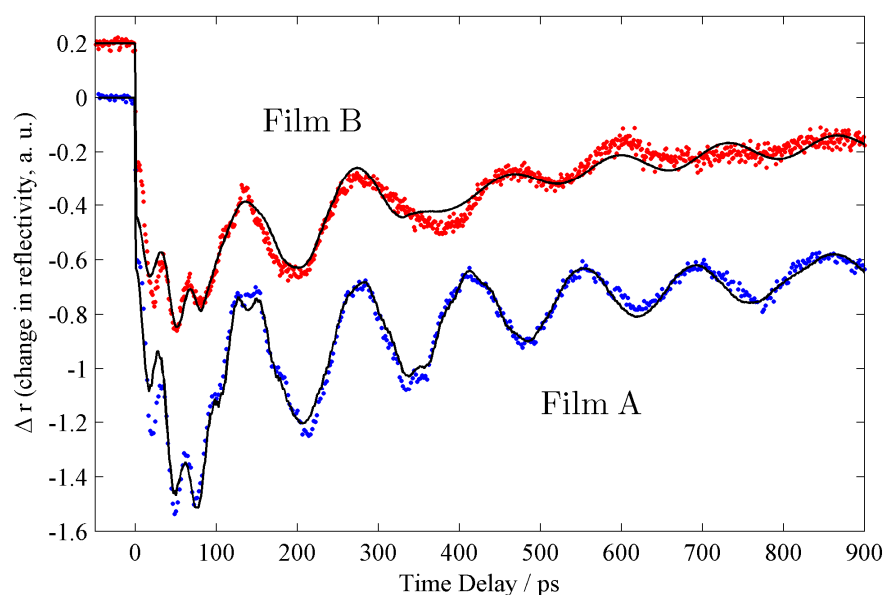

Figure S1: *Dots: Transient optical reflectivity of perylene films A and B, of different thickness, ~800nm and ~300 nm, respectively. The films were excited at 4.66 eV (266 nm) and probed at a wavelength of 3.01 (400 nm). Black lines: Fitted reflectivity curves on basis of the theory presented by Thomsen et al.[1].*

perylene crystal, were indicated by two overlaid frequencies in the transient reflectivity signal (Figure S 1). The oscillation period T is related by $T = \lambda /(2c_s \sqrt{n^2 - \sin^2 \theta})$ to the speed of sound c_s, the refractive index of the film material n, and the incidence angle θ of the probing light with wavelength λ. The speeds of sound found for both frequencies, assuming a refractive index of n=1.5, were 955 m/s and 4460 m/s, which match the values for TA and LA modes in the unit cell's c*-direction observed in a single crystal[2], respectively.
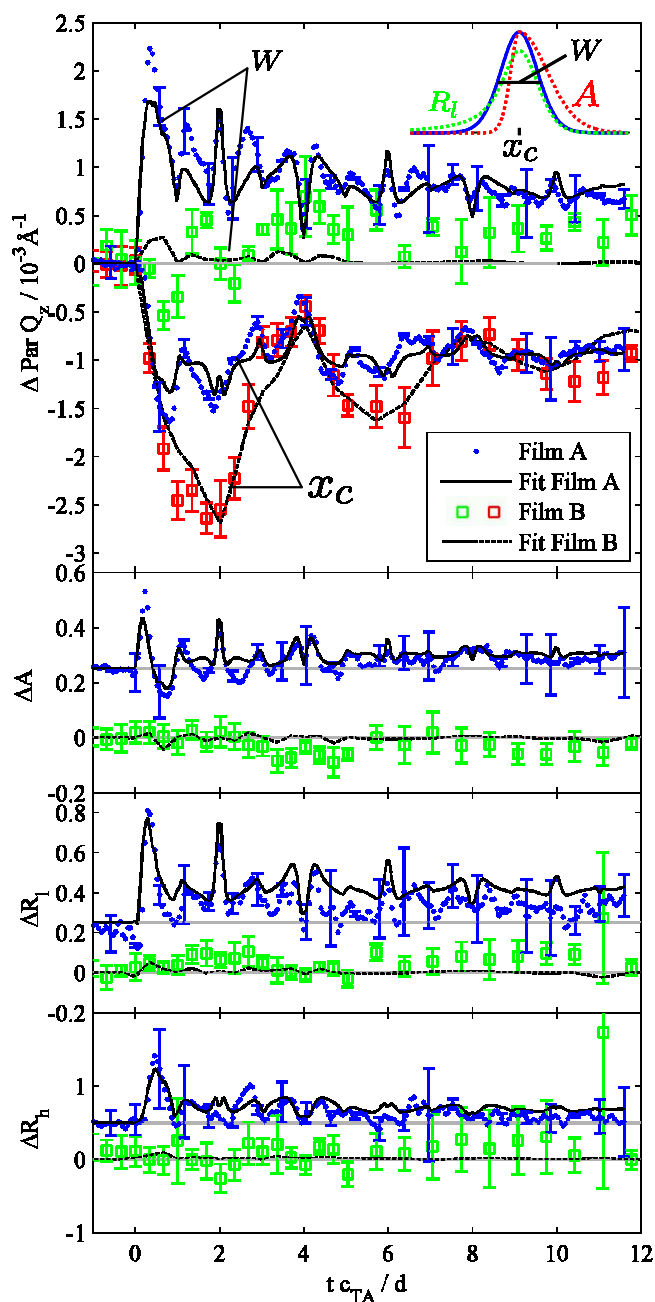
Figure S2: *Dots: Transient optical reflectivity of perylene films A and B, of different thickness, ~800nm and ~300 nm, respectively. The films were excited at 4.66 eV (266 nm) and probed at a wavelength of 3.01 (400 nm). Black lines: Fitted reflectivity curves on basis of the theory presented by Thomsen et al.[1].*

Solution of strain field refinement

In the time-resolved grazing incidence diffraction experiment, the temporal evolution strong $\overline{2}21$-reflection was analyzed to a high level of detail. The measured peak profiles were fitted by the split Pearson VII function[3] (Figure S2, inset). The

time-resolved changes of the all peak shape describing parameters show a complex oscillatory behaviour for perylene Film A (case (i) in article). In case of Film B (case (ii) in article), only the peak position (x_0) shows significant changes after excitation. Global fits of the expected changes of the X-ray signal to all shape parameters according to a simplified mechanical model (see article) show a good agreement with the observations (Figure S2). These fits refined the shear displacement u_x of α-perylene ab-lattice planes as function of depth z in the thin film and time. This two-dimensional result as well the strain ($\partial u_x / \partial z$) for both cases (i) and (ii) are shown in Figure S3. In case (i) a strain pulse propagating with time is clearly visible. In case (ii) the pulse width is much wider than the film thickness. In this case the film consists of a small number of almost equally strained regions, which are divided by a step function propagating at the speed of sound. Here, the X-ray signal was simulated by calculating the lattice sum from a displaced α-perylene crystallite. The signal consists of one main peak with side oscillations defined by the crystal size. In case (i), the highly non-uniform strain pulse leads to complex changes in the side oscillations of that signal which correspond to the changes of peak shape obtained after convolution with the experimental resolution function. In case (ii), where the absorption depth μ is larger than the film thickness, the crystal is almost uniformly strained which leads to a simple movement of the Bragg-reflections in reciprocal space.
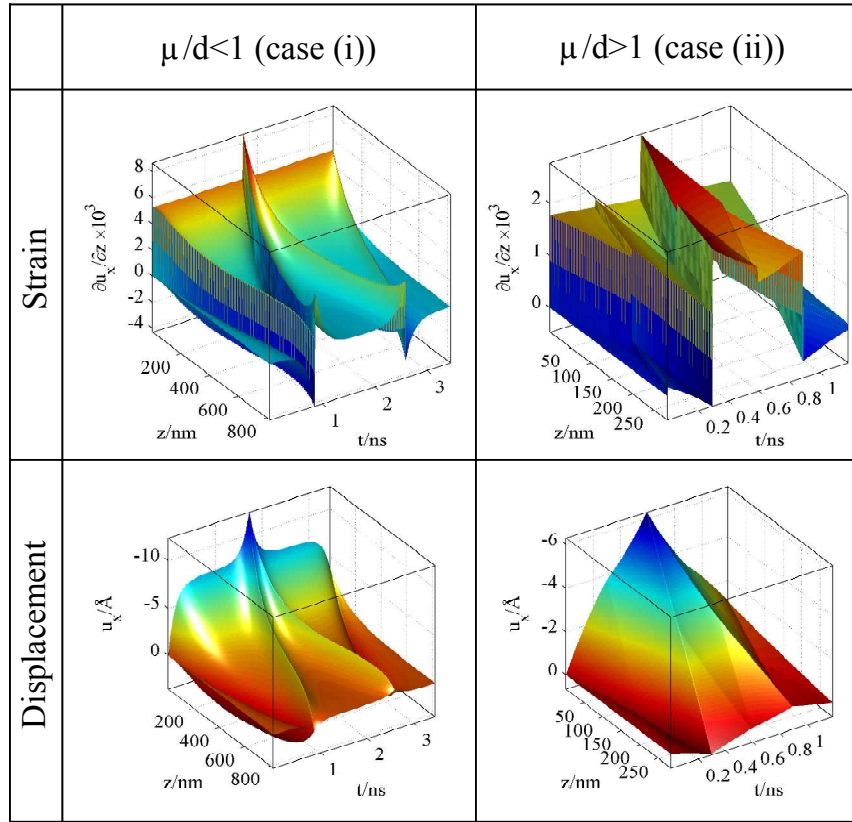
Figure S3: *Refined displacement field $u_x(z,t)$ and strain field $\partial u_x / \partial z$ resulting from the fits shown in Figure S2.*

References

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