4.1 Domain Structure and Nonlinear Optical Properties of Mixed DCANP/CdA Langmuir Films

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The optical properties of mixed and pure Langmuir films of 2-docosylamino-5-nitropyridine (DCANP) LB molecules and arachidic acid (AA) have been investigated with polarisation and second harmonic (SH) microscopy. Specifically the transition from the gaseous to the solid phase was characterised.

When spreading a Langmuir film of DCANP on the water subphase we observed the DCANP molecules spontaneously arrange in spherulites, 2D aggregates of approximately 70 µm in diameter. Within these spherulites, the DCANP molecules tend to align along radial "beams" emerging from the center of each spherulite. In between two "beams" very few molecules are found which underlines the critical shape of those structures. Upon compression, we observe interpenetration of the individual spherulites as soon as the surface area per molecule drops below 30 $Å^2$. Since compression of the spherulite is uniform and symmetric all over the LB trough (neglecting border effects) the spherulites maintain their 2D rounded shape, while simultaneous broadening of the "beams" can be observed in the SHG microscope. Even further compression of the DCANP molecules finally pushes the monomers into the regime of van der Waals repulsion with a steeper gradient of the lateral force with respect to molecular area (phase diagram).

Since the AA molecules are invisible to both the polarisation and the SHG microscope, no direct evidence for the physical arrangement of the AA molecules within the DCANP spherulites can be given. Nevertheless, no change in spherulite structure was observed for AA concentrations of up to 20%. The optical SHG intensity, however, was found to decrease linearly as reported earlier. This suggests the AA molecules act as space filling molecules in between the DCANP "beams". The optical contrast, in addition, can be understood in terms of dilution of the DCANP phase due to the presence of the AA molecules.