LIQUID CRYSTALLOGRAPHY IN MICROCHANNELS

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ABSTRACT
We report that high density monodisperse liquid particles (2μm diameter oil-droplets) self-organize to 3D close-packed face-centered cubic arrangements, when flowing together with water (continuous) phase in a microchannel (10μm×100μm or 10μm×50μm). The resulting remarkably rigid and stable close-packed structure exhibits itself in square and hexagonal patterns, depending on the specific flow conditions. Generally, we observe two (coexisting) 'crystallographic' orientations of the close-packed structure, and distinguish dislocation lines and defects.

KEYWORDS: Liquid Crystallography, Microchannel, Self-Organize, Close-Packed

INTRODUCTION
Fluid systems offered fertile ground for pattern formation studies [1]. The creation and flow of micro droplets (bubbles) in microfluidic systems has attracted considerable attention. Several groups have investigated the formation and periodic arrangements of droplets (bubbles) [2-4]. The complex structures generally result from the system's drive to minimize the local interfacial energy. The self-organisation of the fluid particles is determined by the particle size, the channel geometry, the volume fraction and the relative fluid pressures. All reported fluid particles have a size comparable to the microchannels and hence patterns occured in (2D) monolayers. Here, we present the 3D (multilayer) crystallography of small monodisperse droplets in microchannels.

EXPERIMENTAL
The devices were fabricated in borofloat (glass) wafers using standard photolithography [5]. The channel layout has 500nm deep and 10μm wide channels meeting head-on and flowing out in a 10μm deep and 50μm or 100μm wide microchannel (Fig. 1).

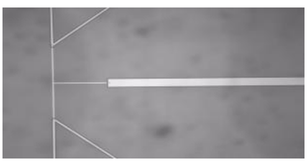

Figure 1. Snapshot of the nano-microfluidic structure.

Two wafers were thermally bonded and diced to 10mm×20mm sized chips. Test chips were fit in a chip holder and connected to gas-tight syringes driven by a syringe pump. The water phase (white) was made fluorescent by dissolving fluorescein sodium salt (0.01M) in DI water and the oil phase (black) was

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hexadecane. The flow was visualized by an inverted microscope and recorded using a CCD camera.

RESULTS

Monodisperse oil droplets (2μm diameter) were generated in the nano-microfluidic channel. The oil-droplets organized into well-defined close-packed superstructures of 4-5 layers thickness, showing a hexagonal or square pattern. Fig. 2 shows microscopic images and their corresponding FFT (Fast Fourier Transform) of hexagonal and square arrangements of the droplets at the channel wall plane. The observed patterns expose different "crystallographic" orientations of the face-centered cubic (fcc) droplet arrays (Fig. 3). The square and hexagonal patterns correspond to the fcc (100) and fcc (111) planes, respectively, of this close-packed structure.

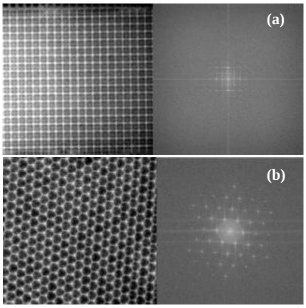

Figure 2. Self-organized patterns (left) and their corresponding FFT (right): (a) square and (b) hexagonal.

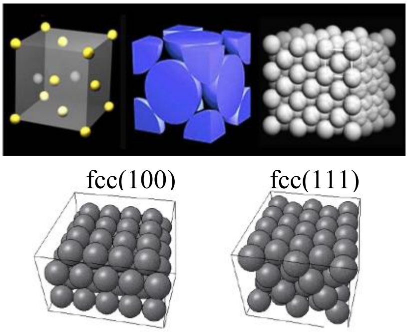

Figure 3. The face-centered cubic (fcc) arrangement. The square pattern corresponds to fcc(100) and hexagonal organization corresponds to fcc(111) planes.

Moreover, we observed coexistence of square and hexagonal patterns, as well as 'grain' boundaries, dislocations and vacancy defects in the patterns (Fig. 4). The latter are exposed as the bright spots in the patterns (both in square and hexagonal arrangements). In general, we observed that in narrower microchannels and at lower flow rates the square arrangement is preferred, i.e. the equilibrium situation is one in which the (100) plane is preferred at the channel wall over the hexagonal (111) plane. We explain this in terms of the hydrophilic nature of the channel wall; the (100) plane has a lower oil-droplet packing density (79%) than the (111) plane (91%). Structures formed proved stable over several days.

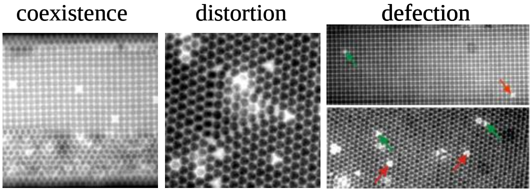

Figure 4. The coexistence of two patterns, distortion, and defects in the self-organized structures. The arrows indicate defects in the top layer or vacancies which are in the layer beneath the top layer.

This study, we hope, can bring us to understand 3D periodic close-packed 2-phase metastable systems and apply them in real life. The structures could for example be useful for liquid chromatography.

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