



Quench depth dependence of liquid crystal nucleus growth: A time resolved statistical analysis

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Abstract

The growth and coarsening of liquid crystalline (LC) germs, nucleating at isothermal conditions from the isotropic melt after a temperature quench, was investigated experimentally and described by application of irreversible thermodynamics. The influence of quench depth, i.e. super-cooling, on the phase ordering process was studied in detail. The analysis of time resolved statistical size distributions of LC germs reveals two basic phase ordering processes involved: (i) nucleus growth and domain coalescence at short times and (ii) domain coarsening via diffusion (Ostwald-ripening) at long times. In the statistical size distributions two individual germ ensembles can be identified. Both of these ensembles exhibit a qualitatively similar behaviour in the course of time, decreasing aggregation energy and increasing mean domain diameter with increasing quench depth. Both ensembles are further shown to obey growth laws that are theoretically predicted for non-conserved order parameter systems.

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1. Introduction

Nucleus growth and coarsening phenomena have attracted much attention in condensed matter

physics and material science for many years, both on a fundamental theoretical level [1] as well as through experimental work [2]. The reason for this interest is rooted in the fact that material properties, may these be related to elastic, electric, magnetic, optical or even clinical behaviour, often depend on the thermal conditions under which the materials were formed and their subsequent tempering treatment, i.e. their thermal history.

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Examples can be found from areas as diverse as the production of metal alloys to the isolation of pure pharmaceutical products. Over decades the main interest remained focused on traditional condensed matter phases, the processes of crystallisation or the phase separation of binary liquids. Little attention has so far been paid to the phase ordering process of intermediate states of matter, especially liquid crystalline (LC) systems. This is somewhat surprising, because the phase formation process of liquid crystals (LCs) can easily be followed in situ by conventional polarising microscopy [3].

LCs [4,5] are partially ordered systems, thermodynamically located between the disordered isotropic liquid and the three-dimensionally ordered solid. They exhibit the flow properties of a liquid combined with a certain degree of order, leading to anisotropic physical properties. The simplest of the large variety of LC phases is the nematic, or in its chiral form also called the cholesteric phase. The nematic/cholesteric phase displays solely orientational order of the long axis of elongated, rod-shaped molecules and absence of any positional correlation of the molecular centres of mass. LCs are spontaneously, self-organised materials. The experimental study and fundamental understanding of their phase ordering process is of importance not only with respect to novel technological applications of for instance unconventional LC phases, but also concerning self-organisation of biological systems, such as cell membranes, which are effectively LC bi-layers.

Studies of phase ordering processes and coarsening behaviour are generally conducted by quench experiments across a phase transition, where a thermodynamic variable of state is rapidly changed to produce a non-equilibrium situation. The average dynamics of the phase ordering process can be described by growth laws of the form $L(t) \sim t^n$, where L is a characteristic length, t the time and n the growth exponent. For a non-conserved order parameter (NCOP) system like a LC at the isotropic to LC phase transition, the growth exponent is expected to vary from $n = \frac{1}{2}$ for zero quench depth ΔT [6,7], i.e. vanishing difference in the Gibbs free energy $\Delta F \rightarrow 0$ between the high and the low-temperature phase, to $n = 1$ for

large super-cooling ($\Delta T \sim \Delta F \rightarrow \text{large}$) [1,8]. This behaviour has indeed been observed experimentally in the few studies reported for different LC phases so far, the blue phase [9], the nematic phase [10], the cholesteric phase [11,12], a twist grain boundary (TGBA*) phase [13], and smectic phases [10,14]. In the latter case growth laws have been shown to also relate to anisotropic nucleus growth [14,15]. LCs are thus expected to follow the same scaling laws as observed for other condensed matter systems.

Despite the successful description of LC nucleus growth by universal growth laws, little attention has been paid towards the time development of the actual *size distribution* of LC nuclei during the growth and coarsening processes. A first time resolved statistical analysis of LC growth employing the model of reversible aggregation [16–18] was carried out for varying confinement conditions [19]. These studies were carried out at constant quench depth. We complement and extend these investigations towards varying quench depths ΔT , i.e. a systematic variation of the difference in the Gibbs free energy ΔF between the high-temperature isotropic and the low-temperature LC phase.

2. Experimental procedure and theoretical model

The LC used in this study was the commercial mixture ZLI-5014-100, formally available from Merck, Darmstadt. We deliberately chose a mixture for this study, because it exhibits a two phase region at the isotropic to cholesteric transition, which allows investigation of both the short time nucleus growth ($\sim 0\text{--}60$ s) as well as the long time coarsening regime ($\sim 1\text{--}60$ min) for the quench depths achievable. Growing germs were of circular shape, simplifying the determination of their respective size distributions. The phase sequence of the material is given by Iso. 70 N* 68 SmA* 64.5 SmC* -11 Cryst. The two phase region at the Iso–N* transition has a width of approximately 1 K and the stated quench depths ΔT were taken as the temperature difference between the actual temperature and that of the occurrence of the first nuclei at extremely

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