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Order parameter dynamics near the Lifshitz point in a ferroelectric liquid crystal

I. Muševič^{a,*}, B. Žekš^a, R. Blinc^a, M.M. Wittebrood^b, Th. Rasing^b

^a *J. Stefan Institute, Jamova 39, 61000 Ljubljana, Slovenia*

^b *Research Institute for Materials and High Field Magnet Laboratory, University of Nijmegen, The Netherlands*

Abstract

The order parameter dynamics has been measured by photon autocorrelation spectroscopy close to the λ -line of a ferroelectric liquid crystal in a transverse magnetic field. It is shown that the critical dynamics can be observed and analysed in the vicinity of the Lifshitz point in this system. Preliminary results indicate the existence of a nearly gapless phason mode for small magnetic fields, whereas above 10 T phason excitations exhibit a finite frequency gap.

The Lifshitz point, as first introduced by Hornreich et al. [1], is a triple point where a modulated ($q \neq 0$), a homogeneously ordered ($q=0$) and a disordered phase coexist. The concept of a Lifshitz point was first introduced into the field of liquid crystals by Michelson [2], who pointed out that such a triple point could exist in the (H , T) phase diagram of a ferroelectric liquid crystal when a magnetic field H is applied in a direction perpendicular to the helical axis. Experiments in magnetic [3] and high frequency electric [4] field have revealed some very interesting features of (H , T) and (E , T) phase diagrams of ferroelectric liquid crystals like reentrant phenomena [3, 4] as well as the observation of a Lifshitz point (H_L , T_L) in a mixture of pure and chiral DOBAMBC [5]. Whereas all known studies of (H , T) and (E , T) phase diagrams were concentrated on the measurements of static properties, like the dielectric constant or the period of the helical modulation, very little is known about the collective dynamics of these systems in external magnetic or high frequency electric fields. An exception is the light-scattering experiment of the collective dynamics of the SmC* phase in high magnetic fields [6] that was

performed far below the λ -line. Here a band structure of the order parameter excitation spectrum was observed for the first time and this was explained on the basis of symmetry breaking by a magnetic field. One of the important results of that study was the manifestation of a fascinating interplay between the symmetry breaking in a system and the existence of zero-frequency, symmetry restoring phason modes. It was predicted [7] that such a zero-frequency, symmetry restoring mode can exist only for fields smaller than the Lifshitz field, whereas beyond the Lifshitz field a finite gap should be present in the phason excitation spectrum.

To recall the arguments for that conclusion briefly, let us consider symmetry changes in the (H , T) phase diagram of a ferroelectric liquid crystal in a transverse magnetic field, as shown schematically in Fig. 1. According to the Goldstone theorem, a zero-frequency, symmetry restoring mode appears whenever a continuous symmetry is spontaneously broken in a system. In our case, this may occur along the $T_\lambda(H)$ line, where the symmetry of the SmA phase is spontaneously broken upon reducing the temperature. For $H=0$ the point symmetry of the SmA phase is D_∞ and the translation symmetry is continuous on the length scales of interest. By crossing the λ -line the point group reduces to C_2 whereas the translation

* Corresponding author.

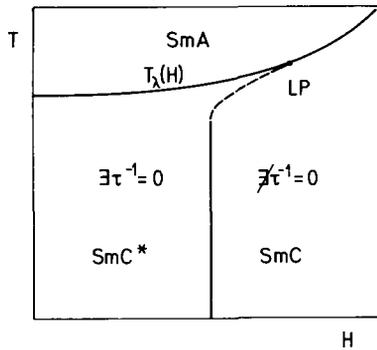


Fig. 1. Schematic view of the (H, T) phase diagram of a ferroelectric liquid crystal in an external magnetic field applied perpendicularly to the helix. Here LP denotes the Lifshitz point (H_L, T_L) . Gapless phason is expected to exist only in the region of the modulated SmC* phase.

symmetry is still continuous helical. We therefore expect the existence of a gapless Goldstone mode as a result of the spontaneous breaking of the continuous rotational symmetry of the SmA phase. For $0 < H < H_L$ the point group of the SmA phase in a transverse field is reduced to D_2 , whereas the translation symmetry is continuous. This continuous translation symmetry is spontaneously broken into the discrete translation symmetry of a magnetically deformed SmC* phase, which in this case is the reason for the existence of a gapless mode in the low-temperature phase. On the contrary, the phase transition from the SmA to the unwound SmC phase involves no continuous symmetry breaking because both phases are continuously translationally invariant, whereas both point groups are discrete (D_2 and C_2 , respectively). This means that a zero-frequency mode should not exist in the unwound SmC phase.

It has been shown experimentally [8] that the Goldstone mode of the SmA to SmC* transition is the gapless phason mode, that can be quite easily observed in a quasielastic light-scattering experiment. On the other hand, the dynamics of the order parameter near the SmA to the SmC* transition seems to be well understood from numerous optical and dielectric studies [9], that show critical slowing down of the soft mode in the SmA phase as well as the splitting of the soft mode into an amplitudon and a phason mode below T_c . This preliminary quasielastic light-scattering study of the order parameter dynamics near the λ -line has been performed in order to clarify whether it is experimentally feasible to observe and analyse thermally excited modes of ferroelectric liquid crystals in high magnetic fields.

We have used a mixture of purely chiral (35 wt%) and racemic (65 wt%) ferroelectric liquid crystal 4-(2'-methyl-butyl)phenyl 4'-n-octylbiphenyl-4-carboxylate (CE-8).

For this material we know from our previous studies that the critical magnetic field for helix unwinding is about 8 T at the temperature of 3 K below T_c . High optical quality, 120 μm thick samples with perfect silane-induced homeotropic alignment were placed in a thermostated, single-stage oven in the center of the magnet. The temperature stability of the oven was better than 5 mK and was achieved by carefully tuning the control parameters of the lock-in based temperature controller. The set-up for the quasielastic light-scattering experiments in high magnetic fields is described elsewhere [6,7]. The scattering geometry was set to ordinary–extraordinary scattering and we have chosen the scattering wave vector close to the wave vector of the modulation of the SmC* phase, thus probing the dynamics in the neighborhood of the Bragg peak.

In the SmA phase we could immediately observe rather strong signal of the soft mode. The relaxation rate showed a critical slowing down when we were approaching the λ -line. We could clearly resolve this signal at any field up to the maximum available magnetic field of almost 15 T. Far from the λ -line in the SmA phase ($T > T_c + 100$ mK) the decay of the autocorrelation function was single-exponential, whereas very close (20–30 mK) to the phase transition a small additional signal with a longer relaxation rate could be detected. The relaxation time of this smaller signal was approximately a factor of 5 longer than the relaxation time of the dominant signal. It is not clear at this moment what is the origin of this signal. Such a behaviour can in principle originate from (i) an additional relaxation mode in the SmA phase, (ii) poorly defined heterodyne regime of the quasielastic detection or (iii) multiple light scattering. Unfortunately we were not able to keep the experiment in the regime of heterodyne detection, which would eliminate assumption (ii). Good heterodyne detection is usually achieved by coherently mixing quasielastically and elastically scattered light from sample imperfections. In our case the intensity of stray light, elastically scattered from sample imperfections, was extremely low because of the high optical quality of the sample. This results in a low strength of the local oscillator field and poorly defined detection regime in between homodyne and heterodyne optical detection, with a normalized peak autocorrelation function $g^{(2)}(0) = 1.2$ –1.1. It is well-known that in this case one can obtain a polydisperse decay of the autocorrelation function, where the relaxation rates of decay components differ by a factor of two. The data were then analysed with a double-exponential fit and only the dominating signals are shown. Figs. 2(a) and (b) present the results of these measurements performed at $H = 0$ and $H = 10$ T. One can clearly observe the critical slowing down of the soft mode in the SmA phase as well as the splitting of the soft mode into the amplitudon and

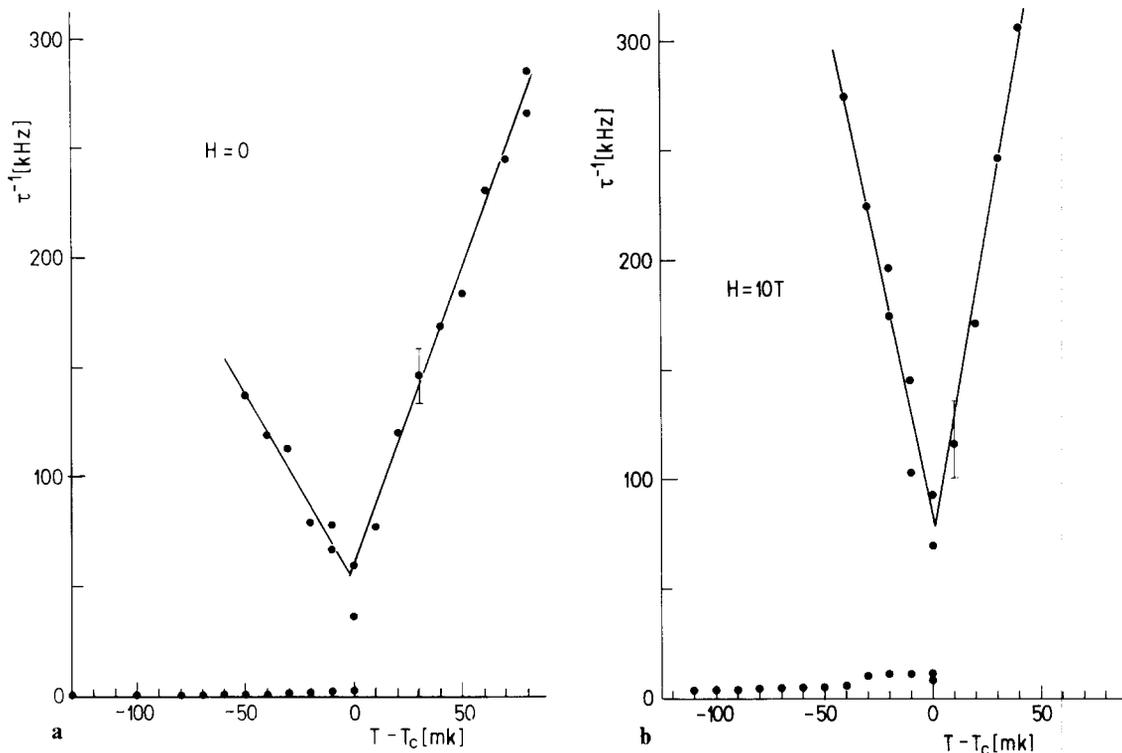


Fig. 2. Order parameter relaxation rates near the λ -line in a mixture of chiral and racemic liquid crystal CE-8 at (a) $H = 0$ and (b) $H = 10$ T. For details, see the text.

the phason mode below T_c . There are two distinct issues that can be observed in these two measurements. The first one is an apparent increase of the slopes of the soft and amplitudon modes at 10 T as compared to the zero-field data. At present we are not able to confirm whether this is the result of a real influence of the field on the soft mode dynamics or is just an artifact that originates from poorly defined detection regime, changing with a field. The second observation is an increase of the phason relaxation rates almost with an order of magnitude. At zero-field, the phason relaxation rates decrease from few kHz at T_c down to less than 100 Hz at $T = T_c - 50$ mK. This temperature change can be explained by the fact that the helical period changes with temperature, so that the minimum of the phason dispersion changes, too. On the contrary, at 10 T the phason relaxation rates are of the order of 5–10 kHz, which is an order of magnitude higher than the zero-field relaxation rates. It is also very important to note that there is no low-frequency signal (below kHz) at 10 T, indicating that we are beyond the Lifshitz field for this material, where the gapless phason

cannot exist. This indicates that in the unwound SmC phase the gapless acoustic phason branch has disappeared and only the optic-like phason branch remains. However, in order to prove that we are really above the Lifshitz field, one has to measure a complete phason dispersion relation.

In conclusion, our preliminary quasielastic light-scattering data indicate that it is experimentally possible to study the order parameter dynamics near the Lifshitz point in the (H, T) phase diagram of a ferroelectric liquid crystal. This is, to our knowledge, the only known physical system where one can study the order parameter dynamics and dispersion in the vicinity of the Lifshitz point. The reason for this unique property is the fact that the length scales involved in the (H, T) phase diagram are of mesoscopic character (modulation period in the μm range) and the corresponding order parameter relaxation rates are in the kHz region. The results of our studies of this problem with a reconstructed setup (double temperature control and controllable optical detection regime) will be published in the near future.

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