Phason Dispersion and Magnetic-Field-Induced Band Gap in a Ferroelectric Liquid Crystal

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The phason dispersion and the dielectric response of a ferroelectric Sm-C* liquid crystal in an external magnetic field have been measured. A magnetic-field-induced splitting of the phason branch into an acousticlike and an opticlike mode was observed, indicating a crossover between the plane-wave-like and solitonlike phason dynamics. The resulting band gap at \( q = 0 \) is a direct consequence of the broken continuous helicoidal symmetry of the Sm-C* phase in a transverse magnetic field.

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In the Sm-C* phase of a ferroelectric liquid crystal, rodlike molecules with a transverse dipole moment are arranged in liquidlike smectic layers. The director \( \mathbf{n}(r) \) that describes the time-averaged orientation of the long molecular axis is tilted with respect to the layer normal at an angle \( \theta \) and precesses with a period \( p_0 \) on moving from one layer to another. Usually \( p_0 \) is much larger than the interlayer distance so that the system can be considered as a helicoidal continuum. The symmetry of each smectic layer is \( C_2 \).

In the long-wavelength limit the spectrum of elementary excitations consists of two branches [1,2]. The amplitude branch represents the variation in the magnitude of the tilt angle and is observable only close to the Sm-C* \( \rightarrow \) Sm-A transition. The phason branch represents a collective plane-wave-like excitation of the phase \( \Phi(r,t) \) of the director field. Because of the continuous helicoidal symmetry, the phason excitation displays a plane-wave behavior with a parabolic dispersion relation.

A static magnetic field \( \mathbf{H} = (0, H, 0) \) perpendicular to the helical axis \( \mathbf{e}_z \) tends to align the molecules in the direction of the field if the anisotropy of the magnetic susceptibility \( \Delta \chi = \chi_\parallel - \chi_\perp \) is positive. The helical structure is distorted in a solitonlike manner and this distortion is accompanied by an increase of the period of the magnetic field has here a similar role as the “lock-in” lattice term in incommensurate solids. The difference is that here the magnetic field can be continuously varied and thus the “soliton” regime and the magnitude of the periodic potential can be externally controlled at any given temperature which is not the case in incommensurate systems.

In this Letter, we report what we believe to be the first experimental observation of the splitting of the phason branch into an acousticlike and an opticlike mode. The experiment was performed in the Sm-C* phase of a ferroelectric liquid crystal and the splitting was induced by an external transverse magnetic field. The phason dispersion curves were determined by quasielastic light scattering spectroscopy, whereas the magnetic-field-induced band gap was measured by both light scattering and dielectric relaxation spectroscopy. The splitting between the two branches was observed even for very small fields, where the change of the period of the helix is too small to be detected.

The dynamics of the phase fluctuations [1,2,4–6], propagating along the helical axis of a distorted Sm-C* phase, can be derived from the Landau thermodynamic potential [7,8]. Expressing the Sm-C* order parameter \( \xi \) as

\[
\xi_1(z,t) = \theta \cos \Phi(z,t), \quad \xi_2(z,t) = \theta \sin \Phi(z,t),
\]

one obtains in the constant amplitude approximation [3] the phase-dependent part of the nonequilibrium free-energy density as

\[
g(\Phi) = \Lambda \theta^2 \frac{\partial \Phi}{\partial z} + \frac{1}{2} K_{33} \theta^2 \left( \frac{\partial \Phi}{\partial z} \right)^2 - \frac{1}{2} \Delta \chi H^2 \theta^2 \sin^2 \Phi.
\]

Here \( \Lambda \) is the Lifshitz term, \( K_{33} \) is the torsional elastic constant, and \( \Delta \chi \) is the diamagnetic anisotropy. After minimizing \( g(\Phi) \) with respect to \( \Phi(z) \), one obtains the static solution \( \Phi_0(z) \) of the sine-Gordon equation in the...
well-known form of a $\pi$-soliton:

$$\sin \Phi_0(z) = \sin(u,k).$$

(3)

Here $u = z/\xi$ is the argument of a Jacobian elliptic function of modulus $k$, and $\xi$ is the magnetic coherence length, $\xi^2 = K_{33}/\Delta H^2$. The modulus $k$ is determined by the external field and satisfies the equations $k = (H/H_c)E(k)$ and $H_c = (\pi^2/\rho_0)(K_{33}/\Delta H)^{1/2}$. Here $E(k)$ is the complete elliptic integral of the second kind.

By taking $\Phi(z,t) = \Phi_0(z) + \delta \Phi(z,t)$, where $\delta \Phi(z,t) = \psi(z)e^{-i(r/H)}$, one obtains after applying the Landau-Khalatnikov equations of motion, linearized in $\psi(z)$, Lamé's equation of order one for the eigenvectors of the phase fluctuations amplitude [4-6] $\psi(u)$:

$$\frac{d^2 \psi}{du^2} + [h - k^2 \sin^2(u,k)] \psi = 0.$$  

(4)

Here $h = k^2 [\xi^2 \gamma/(\Gamma H K_{33})] + 1$ and $\gamma$ is the viscosity of the Sm-C$^*$ phase. General solutions of Lamé's equation (4) in the form of the Bloch waves are known [6,9,10], and the corresponding wave vector $k$ can be ascribed on the basis of symmetry transformation properties of these eigenfunctions. It should be stressed that in the experiment one observes the excitations of the order parameter $\xi$, whereas Eq. (4) describes the excitations of the phase $\delta \Phi_0(z,t)$. It can be shown from Eqs. (1) and (3) that the phase excitation $\delta \Phi_0$ with a wave vector $k$ and the relaxation rate $\tau^{-1}$ is observable as an excitation of the order parameter $\xi$ at the wave vectors $q = k \pm q_c$, where $q_c(H) = 2\pi/p(H)$. This has the consequence that the center of the phason BZ (i.e., $\kappa = 0$, see below), introduced by the periodic potential $k^2 \sin^2(u,k)$ in Eq. (4), is observable near the first Bragg peak at $q = \pm q_c$ in the experiment.

The eigenfunctions of particular interest are those with the wave vectors $q = 0$ and $q = \pm q_c(H)$, corresponding to the edges and to the center of the BZ. These eigenfunctions $\psi^{(1)}$ and their corresponding eigenvalues $\tau^{-1}$ are

$$\psi^{(1)} = \text{dn}(u,k), \quad \tau^{-1}(q = \pm q_c(H)) = 0,$$

(5)

$$\psi^{(2)} = \text{cn}(u,k), \quad \tau^{-1}(q = 0) = \frac{1 - k^2 \Delta \chi}{k^2} H^2,$$

(6)

$$\psi^{(3)} = \text{sn}(u,k), \quad \tau^{-1}(q = 0) = \frac{1 - k^2 \Delta \chi}{k^2} H^2.$$  

(7)

Here $\text{dn}(u,k)$, $\text{cn}(u,k)$, and $\text{sn}(u,k)$ are the Jacobian delta, cosine, and sine amplitudes, respectively. At $H \rightarrow 0$, $k \rightarrow 0$, so that $\tau^{-1}(q = 0) = \tau^{-1}(q = 0) = (K_{33}/\gamma)q_c^2$.

What is the physical interpretation of these solutions? $\psi^{(1)}$ is a Goldstone mode, recovering the broken continuous ($D_\omega$) symmetry of the Sm-A phase. For $H = 0$ this mode amounts to a uniform rotation of the whole helix, or, what is the same, to a sliding of the helix. For $H \neq 0$ this mode represents a uniform translation of the soliton lattice as a whole, resulting in a gapless mode at $q = \pm q_c$. The solution $\psi^{(2)} = \text{cn}(u,k)$ represents an acousticlike phase excitation that has the nodes in the middle of the commensurate regions between the soliton-like domain walls. Thus, $\psi^{(2)}$ shifts the centers of these “domain walls.” The relaxation rate of this mode decreases with increasing magnetic field. The solution $\psi^{(3)} = \text{sn}(u,k)$ represents an opticlike phase excitation that changes the width of the domain walls. The corresponding relaxation rate increases with increasing magnetic field.

As a result of quadratic diamagnetic coupling, the phason excitation sees in the presence of $H \neq 0$ a periodic potential $k^2 \sin^2(u,k)$ with periodicity $p/2$. This symmetry breaking introduces a Brillouin zone defined by $-2\pi/p \leq k \leq 2\pi/p$ and implies a Bloch form of the phason eigenfunctions. The coupling between the phason modes at $k = \pm q_c$ causes a gap $G(H)$ to appear at the edges of the BZ, separating the acousticlike mode $\psi^{(2)}$ from the opticlike mode $\psi^{(3)}$. In the experiment, the phason band gap $G(H)$ is observable at $q = 0$ as shown in Fig. 1:

$$G(H,q = 0) = r^{+1}(q = 0) - r^{-1}(q = 0) = (\Delta \chi/\gamma)H^2.$$  

(8)

As shown by Sutherland [10], in such a system there is only one gap.

In the Sm-C$^*$ phase, the fluctuations of the phase $\Phi(z,t)$ of the order parameter induce the fluctuations of the spontaneous dipole moment $\rho(r,t)$ and the local dielectric tensor $\epsilon(r,t)$. Whereas quasielastic light scattering spectroscopy allows for the measurement of the phason dispersion [1,2], dielectric spectroscopy will give information on the phason dynamics at $q = 0$. It can be shown from Eqs. (6) and (7) that the acousticlike mode $\psi^{(2)}$ represents a fluctuation of the space-averaged spontaneous electric polarization $\delta \rho = (\delta \rho,0,0)$, which is perpendicular to the magnetic field $H$. In a similar way, the opticlike phason $\psi^{(3)}$ represents a fluctuation of space-averaged polarization $\delta \rho = (0,\delta \rho,0)$ in the direction of the magnetic field.

The experiment was performed in a mixture consisting

FIG. 1. The effect of an external magnetic field on the phason dispersion in the Sm-C$^*$ phase of a ferroelectric liquid crystal.

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of 35\% of pure chiral liquid crystal 4-(2'-methylbutyl)-
phenyl 4'-octylibiphenyl-4-carboxylate (CE-8) and 65\% of
racemic liquid crystal CE-8R. The phason dispersion
curves were measured with a miniature quasielastic light
scattering spectrometer, fitting into a 60-mm Bitter mag-
net. A He-Ne ordinary polarized laser beam was slightly
focused to a 50-\mu m spot, illuminating a 120-\mu m-thick,
homeotropically aligned liquid-crystal sample, placed in a
thermostated oven (\pm 0.1 K) in the center of the magnet.
The scattered extraordinary light was collected into a
multimode fiber and fed into a photomultiplier, placed far
down away from the magnet. The axis of the collecting optics
was positioned at an angle 1^\circ from the incoming light
direction, thus assuring a heterodyne detection of the au-
tocorrelation function of the scattered light intensity. In
addition, the field dependence of the period of the helix as
well as the dielectric response was determined. The
dielectric response was measured with \( \mathbf{E} \mathbf{H} \perp \mathbf{q} \), thus
probing the optileclike phason mode at \( q = 0 \). For the ex-
perimental details, see Ref. [1].

The phason dispersions were measured 3 K below \( T_c \) at
several magnetic fields. The magnetic field was swept
slowly to the desired value, and then the system was left
for 30 min to reach the equilibrium state. Phason disper-
sions as measured at 0, 6.65, and 8.1 T are shown in Fig.
2. One can clearly observe the change in the shape of the
phason dispersion curves, being most pronounced at \( q = 0 \).
The best parabolic fit to the zero-field dispersion gives the
value \( K_{33}/\gamma = 1.1 \times 10^3 \ \text{\mu m}^2 \text{\text{~s}}^{-1} \) and \( p_0 = 5.7 \ \mu m \). A par-
abolic fit of the phason dispersions at 6.65 and 8.1 T
shows a systematic decrease of the constant \( K_{33}/\gamma \), to-
gether with a shift of the center of the dispersion, corre-
sponding to a change of the helical pitch. The minimum
of the phason dispersion at 8.1 T is shifted to \( q \neq 0 \), clearly
indicating the appearance of the homogeneous Sm-C*
phase. On the basis of these observations, we conclude
that in the quasielastic light scattering experiment below
\( H_c \) we observe the dominant acousticlelike phason branch.
The intensity of the higher-frequency optileclike phason
branch is covered by the high intensity signal of the
acoustic branch and is observable only as a weak signal
above \( H_c \), where the acoustic branch disappears.

Figure 3(a) shows the magnetic-field dependence of the
normalized phason relaxation rates at \( q = 0 \), as obtained
from quasielastic light scattering and dielectric response.
One can clearly observe a large decrease of the relaxation
rates of the acousticlelike phason mode at fields far below
the critical field. At the same time, we can observe a
strong increase of the relaxation rates of the optileclike
phason mode, as determined from the dielectric response.
Figure 3(b) shows the magnetic-field dependence of the
period of the helix, measured at the same temperature.
By comparing the magnetic-field dependences of the two
phason modes to the magnetic-field dependence of the
period of the helix, one sees a striking difference. Whereas
one can hardly see any change in the period up
to 6 T, the magnitude of the gap separating the optic and
the acoustic phason branches is of the order of the zero-
field phason relaxation rate, i.e., 1.5 kHz. This is a clear
indication of a crossover behavior of the phason dynamics
between the plane-wave dynamics in the unperturbed hel-
coiloidal structure and the solitonlike dynamics in a distort-
ed helicoideal Sm-C* structure.

In Fig. 4 we present the magnetic-field dependence of the
gap \( G(H,q = 0) \) as determined from optic and dielec-

![Figure 2](image1.png)

**FIG. 2.** Dispersion of the acousticlike phason branch as
determined at different magnetic fields. Solid lines represent
the best parabolic fits \( r^{-1} = (K_3/\gamma)(q - q_c)^2 \). At \( H = 0 \) we get
\( K_{33}/\gamma = 1.1 \times 10^3 \ \text{\mu m}^2 \text{\text{~s}}^{-1} \) and \( p_0 = 5.7 \ \mu m \), whereas at \( H = 6.65 \)
T we get \( K_{33}/\gamma = 0.76 \times 10^3 \ \text{\mu m}^2 \text{\text{~s}}^{-1} \) and \( p = 6.2 \ \mu m \). Inset:
The phason dispersion at 8.1 T with \( K_f/\gamma = 0.6 \times 10^3 \ \text{\mu m}^2 \text{\text{~s}}^{-1} \)
and \( q = 0 \).

![Figure 3](image2.png)

**FIG. 3.** Magnetic-field dependence of the (a) normalized re-
lexation rates of the optic and acoustic phason modes at \( q = 0 \)
and of the (b) period of the helix. Solid lines in (a) represent
the best fits by Eqs. (6) and (7) with \( \Delta x/\gamma = 48 \ \text{T}^{-2} \text{~s}^{-1} \) and
\( H_c = 8.1 \) T for the optic phason mode \( r^{-1}(q = 0) \) and \( \Delta x/\gamma = 42 \ \text{T}^{-2} \text{~s}^{-1} \) and
\( H_c = 9.7 \) T for the acoustic phason mode \( r^{-1}(q = 0) \). The solid line in (b) represents the constant ampli-
tude approximation fit [3,5] with \( p_0 = 6.2 \ \mu m \) and \( H_c = 8.6 \) T.
FIG. 4. Magnetic-field dependence of the gap $G(H)$, separating the optic and acoustic phason modes at $q=0$, $T_c - T = 3$ K. The difference between the normalized optic and acoustic phason relaxation rates was multiplied by the mean value of 1.5 kHz for both relaxation rates at zero field (see text) to obtain $G(H)$.

In conclusion, we have observed for the first time the magnetically induced splitting of the phason branch in the Sm-C* phase of a ferroelectric liquid crystal into an acoustic-like and an optic-like mode. The appearance of a single gap in the phason excitation spectrum is here connected with the crossover from the plane wave to the soliton lattice modulation regime, and is the result of the breaking of the original continuous symmetry by the external field. We believe that the above observations and conclusions may lead to a better understanding of the dynamical properties of modulated structures in external fields and incommensurate systems in general.

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