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Light Scattering and Gapless Excitations in Antiferroelectric Liquid Crystals

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The phason dispersion in the antiferroelectric SmC^*_A liquid crystalline phase of 4-(1-methylheptyl-oxycarbonyl) phenyl 4′-octyloxybiphenyl-4-carboxylate has been studied by time-resolved quasielastic light scattering spectroscopy. The phason branch is found to be gapless as predicted for a Goldstone mode recovering the broken continuous symmetry. The dispersion has a minimum at $q = 2q_c$, where q_c is the wave-vector of the helicoidal SmC^*_A phase. The results are consistent with the alternating-tilt model of antiferroelectric liquid crystals and the first order approximation to the optical properties of birefringent and modulated superstructures.

Keywords: Antiferroelectric; liquid crystal; fluctuations

PACS numbers: 61.30.Gd, 64.70.Md, 64.70.Rh

The liquid crystalline state is characterized by long range orientational order of the average direction of the long molecular axis. Similar to the case of a Heisenberg ferromagnet it is a state of a spontaneously broken continuous orientational symmetry of the high temperature isotropic liquid crystalline phase. As a consequence, in many liquid crystalline phases the spectrum of collective excitations of the director field is expected to be gapless in the long-wavelength limit. This should allow for the existence of symmetry recovering, zero frequency Goldstone modes, which are known to occur in particle physics. A particularly interesting Goldstone mode should exist in the chiral antiferroelectric SmC^*_A phase, which is a state of spontaneously broken D_∞ symmetry of the chiral SmA phase.

The antiferroelectric liquid crystalline SmC^*_A phase [1, 2] is characterized by an alternation of the tilt direction of the average molecular orientation

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and the direction of the in-plane spontaneous polarization $\vec{P}$ by nearly 180° on going from one smectic layer to another (Fig. 1a). Two neighboring layers thus form an antiferroelectric unit cell with two antiparallel electric dipoles and a zero value of the equilibrium electric polarization $\vec{P}_o(\vec{r}) = \vec{P}_i + \vec{P}_{i+1} = 0$. Because of chirality, the directions of the spontaneous tilt and the in-plane polarization slowly precess around the layer normal as one moves along the direction perpendicular to the smectic plane. This causes a small deviation from the 180° alternation in the tilt between two consecutive layers and the formation of a modulated, helicoidal structure. Since the basic structural unit of the SmC^*_A phase are two neighboring layers, we have here in fact a double-twisted helicoidal structure, formed by two identical ferroelectric SmC^* helices gearing into each other as shown in Figure 1b. The periodicity of this helical modulation is of the order of the wavelength of the visible light and is in general incommensurate to the basic antiferroelectric unit cell of the SmC^*_A phase.

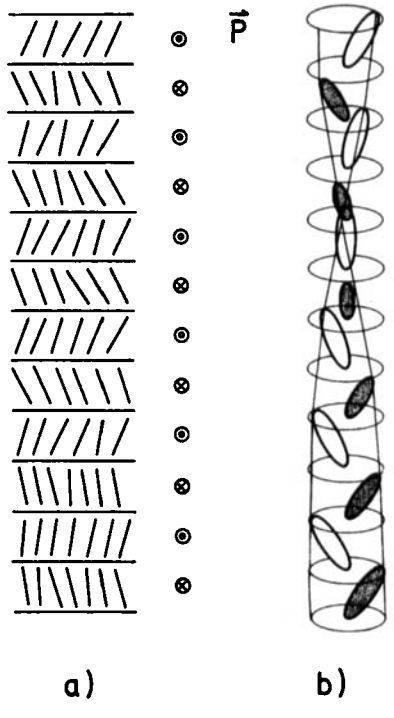

FIGURE 1 (a) The alternating tilt model of the homogeneous antiferroelectric SmC^*_A phase of liquid crystals and (b) the antiferroelectric double helix.

The theory of incommensurate systems [3] predicts the existence of a gapless phason mode in the incommensurate phase recovering the broken translational periodicity of the high temperature phase. In the antiferroelectric SmC_A^* phase, we have a spontaneously broken continuous rotational symmetry and the phason mode represents the sliding, or what is equivalent, the slow rotation of the double twisted helicoidal modulation wave, which tries to restore the symmetry, lost at the $SmA \rightarrow SmC_A^*$ transition. Since the chiral SmA phase has the continuous D_∞ symmetry, whereas the symmetry of the SmC_A^* phase is discrete, the symmetry recovering phason is here predicted to be a truly gapless Goldstone mode.

Whereas the presence of acoustic-like phason modes in structurally incommensurate systems has been by now clearly demonstrated [4], a truly gapless phason branch in a liquid crystalline phase with a discrete lattice symmetry has been observed only recently [5]. A time-resolved light scattering experiment was performed in the antiferroelectric SmC_A^* phase, revealing for the first time a gapless nature of phase excitations in this phase. Although the dynamics of the antiferroelectric phase is expected to be richer than the dynamics of ferroelectric liquid crystals, where the basic structure is quasicontinuous rather than discrete, the experiment clearly revealed a fundamental similarity of the order parameter dispersion relations in the ferroelectric [6] and antiferroelectric liquid crystalline phases. Recently, a phason dispersion measurement has also been performed in the SmC_γ^* phase [7], where however the results seemed to show a finite value of the phason relaxation frequency at zero scattering wave-vector, $\vec{q} = 0$.

The thermodynamic properties and phase transitions between the antiferroelectric SmC_A^* phase and the related ferri-, ferro-and paraelectric phases have been theoretically analyzed by Orihara and Ishibashi [8] and later by Žekš *et al.* [9] within the framework of a Landau theory. Similar to solid antiferroelectrics [10] the order parameters of the phase transitions between these phases have been conveniently chosen as linear combinations of the tilt vectors $\vec{\xi}_i$ and $\vec{\xi}_{i+1}$ in the two neighboring smectic layers i and $i+1$

$$\vec{\xi}_f = \frac{1}{2}(\vec{\xi}_i + \vec{\xi}\xi_{i+1}) \tag{1a}$$

$$\vec{\xi}_a = \frac{1}{2}(\vec{\xi}_i - \vec{\xi}\xi_{i+1}) \tag{1b}$$

thus describing the ferroelectric ($\vec{\xi}_f$) and antiferroelectric ($\vec{\xi}_a$) ordering, respectively. By writing down scalar invariants of the two order parameters

[8] with respect to the D_∞ symmetry of the paraelectric *SmA* phase, one obtains by minimization regions of stability of the para-, ferri-, ferro- and antiferroelectric phases. These phases are characterized by different equilibrium values of the order parameters $\vec{\xi}_f^0$ and $\vec{\xi}_a^0$. For example, in the paraelectric *SmA* phase $\vec{\xi}_f^0 = \vec{\xi}_a^0 = 0$, in the ferrielectric phase $\vec{\xi}_f \neq 0$ and $\vec{\xi}_a \neq 0$, in the ferroelectric phase $\vec{\xi}_f = 0$ and $\vec{\xi}_a = 0$, whereas in the antiferroelectric phase $\vec{\xi}_a \neq 0$ and $\vec{\xi}_f = 0$.

The spectrum of elementary excitations of the director-polarization field in an antiferroelectric liquid crystal has been first discussed by Žekš, Blinc and Čepič [9]. They have shown that the onset of antiferroelectric order at the phase transition point is accompanied by a slowing down of the soft mode, that has an antiferroelectric (non-polar) character. In contrast to ferroelectric liquid crystals, where the soft mode represents the condensation of a plane wave excitation with a wave-vector $\vec{q}_f$ which is *close to the center* of the Brillouin zone, the antiferroelectric soft mode here represents the condensation of a plane wave with the wave-vector $\vec{q}_d$ *near the edge* of the Brillouin zone. Here $|\vec{q}_f| = 2\pi/p$, and p is the period of the helix of the ferroelectric *SmC** phase, whereas $|\vec{q}_d| = \pi/d$ and d is the smectic interlayer distance. The ferroelectric and antiferroelectric fluctuations in the *SmA* phase are illustrated in Figure 2. We have here defined a Brillouin zone because of the 1-D periodicity of the smectic layers. For the SmA phase the Brillouin zone is $(-\pi/d, \pi/d)$.

In the antiferroelectric SmC_A^* the doubly degenerate branch of tilt excitations splits into amplitude and phase dispersion branches. The amplitude branch represents the fluctuations of the magnitude of the tilt angle θ and has a finite frequency gap for all temperatures except at the $SmA - SmC_A^*$ phase transition temperature T_{ca}. On the contrary, the branch of phase excitations, which represent thermal excitations of the phase profile $\Phi(z, t)$ is expected to be gapless for all temperatures below T_{ca}. This is a result of a breaking of the continuous symmetry of the *SmA* phase and the appearance of nonzero tilt in the antiferroelectric SmC_A^* phase.

It is interesting to see the structure of the amplitude and phase excitations in the antiferroelectric SmC_A^* phase, as derived from the Landau theory [9]. As it is shown in Figure 3, we have at the center of the Brillouin zone a noncritical ferroelectric (polar) amplitude mode (a), and a gapless antiferroelectric (nonpolar) phase mode shown in (b). The ferroelectric amplitudon represents a small increase of the tilt in the i-th layer and a small decrease of the tilt in the neighboring layers. As a result, there will be a small, spatially homogeneous polarization $\delta\vec{P}(t)$, fluctuating in time and the mode will be therefore polar. On the other hand, the antiferroelectric phason (b) repre-

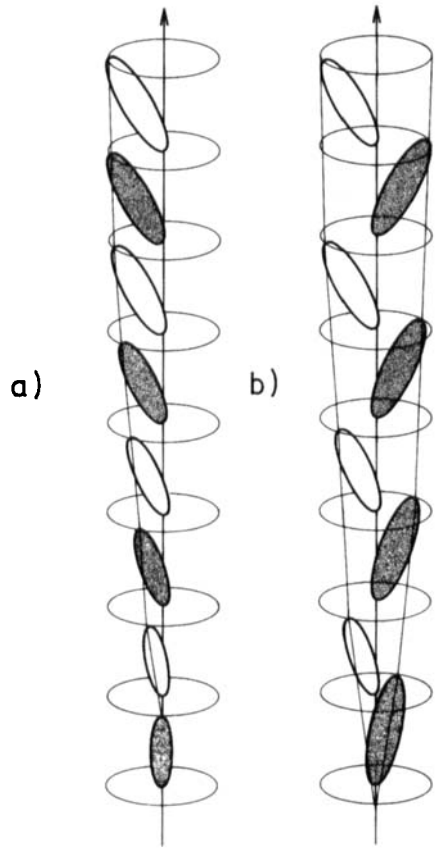

FIGURE 2 The ferroelectric (a) and antiferroelectric (b) fluctuations in the *SmA* phase of chiral antiferroelectric liquid crystal. Whereas the ferroelectric fluctuations are noncritical, the antiferroelectric soft mode critically slows down and condenses at the edge of the Brillouin zone. The drawings are schematic.

sents a small local rotation of the molecules around the layer normal in each smectic layer and appears as a small rotation of the helix as a whole. Because this costs no energy, the corresponding relaxation time is infinite. This is therefore the zero-frequency, symmetry restoring Goldstone mode of the transition. In contrast to the polar Goldstone mode in ferroelectric liquid crystals, the Goldstone mode in antiferroelectric liquid crystals is of non-polar nature, because the overall rotation of a sample as a whole induces no time fluctuating polarization. Similarly we observe that the antiferroelectric amplitudon at the center of the Brillouin zone represents

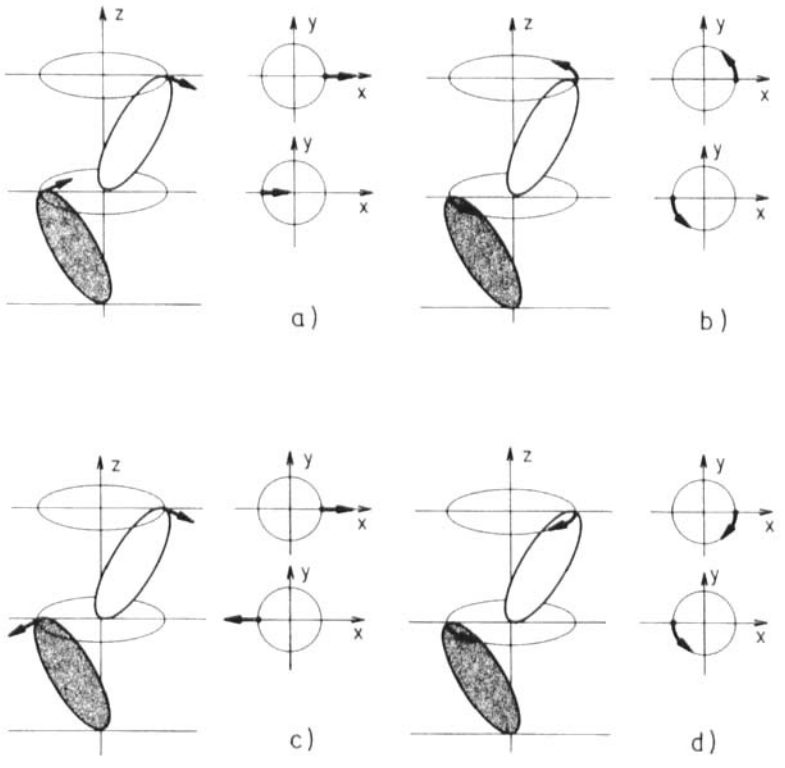

FIGURE 3 Characteristic excitations in the SmC^*_A phase: (a) the ferroelectric amplitudon and (b) the Goldstone mode (antiferroelectric phase mode) near the center of the Brillouin zone (c) the non-polar antiferroelectric amplitudon and (d) the ferroelectric phase mode. Excitations (c) and (d) also take place at the center of the Brillouin zone.

a non-polar fluctuation of the magnitude of the tilt angle (see Fig. 3c), whereas the phase mode at the center of the Brillouin zone represents a polar fluctuation, as shown in Figure 3d.

The dynamics of the antiferroelectric phase modes near the center of the Brillouin zone in the SmC^*_A phase can be derived from the phase dependent part of the nonequilibrium free energy density in the constant amplitude approximation, where the fluctuations of the magnitude of the order parameter are neglected

$$g(z,t) = -\wedge_a \theta_0^2 \left(\frac{\partial \Phi(z,t)}{\partial z}\right) + \frac{1}{2} K_{3a} \theta_0^2 \left(\frac{\partial \Phi(z,t)}{\partial z}\right)^2 \tag{2}$$

The above expression is similar to the free-energy expansion for the phason excitations in the ferroelectric SmC^* phase [6]. Here, Λ_a and K_{3a} are the Lifshitz and torsional elastic constants, associated with the antiferroelectric order parameter $\vec{\xi}_a$, which is expressed as

$$\vec{\xi}_a(z,t) = \theta_0(\cos\Phi(z,t), \sin\Phi(z,t)) \tag{3}$$

and θ_0 is the tilt angle in the antiferroelectric phase.

Following the Landau-Khalatnikov equations of motion for the nonequilibrium phase profile $\Phi(z,t) = q_c z + \delta\Phi(z,t)$ in the unperturbed SmC_A^* phase, one obtains overdamped and monochromatic plane-wave solutions for the phase excitations.

$$\delta\Phi_q(z,t) \propto e^{-t/\tau(q)} \cdot \cos(qz + \phi) \tag{4}$$

where ϕ is arbitrarily chosen. The corresponding phason dispersion relation is gapless and parabolic

$$\tau^{-1}(q) = \frac{K_{3a}}{\gamma} \cdot q^2 \tag{5}$$

It should be noted that in view of the relation $\Phi(z,t) = q_c z + \delta\Phi(z,t)$, the above expressions are written in a frame, which rotates together with the helix. As a result, the phason dispersion has a minimum at finite $\vec{q}$ in the reciprocal space, which is a well established result, characteristic of modulated structures. One should also stress that in the low-frequency and long-wavelength limit $\vec{q} \to 0$, the antiferroelectric phason excitations in the SmC_A^* phase are non-polar, i.e. there is no fluctuating electric dipole moment, $\delta\vec{P}(z,t) \equiv 0$. This has the consequence, that contrary to the case of ferroelectric liquid crystals, the antiferroelectric eigenmodes with $\vec{q} \to 0$ cannot contribute to the linear response of the SmC_A^* phase in a dielectric experiment. On the other hand, these eigenmodes give rise to strong fluctuations of the dielectric tensor and should be observable in the quasielastic light scattering experiments.

In a quasielastic light scattering experiment, one measures the time-autocorrelation function $G(t') = \langle I(0) \cdot I(t') \rangle$ of the scattered light intensity $I(t)$. Here the monochromatic light propagates through the crystal as an optical eigenmode $|\vec{k}, i\rangle$ with the wave-vector $\vec{k}$ and polarization i and is scattered by the thermally excited fluctuations of the dielectric tensor $\delta\underline{\varepsilon}(\vec{r},t)$

into the optical eigenmode $|\vec{k}', f\rangle$ with the wave-vector $\vec{k}'$ and polarization f. In the single scattering (Born) approximation, the time-autocorrelation function of the scattered light intensity at a scattering wave-vector $\vec{q} = \vec{k}_i - \vec{k}_f$ is

$$G(t') \propto \langle \delta\varepsilon_{if}(\vec{q},0) \cdot \delta\varepsilon^*_f(\vec{q},t') \rangle \tag{6}$$

where $\delta\varepsilon_{if}(\vec{q},t)$ denotes the Fourier component of the fluctuating part of the dielectric tensor at the scattering wave-vector. For a light scattering experiment in spatially inhomogeneous liquid crystalline phases the following issues are important:

(i) One must know the structure of optical eigenmodes in the crystal as well as their dispersion relation in order to determine the wave-vectors $\vec{k}$ and polarizations of the incoming and scattered beam, respectively. Although this seems to be an analytically unsolvable problem for spatially modulated and birefringent liquid crystalline phases, it has been shown in recent years [11,12] that a simple perturbative approach to the optics can be used here. Within this approach the optical properties of birefringent and modulated structures are described by the *space-averaged* dielectric tensor. The corresponding approximate optical eigenmodes are just linearly polarized ordinary and extraordinary waves with linear dispersion relations. It has also been shown that this approach is valid for most practical purposes and breaks down for very large tilt angles or for the propagation of light along the helical axis or at the Bragg angle. Keeping in mind that the interlayer distance of the SmC^*_A phase is several orders of magnitude smaller than the wavelength of light, the dielectric tensor can be approximated by its average value within the antiferroelectric unit cell

$$\underline{\varepsilon} = \frac{1}{2}(\underline{\varepsilon}_i + \underline{\varepsilon}_{i+1}) \tag{7}$$

Here, $\underline{\varepsilon}_i$ denotes the local dielectric tensor in the i-th layer, which is obtained [11] by two successive rotations of the tensor with the principal axis $\varepsilon_i, i = 1,2,3$, starting from the configuration, where the eigenframe (1,2,3) coincides with the laboratory frame (x, y, z). First the tensor is rotated through the tilt angle θ around the y axis followed by a rotation through the phase angle $\Phi(z)$ around the z axis. The resulting dielectric tensor, averaged over two neighboring layers is in the limit of

small tilt and local uniaxiality ($\varepsilon_1 = \varepsilon_2$)

$$\underline{\varepsilon} = \begin{bmatrix} \varepsilon_1 + \frac{1}{2}(\varepsilon_3 - \varepsilon_1)\theta^2 & 0 & 0 \\ 0 & \varepsilon_1 + \frac{1}{2}(\varepsilon_3 - \varepsilon_1)\theta^2 & 0 \\ 0 & 0 & \varepsilon_3 - (\varepsilon_3 - \varepsilon_1)\theta^2 \end{bmatrix}$$

$$+ \frac{1}{2}(\varepsilon_3 - \varepsilon_1)\theta^2 \begin{bmatrix} \cos 2\Phi(z) & \sin 2\Phi(z) & 0 \\ \sin 2\Phi(z) & -\cos 2\Phi(z) & 0 \\ 0 & 0 & 0 \end{bmatrix} \tag{8}$$

It is not surprising that the dielectric tensor of the SmC_A^* phase is analogous to the dielectric tensor of a chiral nematic phase [13], because both phases are nonpolar and chiral.

(ii) The fluctuating part of the dielectric tensor, which is responsible for the quasielastic light scattering, is in the limit of a small tilt angle and small amplitudes of phase excitations directly proportional to the phase excitation $\delta\Phi_q(z,t)$:

$$\delta\underline{\varepsilon}(z,t) = \frac{1}{2}(\varepsilon_3 - \varepsilon_1)\theta_\circ^2 \begin{bmatrix} -\sin(2q_c z) & \cos(2q_c z) & 0 \\ \cos(2q_c z) & \sin(2q_c z) & 0 \\ 0 & 0 & 0 \end{bmatrix} \cdot \delta\Phi_q(z,t) \tag{9}$$

From the above expression one observes that the phason excitation $\delta\Phi_q(z,t)$ with the wave-vector q is observable via fluctuations of the dielectric tensor as an excitation with the wave-vector $q \pm 2q_c$. The dispersion relation for the phason excitations in the SmC_A^* phase has in the laboratory frame a minimum at the scattering wave-vector $q_s = 2q_c$:

$$\tau^{-1}(q) = \frac{K_{3a}}{\gamma}(q \pm 2q_c)^2 \tag{10}$$

There are several time-resolved, quasielastic light scattering experiments reported for the SmC_A^* phase [5,7] and we shall here only describe the observation of a truly gapless phason dispersion. The experiment was performed [5] in the SmC_A^* phase of 4-(1-methylheptyloxycarbonyl)phenyl4'-octyloxybiphenyl-4-carboxylate (MHPOBC), slightly below the $SmC_\gamma - SmC_A^*$ phase transition point. Well aligned homeotropic samples of thickness 50 μm have been used in an experimental arrangement, described

elsewhere [6]. In view of the rather small values of the period of the helix near the $SmC_\gamma - SmC_A^*$ phase transition point in this substance, which corresponds to a large critical wave-vector q_c, the phason dispersion relation was measured in a back-scattering, depolarized geometry, as shown in the inset to Figure 4. The light-collecting optics was positioned at a small angle (2°) with respect to the light reflection from the sample surface, thus assuring a heterodyne detection regime. This small deviation of the detector angle from direct reflection introduces a small transverse component q_x of the wave-vector which has been treated as a small perturbation to the dispersion relation [14].

The dispersion relation for phase excitations propagating along the helical axis of the SmC_A^* phase of MHPOBC-as obtained by the quasielastic light scattering in a back-scattering geometry is shown in Figure 4. The solid line represents the best fit to Eq. (10) giving $K_{3a}/\gamma = 5.8 \cdot 10^{-10} m^2 s^{-1}$. From the contribution of the transverse component q_x of the wave-vector to the dispersion relation we can estimate $K_{+a}/\gamma \approx 1 \cdot 10^{-9} m^2 s^{-1}$. Here $K_{+a}/\gamma = 1/2(K_s + K_B)$ is an effective transverse elastic constant [14], K_s and

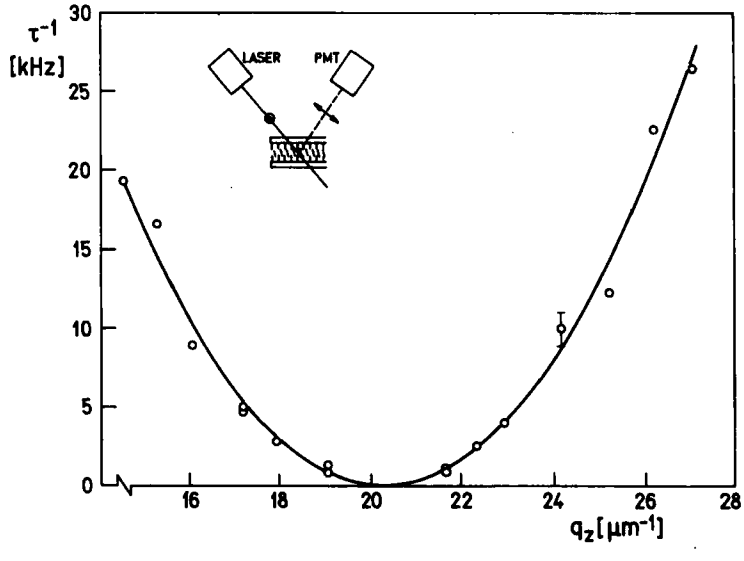

FIGURE 4 Phason dispersion in the SmC_A^* phase of MHPOBC, 0.1K below the $SmC_\gamma^* - SmC_A^*$ transition. The solid line represents the best fit to the Eq. (10) with $K_{3a}/\gamma = 5.8 \cdot 10^{-10} m^2 s^{-1}$ and $K_{+a}/\gamma \approx 1.10^{-9} m^2 s^{-1}$. The inset shows the scattering geometry. The magnitude of the scattering wave-vector q_z, along the z-direction was calculated according to the uniaxial optical model of the SmC_A^* phase, with refractive indices $n_o = 1.5$ and $n_e = 1.625$, according to Ref. 20.

K_B are splay and bend elastic constants respectively and γ is the corresponding viscosity. As one can see from Figure 4, the phason dispersion is indeed gapless within the limits of the experimental error and centered at the scattering wave-vector $q_s = 20.3\ \mu m^{-1}$. For $q_s = 2q_c$ this corresponds to a helix with a wave-vector $q_c = 10.2\ \mu m^{-1}$ or a period of 0.62 μm. The period of the helical structure in the SmC_A^* phase of MHPOBC can also directly be obtained by the selective reflection method as has been measured by A.D.L.Chandani *et al.* [1]. From the position of the selective reflection peak near the $SmC_\gamma - SmC_A^*$ phase transition point, taking into account an average refractive index of the SmC_A^* phase of MHPOBC and assuming a form of the dielectric tensor given by the Eq.8, one obtains the period of the helix of MHPOBC $p = 0.6$ μm, which is in a remarkable agreement with our experimental observations.

The value of K_{3a}/γ, as obtained from Figure 4 is of the same order of magnitude as that obtained in quasielastic light scattering experiments in the ferroelectric liquid crystals CE-8 and DOBAMBC [6,14]. In those experiments, one would usually observe a ratio K_+/γ which is one order of magnitude larger. This is in contrast with the situation in MHPOBC, where K_{+a}/γ is only a factor of two larger than K_{3a}/γ. Such a low value of K_{+a}/γ in antiferroelectric SmC_A^* could be the result of the absence of a spontaneous polarization $\vec{P}(\vec{r})$ in this phase. It was argued [15,16,17,18] and recently observed [19] that the presence of a fluctuating dipole field $\vec{P}(\vec{r}t)$ significantly influences the magnitude of the transverse (or in-plane) elastic constant K_+ due to bend fluctuations of the $\vec{C}$-director field, which represent splay-like fluctuations of the spontaneous polarization field $\vec{P}(\vec{r})$. This results in the appearance of a fluctuating of space charge density $\rho(\vec{r},t)$ and renormalizes the bend elastic constant K_B due to the electrostatic self-energy of the charge distribution [18,19].

The value of K_{3a}/γ as obtained from Figure 4, is in excellent agreement with the value of $K_{3a}/\gamma \approx 6.10^{-10} m^2 s^{-1}$, as obtained by H.Sun *et al* [7] in the SmC_A^* phase of MHPOBC at $q_s \to 0$. Their forward scattering geometry allowed only for the determination of phason relaxation rates in the limit of small scattering wave-vectors, $q_s \to 0$. In this limit they obtain relaxation rates in the SmC_A^* phase of MHOPBC of the order of $200 \sim 300$ kHz. By extrapolating our data in Figure 4. to $q_s = 0$, we obtain phason relaxation rates of the order of $\tau^{-1}(q=0) \approx 250$kHz, which is in excellent agreement with the results of H. Sun *et al.*

In conclusion, our time-resolved quasielastic light scattering experiments clearly demonstrated that a dispersion relation for phason excitations, propagating along the helical axis of the SmC_A^* phase is gapless, as expected

for a Goldstone mode restoring the broken continuous symmetry of the *SmA* phase. It has a minimum at the wave-vector $q = 2q_c$, where q_c is the wave-vector of the unperturbed SmC_A^* structure. This observation is consistent with the proposed form of the dielectric tensor in the SmC_A^* phase and the alternating-tilt model of antiferroelectric SmC_A^* liquid crystals and seems to be the first demonstration of the gapless nature of the phason mode in any incommensurate system with discrete lattice symmetry.

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