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Collective dynamics of a ferroelectric smectogen in geometrical confinement

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We investigate the influence of geometric confinement on the dynamics in a ferroelectric smectic C* material. Molecular and collective relaxation processes are studied by means of broadband dielectric spectroscopy. In microporous material (pore size 200 nm) the Goldstone mode (GM) is still observed, but it is shifted to lower frequencies compared with bulk samples. In nanoporous material (pore size 7·5 nm), the collective reorientation modes are completely suppressed. In all samples, the molecular processes seem to be uninfluenced by the confinement.

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1. Introduction

The effects of geometrical confinement on liquid crystalline order and dynamics have been widely studied during recent years [1]. Interactions of the mesogens with the confining surfaces can not only determine the orientation of the liquid crystal (LC) material, but the geometrical restrictions can also induce or suppress liquid crystalline order. Interactions of liquid crystals with solid surfaces and influences of restricting geometries are of considerable academic and technical interest. Adsorption of mesogens in porous materials provides a convenient way to study such effects. Most investigations have been performed with nematogens. Among the various adsorbate systems studied in the past, listed in order of decreasing cavity sizes, are polymer dispersed liquid crystals (PDLC, see e.g. [2]) consisting of spherical or ellipsoidal LC droplets in a polymeric matrix, organic and inorganic filters with well defined cylindrical parallel pores like ANOPORE or NUCLEPORE membranes (see, e.g. [3]), porous glasses with narrow pore size distributions but random orientations of the cavities (see, e.g. [4–8]), and silica aerogels (see, e.g. [9]) with irregularly shaped cavities of different sizes. In particular, the existence of the smectic layer structure in the latter has been established by X-ray scattering techniques [10]. Even smaller, regular cavities could be provided by MCM-41 type materials.

PDLC systems are particularly well investigated. Due to their electrically controllable optical properties, they are promising materials for technical application. ANOPORE and NUCLEPORE filter materials have been used to determine surface induced nematic order and surface-like elastic constants, and to study geometrical structures of the director field with non-trivial boundary conditions. In both systems, the pore sizes are very large compared with molecular dimensions or the correlation length of the liquid crystalline order parameter. The structure of the nematic director field in these systems is determined to a great extent by the boundary conditions, but influences on nematic order and phase transitions are comparably weak.

Sol–gel glasses can be manufactured with pore sizes of a few nanometers. In addition to influences on the orientation of the adsorbed LC phases, confinement in such materials also has essential effects for nematic and smectic order and the range of existence of the LC phases. A certain disadvantage of porous glasses in the experiments is the random orientation of the pore matrix compared with the straight, oriented channels in ANOPORE and NUCLEPORE filters. Liquid crystals confined to sol–gel glasses have been previously studied, e.g. dielectrically [5–7], by optical Kerr effect measurements [8] and with ^{2}H NMR [4]. These systems provide satisfactorily large filling factors (about 50%), high porosity and a relatively narrow pore size distribution. Nanoporous glasses are transparent and dielectrically inactive.

In previous work, we have performed dielectric and NMR investigations on a confined nematogen (5CB) in ANOPORE filters [11] and porous glass [7, 12], and measured orientational order and molecular dynamics. The isotropic–nematic phase transition is considerably suppressed with respect to the bulk phase. When the sample is cooled from the isotropic phase, the nematic ordering starts smoothly at the phase transition and the order parameter remains lower than in the bulk

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phase at comparable temperatures. The confined nematic phase can be considerably supercooled, more than 50 K below the bulk crystallization temperature. The molecular dynamics in the pores is as fast as in the bulk phase. An additional retarded dielectric process (about two decades slower than the molecular dynamics in the bulk) is detected; it can be attributed to relaxation in a molecular layer directly attached to the pore surfaces. In addition, dielectric studies of isotropic liquids adsorbed in porous sol–gel glass have been performed [13, 14]. They support the correct assignment of the dielectric relaxation processes in confined material.

In this work we focus on the collective dynamic properties of confined smectic samples. A pioneering investigation of the behaviour of confined ferroelectric LC phases was reported by Aliev [6]. He studied silica porous glasses with pore sizes of 1000 Å (referred to as macropores) and 100 Å (micropores). In macropores, he observed a reduction of the smectic A to smectic C phase transition temperature by about 15 K with respect to the bulk phase, and the existence of soft and Goldstone modes. The viscosity for the soft mode was about 15% higher than in the bulk. In micropores, neither soft mode nor Goldstone mode was detected. Collective dynamic modes in ferroelectric liquid crystalline material confined in aerogels have been previously studied by Xu *et al.* [15] who determined the relaxation frequencies and dielectric relaxation strength of the Goldstone mode and found an additional dielectric relaxation process in the confined phase which they attributed to an LC interfacial layer at the inner surfaces of the system.

We use the standard substance DOBAMBC confined to 0·2 μm ANOPORE filters and nanometer pores of sol–gel glass to study the influences of different pore sizes. Broadband dielectric spectroscopy is applied which is particularly useful in studying the molecular mobility and collective dynamics over a large frequency range.

2. Samples

Commercial porous glass was obtained from Geltech Inc., U.S.A., with pores of size 7·5 nm. The glass cylinders of approximately 1 cm diameter and 1 cm length were cut to thin slices by means of a diamond string saw. The slices were filled with mesogenic material using a preparation technique described earlier [7, 12]. The porous glass slices were evacuated and heated for two days to remove volatile impurities before filling in vacuum with the mesomorphic material in the isotropic phase. The ANOPORE filters from Anotech consist of 60 μm thick sheets with 0·2 μm diameter cylindrical pores perpendicular to the surface. They were cut to small (1 cm diameter) disks and filled with the mesomorphic material without special previous surface treatment. All samples were carefully dried between filter papers after filling, in order to remove excessive material at the outer surfaces. The measurements were performed on cooling from high to lower temperatures. In this way we avoided liquid crystal material being squeezed out of the pores after thermal expansion.

Dielectric measurements were performed in the frequency range 10^{-2}–10^{6} Hz using a Solartron–Schlumberger frequency response analyser FRA 1260, and a Novocontrol active sample cell BDC-S. The sample disks were covered on both sides with thin aluminium foil to ensure good electrical contact. Both the porous glass and the ANOPORE filters are dielectrically inactive. This was tested in previous measurements of the non-filled porous materials. Temperature control was better than ±0·05 K.

The commercial substance 4-*n*-decyloxybenzylidene-4′-amino-2-methylbutylcinnamate (DOBAMBC) was obtained from Aldrich and used without further purification. The phase sequence of bulk DOBAMBC is

$$\begin{array}{ccccccc} \mathrm{I} & \leftrightarrow & \mathrm{SmA} & \leftrightarrow & \mathrm{SmC}^* & \rightarrow & \mathrm{SmI}^* \\ & 386\ \mathrm{K} & & 366\ \mathrm{K} & & 334\ \mathrm{K} & \end{array}$$

3. Dielectric spectra

We extract the information on the dynamic processes in the sample from the imaginary part $\varepsilon''(\omega)$ (dielectric loss) of the complex dielectric function $\varepsilon^* = \varepsilon' + \mathrm{i}\varepsilon''$. Figure 1 shows some characteristic dielectric loss spectra of DOBAMBC in different types of geometrical confinement. As we are particularly interested in the collective dynamics, we have concentrated only on the low frequency part of the spectra up to 1 MHz. More molecular relaxation processes were detected in the frequency range 10^{6}–10^{9} Hz [16]. The frequency dependence of ε^* can be described by a superposition of Havriliak–Negami [17] model functions and a conductivity contribution:

$$\varepsilon'' = \frac{\sigma_0}{\varepsilon_0}\frac{1}{\omega^{s}} - \sum_{k=1}^{N} \mathrm{Im}\left\{\frac{\Delta\varepsilon_k}{[1+(\mathrm{i}\omega\tau_k)^{\alpha_k}]^{\beta_k}}\right\} \qquad (1)$$

with the dielectric strengths $\Delta\varepsilon_k$ and relaxation times τ_k of each individual process k involved in dielectric relaxation, the vacuum permittivity ε_0, and the conduction parameter σ_0. The exponents α and β are empirical fit parameters which describe a symmetric and unsymmetric broadening, respectively, of the relaxation peaks. The fit of the spectra could be performed with two relaxation processes in all samples. The first term on the right hand side of equation (1) describes the motion of free charge carriers in the sample. In the case of an Ohmic behaviour ($s = 1$) σ_0 is the Ohmic conductivity of the smectic material. Details of the analysis of the dielectric spectra are given in [7, 12].

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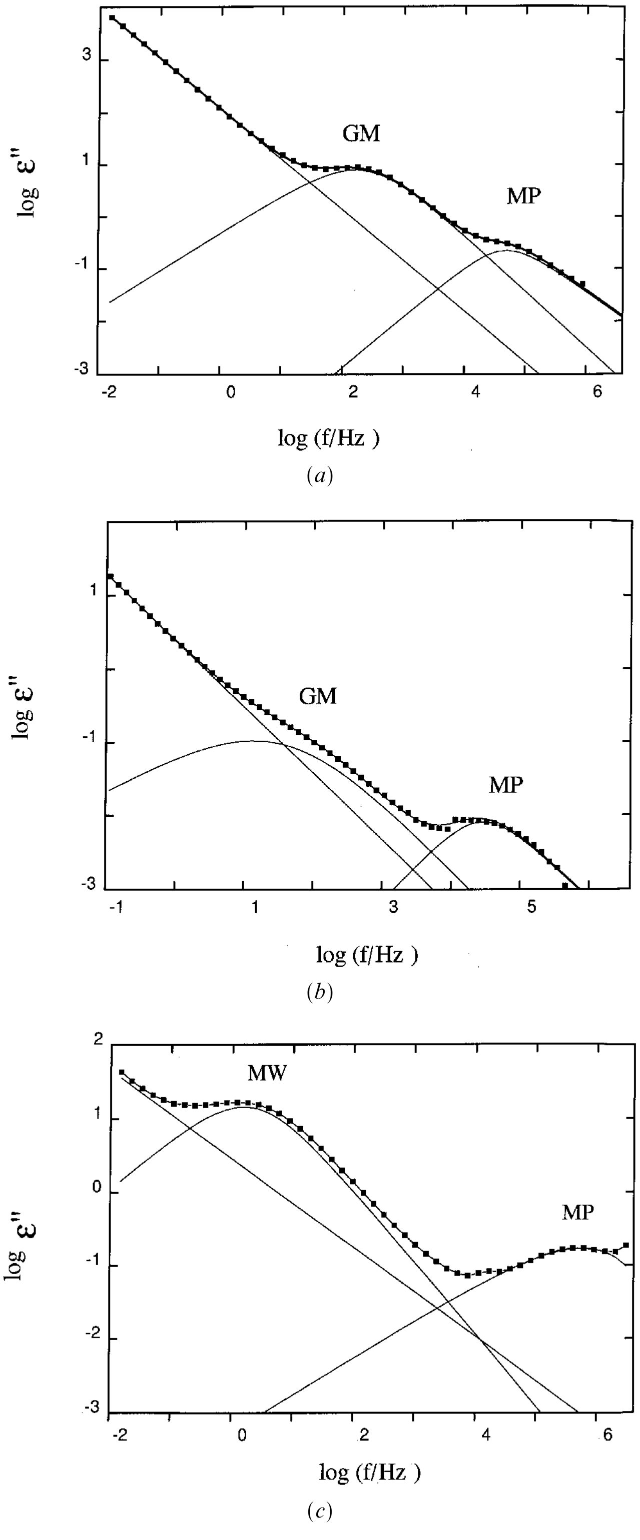
4. Experimental results

Figure 2 shows a 3D graphics representation of the temperature and frequency dependent dielectric loss in a bulk sample. One recognizes the pronounced conductivity wing in the low frequency parts of the spectra. The slope is linear with $1/\omega$ and the conductivity σ_0 is a function of temperature which shows a small step at the nematic–isotropic transition. At high frequencies, a molecular reorientation process with a comparably small relaxation strength is seen. This is present in all phases and thermally activated with $\tau \propto \exp(E/RT)$. The activation energy E is about 82 kJ mol^{-1}. Individual molecular motions are responsible for the relaxation of the dipolar polarization in the external field. In the mesophases there exist several individual relaxation processes corresponding to rotational motion of the molecules about their long axes, librations of this axis and flips of the molecule about the short axis. While most of these processes appear in the frequency range above 1 MHz [16], the latter reorientation is comparably slow and leads to the particular dielectric relaxation peak observed in our spectra. At the transition from an isotropic to the smectic phase we expected a slight step in the relaxation rate to lower frequencies, as found at the T_{NI} transition (e.g. [7]), when the isotropic reorientation changes to the slower reorientation about the molecular short axis in the smectic phase, but such a step was too small to be detectable.



At frequencies in the kHz range the Goldstone mode is found. Its relaxation strength is higher by about two orders of magnitude than that of the molecular process, which makes the identification as a collective process unambiguous. The Goldstone mode is present only in the smectic C* phase, below 366 K. Its relaxation frequency has almost no temperature dependence, and it occurs in the typical range found in other ferroelectric smectics (e.g. [15]). The soft mode is basically hidden in the large conductivity wing.

At 333 K, the dielectric relaxation suddenly drops by approximately two orders of magnitude at all frequencies. This change in the dynamic properties indicates the transition of the bulk sample into the smectic I* phase.

Figure 1. Dielectric loss spectra of the SmC* phase of DOBAMBC in different environments. The fitted relaxation processes and conductivity contributions are shown as solid lines. (*a*) Bulk sample at 363 K (SmC*) with two relaxation processes, a molecular process (MP) at high frequencies and the Goldstone mode (GM) of the *c*-director on the tilt cone. (*b*) Untreated ANOPORE powder sample, pore diameter 200 nm, at 353 K (see text); the high frequency peak is again the molecular process (MP) and the low frequency process is the Goldstone mode (GM). (*c*) Porous glass with 7·5 nm pore diameter at 363 K, with the molecular process (MP) at high frequency and a low frequency Maxwell–Wagner (MW) process.

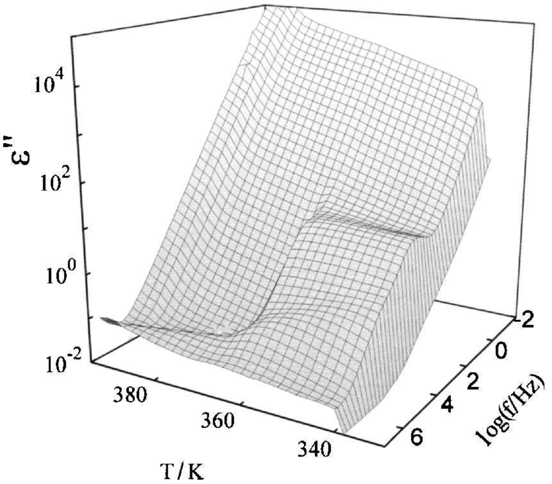


Figure 2. Dielectric loss of bulk DOBAMBC as a function of temperature and frequency. The relaxation processes seen are the Goldstone mode at low frequencies (≈1000 Hz) in the ferroelectric phase and the molecular process in all phases.

When the dielectric spectra of ANOPORE confined DOBAMBC (figure 3) are compared with the bulk spectra, one sees that the dielectric process related to the Goldstone mode has disappeared completely, while conductivity and molecular relation contributions are practically the same as in the bulk sample. The reason for that need not be a change in the dynamical properties due to confinement. It might well be (and it is in fact) caused mainly by the aligning properties of the ANOPORE channels. Untreated ANOPOREs orient most nematic liquid crystals axially along the pores. The smectic A layers which form when the sample is cooled from the isotropic to the smectic phase arrange normal to the pore channels. Consequently, the Goldstone mode is dielectrically inactive when the electric field is applied parallel to the pore axes. The same applies to the soft mode at the A–C transition. This potential effect cannot be distinguished by the present experiment from an actual suppression of the GM. The application of the electric field perpendicular to the pores of an ANOPORE disk, which could be used to detect the Goldstone mode in the stacked layers, requires special interdigitated electrodes which cause a remarkably reduced signal and a number of other problems for the dielectric experiments. It is also impossible to slice the brittle filter material and to stack it perpendicularly in the capacitor.

We have therefore used another technique to probe the presence of the Goldstone mode in the SmC* phase. The ANOPORE filter sheet was loaded with the liquid crystal as in the previous experiment, but then, before putting the material into the capacitor, the sample was ground to a powder of small grains, pressed and put between the electrodes of the set-up. We do not expect that the layers are isotropically oriented in the ANOPORE powder, but at least a part of the large number of ANOPORE grains will be oriented with pore axes oblique to the electric field. Figure 4 shows the dielectric spectrum measured with the ANOPORE powder. A reappearance of a dielectric loss peak between the molecular relaxation process and the conductivity wing is observed. Two features of this process help to identify it as the Goldstone mode. First, the process is present solely in the smectic C* temperature range; it sets in at 366 K. The SmA–SmC* phase transition temperature coincides with the bulk value. Secondly,

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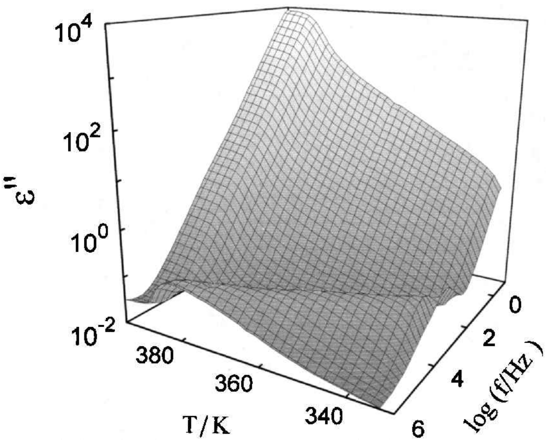

Figure 3. Dielectric loss of DOBAMBC in ANOPORE filter sheets as a function of temperature and frequency: only the molecular relaxation process is present.

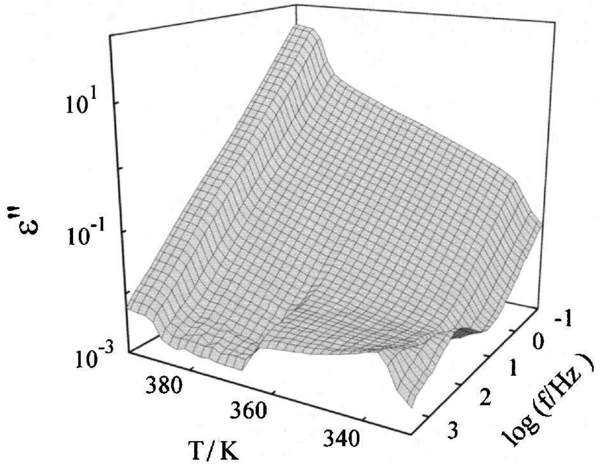

Figure 4. Dielectric loss of DOBAMBC in ANOPORE powder as a function of temperature and frequency. The molecular relaxation process is present as in figure 3, but we have clipped the frequency window to show the appearance of the Goldstone mode in the confined SmC* phase.

relaxation strength and frequency are both nearly temperature independent, which is also a characteristic of the bulk Goldstone mode. The relaxation frequency is shifted to lower frequencies by less than one decade with respect to the bulk phase. We can discount that the process is generated by small amounts of liquid crystal which might have been squeezed out of the pores during the process of grinding the ANOPORE sheets. Such material would behave in a bulk-like way and we would expect the Goldstone mode contribution exactly at the bulk frequency.

The dielectric spectra of the nanometer-confined mesogen are depicted in figure 5. They differ from the spectra of the bulk phase (figure 2) in three essential details. The first is the appearance of a thermally activated loss process at low frequencies in the conductivity wing. This process is present in all phases and its characteristic frequency has the same temperature dependence as the conductivity $\sigma_0(t)$. We identify this process as a Maxwell–Wagner (MW) contribution. It is not related to molecular or collective dynamics of the smectogen but is a general feature of confined liquids in very small pores, originating from the restricted mobility of free charge carriers in pore channels which are not parallel to the electric field. The second feature is the complete disappearance of the Goldstone mode contribution in the nano-confined sample. In the porous glass, the pore channel distribution is isotropic and even if a preferred smectic layer orientation existed in the pores, a large part of the sample would be oriented such that the Goldstone mode is dielectrically active. Therefore, the observed disappearance of the Goldstone mode contribution in the dielectric spectra cannot be an orientational effect as in the oriented ANOPORE sample. It is a direct effect of the confining geometry. The third difference is the disappearance of the step at the bulk transition SmC*–SmI*. The sample can be supercooled below 300 K without any indication of a step in the dielectric curves.

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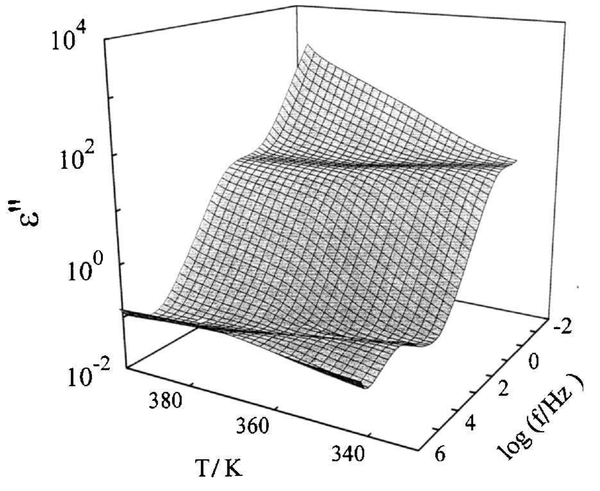

Figure 5. Dielectric loss of DOBAMBC in ANOPORE powder as a function of temperature and frequency. The relaxation processes seen are the low frequency Maxwell–Wagner process and the molecular process; both appear at all temperatures.

After this qualitative discussion, we present the relaxation rates and relaxation strengths of the DOBAMBC samples in different geometries in figures 6 and 7, respectively. The data were extracted from a fit of the

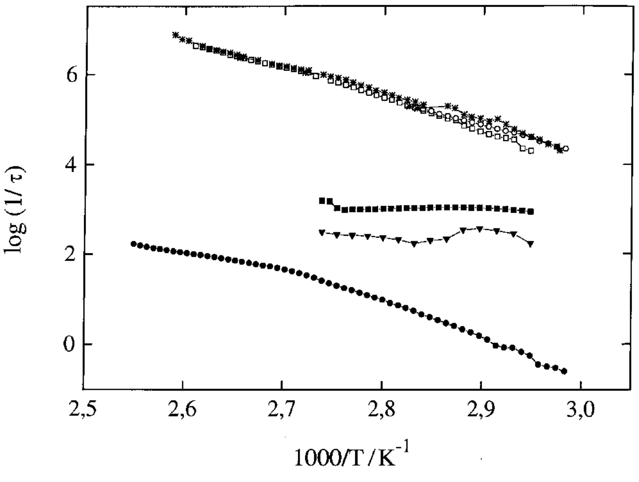

Figure 6. Relaxation frequencies of the individual processes in DOBAMBC extracted from figures 2–5. The denotation is: MW in porous glass (●), GM in powdered ANOPORE (▼), bulk GM (■), bulk MP (□), MP in ANOPORE filters (○), MP in porous glass (*).

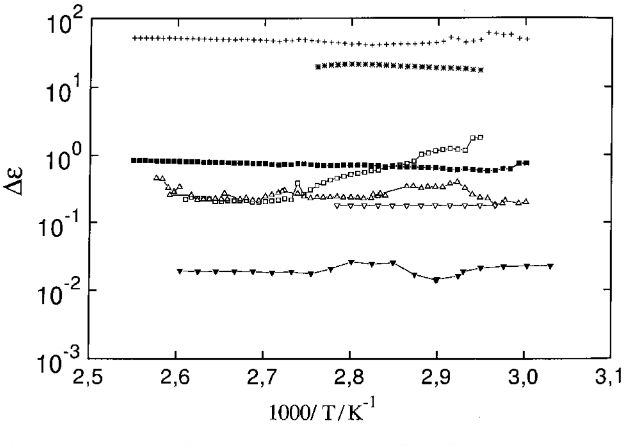

Figure 7. Relaxation strengths of the individual processes in DOBAMBC, extracted from figures 2–5. The denotation is: MP in powdered ANOPORE (▼), GM in powdered ANOPORE (▽), MP in ANOPORE (△), MP in porous glass (□), bulk MP (■), bulk GM (*), MW in porous glass (+). Different processes in individual samples can be compared quantitatively while comparison of different samples is correct only to the order of magnitude. For the GM of the ANOPORE powder sample, one has to consider that the orientational distribution of the ANOPORE grains is not known exactly.

experimental spectra to equation (1). One observes that the molecular dynamics are relatively uninfluenced by the confining geometries. The relaxation frequencies are unchanged with respect to the bulk sample. Even in the nanometer glass pores, there is no noticeable slowing down of the molecular dynamics.

The Goldstone mode appears only in the bulk and the ANOPORE powder sample in the temperature range of the SmC* phase. Its relaxation rate is fairly temperature independent. In the bulk sample, the inverse relaxation rate $1/\tau$ is about $10^3\ \mathrm{s}^{-1}$, while in the confined sample it slows down by a factor of 5 to about $200\ \mathrm{s}^{-1}$. A similar decrease of the GM frequency has been reported for another system, the ferroelectric liquid crystal W314 [18] adsorbed in aerogel. The measured GM frequency was reported to decay from 4 kHz in the bulk sample to less than 650 Hz in the confined material [15], that is approximately by the same ratio as in our samples. The authors explain this retardation by an extremely high viscosity for the collective modes in the confined material as compared with the bulk.

The dielectric strengths $\Delta\varepsilon$ (figure 7) indicate that all or at least a large portion of liquid crystalline material contributes to the fast molecular relaxation. We could not detect any surface attached layer with retarded dynamics as was found in previous experiments with the nematogen 5CB in porous glass [7, 12].

Comparison of the dielectric strengths of different samples is not very accurate because of difficulties in the exact determination of the sample geometry of the confined material. In particular, dielectric strengths of the powdered ANOPORE sample are meaningful only for a comparison of the Goldstone and molecular modes in that individual sample, and not in an absolute comparison with other samples, as the filling factor of the capacitor and the orientational arrangement of the grains are not exactly known.

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5. Summary

We summarize the results as follows. Dielectric experiments show that the molecular dynamics of the free molecules in micrometer and nanometer pores are as fast as in the bulk phase. We did not detect any retarded layer of directly surface-attached molecules in the nanometer confined material.

Substantial differences are observed for the collective dynamics. The Goldstone mode which is characteristic for the SmC* bulk phase can also be found in micropores of 0·2 μm. The SmC* layer structure in untreated ANOPOREs is stacked along the pore axis and consequently the spontaneous polarization is in the plane of the ANOPORE sheet. Therefore the Goldstone mode is not observed in the ANOPORE confined mesogen when the electric field is applied along the pore axes. In powdered ANOPORE material we have unambiguously identified the Goldstone mode. From the temperature range in which the Goldstone mode is observed in the dielectric spectra we conclude that the smectic A–smectic C* phase transition temperature in the micropores is the same as in the bulk material.

Compared with porous glass with randomly oriented interconnected pores, the channels in our ANOPORE powder sample have practically infinite length (60 μm≫7 nm). The motion of free charge carriers is unrestricted and therefore we observed no contributions from the Maxwell–Wagner effect. (MW appears in dielectric spectra of macroporous glass [6] in the same frequency range as the Goldstone mode and masks the latter at least partially.) The absence of a Maxwell–Wagner peak allows us to make quantitative statements on the dynamics of the Goldstone mode. It is shifted to lower frequencies by a factor of about 5 with respect to the bulk process. This behaviour is similar to the observations of Xu *et al.* [15] who also reported a slowing down of the Goldstone mode relaxation frequency in aerogel confined material by slightly less than one decade.

In nanometer porous glass, the Goldstone mode is completely suppressed. We observe a low frequency Maxwell–Wagner process, which is thermally activated and appears at all temperatures investigated. Our results confirm some previous experiments by Aliev [6] for macroporous and microporous glasses filled with an enantiomorphic smectogenic mixture (SCE12 from Merck Ltd, UK). Aliev reported a suppression of the Goldstone mode in nanoporous material. In their experiments performed with smectogens adsorbed in silica aerogels, Xu *et al.* [15] found an additional dielectric process in the ferroelectric smectic C* phase of their mesogens. The dielectric strength of this process was substantially larger than that of the molecular process. In our porous glasses, we have found no comparable process.

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