Creep dynamics of structural defects in ferroelectric liquid crystals with chevron geometry

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Electric-field induced motions of zigzag walls forming defects in the chevron structure of surface stabilized ferroelectric liquid crystals are investigated. An explicit experimental evidence, including direct microscopic observation, is reported that the walls display viscous creep motions at small length scales, being dependent on the amplitude U and frequency f of an applied voltage. The relaxation-to-creep transition is analyzed using both the electro-optical response and dynamic hysteresis data recorded at different values of U and f. It is shown that, at any fixed temperature below the chevron-to-bookshelf transition point, there exists a critical line in the U-f plane, separating the relaxation and creep dynamic regions. In contrast to field-activated nonlocal excitations discovered in various disordered media, the creep motions of zigzag walls are found to occur not only at low or very low field frequencies.

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I. INTRODUCTION

The complex structure of disordered media, despite their large diversity, mostly originates from a common mechanism of a competition between elasticity and disorder. A simple consequence of this competition is the formation of metastable areas (or domains) of different order degrees, separated by hypersurfaces (or domain walls). Understanding temperature and/or field activated motions of such defect interfaces is a challenging research problem of a great importance in developing innovative technologies.[1–6] Although the dynamics of defects has intensively been studied both experimentally[2,7–9] and theoretically,[10–13] mainly in the framework of a phenomenological approach to analyze creep and depinning processes, the propagation of defect walls is still not completely explained, especially at the microscopic level. In particular, a stretched exponential dependence of the wall velocity on the applied field, being the main characteristics of creep jumps over the energy barriers between metastable states, has experimentally been demonstrated to be described by the dynamic exponent being dependent only on the system dimension and the kind of the pinning potential.[1,4,14,15] However, there are no suggestions whether universality classes of creep phenomena exist or not. Furthermore, the nature of relaxation-to-creep and creep-to-sliding (depinning) transitions in the presence of ac external fields remains rather unclear.[8,9,13] Thus, to gain a better insight into these questions, studies of defect wall dynamics in other systems are advisable.

The aim of this paper is to report experimental results of studying viscous creep motions of structural defects in liquid crystals. More specifically, these creep motions refer to the dynamics of the so-called zigzag walls[16–19] in thin surface stabilized ferroelectric liquid crystals (SSFLCs) of the chevron structure. Due to a high-speed electro-optic switching between bistable orientational states of molecules in the chevron SSFLC, such materials are very important both for display and nondisplay applications, in spite of a rather large instability of chevron structures and their strong tendency to form macroscopically large defects.[19] In contrast to purely geometric interfaces in various systems studied until now, the zigzag walls are "physical" objects, having nonzero thicknesses, as illustrated in Fig. 1. These walls exhibit unfolded (bookshelf) layer structure, which is typical for the high-temperature phase rather than for the low-temperature folded phase.[19] Consequently, the character of the field-induced dynamics of molecules within zigzag walls and chevron regions can be very different. At weak enough ac external electric fields, molecules forming both folded and unfolded smectic layers can only undergo a collective motion.[20] However, for growing field amplitudes, the molecular motion can be expected to begin disabling its collectivity in the first instance within the zigzag walls. This can lead to displacements of the defect walls or their fragments. In the creep regime (at intermediate field amplitudes), the wall motions are confined to small regions of systems, and thus have a nonergodic character. However, in the sliding regime (at sufficiently strong fields), the wall motions occur at all length scales, up to length scales of whole systems, displaying ergodicity. To verify this proposed conception of zigzag wall motions, the electro-optic response and the area of the hysteresis loop have been measured for thin samples of liquid-crystalline mixture Felix 15-100, in wide ranges of amplitudes and frequencies of applied voltages. Moreover, microscopic observations of defect motions under relatively high voltages have been performed.

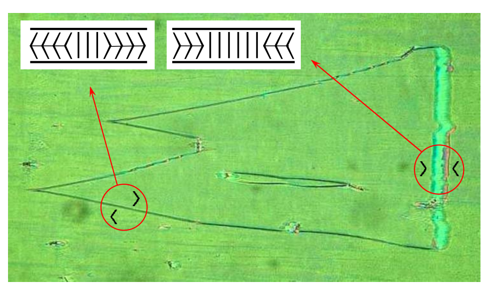

FIG. 1. (Color online) Microphotograph of a zigzag defect forming a closed loop in a cell filled with the Felix 15-100 mixture. The arrangement of folded smectic layers is symbolically indicated inside circles. Structures of thin and thick walls of the zigzag defect are schematically shown in left and right insets, respectively, within the chevron plane (perpendicular to the layer plane and to sample plates).

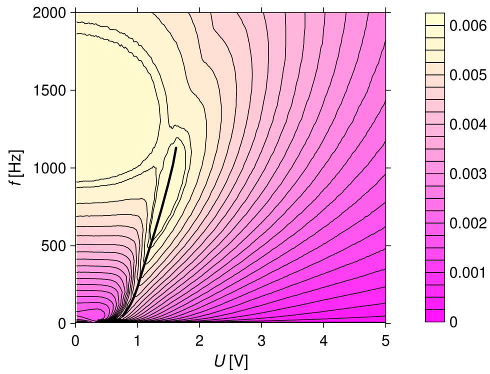

FIG. 2. (Color online) Contour map showing the U and f dependences of the imaginary part ε'' (in arb. units) of the electro-optic response for a chevron SSFLC sample. The thick line represents a projection of an edge line on the U-f plane. This line indicates a sharp change of the slope of ε''.

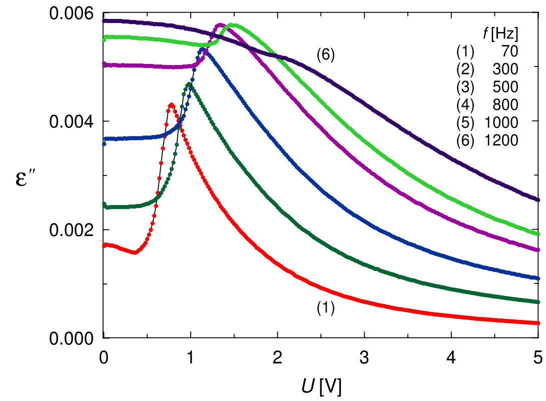

FIG. 3. (Color online) Dielectric loss as a function of the voltage amplitude for different voltage frequencies.

II. RESULTS AND DISCUSSION

Measurements of the electro-optic response spectra have been carried out at fixed temperature of 50 °C, for sinusoidal voltages of amplitudes U (rms) below the switching threshold and frequencies $f > 5$ Hz, for which the contribution due to ionic currents is negligible.[21] Figure 2 illustrates the U and f dependences of the dielectric loss ε'' determined for a sample with thickness $d=5$ μm. It is seen that, besides a significant contribution to the collective relaxation reorientations of molecules at weak U and intermediate frequencies covering roughly the range 700 Hz $< f <$ 2 kHz, there appears also a subspectrum at relatively low frequencies $f < 700$ Hz in the regime of weak U. Such a low-frequency subspectrum displays large complexity and can be attributed to localized relaxation processes within zigzag walls at various size scales.[20] As the voltage amplitude increases, the level lines on the contour map, which at first are nearly straight, parallel to the U axis, change subsequently their directions, especially quickly in the vicinity of the edge line drawn in Fig. 2, indicating a strong nonlinear behavior of the system. Since ionic currents considerably affect the dielectric response as $f \to 0$, the low-frequency endpoint of this line cannot be reliably determined. The high-frequency endpoint is located at $f_{\max} \approx 1100$ Hz. At each remaining point of the edge line, the slope of the dielectric loss function rapidly changes along the U axis, as shown in Fig. 3. The resulting transformation of the functional form of ε'' reflects the disappearance of collective motions of molecules within those sample fragments which are less stable than others, i.e., within zigzag walls. Thus, in the regime of high enough U, much less, however, than the switching threshold, and in the regime of $f < f_{\max}$, the molecules forming zigzag walls undergo nonlinear, noncollective excitations. These excitations perturb motions of molecules arranging neighboring chevron layers and can destabilize borders between defect walls and the folded layers leading, in consequence, to displacements of the walls or their borders.

The zigzag walls can undergo field-induced motions of two kinds. The first kind, called as creep, consists in drift motions of defect borders over small length scales.[8,11] Such creep motions can be considered as being localized in very small system regions.[20] These motions of defect borders are activated above threshold voltage U_c, dependent on f. Then, the critical line $U_c(f)$, defined as a projection of the edge line (see Fig. 2) onto the U-f plane, separates relaxation and creep dynamic modes. As demonstrated in Fig. 4, this line can be described by a shifted power-law relation $U_c(f) = c_0 + c_1 f^{\alpha}$, where c_0, c_1, and α are independent of f. The exponent α has been found to be $\alpha = 0.5 \pm 0.05$ for samples differing in the thickness and defect distribution. This suggests the universality of the exponent for chevron SSFLC. The second kind of motions of zigzag defects, known as sliding, consists in the propagation of the defect walls on very large length scales.[20] The sliding movements are activated at high voltages, generally much larger than the critical value $U_c(f_{\max})$.

The polydispersive character of frequency spectra of the dielectric permittivity of the defected chevron samples, already visible in the contour map of ε'', manifests itself in the Cole-Cole plots drawn in Fig. 5 for different voltages $U < U_c(f_{\max})$. These plots exhibit two distinctive features: a translocation of the relatively low-frequency subspectrum, corresponding to field-induced dynamics of defect walls, to-

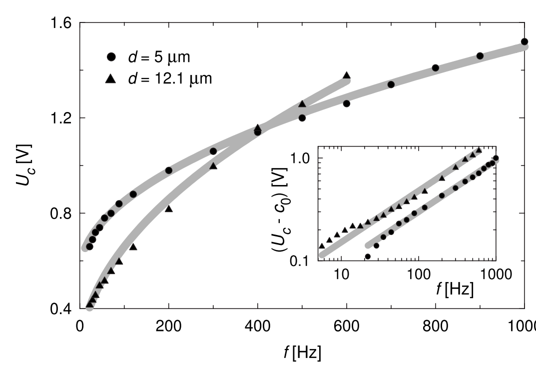

FIG. 4. The f dependence of U_c. The filled circles and triangles represent experimental data obtained for cells of the thicknesses $d=5$ μm and $d=12.1$ μm, respectively, while the thick lines are plots of the function $U_c(f)$, with the best fitted values of c_0, c_1, and the index $\alpha=0.5$. The inset shows the log-log plots of the subtracted function $U_c - c_0$.

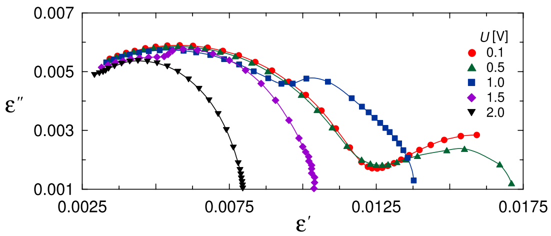

FIG. 5. (Color online) The Cole-Cole plots experimentally obtained with different voltage amplitudes for the same sample, for which data of Figs. 2 and 3 were found.

ward higher-frequency subspectrum, connected with collective excitations of molecules in chevroned system areas, for large enough growing U but less than $U_c(f_{\max})$, and the disappearance of this subspectrum at rather high voltages $U > U_c(f_{\max})$. Both the effects give an evidence of a strong nonlinearity of molecular motions within the defect walls. However, the rapid vanishing of the lower-frequency subspectrum in the regime of high U is a manifestation of a large randomness and thereby a large noncollectivity of molecular reorientations. In fact, random, to a large extent, rotational motions of molecules cannot yield very significant contributions to the electro-optic response.[21] The absence of a low-frequency and ultralow-frequency branch of the Cole-Cole plots shown in Fig. 4 is in sharp contrast to respective results obtained for various solid-state multidomain systems.[3,8,9,22–24] In all these systems, thermally activated creep motions are caused by polarization or magnetization switching at interfaces between domains. Such local switching processes can essentially occur at arbitrary low field frequencies. As the switching processes, and thereby the resulting creep motions, have generally a cooperative character, they yield considerable contributions to the frequency response functions. On the contrary, creep motions in chevron SSFLC are not associated with switching processes but are provoked by noncollective molecular rotations of relatively small amplitudes.

The relaxation-to-creep transition in the chevron SSFLC can also be investigated by analyzing the ferroelectric hysteresis loop. In particular, one would expect that the appearance of creep displacements of zigzag walls under the oscillating field $E(t)=(U/d)\sin(2\pi ft)$ will affect the hysteresis-loop area, given by[25] $A=(2\pi fU/d)\oint P(t)\cos(2\pi ft)dt$, where $P(t)$ denotes the polarization of a light illuminated subsystem. In general, this quantity can take nonzero values for both relaxation and creep regimes. Nevertheless, the functional dependence of the hysteresis-loop area on U and/or f can be expected to change abruptly at $U=U_c(f)$, where $f < f_{\max}$. Such a behavior of A is really observed in the case of the U dependence, as illustrated in Fig. 6. The positions of cusps visible in multiple plots in this figure agree very well with values determined by $U_c(f)$ for respective voltage frequencies. The effect of the transition between relaxation and creep dynamic phases on the frequency dependence of A is less distinct. It proves that A, considered as a function of f, possesses a local maximum at $f=U_c^{-1}(U)$, provided that $U < U_c(f_{\max})$. However, each of the local maxima appearing for different U is very smeared, as shown in Fig. 7 for

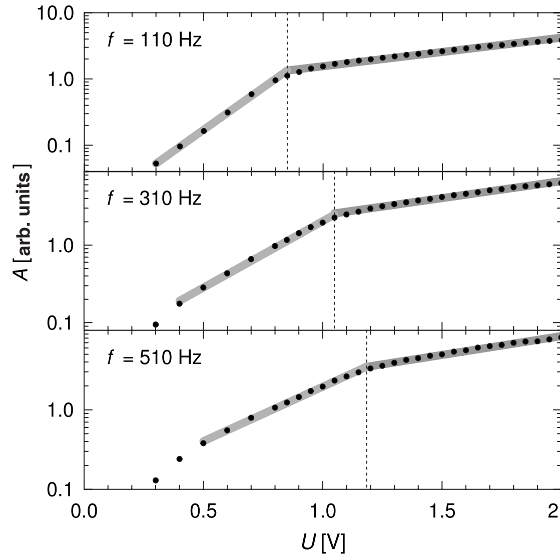

FIG. 6. Dependence of the area of the dynamic hysteresis loop on the voltage amplitude for different frequencies. The solid lines represent exponential functions fitted to experimental data (filled circles), separately for ranges of U below and above cusp positions (indicated by dashed lines).

$U=0.5$ and 1.1 V [note that the third plot in this figure is drawn for $U=1.8 > U_c(f_{\max})$].

III. DIRECT OBSERVATIONS OF CREEP MOTIONS

The field-induced bookshelf-to-chevron (or conversely) transformations of smectic layers, occurring at borders of zigzag walls, give rise to changes of color and intensity of light passing through the studied cells. For sufficiently large voltage amplitudes, these transformations can propagate in macroscopically large length scales, still remaining in the creep regime. Then, the resulting creep motions of defect walls can easily be observed with a standard optical microscope. A typical such motion of a defect wall is shown in Fig. 8, where microphotographs of the thick wall being a fragment of a closed-loop defect are presented for the voltage frequency $f=600$ Hz and the amplitudes $U=4.5$ V and $U=5.5$ V. These micrographs show that the motion of the

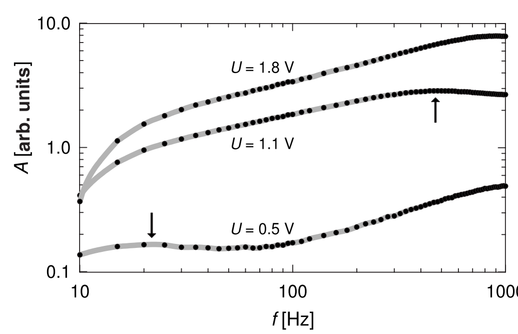

FIG. 7. Frequency dependence of the area of the dynamic hysteresis loop for different voltage amplitudes. The filled circles correspond to measurement data. The arrows indicate maxima connected with relaxation-to-creep transitions.

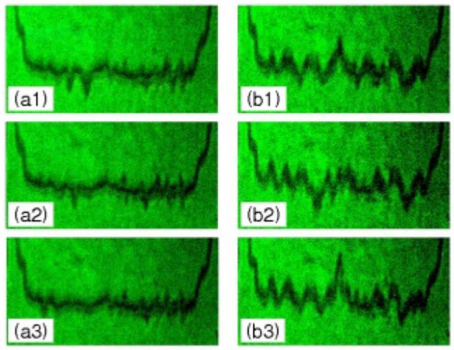

FIG. 8. (Color online) Microphotographs of a zigzag wall taken at time distances about 1 s [(a1)–(a3)] for $f=600$ Hz and $U=4.5$ V [(b1)–(b3)] and for $U=5.5$ V.

thick wall is pinned by more stable thin walls (seen at the left and right ends of the thick wall) and that the motion is confined to a small region of the sample.[26] It is seen that temporary walls borders are rougher at higher than lower voltage amplitudes. However, the degree of instantaneous roughness of thick walls as well as the extent of their displacements depend, in general, not only on the amplitude but also on frequency of applied voltage. At sufficiently large U and appropriate f, the thick walls transform into thin zigzag walls, which then proceed to slide and eventually decay.[20] The slidings of walls are not confined to small regions of a system, but can propagate in large length scales, even in the length scale of the whole system. Since threshold values of U and f, at which the sliding motions of defect walls appear, depend on the shape and size of the walls, the transition between creep and sliding dynamic regimes is very blurred, contrary to the relaxation-to-creep transition.

IV. CONCLUDING REMARKS

In conclusion, a new scenario of the dynamic transition of defect walls from relaxation to creep state under the influence of an applied alternating field has been proposed. Due to specific properties of SSFLC of the chevron structure, the considered creep dynamics of defect walls considerably differs from that being typical for defects occurring in solid-state systems. More complete description of creep motions of zigzag walls needs further research, first of all, needs an explanation of the role of temperature in activating the creep and sliding motions. An important question is whether dynamic phase transitions in chevron liquid crystals have universal character, similar to the transition between dynamic modes of domain walls in solid state systems.[27]

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[26] Movie showing creep motions of a thick zigzag wall in a sample of the thickness $d=5$ μm, at a fixed frequency and different amplitudes of an applied voltage, is available at http://www.ifmpan.poznan.pl/zp6/film.htm.
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