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Magnetic field-induced instabilities in cholesteric liquid crystals: periodic deformations of the Grandjean texture

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The effect of a magnetic field applied perpendicular to the helical axis of a cholesteric liquid crystal mixture with positive diamagnetic anisotropy has been studied by laser diffraction and microscopy. A rapid decrease of the field below a threshold H_s generates first a metastable periodically striated texture. A further decrease of the field below a second threshold H_0 transforms this into a metastable square grid texture. These periodic textures gradually convert into a stable planar texture. The periodic deformation structures can be explained by a simple model based on the hindered relaxation of the partially unwound cholesteric helix under certain boundary constraints.

1. Introduction

If a magnetic field is applied to a non-aligned cholesteric liquid crystal mixture with positive diamagnetic anisotropy, an alignment of the helical axes perpendicular to the field takes place. On increasing the field strength the helical structure unwinds. At a certain critical field H_c the pitch becomes infinite, i.e., a cholesteric to nematic transition occurs [1–3]. We must distinguish between two possibilities for unwinding the helix. (*a*) For a free helical structure which is not affected by any boundary constraints, the pitch increases continuously with increasing field (see figure 1, curve A) [4]. (*b*) If the helix is fixed by means of molecular anchoring to the substrates, the pitch increases stepwise with increasing field (see figure 1, curve B), since in this case the pitch can have only discrete values (see figure 2). This stepwise unwinding of the helix, predicted by Dreher [5], was first observed in a non-uniform electric field by Belyaev and Blinov [6].

2. Experimental

A mixture of nematogenic 4-cyano-4′-*n*-heptylbicyclohexyl (CCH7) and 4-cyano-4′-*n*-heptylbiphenyl (CB7) doped with chiral cholesterylpropionate (CP) was used. A mixture of 82·4 mol % CB7, 16·3 mol % CCH7 and 1·3 mol % CP exhibits a positive diamagnetic anisotropy ($\Delta\chi > 0$) and a pitch of about 7 μm. A planar cholesteric texture (Grandjean texture) was obtained by rubbing the substrates and compressing the filled cell. The sample was placed between the poles of an electromagnet (Drusch, EAF 30 E) with the field direction perpendicular to the helical axis ($\mathbf{p} \perp \mathbf{H}$).

The effect of the magnetic field on the liquid crystal was studied by laser diffraction with a He–Ne laser (Spectra Physics). For visual observation of the sample a microscope with a phototube but with no polarizers was inserted between the magnetic poles. The sample was placed in a thermally insulated copper chamber and heated with circulating silicone oil.

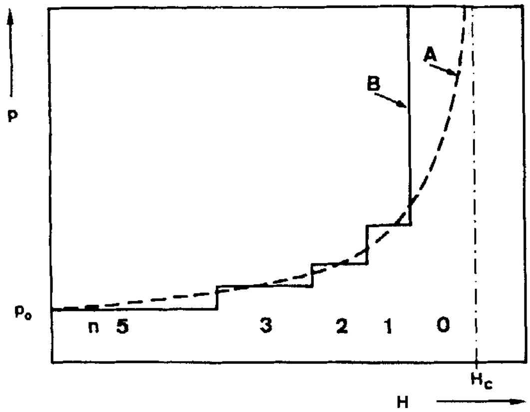

Figure 1. Dependence of the helical pitch p on the magnetic field H. (A) Without boundary constraints and (B) with boundary constraints (n is the number of half-pitches).

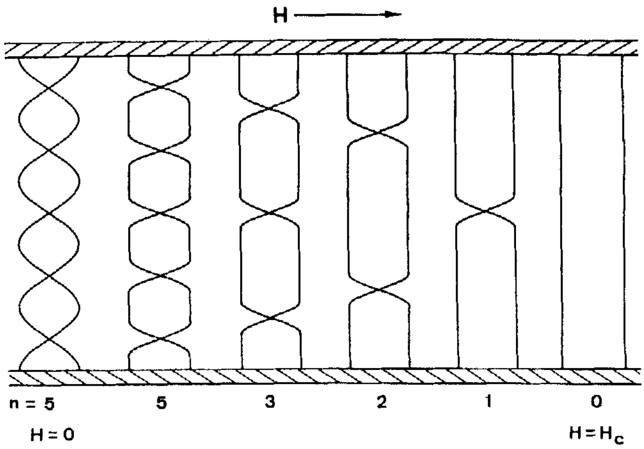

Figure 2. Sketch of the unwinding of a boundary fixed cholesteric helix by a magnetic field (n is the number of half-pitches).

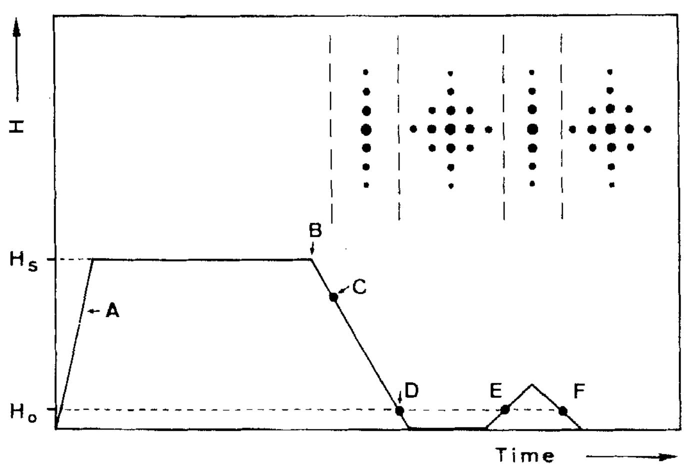

Figure 3. Schematic illustration of the appearance and transformation of a laser diffraction pattern in a magnetic field.

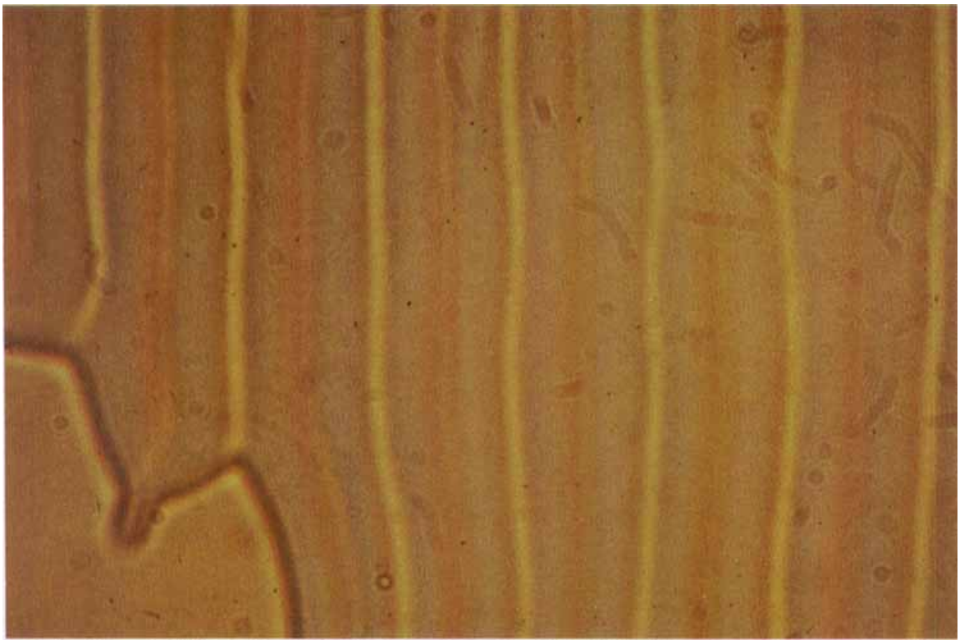

Figure 4. Microphotograph of the periodically striated texture ($H_0 < H < H_s$).

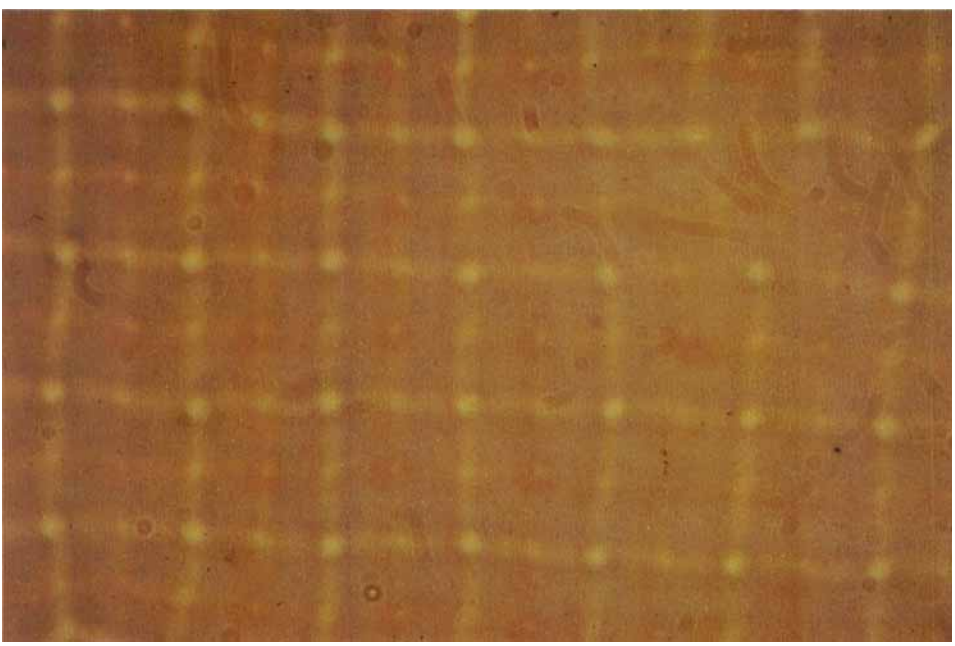

Figure 5. Microphotograph of the square grid texture ($0 < H < H_0$).

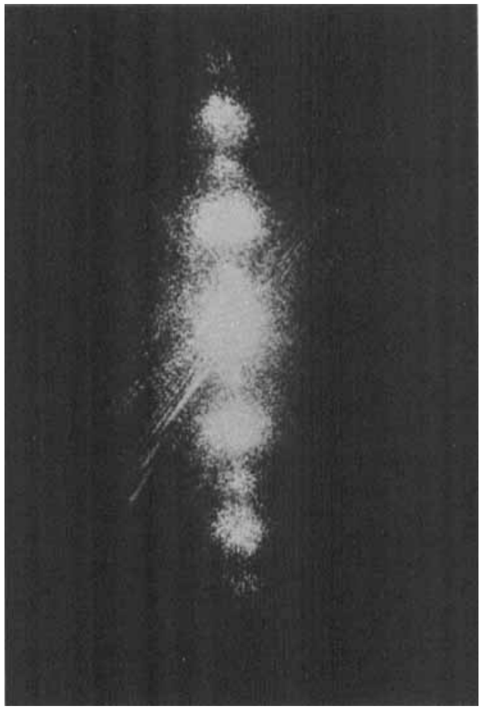

Figure 6. One dimensional laser diffraction pattern with incidence parallel to the helical axis ($H_0 < H < H_s$).

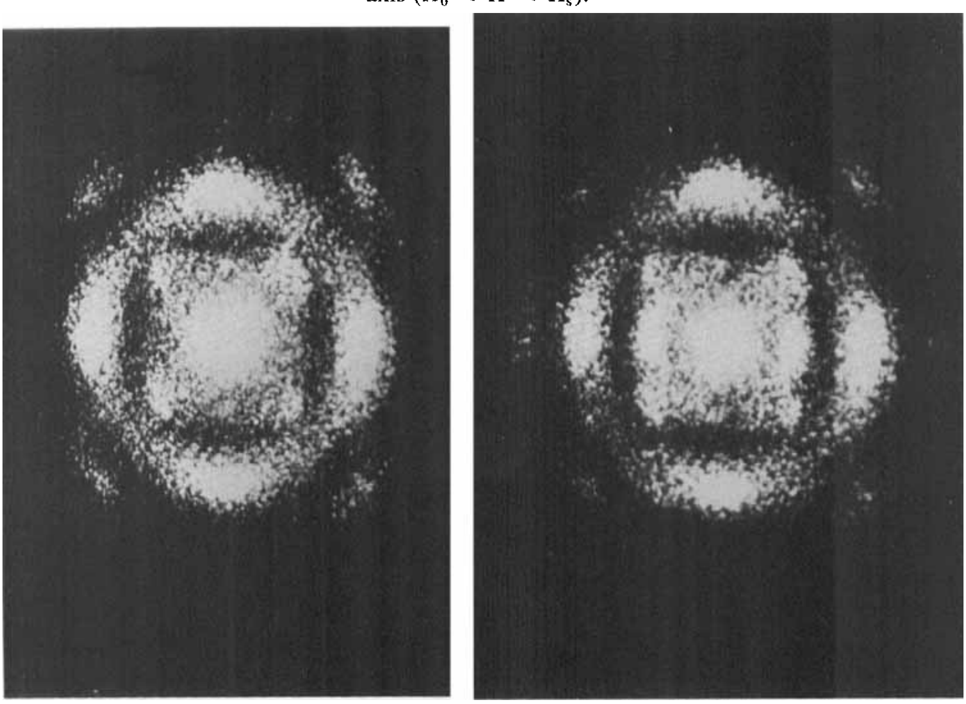

Figure 7. (*a*) Two dimensional laser diffraction pattern with parallel incidence ($H = 0$). The intensity of the diffraction spots of even order are higher than those of odd order. (*b*) Two dimensional laser diffraction pattern with oblique incidence ($H = 0$). The diffraction spots in the plane parallel to the field direction appear with as high intensity as the even order ones.

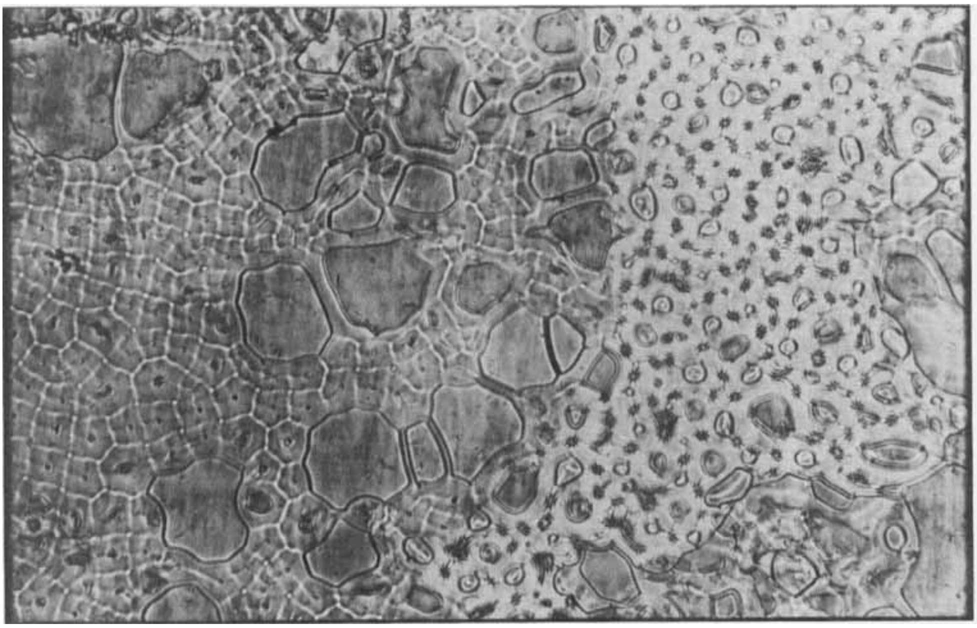

Figure 8. The metastable textures transform into a stable Grandjean texture in time; some regions first transform into a fingerprint texture and then into a planar texture with remaining line defects.

3. Results

The magnetic field, applied perpendicular to the helical axis of a planar texture, exerts a torque which tends to align the director parallel to the field direction if $\Delta\chi > 0$. Thus, the helical pitch increases with increasing field strength. This unwinding of the spiral is not detectable by laser diffraction or microscopy, because the observation direction is parallel to the helical axis. However, with increasing field up to a field strength H_s ($\approx 2{\cdot}3$ T) the appearance and divergence of disclination lines in the Grandjean texture was observed microscopically (see figure 3, point A). These disclination lines separate regions with different numbers of half-pitches n, due to the partial unwinding of the helix by the magnetic field.

After keeping the sample for some time above the threshold H_s the field was decreased rapidly to a lower strength H ($H_s > H > H_0$). As a result, a metastable striated texture appears (see figures 3 (point C) and 4). The striations, alternately light and dark, run parallel to the field direction and the sample surface. This structure produces a one dimensional laser diffraction pattern with spots of alternating intensity (see figure 6). Further decrease of the field below H_0 ($\approx 0{\cdot}3$ T) causes a texture with two interlaced square grids to appear, resulting in a two dimensional laser diffraction pattern (see figures 3, 5 and 7). Increasing the field above H_0 the two dimensional pattern transforms into the one dimensional one again (see figure 3, point E). By repeatedly decreasing the field the two dimensional pattern reappears with a lower intensity (see figure 3, point F). The metastable periodic textures gradually transform into the stable Grandjean texture (see figure 8).

With parallel incidence of the laser beam to the helical axis, the diffraction spots of odd order ($m = 1, 3, \ldots$) are either of weaker intensity than those of even order ($m = 2, 4, \ldots$) or absent (see figures 6, 7(a) and 10). The separation of the

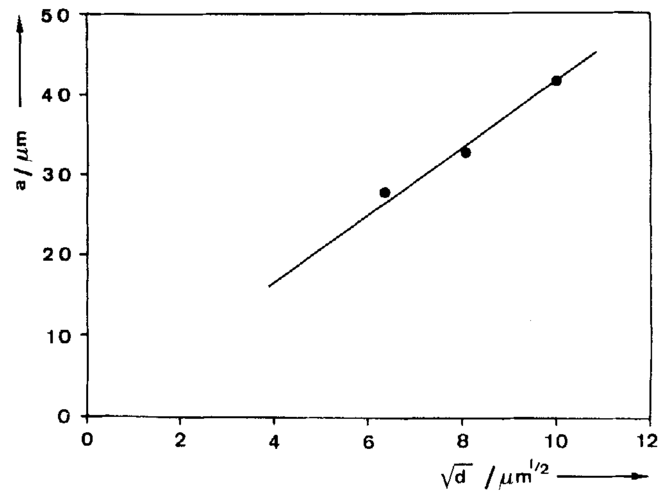

Figure 9. Lattice constant *a* of the grid versus the square root of the sample thickness *d* (*a* is the distance between two equal points of the periodic structure).

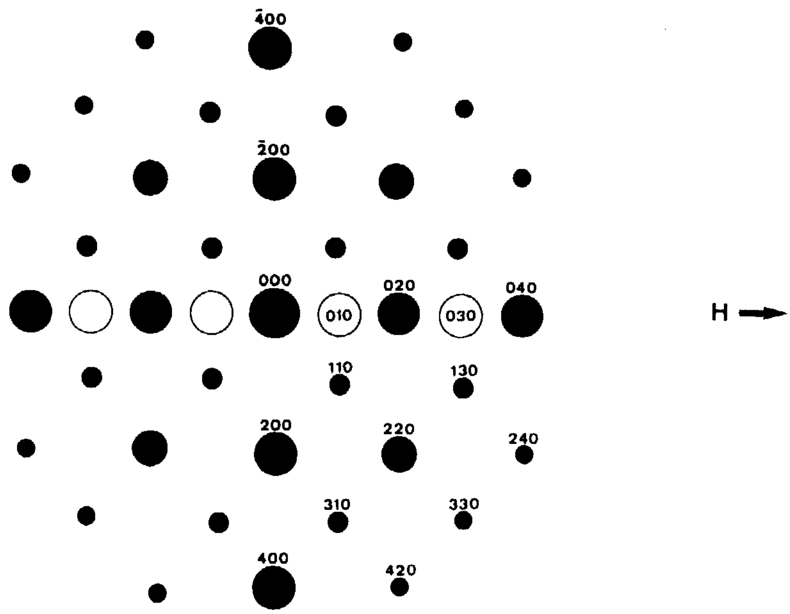

Figure 10. Sketch of the laser diffraction pattern with the associated Miller indices ($H = 0$). ●, Incidence parallel to the helix axis; ○, additional diffraction spots obtained by oblique incidence.

deformation lines has also been measured with the microscope. It turns out that in the absence of the odd order diffraction spots the periodicity of the even order ones (see figure 10) corresponds to the half-distance of two equal deformation lines ($a/2$) of the periodic structure in figures 4 and 5.

At the oblique incidence, achieved by a small rotation of the sample around the axis orthogonal to the field and laser direction, the odd order diffraction spots in the plane parallel to the field direction appear with an intensity as high as the even order ones (see figures 7 (*b*) and 10). The periodicity of the diffraction pattern corresponds to the distance between two equal deformation lines (*a*) of the striated as well as the square grid texture.

To obtain further information about the metastable periodic structures, the dependence of the spatial periodicity *a* on the sample thickness *d* has been investigated. As shown in figure 9, the periodicity *a* is proportional to the square root of the sample thickness *d*. This relation is in agreement with the behaviour of a square grid pattern induced in cholesterics with $\Delta\chi > 0$ by a magnetic field applied *parallel* to the helical axis. For this case it has been calculated that $a \approx (p_0 d)^{1/2}$ [7, 8]. This result indicates that the origin of the periodic structures is not an intrinsic property of the system, but is due to elastic moments affected by boundary constraints.

4. Discussion

4.1. *Origin and model of the periodic instabilities*

Rapid decrease of the magnetic field causes the cholesteric helix which is partially unwound between the anchoring substrates to relax to the initial number of half-pitches. This relaxation is hindered by the boundary constraints. As a result, the excess twist free energy causes a mechanical strain, resulting in an additional periodic deformation of the director field perpendicular to the optic axis. The director rotates helicoidally around one or two axes inclined to the optic axis. This is possible by a progressive tilt of the molecules with respect to these axes, yielding in an undulation of the quasi-nematic planes of cholesteric structure the so-called cholesteric planes (cf. figures 11 and 12) [9].

In the field the wave-like deformation of the cholesteric planes runs perpendicular to the field direction and generates the striated structure. Thus, the striated structure is the result of the competition between the field-induced torque and the elastic moments. Without an external field as well as at lower field strength $H < H_0$, an additional undulation parallel to the field direction appears which generates the square grid structure. This deformation structure is due to the action of only the elastic moments.

The one and two dimensional undulations of the cholesteric planes are illustrated schematically in figures 11 and 12. The two dimensional undulation and the helical twist generates a tetragonal lattice structure ($a = b \neq c$) in which the periodicity in the *c* direction is determined by the helical pitch (cf. figure 12 (*a*)). The undulation causes a periodic variation of the refractive index, leading to a focussing effect which becomes visible as striations or a square grid, respectively.

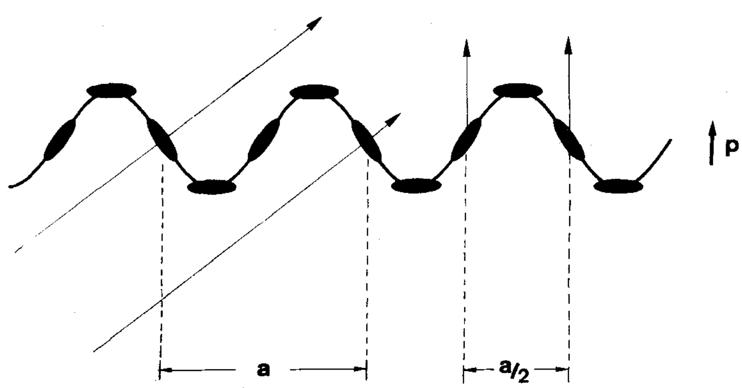

Figure 11. Wave-like modulation of a cholesteric plane with parallel incidence: periodicity = $a/2$ and oblique incidence: periodicity = a.

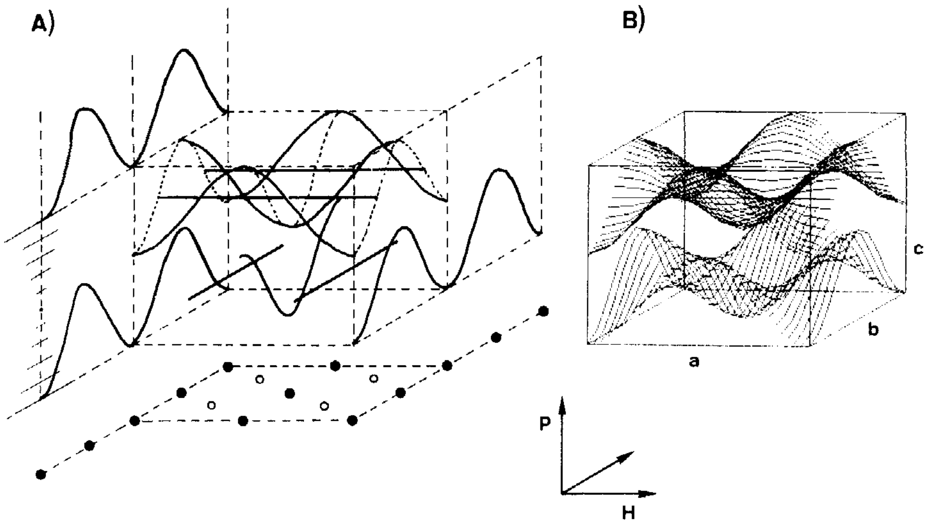

Figure 12. Two dimensional undulation of cholesteric planes: (*a*) schematic illustration of the deformed helical structure and (*b*) computer graph of the deformed structure (by courtesy of Mr F. Porsch).

The simple model can also explain the different intensities of the diffraction spots with parallel and oblique incidence with respect to the helical axis **p**, sketched in figure 11. At parallel incidence to the helical axis and perpendicular to the undulation planes the periodicity is $a/2$. With oblique incidence, however, the double periodicity a occurs.

An important argument for this distortion model is that it can be derived directly from the model of the partially unwound as well as from the unperturbated cholesteric helix (see figure 2).

4.2. *Comparison with other experiments*

If **H** is applied *parallel* to the helical axis of a planar texture, static, periodically striated as well as square grid structures have been observed. At higher field strengths a 90° rotation of the helical axis and the cholesteric to nematic transition occur [9–11]. These well-known instabilities only exist in static fields and are quite different from the metastable pattern which develops after a rapid decrease of the field as described here.

Similar striated and grid-like patterns have been obtained in a planar cholesteric between two-rubbed substrates by mechanical untwisting of the helix around the helical axis as well as by a temperature induced pitch decrease [9, 12, 13]. Rapidly decreasing a non-uniform electric field also produces spatially modulated structures by relaxation of an unwound cholesteric structure under boundary constraints [6, 14]. In this case the striations are parallel and/or perpendicular to the rubbing direction. In the experiments described here, however, the striations run parallel and/or perpendicular to the field direction. The physical nature of the periodic structures generated by mechanical untwisting, temperature variation and relaxation of an unwound helix in an electric field seems to be comparable with those of the periodic instabilities described in § 3. Therefore, it can be concluded that the origin of metastable striated and square grid patterns is caused by elastic deformation of the cholesteric structure during the relaxation of a partially unwound helix to a shorter pitch under boundary constraints.

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