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Total Internal Reflection—A Method for Determining of LC Anchoring and Orientation on Various Substrates

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The general problems being important for the determination of the liquid crystal surface anchoring and liquid crystal surface orientation of nematics, large-pitch cholesterics, and some smectics by means of attenuated total internal reflection (ATIR) of a visible light beam have been outlined. The advantages and disadvantages of ATIR for measuring of the surface energy have been discussed and compared with those of other well-known methods. The possible influence of additional orienting and/or conducting layers on the reflectivity and polarization of the incident wave are also briefly discussed.

I. INTRODUCTION

The liquid crystal orientation (LCO), either at the substrate or in the depth of a deformed nematic (N), was determined by means of attenuated (or frustrated) total internal reflection (ATIR) of a visible light beam for the first time in 1934 by Fréedericksz and Zwetkoff.[1] For nearly 40 years,[2] however, this useful method for investigation of various surface and bulk LC realignments has been forgotten. The revival of this idea began with the construction of a number of optical means based on the ATIR, such as light deflectors,[3,4] optical waveguides for modulation and switching of the light,[5,6] thin NLC film waveguides for optical deflection,[7] and field-realigned NLC optical waveguides.[8] On the other hand, during the past eight years the ATIR has been also utilized for studying of the LC realignment near variously-treated solid substrates closely related with the surface anchoring[9-15] and for determining of the optical indices of various Ns.[15,16] Recently, the ATIR of nonpolarized light has been utilized for the construction of LC elements for LC fiber-optic switch.[17-20] The ATIR method can be successfully applied for measuring of maximal LC deformations caused either by electric, magnetic or elastic forces,[10,21,22] or by fluid flow.

The aim of this paper is to outline the general problems being important for determining the liquid crystal surface anchoring (LCSA) and liquid crystal surface orientation (LCSO) on various substrates by ATIR, including the elucidation of the possible advantages and disadvantages of this method for measuring of the LCSA in comparison with other well-known methods, the estimation of the possible influence of additional orienting and/or conductive layers on the reflectivity and polarization of the incident wave, the evaluation of the range of the applicability of the ATIR for measuring of different LCSA and LCSO as follows:

a) θ-polar SO and SA in various monotone and non-monotone deformed N layers;

b) φ-azimuthal SO and SA in purely-twisted Ns or cholesterics (Chs) with a large pitch which should be comparable to the LC thickness;

c) θ, φ -polar, azimuthal SO and SA in twisted N cells or large-pitch Chs.

Finally, the possible application of the ATIR method for measuring of either LC material parameters or inclination of the molecules in large-area Sm A or Sm C monodomains has been briefly mentioned. Simultaneously with these important questions, a number of problems which are of interest, such as the penetration of the evanescent wave in the LC medium, the depolarization of the incident wave, the influence of the thermal fluctuations, etc., have been emphasized.

a. Surface energy, treatment of confining the LC glass plates, and possible ways for measuring the strength of the LC surface anchoring

It is well-known that all the Ns and large-pitch Chs can be easily influenced by the various treatment of the glass plates confining the LC. In the last few years, one notes interest for obtaining large-area Sm A and Sm C monodomains.[22-28] In this case, however, not only the surface treatment and the substrate-LC interactions are important, but also the type of the phase transition, i.e. N–Sm A, N–Sm C, N–Sm A–Sm C, etc., the possible ways for the smectic formation, i.e. under cooling or heating, and the apparent need from the application of orienting ac electric or magnetic fields with sufficient strength.

The surface orientation and anchoring of the LC are of great importance for the main characteristics of the LC electro-optical devices. It is well-known HOW it is possible to obtain weak or strong anchoring, planar, tilted, or homeotropic orientation of the LC being closely related with the complex simultaneous action of physico-chemical, elastic, flexoelectric, and other forces. For our aim, it is important to point out only the POSSIBLE WAYS for measuring the strength of the surface anchoring of the LCs under consideration, and to compare the possibilities of these methods with those of the ATIR method. Further, we shall be interested only in thin (10–50 microns) LC cells and shall utilize the convenient extrapolation length $b = K/W_s$ introduced by de Gennes, where K is the mean elastic constant, and W_s is the surface stiffness constant.[22,29]

We must emphasize the following three main cases:

a) $b > d$, where d is the LC (or cell) thickness.

The measurement of the surface energy in this case of very weak surface anchoring is convenient by the estimation of the width of Bloch or Néel walls existing in such LC films.[30,31,32] The ATIR method is not convenient since it gives the LCSA and LCSO by a Z (i.e. perpendicular to the glass plates) penetration of the evanescent wave (it is clear that the LC orientation is changed only in the X,Y plane, i.e. in the plane of the glass plates). On the other hand, the inclination of the LC molecules in the very Bloch or Néel walls now is impossible to measure by ATIR due to technical causes.

b) $b \sim d$.

The measurement of the LCSA and LCSO can be performed successfully in this case, either by a measurement of the width of the various surface disclinations,[30,33,34,35] or by a utilization of the ATIR method. It should be noted, however, that the utilization of the surface disclinations is more convenient since first: this method can be applied for LC cells being worked as LC indicators or elements. This is not possible for the ATIR method, with one exception of ATIR LC display devices requiring the utilization of high-optical index flint or other glass prisms, and second: the application of the ATIR method is embarrassed for high-temperature Ns.[10] However, the ATIR method is more simple and can give repeatable LCSA and LCSO values. Furthermore, the comparison of the eventual values of the surface energy and orientation being obtained by these two separate methods is of apparent interest. Of course, let us note that the well-known Fréedericksz transition, very well studied both theoretically and experimentally, can be also applied for the measurement of the surface energy in this case. However, this method is integral, depending strongly on the homogeneity of the LC orientation, and providing for good knowledge for all the LC constants as well as for the possible influence of the impurity content, the gradient in the electric, magnetic or other fields, the existence of bulk and surface defects, etc.

c) $b < d$.

In this case of a relatively strong anchoring of the LC, the ATIR method is very useful and has much more advantages relative to the Fréedericksz one; however, it must be applied only for those cases when the ATIR measurements of the reflectivity and/or transmissivity of the incident light can give the correct answer for the value of the surface anchoring and orientation. There are a number of cases when the interpretation of the experimental results is either very cumbersome or is not completely investigated.[10] Furthermore, the LC orientation in the depth of the layer is possible to be investigated by the change of the incident angle which is not possible by any other means for investigating the LC deformations.

b. Possible influence of the surface orienting and conductive layers on the reflectivity and polarization of the incident light

In this part of the paper we shall consider the possible influence of the inhomogeneities of thin SiO or other oxide, metal films deposited under vacuum evaporation on the glass plates[23,29,36–40] or additionally treated by a monomolecular surfactant layer[41,42] as well as the role of the regular or nonregular grooves made by rubbing or by other technique on the glass plates[43–48] on the reflectivity and polarization of the incident light.[49] It is easy to understand that one should expect scattering and depolarization of the incident light.[49,51] According to Al'pert *et al*,[50] this problem can be successfully solved for the sake of the WEAKENING function $W(X,Y,Z)$ multiplying the non-perturbed electric field components for one completely-smooth surface. This problem is not solved for the case of the surfaces utilized in the LC device technique. It is easier for the case of rubbed surfaces when the period of the grooves is in the range of the incident wave-length. For instance, the formulae obtained by Al'pert *et al*[50] clearly point out that the depolarization of the incident light is absent for the case of one-dimensional grooves, i.e. when $Z = Z(X)$ and Z,X is the incident plane. For the other one-dimensional case $Z = Z(Y)$, as well as for the two-dimensional case $Z = Z(X,Y)$, one notes obvious depolarization of the incident light. In our opinion, this problem can be solved more easily by convenient experimental measurements such as the total internal reflection microscopy measurements utilized by Temple[52] (let us note that this method for the case of LCs has been utilized for the first time by Rivière).[53] It is clear that the scattering due to the roughnesses of the additional orienting and conducting, very thin films should be added to that due to the divergence of the laser beam (or light), the thermal fluctuations, and the static surface defects.[10] On the other hand, the depolarization effects might be more important for the correct performance of the ATIR measurements.

Other thin films which can be of significance for the determination of the ATIR conditions are the conductive ones. These films, as well as the orienting, usually have been neglected due to their small thickness relative to the wave-length. Indeed, the obtaining of the ATIR components of the electric field is not difficult when the very well-known (to date) matrix method (nearly all the papers dealing with this question have been cited in Ref. 10) is used.[14,54] However, the physical interpretation of the final results is difficult. We only know that the conductive layers are of importance for the transmission or the reflection of the incident wave for the cases when their optical index is well above the optical indices of the LCs.[55,56] The analytical solution of these cases would give formulae being able to show one eventual change in the ATIR conditions due to the inclusion of additional thin orienting and/or conductive layers.

c. Possible application of the ATIR method for determining of the surface orientation and anchoring of nematics, large-pitch cholesterics and some of the smectics

Let us estimate the possibilities of the ATIR method for obtaining of LCSA and LCSO of differently-realigned Ns, large-pitch Chs and some of the smectics.

a) The ATIR experimental set-up.

The experimental set-up for the realization of ATIR measurements is well-known.[10,12,16] It consists of a measuring cell including a high-optical index ($n > 1{,}9$) glass prism coated with a number of conductive orienting, etc.; very thin films from several-tens to several-hundreds Å thick; followed by LC with different thickness (usually below several tens of microns) and a second high-optical index or usual glass plate coated with the same additional films (Figure 1).

The NLC can be deformed along Z (i.e. perpendicular to the glass plates) as follows:

a) a monotone θ-polar deformation being changed either from homeotropic or tilted to planar, or from planar or tilted to homeotropic;[57]

b) a θ-polar deformation with a maximum being in the depth of the LC layer which is changed either from homeotropic or tilted at the boundaries to planar in the middle part of the LC cell, or from planar or tilted at the boundaries to homeotropic in the middle part of the layer.[58,59]

Note that cases a) and b) can be complicated by an additional φ-azimuthal deformations being characteristic for twisted nematic (TN) layers[60]

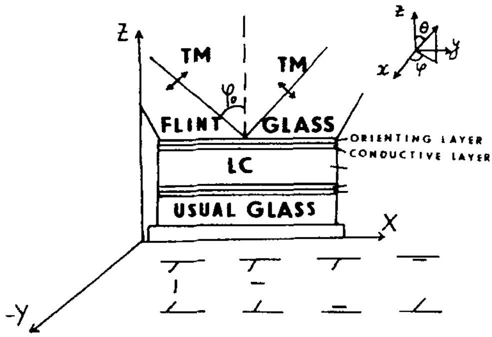

FIGURE 1 Schematic representation of the attenuated total internal reflection measurable cell, the various LC θ deformations and the incident light beam.

or for large-pitch Chs.[61] Furthermore, the LC can be large-area Sm A or Sm C monodomains with well-defined orientation of the molecules.[22–27]

The performance of the ATIR measurements provides for a light source (usually a good-quality laser), and the light has to be directed towards the interface flint glass-prism additional orienting or conducting layers — LC at oblique incidence (Figure 1). It is preferable to use a TM wave with p polarization, or a TE wave with s polarization since the performance and the eventual interpretation of ATIR of coupled TM-TE modes would be very complicated.

The combination of this standard ATIR set-up with a usual microscope gives the very promising ATIR microscopy[52,53] (see Figure 2). The advantages of this method are mainly in the possibilities simultaneously to perform the ATIR measurements and to observe the detailed LC pictures under the microscope. It is possible in this way to overcome a number of difficulties related to the nonhomogeneity in the LC orientation (domains, defects, etc.) being in the laser spot region. On the other hand, the observer may usually determine very exactly the beginning of the total reflection of the light.

b) ATIR method for determining of the LC anchoring and orientation of θ-polar non-deformed and deformed nematics.

The ATIR method may give exactly the LCSA and LCSO of all the non-deformed planar, homeotropic and tilted LC layers, and of all θ-deformed N layers when the orientation is changed from homeotropic or tilted to planar. This fact is due to the behavior of the evanescent wave being attenuated very near (in the range of one wave-length) to the interface glass-prism — LC owing to the validity of the surface ATIR condition for the entire LC layer. The measurement of the surface anchoring parameter

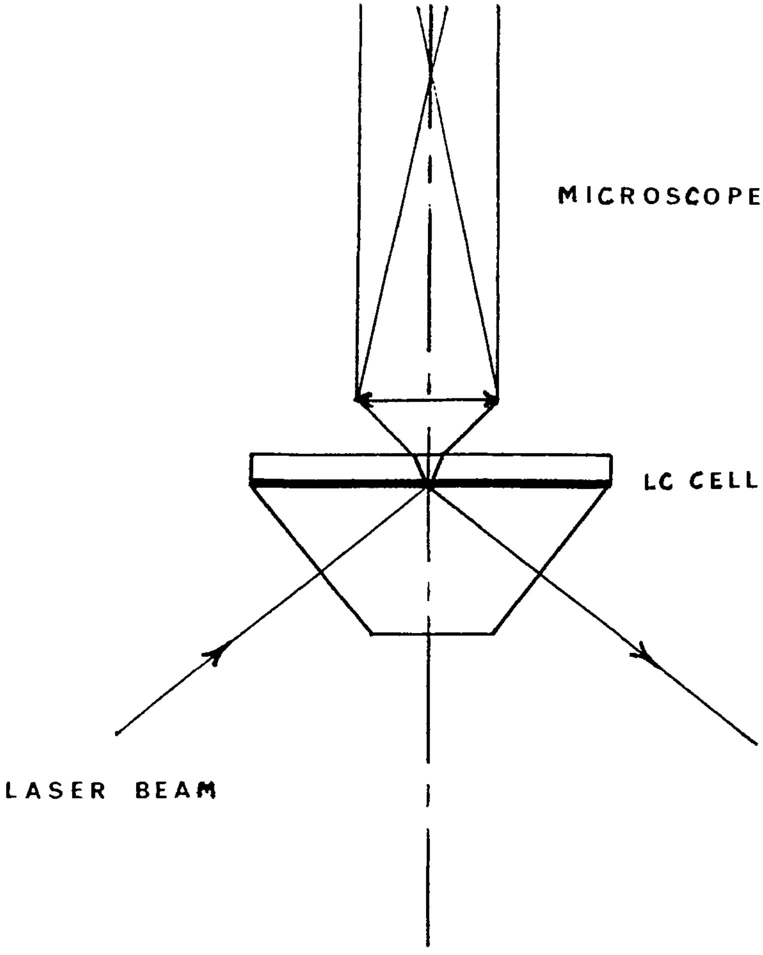

FIGURE 2 ATR Microscopy.

is possible by two or more values of the LCSO (expressed by the surface deformation angles θ_s) which might be obtained either for different LC thicknesses of the layers under investigation or for different external forces (electric or magnetic fields, flow, etc.) applied across the LC cells.[10,22,62] Another, much more complex case, is when the LC orientation is changed from planar or tilted to homeotropic. The difficulties arise mainly from the fact that the ATIR condition might be satisfied at the boundary (or in one intermediate LC layer with thickness depending on the incident angle, the optical indices and the strength of the LC deformations, etc) followed by a region where this condition is not satisfied. It is easy to understand that the solution of this case depend on many conditions. For example, we can point out the large discrepancy between the experimental and theoretical values of the surface angle of such a LC layer obtained by Sprokel *et al*[14] and attempted to be explained by appropriate weak anchoring of the LC layer.[63] Accordingly, there are several ways for overcoming the difficulties connected with this special case of LC variation, which are as follows:

b1) The first one can be successfully applied for thin (below several tens microns) LC layers with symmetrical deformations having a maximum in the middle of the cells (the asymmetrical case, although more complicated, can be also resolved by investigating two symmetrical LC cells with LC anchoring corresponding to either of the two different ones). It consists of the utilization of the following crude relation (when the light penetrates just to the middle of the cells):[21,64]

$$N^2 \sin^2\varphi_0 = n_e^2 \sin^2\theta_m + n_0^2\cos^2\theta_m$$

where N is the glass-prism optical index, n_0 and n_e are the principal LC optical indices, φ_0 is the incident angle, and θ_m is the maximal LC deformation; for determining the maximal deformation after ATIR from the entire LC cell. The variation of the maximal angle with the external forces can simply give the strength of the surface anchoring.[62] On the other hand, the well-known relations between the strength of the surface anchoring, the surface and maximal deformation can be utilized for obtaining LCSO.[59,62]

b2) The second way can be applied for thick cells with LC deformations possessing also maximum by utilizing the attenuation of the incident wave in the well-known de Gennes' coherent LC length. The determination of LCSA and LCSO in this case is more complex and to date is unresolved.

b3) The third way is to offer an appropriate qualitative interpretation of the eventual changes in the transmission or reflection of the incident light after ATIR from the first several molecular layers. This case is also complex and non-resolved.

The cases of monotone deformations can be solved more easily by construction and ATIR investigation of asymmetrically weak-strong (homeotropic-planar or tilted-planar) or strong-weak (homeotropic-tilted) LC layers by appropriate treatment of the glass plates with lecithin, soap and other surfactants.

Another important problem is the measurement of the LCSA and LCSO of tilted N layers. The ATIR method can be successfully applied either for tilted-planar layers at various thickness or reversely-tilted layers[65,66] and appropriate utilization of elastic-transition formulae.[57]

c) ATR method for determining the LC orientation and anchoring of φ-azimuthally-deformed nematics.

Of interest is to mention that the first ATIR measurements have been performed on pure-magnetically-twisted N layers.[1] Although the usefulness of ATIR method for measuring pure twist surface and bulk LC orientations have been pointed out by many authors;[2,67,68] today there are still many unsolved theoretical and experimental problems for this case. On the other hand, the measurement of the surface azimuthal orientation and anchoring is of importance for the LC display devices.[69–77] The ATIR method, according to us, can be easily applied experimentally as well as can be theoretically investigated in detail when the linear polarization of the incident light is not changed into eliptical one, i.e. when the direction of the incident polarization follows the azimuthal orientation of the LC at the interface glass prism -LC and in the depth when the following inequality

$$\left(\frac{d\varphi}{dz}\right)\left(\frac{\lambda}{2\pi(n_e - n_o)}\right) \ll 1$$

is valid (φ is the azimuthal deformation angle, λ is the wave-length of the light passing through the LC medium).

d) ATIR method for determining the LC orientation and anchoring θ, φ polar-azimuthal deformations in twisted nematics and in large-pitch cholesterics.

This case is much more complex relative to the previous ones; however, the measurement of the θ-polar surface angle closely related with the surface anchoring in such LC cells is of great importance for the viewing characteristics of the N-twisted displays. Again of significance is the depolarization of the incident linear polarized light.[78–80] Although the matrix solution is well-known for this case,[81] the ATIR problem should be resolved by analytical means, knowing the exact dielectric tensors even for the more complex cases[82] and the dispersion relation which can give the effective optical index and the ATIR condition even for the case of the geometrical optics.[83] Indeed, this case would be complicated owing to two unknowns, namely the θ-polar and the φ-azimuthal surface angles, the latter of which can be determined by other optical means.

The surface energy is of importance only for large-pitch Chs[84] and the ATIR method can be successfully applied for measuring this energy through the Ch-N transition.[84]

e) ATIR method for determining the LC orientation and anchoring of large-area Sm A and Sm C monodomains.

The Sm A and Sm C orientation have been measured to date by conoscopical methods[24,25,85] or by Fréedericksz transition.[26] The ATIR method can be applied for measuring either the inclination of the smectic molecules in large-area (several mm^2) Sm A and/or Sm C monodomains or their optical indices. This method is very simple and could be applied for exact measurements of the Sm C tilt angle around the N-Sm C, Sm A — Sm C[86,87] or near the N-Sm A — Sm C multicritical point.[88] On the other hand, the ATIR method can be successfully applied for the measurement of the temperature dependence of the Sm C tilt, characteristic for all the smectic LC arising from Sm A under cooling.

CONCLUSION

The light propagation in an anisotropic, inhomogeneous medium such as a deformed LC is a very incompletely-known subject. Only a few cases can be solved analytically. The matrix methods, on the other hand, cannot give usual ATIR formulae for measuring the LC surface anchoring and orientation. They always should be accompanied by approximate analytical methods such as the WKB method and others. Of importance is the resolution of a number of technical questions such as the quality of the laser and the glass-prisms, the divergence and the depolarization of the light, the deviation of the incident light from plane waves, the influence of the thermal fluctuations, and of the many additional layers being important for the LC display devices. Other groups of problems are connected with the evaluation of the usefulness of the ATIR method for measuring the surface anchoring and orientation of Ns with different orientation, large-pitch cholesteric and some smectics. Many experimental and theoretical problems are not solved yet up to date and await their further elucidation. The utilization of the total internal microscopy is a step of great importance for the application of the ATR method. Other more difficult questions for measuring the LC anchoring and orientation of LC domains and defects in the range of several hundred microns are connected mainly with technical difficulties. However, their resolution would lead to the much more and deep ATIR application for the investigation of the surface anchoring and orientation.

Our brief outlook clearly shows that the ATIR method is a unique one for some cases which can give excellent results when measuring the LC anchoring and orientation.

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