Dynamics and Electrooptics of Vertically Aligned Nematics With Induced Pretilt on SiO_x

Eva Otón, Sol López-Andrés, Carlos Carrasco-Vela, Beatriz Cerrolaza, Noureddine Bennis, and José Manuel Otón

***Abstract*—The response time and contrast of vertically-aligned negative nematics depend critically on the aligning surface and its induced pretilt. Pretilt can be tuned up using obliquely evaporated inorganic oxides as alignment surface.**

A thorough study of pretilt angles induced by aligning surfaces of thermally evaporated SiO_x has been carried out on several commercial and experimental negative nematic mixtures. Morphology of SiO_x with different evaporation angles has been studied by Field Emission Scanning Electron Microscopy. Some morphological variations appear at specific evaporation angles. These are related to the pretilt induced by the surface and to the dynamic response of the device.

***Index Terms*—Field-emission scanning electron microscopy (FESEM), liquid crystals, SiO_x, vertically aligned nematic (VAN), vertical alignment.**

I. Introduction

VERTICALLY aligned nematic (VAN) liquid crystals with a negative dielectric anisotropy are useful in many applications requiring fast response time and excellent contrast ratio. VAN displays have become popular in consumer electronics because of their excellent contrast and low response time (often reaching video frequency). At present, a number of solutions based on VANs (MVA, PVA, etc.) can be found in commercial products, like large-area direct-view TV sets and high-end projection displays [1], [2]. VANs, on the other hand, are excellent candidates for a number of photonic devices—beamsteerers, tunable lenses, prisms, or filters—whose action on an impinging light beam is restricted to phase delays, i.e., without modifications of the light intensity or the state of polarization. Indeed, a VAN cell upon switching can be employed as an analog phase-only linear retarder i.e., produces arbitrary phase delays depending on voltage, without modifying the state of polarization of a linear incoming light.

In either case, the display performance is ultimately determined by its contrast (or visibility) and its dynamic response. Both parameters depend on the pretilt angle induced by the aligning surface on the liquid crystal molecules; therefore pretilt must be controlled [3]. Pure homeotropic alignment (pretilt angle = 0°) would produce theoretically a contrast only limited by the light leakage of the polarizers. However is useless for actual devices since the switching torque is null (in practice, switching time is exceedingly long), and the switching direction is undefined. Therefore, a non-null induced pretilt angle is required for the material to switch along a predefined direction. As pretilt is increased, the response time improves but contrast ratio is compromised.

It has been shown that pure vertically alignment can be obtained by doping the host liquid crystal with other materials [4]. However, pretilt angles cannot be controlled with this method. Recently, vertically alignments have been achieved by the use of ion and plasma beams [5], as well as depositions by a filtered cathodic arc process, leading to a different variation of pretilt angles [6].

In this work, the aim has been to generate a controlled pretilt angle depending on the evaporation angle and determine a relation between SiO_x surface structure and pretilt. The generation of pretilt angles using SiO_x as alignment layer has been studied. SiO_x layers were obtained by oblique PVD (Physical Vapor Deposition). SiO_x surface morphology has been studied using scanning microscopy techniques to find a relation between surface structure and liquid crystal pretilt angle. Different evaporation angles produce different pretilts or even different types of alignments. These pretilts have been measured along with their corresponding variations of contrast and dynamic response. Low pretilt angles, i.e., nearly homeotropic or vertical orientation, guarantee the best contrast ratios, due to perfect vertical alignment, but response time is remarkably longer. On the other hand, devices having high pretilt angles show excellent response times but poor contrast ratio. Therefore, pretilt needs to be tightly controlled in order to achieve a trade-off between good contrast ratio and fast response time [7].

II. Experimental

Two kinds of experiments have been done. Several batches of ITO-coated glasses with evaporated SiO_x at different angles were prepared for SEM microscopic analysis. The analysis occasionally damages the SiO_x; therefore, it was decided not to use the same batches for cell manufacturing. Instead, parallel batches of every orientation and manufacturing condition were prepared. The second batch was employed for cell manufacturing. Single pixel cells were assembled in antiparallel orientation. The cell gap was 4 μm in all cases. A nematic commercial mixture MLC 6608 (Merck) with a negative dielectric anisotropy was filled at isotropic state and allowed to cool down following a standard manufacturing protocol.

A. SiO_x PVD (Physical Vapor Deposition)

Several evaporations have been done using PVD technique. Selected angles covered a range from 20° to 86°. Each evaporation for every group of samples consisted of: first, deposition of 100Å
speed of 1Å/s, then, selecting the desired angle, evaporation at 1 Å/s and 100 Å thick. It must be mentioned that our evaporation chamber features several modifications including an external goniometer actuator attached to the sample. Therefore, the orientation of the batch can be modified and the second evaporation can be performed without breaking the vacuum.

SiO_x surface structures were analyzed by FESEM (Field Emission Scanning Electron Microscopy), starting at 20° and studying surface variations in each sample with a difference of 10°. Zoom had to be set at x60000 due to the extremely high uniformity and flatness of the surfaces.

B. Pretilt Measurements

Pretilt measurements have been performed in the whole series of manufactured cells with SiO_x layers deposited at different evaporation angles. Pretilt angles were measured using an ellipsometric method described elsewhere. The ellipsometric system is able to detect minute changes in the state of polarization (SOP) of the incoming light, thus resulting very sensitive for pretilt measurements. The ellipsometer actually measures the tilt angle of the liquid crystal molecules across the cell. Measurements are indirect, i.e., the tilt is obtained as the best fit of the liquid crystal profile producing a given set of SOP variations. The method is quite reliable since data acquisition is performed at several impinging angles and wavelengths. A full discussion of the method can be found in [8].

Within this experimental context, pretilt is simply the tilt of the liquid crystal adjacent to the glass surfaces. The precision can be further improved by repeating the procedure for several cells of every evaporation angle. Pretilt is referred to the surface normal, i.e., 0° means perpendicular and 90° means parallel to the glass surface.

C. Electrooptic Characterization

Quasi-static and dynamic electrooptical characterization of the cells was carried out by recording the time resolved optical transmission of the samples between crossed polarizers while being addressing with driving voltage signals. Cells are placed in a microscope (Nikon Optiphot Pol 2) provided with a programmable hot plate and a rotating stage. All cells were measured at 35 °C. The output light is collected in an optical fiber bunch bringing the signal to a fast response photodiode (Hamamatsu). The photodiode signal is stored in a digital oscilloscope (Hewlett Packard) and sent to a computer. The same computer controls the electronic driving of the cell via a LabView application. Cells were addressed using a 10 kHz AC carrier modulated by a 1 Hz square signal with variable amplitude.

III. Results and Discussion

A. SiO_x Surface Structure—General View From 20° to 86°

Observed surfaces correspond to 20°, 30°, 40°, 50°, 60°, 70°, 80°, 82°, 84°, 85°, and 86°, see Fig. 1.

From 20° to 40°, the uniformity of the surface morphology is very high, with little or no features. Non-polyhedral particles are found; morphology appears to be a 'droplet-like' structure. Droplets are rounded shaped though not regular, with no apparent orientation and placed randomly. Droplet diameter is variable between 100 and 250 nm approximately. Generally speaking, the particle size increases as evaporation angle increases.

From 50° to 80°, surface structures show some variations. Particles or droplets become more defined, with sharper delineated boundaries. They also have a more rounded shape and their size appears to be larger than for lower angles. Near 80°, surface structure starts being less uniform having little holes or cracks between particles.

A sudden change in morphology occurs for angles greater than 80°. A more detailed study was done for samples with evaporation angles from 80° to 86°, as its morphology showed a remarkable alteration: structure and particle size are different, and particle distribution highly differs. In this case surface was less uniform than the surface observed at lower evaporation angles, showing irregularities along the whole sample and a more detailed structure. The particle size radically changes, showing uniformity and diameters from 30 to 50 nm.

In these cases, a progressive evolution of the surface structure was observed, changing from 'droplet-like' structure to spherulitic morphology. Evaporations for 82° and 84° show intermediate morphology states, coexisting both droplet and spherulitic structures. 85° and 86° show, a spherulitic or a granulate morphology, with some spherical particles as well, with sharper edges, smaller particles and presence of holes at high angles. Holes have a triangular shape, according to a sphere stacking structure. Additionally, structure at higher angles is less compact, has bigger holes and spherical isolated particles are present. Alignment of particles and holes perpendicular to evaporation direction is observed. Thicker SiO_x layers lead to an oblique columnar structure [9].

B. Liquid Crystal Alignment

A series of liquid crystal cells were manufactured with SiO_x as alignment layer, changing evaporation angle in 10° steps for non-variable alignments and in 2° when alignment showed a bigger variation. Table I shows the measured pretilt angle of the liquid crystal molecules depending on SiO_x evaporation angle.

The liquid crystal alignment strongly depends on the evaporation angle set on the outer glass plates of the cell. As a rough approximation, liquid crystal cannot be aligned vertically if evaporation angle is lower than 50°. Vertical alignment shows up for evaporation angles greater than 50°.

Samples with evaporations from 20° to 50° present mostly an homogeneous alignment, clear state is present at 45° between crossed polarizers.

Fig. 2 shows several cells observed between crossed polarizers. 20° and 40° cells show lack of aligmnent and scattering. As a rule, cells obtained with deposition angles lower than 50° were useless; no vertical alignment can be obtained.

Alignment in samples with evaporations from 50° to 80° presents the most interesting results. SiO_x evaporated in this

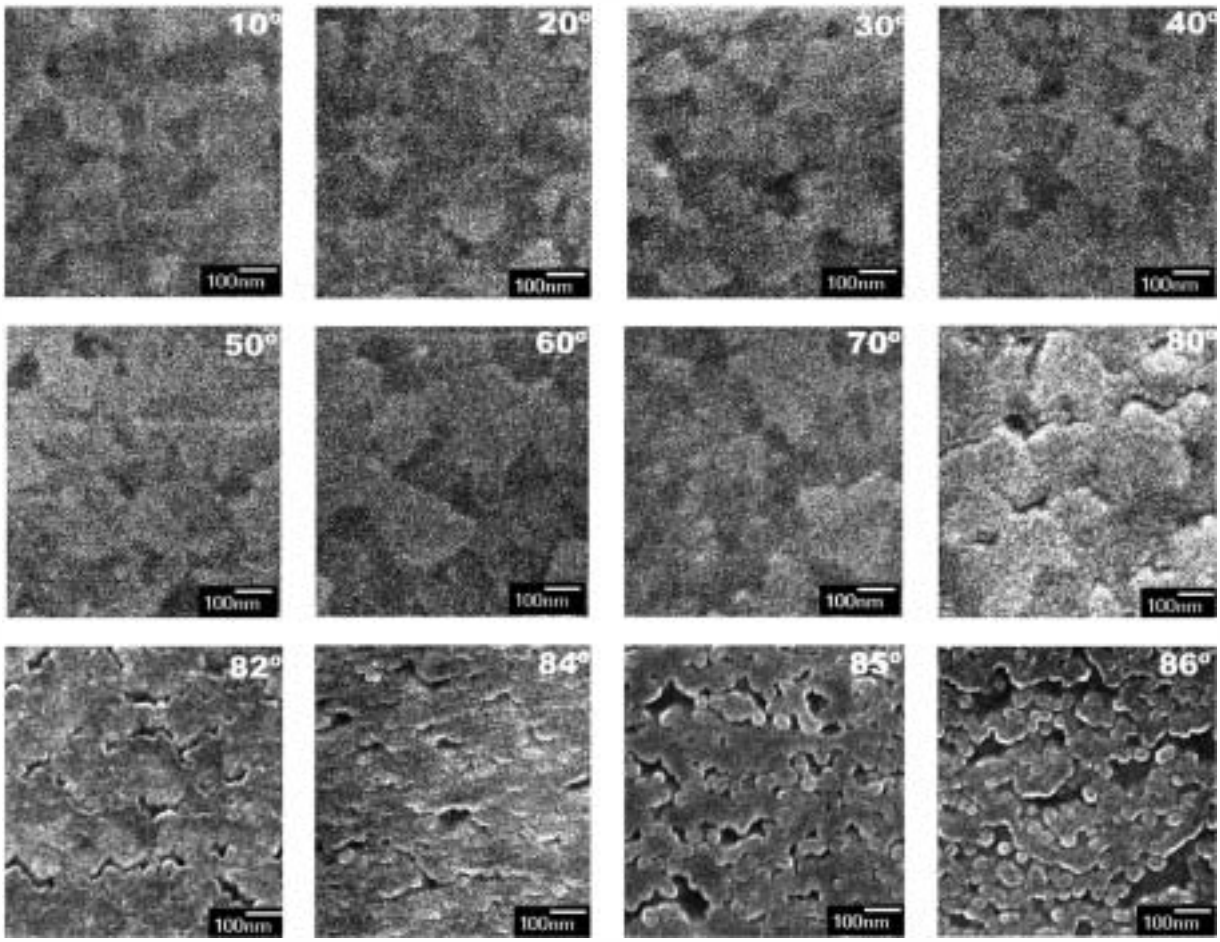

Fig. 1. SiO_x surface morphology, evaporation angles from 20° to 86° analysed by FESEM.

TABLE I
LIQUID CRYSTAL PRETILT ANGLES OBTAINED FOR DIFFERENT SiO_x EVAPORATION ANGLES

Evaporation angle	< 50º	50º-60º	70º	80º	> 85º
LC pretilt angle	Random	~ 1º	~ 8º	~ 26º	~ 90º

range will produce vertically aligned cells and thus, useful devices. It is worth noticing that, as mentioned above, only minute variations in the SiO_x surface morphology are appreciated when comparing surfaces in the 20°–50° range and surfaces in the 50°–80°. These minute variations scarcely visible at very high zoom give rise, however, to a dramatic change in the liquid crystal molecular orientation. It is reasonable to assume that increasing the evaporation angle, the anchoring strength of the surface increases as well. A method for measuring the anchoring strength by ellipsometry has been recently developed, and is currently being applied to this problem.

Cells that have been manufactured with glass plates in the 50°–60° range are black with no transmission variations when rotating the cell between crossed polarizers; this is a typical response of vertical aligned cells. Pretilt angle increases with evaporation angle; consequently, the higher the evaporation angle (from 50° onwards), the higher the cell transmission. Cells in the 60°–80° range show a small light transmission when placing the cell at 45° within the polarizers. This is a result of an increasing pretilt making the cell more and more birefringent as the evaporation angle increases.

Cells with evaporation angles at 50°, 60°, 70° and 80° are shown in Fig. 2. 50° cell shows no transmission (black), therefore pretilt is null or near null, and cell will show a good contrast ratio. 60° cells have the same appearance as 50°. Again, pretilt angle is null and there's no transmission. 50° and 60° liquid crystal alignments are equivalent being their pretilt angle close to zero. Ellipsometric measurements show that the pretilt angle is about 1° at most.

Alignment obtained for 70° cells is different. Homeotropic alignment is achieved as well but a certain transmission in the cell can be seen because 70° cells have a certain pretilt angle and contrast decreases. Pretilt angle as measured by spectral ellipsometry is about 8°.

80° cells exhibit the highest transmission. Pretilt angle increases considerably being close to 26°, which is too high for most applications requiring homeotropic alignment. The contrast ratio is consequently compromised.

Samples with evaporation angles from 80° to 86° showed interesting variations. As mentioned before, SiO_x morphology for angles greater than 80° changes drastically from lower angles. Liquid crystal alignment manifests a variation in the same way. For high angles, pretilt angle is so high that alignment cannot

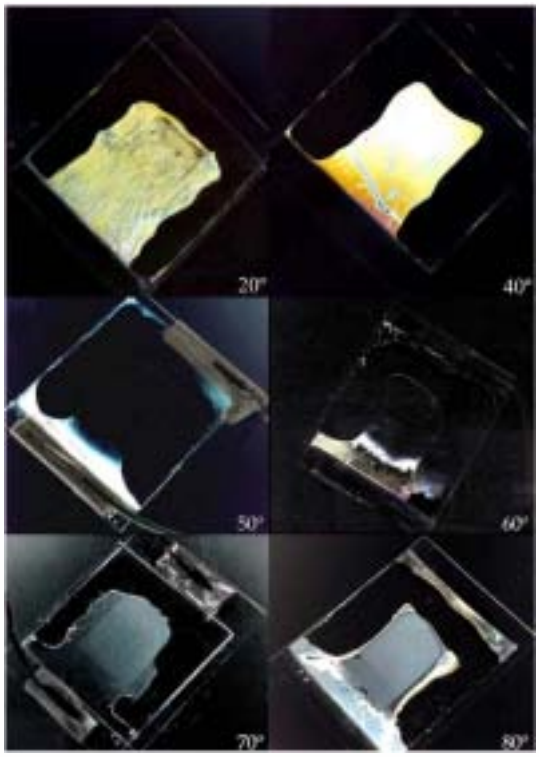

Fig. 2. Displays from 20° to 80°. 20° and 40° evaporation angle show non aligned liquid crystal. 50°, 60°, 70° and 80° evaporation angle surfaces show vertical alignment of the LC, but pretilt increases, as seen by the increased transmission between crossed polarizers.

be considered homeotropic anymore. Evaporation angle of 85° produces an almost homogeneous alignment, molecules having a pretilt close to 90°, i.e., parallel to the glass plates. Contrast in these cells is very low, and switching is barely noticeable. Note that switching would intend to bring the molecules to any position within the glass plates lane. Since the molecules are already on this plane in steady state, no reorientation is required for the liquid crystal to "switch".

1) Modifying Evaporation Parameters: A group of cells with different evaporation parameters was analyzed to check alignment uniformity.

A number of evaporations were made to verify the PVD parameters. A batch with layer thickness 300 Å was prepared to confirm that the 100 Å layer thickness is thick enough to shield any effect from the vertically evaporated layer. Similarly, using a 0,2 Å/s evaporation speed confirmed that the same ranges of alignments as in 1.0 Å/s were obtained. Liquid crystal alignment did not show any variation with the new parameters. It is assumed, therefore, that within a certain range, evaporation angle is the only relevant manufacturing parameter affecting the pretilt in SiO_x aligning surfaces

C. Electrooptic Characterization

All the test cells included in each batch for every evaporation angle were characterized electrooptically in terms of contrast, response time and greyscale. As expected, pretilt angle has a strong influence in the liquid crystal cells, especially in contrast and response time. On average, low pretilt cells, corresponding to 50° and 60° evaporation angle have significantly higher contrast ratios but worse response times than cells having high pretilt angles as obtained with cells of 70° and 80° evaporation angles. The response time, as measured in the whole range of evaporation angles, was in the range of 2–3 ms, while contrast steadily decreased when increasing evaporation angle.

1) Backflow Effect: The rise time of any LC display is usually shorter as the applied voltage increases, so liquid crystal molecules switch faster at higher voltages. In some of the manufactured cells the opposite behavior was found.

For displays having null or near null pretilt angle, the response time increases above a certain voltage value, and rise time achieve very long values. However, fall times are not affected. This effect alters the measurement of response times and is called backflow effect [10]

Above certain voltages, there is a fast rearrangement of the liquid crystal molecules followed by a slow reorientation, which induces the molecules to switch slowly increasing the overall rise time. From the practical point of view, cells showing backflow have a limited voltage range. Above that range, the response time is so large that cells become useless in most applications. It has been found that the presence or absence of backflow depends heavily on the pretilt (it is only shown for near-zero pretilts) and on the cell thickness.

Cells showing backflow can be obtained without this effect reducing the cell thickness while keeping constant the remaining manufacturing parameters. When thickness is reduced, backflow either disappears or is shown at higher voltages

D. Electrooptic Results

Fig. 3 shows greyscale and dynamic response for 50°, 60°, 70°, and 80° displays.

Cells with 50° and 60° evaporation angles have a very low pretilt. They both exhibited backflow when applying voltages greater than 6 V and showed a double-peaked transmission profile and high response times. For example, rise time was 20 ms for 10 V. Nevertheless, in a working range up to 6 V, 50° and 60° displays show very good response time (3 ms) and excellent contrast ratios.

For 70° displays, the backflow effect did not show up. As pretilt angle increases, switching becomes easier so turbulences do not appear and backflow effect is avoided. Liquid crystal behavior is standard for this particular angle. Response times are fast in the whole voltage range and faster than for 50° or 60°. Contrast, however, decreases as a result of the birefringence of the OFF state induced by the pretilt angle.

For 80° displays, the response shown is similar to 70°. In these cells, however, pretilt is very high (about 26°), making the OFF state highly transmissive. Backflow could not be found in these displays either and response times were the fastest due to high pretilt. Rise and fall response times are compared in Table II. Fall times increase as pretilt increases, opposite to rise time. This behavior can be explained by differences in the elastic energy. The higher the pretilt is, the smaller the elastic energy between on and off state becomes and for that reason a longer fall time is expected.

The absence of backflow and the excellent time response should make 70° and 80° displays the best choice for applications. However, contrast is poor, thus precluding their use in display applications. This is not the case, however, for photonic

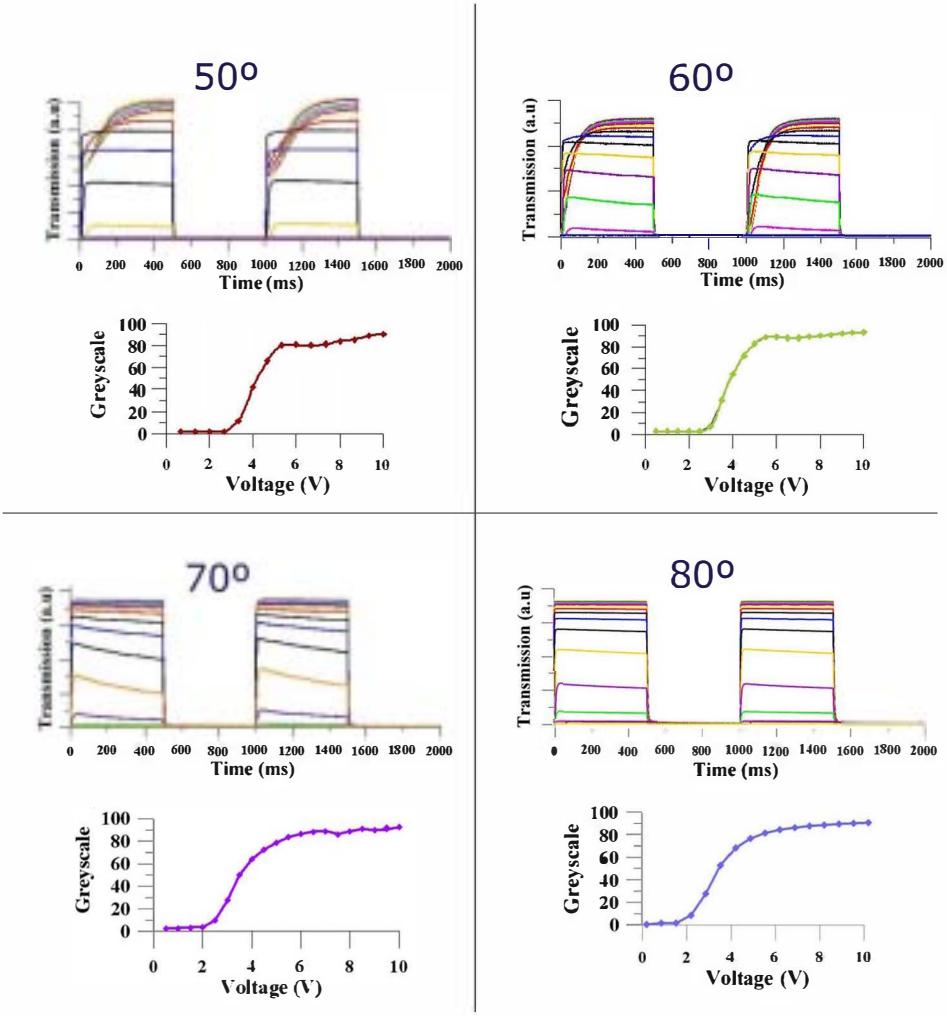

Fig. 3. Electrooptical results for 50°, 60, 70• and 80° displays. Backflow effect can be noticed in 50° and 60° displays.

TABLE II
LIQUID CRYSTAL RESPONSE TIMES FOR VARIOUS DISPLAYS WITH DIFFERENT SiO_x EVAPORATION ANGLES

SiOx evap. angle	50º	60º	70º	80º
Rise time (ms)	3.1	3.4	3.0	1.3
Fall time (ms)	3.5	4.8	6.6	7.5

applications where the variable phase delays of VANs are used. In this case, the only drawback of high pretilt is that the range of delays generated by a given LC material and cell thickness is somewhat reduced. This small disadvantage is superseded in many applications by the excellent dynamic behavior of high pretilt cells.

IV. CONCLUSION

The results demonstrate, therefore, that SiO_x as aligning surface for VAN LCs may generate a full range of pretilts that can be eventually optimized depending on the requirements of the specific application. In principle, display-related applications should use evaporation angles in the 50°–60° range, so that contrast can be maximized. The contrast requirement shall be met at its minimum for the response time to be improved as much as possible. Non-display applications probably would prefer higher pretilt angles to minimize the response time. The slightly lower phase delay range achieved in these cells should not be relevant for the device performance in most cases

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Eva Otón received the B.Sc. degree in chemistry from the Universidad Autónoma de Madrid, in 2005, and the M.Sc. degree in material engineering from the Universidad Complutense de Madrid in 2008.

She joined the Department of Photonic Technology in Universidad Politécnica de Madrid, Madrid, Spain. Her research interest has been focused on vertically aligned nematic liquid crystals and interaction with surface conditioning in SiO_x and SiO_2. She is also interested on applications of adaptive optic devices such as liquid crystal graded index beam steerers.

Sol López-Andrés, photograph and biography no available at time of publication.

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