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Freedericksz transition and beam coupling in liquid crystals with PVK:C_{60} alignment-command layer

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Photorefractive structures consisting of liquid crystals[1] with photoconductive polymer layers have attracted a lot of interest because of their high nonlinearity achieved with low light intensities[2]. In particular, photorefractive polymers such as polyvinyl carbazole (PVK) doped with photosensitiser[3] trinitrofluorene (TNF) or poly(octyl-thiophene) functionalised with dyes showed particularly promising two-beam coupling gain and diffraction.

The reorientation process of liquid crystal molecules, responsible for nonlinear effects, can be induced and controlled by the application of light and electric fields. While it is widely accepted that surface effects play an important role in the formation of photorefractive gratings, the exact nature of their contribution or the role of the system's conductivity has not been determined yet. In particular, surface-mediated effects[4] such as photorefractive surface-charge modulation generated in polymer coatings of cell substrates provide strong nonlinear contribution.

We investigated different, unidirectionally rubbed polymer alignment layers spincoated onto ITO glass. They included: undoped PVK, PVK doped with different concentrations of fullerene C_{60} (PVK:C_{60}) and, as a reference, conventional, rubbed polyimide (PI) layers. We observed that unidirectional rubbing of PVK surfaces aligns liquid crystal molecules with an easy axis in the plane of the substrate and perpendicular to the rubbing direction. Cells were filled with E7 liquid crystal.

We measured a significant increase of DC Freedericksz transition threshold for alignment layers made of pure PVK in comparison with PI and PVK:C_{60}. In cells with undoped PVK layers, no reorientation was observed even when both a DC field and light were applied. Doping of PVK layers with C_{60} made the system extremely photosensitive and reorientation took place in the illuminated area with decreasing threshold of Freedericksz transition for higher light intensities. In the case of undoped PVK, the trapping of charges and the build-up of double electric layers near substrates prevent the reorientation of liquid crystals by an external electric field. When PVK is doped with sensitisers, the effect of photocontrolled reorientation could be explained by the light induced annihilation of the double charge layers and the light-induced change of charge trapping and conductivity.

We will also present data on efficient beam coupling in cells with PVK:C_{60} alignment-command layers and discuss the details of its mechanism, based on results from alignment and Freedericksz transition in this system. The two-beam coupling gratings were written in 30 μm thick planar cells using two linearly p-polarised beams at 633 nm (or 514 nm). The intensities of both incident beams, as well as the build-up of power in the probe beam and the depletion of the pump beam were measured. When we changed the polarity of the applied DC field, the direction of the energy flow was reversed.

We determined the two-beam coupling gain ratio G ($G=I_{12}/I_1$) where I_1 is the intensity of the probe beam in the absence of the pump beam I_2 and I_{12} is the intensity of the probe beam in the presence of the pump beam and deduced the exponential gain parameter (coupling coefficient Γ) to characterise the efficiency of the structure (liquid crystal-polymer-photosensitiser). Even for small grating spacings of 6 μm we achieved a gain G as high as 5, indicating an exponential gain parameter Γ of over 700 cm^{-1}. The results presented so far were measured at the optimum cell tilt angle of 45°, but we noted that a significant beam coupling exists for the whole range of tilt angles between 0 and 45°.

The gain depended on applied electric field, with a maximum obtained for around 20 V and could be observed for very low light intensities, namely for a total incident intensity of 150 $\mu W/cm^2$. In fact, we observed that the net value of gain G varied very little with incident intensity, so even increasing the incident power by an order of magnitude did not yield significant increase in the gain. However, the dynamics of the beam coupling response depended on voltage, namely at higher light intensities, the reorientation processes started at lower voltages than in the case of low incident intensity.

Finally, we will discuss the role of diffraction that accompanies the beam coupling process. A typical two-beam coupling experiment in liquid crystal cells operates in the Raman-Nath (thin) grating regime and as a result several diffraction orders of the beams can be present. It is, therefore, usually quite difficult to separate beam coupling and diffraction. Careful monitoring of both pump and probe beam intensities with and without the other beam can help[5] to estimate the value of net exchange of energy between two beams, but the phase shift has to be determined to verify the nature of the amplified intensity of the probe beam. In our case, we measured the phase shift to be $\pi/2$ and investigated its dependence on the cell's tilt angle.

All in all, liquid crystal structures incorporating PVK:C_{60} not only show excellent photorefractive performance and their suitability for coherent light amplification and patterning, but also allow us to investigate in detail photosensitive surface effects, critical to two-beam coupling process.

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