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A pitch of helicoid of chiral liquid crystals and surface plasmon excitation: A theoretical study

G. Dyankov[a,c], K. Zhelyazkova[a], M. Petrov[b], and B. Katranchev[b]

[a]Faculty of Physics, Plovdiv University "Paisii Hilendarski", Plovdiv, Bulgaria; [b]Institute of Solid State Physics, Bulgarian Academy of Sciences, Sofia, Bulgaria; [c]Institute of Optical Materials and Technologies, Bulgarian Academy of Sciences, Sofia, Bulgaria

ABSTRACT

Surface plasmon resonance (SPR) is widely used in different types of optical detection schemes and for light manipulation. There are a lot of experimental and theoretical investigations about the influence of different media, adjacent to the metal layer, on SPR. However, the influence of liquid crystal (LC) layer is not fully revealed. There are only few publications considering Kretshmann configuration with LC layer sandwiched between the prism and metal layer. Respectively, a lot of questions regarding the influence of LC properties on the surface plasmon resonance (SPR) are still open. The aim of this paper is to fill this gap.

KEYWORDS

chiral liquid crystal; surface plasmon resonance; Bragg reflectioon

1. Introduction

Because of its high optically anisotropy, extremely low power consumption and easily controlled alignment by electrical, light, and acoustic waves liquid crystal (LC) is used as an active medium for controlling variety of physical processes and effects. Thus the unique electro-optical and thermo-optical properties of the LCs have been used in numerous applications [1,2], such as displays, spatial light modulators for adaptive optics in real time, optical imaging, optical switches and attenuators for telecommunications, optical phase arrays for beam steering, *etc*.

Plasmonics has attracted considerable attention because of its potential applications in super-resolution imaging [3], optical cloaking [4], nanofabrication [5], energy harvesting [6] and plasmonic circuitry [7]. Logically, the unique properties of LC, us the large birefringence, have been used for making active plasmonic devices for different applications, such as plasmonic switches [8], active plasmonic color filters [9], plasmonic waveguides [10] and not at last as sensors.

Sambles and co-workers have used surface plasmons for characterization of LC cells properties both experimentally and theoretically [11–16]. These comprehensive studies have demonstrated that the problem about interaction of surface plasmon with an adjacent anisotropic layer does not lose its actuality. Experimental works [8–10] show that the surface plasmon resonance (SPR) can be controlled effectively by changing LC parameters. The experimental results in [11–16] reveal the possibility for determination LC parameters by SPR. However, a device operating as a sensor, based on the SPR interaction with LC layer, has not

CONTACT G. Dyankov gdyankov@iomt.bas.bg
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launched on the market, so far. This demonstrates, probably, that the feasibility of these kinds of interactions is not fully revealed.

To our knowledge there is only one publication [17] considering Kretshmann configuration with a buffer LC layer sandwiched between the prism and the metal layer. Experimentally is studded the surface plasmon excitation as a function of director pretilt, as a function of p- and s- polarization of incoming beam and as a function of voltage applied the LC cell.

We studied theoretically the same configuration in our previous paper [18], however a lot of questions regarding the influence of LC properties on SPR are still open. The aim of this paper is to fill this gap.

We focus our attention on simulation of the influence of LC layer on SPR excitation which eventually could increase the feasibility of the structure. The simulation is performed for *p*- and *s*-polarized incident light and for chiral ferroelectric smectic C (SmC*) and chiral cholesteric LC (C_hLC - chiral nematic N*) phases both consisting of a helical structure. It is well known [19,20] that at 90° tilt angle θ, in SmC* phase, the medium becomes cholesteric. Furthermore, helical pitch p and tilt angle θ are easily controlled by electric field and can be used as a sensor.

Our simulations are based on a theoretical model, obtained by solving Maxwell equations in 4×4 matrix form, for an anisotropic medium [21]. We have considered usually used in the experiments LC cell (see for example [20]). Fig. 1 shows the structure we have simulated. Regarding the parameters of LC cell an LC molecule orientation we use the notations presented in [20].

For prism we have used permittivity $\varepsilon_p = 2.89$, the gold film has thickness $d_{Au} = 50$ nm. For LC we assume $\varepsilon_1 = \varepsilon_2 = 2$ and $\varepsilon_3 = 3$. The permittivity of gold is according [16]. As in [12], here we consider LC layer, no LC cell – LC layer sandwiched between prism and glass plate over which gold is evaporated. We have shown that despite the structure prism/LC layer/gold is not experimentally feasible, we have paid attention to it in purpose to make clear our main ideas. In the case of LC cell the only difference is due to the interference from glass plate which blurs the reflectance.

Keeping in mind that the spectral read out is more feasible, in our simulations we have assumed white light source and spectrometer for SPR excitation and detection, respectively (as shown in Fig. 1). The incident angle is $\varphi = 38°$ ensuring SPR excitation at incident wavelength $\lambda = 632$ nm.

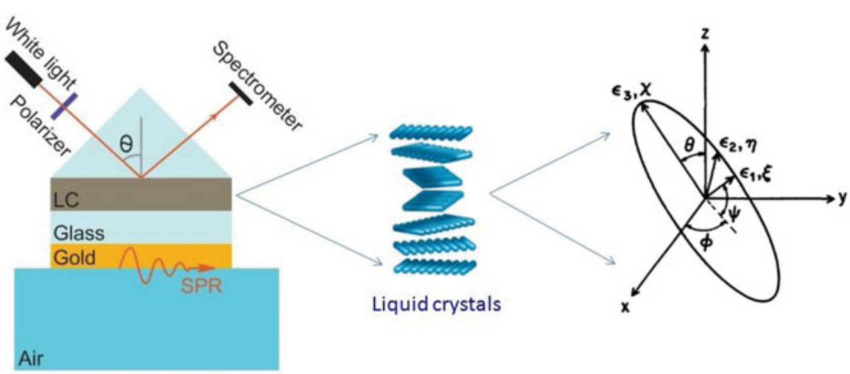

Figure 1. Configuration of the structure and orientation of the principle axes χ, ξ, η is of the local dielectric tensor ellipsoid defined by Euler angles θ, ϕ, ψ in some chiral molecular layer.

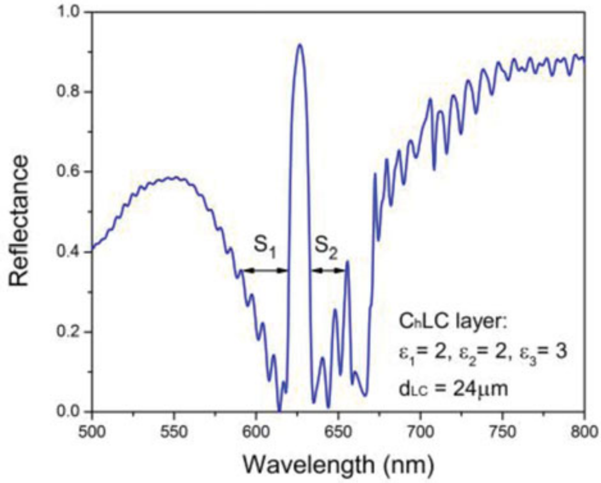

Figure 2. Simultaneous SPR and Bragg reflection excitation. Rpp reflection spectra as a function of wavelength for incident angle $\varphi = 38.15°$ and C_hLC with: p = 1200 nm, Np = 20. S_1/S_2 is the surface of the area locked between left/right side of Bragg reflection curve and resonance curve.

2. Bragg reflection and SPR excitation

We found [18] that LC layer thickness does not influence SPR position, but depends on the tilt angle θ. This gives an opportunity, at fixed tilt angle, to adjust the helical pitch length p and turn numbers Np so, to excite second order Bragg reflection at the same wavelength where SPR is excited. We prefer the second order because it is narrow and does not mask SPR. Because the effectiveness of Bragg reflection is higher at $\theta = 90°$, we have chosen C_hLC for simultaneous SPR and Bragg reflection excitation.

Figure 2 shows the reflection spectrum of structure prism/C_hLC/metal/air. LC layer parameters are shown on the figure. Bragg reflection and plasmon resonance appear simultaneously at the same wavelength. The pitch length is p = 1200 nm and it is twice longer than the SPR wavelength and fulfils the condition for second order Bragg reflection.

Next we are interested in SmC* phase with pitch length around 300 nm. According to the theory [23] the tilt angle influences the bandgap width and strong *s*-*p* and *p*-*s* coupling can occurs around 600 nm. Indeed, our simulations confirmed this fact - at small tilt angle we observe narrow bandgap near to 600 nm and minimal sidelobe modulation. At small tilt angle SPR excitation is effective with *p*-polarized incident light, while *s*- polarization is strongly coupled with *p*- polarization through Bragg reflection. With polarizer set at 45° in reference to *p*- incident plain, the incident light with *s*- and *p*- polarization simultaneously illuminates the structure: *p*- polarization excites SPR, while *s*- polarization undergoes Bragg reflection. The reflection of *p*- polarized reflected light is as Fig. 3 shows.

We have almost the same shape of the spectrum of reflection curve as a function of wavelength as given in Fig. 2. However, excitation conditions are very different.

3. Pitch length and SPR excitation

We shall show now that SPR excitation is effective not only for SmC* with small tilt angle and for chiral cholesteric LC, but also for other values of tilt angle.

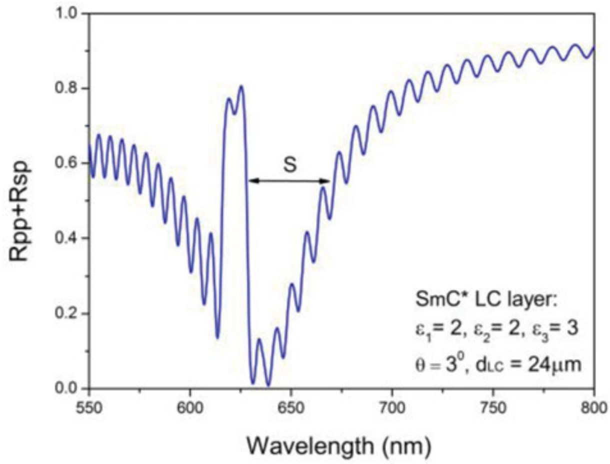

Figure 3. The reflectance of *p*- polarized reflected light at simultaneous excitation with *p*- and *s*- polarization. The reflectance of structure prism/SmC*/gold layer: $\theta = 3°$, p = 300 nm, Np = 80 and LC layer thickness is $d_{LC} = 24$ μm. *S* is the surface of the area locked between right side of Bragg reflection and resonance curve.

We have scrupulously studied the effectiveness of excitation on pitch length. Keeping in mind that SPR position does not depend on LC layer thickness, we studied also the dependence on number of turns at fixed pitch length. The main conclusions are:

- For small tilt angle ($\theta < 3°$) SPR is well expressed for all pitch length;
- For $\theta = 90°$ the best choice is a pitch length (marked as $\boldsymbol{p_{cr}}$) which fulfils the condition for second order Bragg resonance [23]; this case is illustrated by Fig. 2; for $p \neq p_{cr}$ SPR is still observed, but with strong sidelobe due to the interference caused by the birefringence in LC layer;
- For other tilt angles the situation is complicate. SPR is effectively excited if *p*- incident polarization does not couple with others reflected or transmitted polarizations. This depends on the coupling coefficients including different terms of the dielectric tensor [23] which are functions of the tilt angle and pitch. That why for some theirs values SPR is completely destroyed, for others – not.

The polarization conversion in LC layer at tilt angle $3° < \theta < 89°$ is pronounced because the birefringence is large. The conversion we need is from *s*- to *p*- polarization which excites the surface plasmon. The conversion can be accomplished when the last helix, contiguous to the metal layer, has length *p/4* or *3p/4*. We have consider SmC* phase as LC layer with thickness 24μm and $p = 2.4$μm despite other values are feasible too. Fig. 4a shows SPR resonance (marked in thick red) excited with *s*- polarized incident light. The red color marked dip in the curve disappears when the gold layer is removed as should be in the case of SPR. However, the effectiveness of resonance excitation is low – the maximum value of Rsp is about 0.6 while the effectiveness of excitation with *p*- polarization is higher – Rpp is about 0.7 – 0.8. Logically, the simultaneous excitation with *p*- and *s*- polarization gives the best result, shown in Fig. 4b. This excitation can be accomplished experimentally when input linear polarization is set at 45° in reference to *p*- incident plane. LC layer parameters are reported in the inset of Fig. 4b.

In Section 2 we discussed the behavior at small tilt angle of a structure with SmC* phase having pitch length around 300 nm. Now we are interested in the behavior at bigger tilt angle. The simulations show that the bandgap has a maximum width around 600 nm at tilt angle 40° – 50° (Fig. 5a). The bandgap is so width that masks completely SPR. According to [23] incident

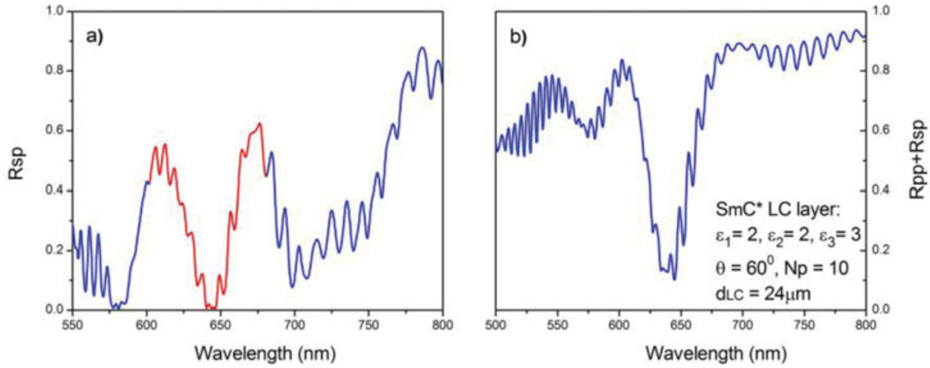

Figure 4. a) SPR excitation at $\theta = 60°$, with *s*- to *p*- polarization conversion, the last helix has a length *3p/4*; b) simultaneously with *s*- and *p*- polarization.

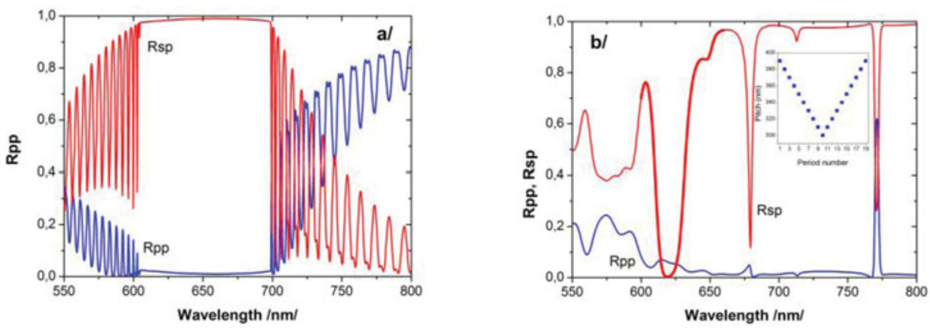

Figure 5. Rpp and Rsp as a function of wavelength for configuration: a) prism/ SmC* layer/ gold layer/ air with $\theta = 50°$, p = 300 nm, Np = 20; b) prism/ SmC* layer/ gold layer/ air with $\theta = 50°$, Np = 19, with pitch chirping (inset).

and reflected light with different polarization can couple producing Bragg reflection. Indeed, Bragg reflection is observed in the range 600 – 700 nm for incident light with *s*- polarization and for reflected light with *p*- polarization (Fig. 5a). It is possible to reveal SPR if the Bragg reflection is destroyed. This can be done if the pitch length is no constant. Keeping in mind that the feasibility of making LC layer requires the same pitch length on the two boundaries of LC layer, we assume linear decreasing of pitch length to the middle of the layer. Then the pitch length increases linearly as shown in the inset of Fig. 5b. We name non constant pitch length "pitch chirping" by analogy with gratings with non constant period. Fig. 5b shows the reflectivity of the structure with pitch chirping. There are no coupling and Bragg reflection for incident *s*- polarization and reflected *p*- polarization. Instead, SPR is observed (marked with thicker line) and excited with *s*- polarized incident light.

Discussion

Liquid crystal assistance in SPR excitation gives some advantages. The first is increased accuracy in SPR spectral shifting determination. SPR spectral position depends on the events arising on the interface gold/air, while the position of Bragg reflection depends only on LC parameters. This gives the opportunity to use Bragg reflection as a reference point again which resonance shifting can be detected. The method we propose is not related to the measurement of the spectral position of one specific point of the curve of plasmon resonance (usually this is the point with the highest slope). In our configuration it is feasible the spectral shift measurement to be accomplished by comparing the areas between the resonance curve and Bragg reflection curve from both its sides S_1 and S_2, as marked in Fig. 2. The lobes on the resonance curve, due to the interference, could be a problem for performing of this operation. However, there are a lot of software tricks giving the envelope of the resonance curve and neglecting the lobes. The accuracy of this method of measurement is much higher than the spectral position determination of one point of plasmon resonance curve.

This method of measurement can be used not only for C_hLC layer, but for SmC* with small tilt angle (Fig. 3). In this case the excitation is provided with *s*- and *p*- polarization illumination simultaneously. What method would be used (with assistance of C_hLC layer or SmC*) depends on the feasibility of LC layer preparation.

Let us discuss now the dependence of pitch length on SPR excitation. It is different for different tilt angles. The most interesting is the case of SmC* with $3° < \theta < 89°$. At what tilt angle the SPR excitation will be effective at desired wavelength depends on the pitch length and on the dielectric anisotropy of the LC molecule. It is possible to find an optimal condition for both tilt angle – pitch at which the resonance is excited with almost the same efficiency with *s*- and *p*- polarized light. It is important to note that in this case the resonances are spectrally shifted. This provides an opportunity for getting narrow SPR with simultaneous excitation with *s*- and *p*- polarized light what is illustrated in Fig. 4b. The narrow resonance increases the accuracy of shift determination because the curve slope is higher.

For SmC* assisted resonance excitation is not necessary to control the polarization of incident light what is other advantage of this structure. It is enough to use linearly polarized light and do not take into account the orientation in respect to the incident plane.

Pitch chirping in space domain reveals new options for destroying Bragg reflection and bandgap and for SPR excitation. We have shown that pitch chirping in space domain could be very effective method for SPR excitation with *s*- polarized light.

One can extend our study to take the information about the influence of pitch chirping in wavelength domain. We have studied it carefully. Despite our expectations were related to a clear influence on SPR excitation, we have observed a minor effect. As expected, the Bragg reflection totally disappears because the order of stratified media is destroyed. The study in this direction will be continued.

Conclusion

In this work we have studied theoretically LC assisted excitation considering Kretshmann configuration with LC layer between the prism and the metal layer. Till now the influence of LC layer properties on SPR, excited on the adjacent metal film, has not been investigated with exception of [17,18]. We have shown that the Bragg reflection and SPR can be observed simultaneously at SmC*with small tilt angle and at C_hLC with a properly chosen pitch. This gives an opportunity to propose new method of SPR shift determination with increased accuracy.

We have shown also, that SPR can be excited with assistance of SmC* layer for a variety of tilt angles. The features of this configuration are manifested by resonance excitation with *s*- and *p*- polarization simultaneously. In this case SPR is narrow what increases the determination accuracy of the resonance shift.

Pitch chirping in the space domain offers new possibility for SPR excitation assisted with LC layer. The question about the influence of pitch chirp on SPR is still open and the work in this direction will be continied.

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