Report on the thesis dissertation by Marcin Muszyńksi

Title: *Self-Organizing Photonic Potentials of Different Dimensions in Liquid Crystal Microcavities with Spin-Orbit and Light-Matter Couplings*

I have read with much interest the thesis dissertation by M. Muszyńksi. It explores a research field at the intersection of soft condensed matter, optics, and quantum physics that is reviving the study of liquid crystals (LC) through a series of breakthrough discoveries. For more than half a century, LC displays and optical devices have been developed using the classical theory of light-matter interaction. The LC order is represented with an asymmetric dielectric tensor, and polarized light propagation, transmission, and reflection across birefringent multilayer films can be simulated using well-established methods such as Berreman 4x4 matrix multiplication. Calculation, however, does not mean explanation. As complexity increases, new phenomena emerge that require a new interpretation framework. In 2019, the group of Prof. J. Szczytko at the University of Warsaw highlighted the formal analogy between the resonance modes of a planar optical cavity filled with a uniform nematic LC and the quantum states of a massive particle in a spin-dependent potential, enabling spin-orbit coupling (SOC). The analogy provided a powerful tool to understand how the mode polarization (analogous to particle spin) affects its wavevector (analogous to the momentum degree of freedom), and vice versa.

The voluminous dissertation by M. Muszyńksi reports on the massive effort undertaken over five years to explore the intriguing consequences of this analogy while also investigating new types of LC-filled microcavities (LCMC) and emergent optical phenomena. This work was developed along two main directions of research: (1) SOC effects on the lasing, exciton-polariton generation, and polariton Bose-Einstein condensation (BEC) of LCMC, (2) Spatially non-uniform, SOC-enabling, synthetic photonic potentials created by LC self-assembly. The PhD work addressed outstanding questions in condensed matter theory (e.g., room-temperature supersolid state, spin currents) in light of possible technological applications (e.g., tunable lasers with electrical control of energy, polarization, and propagation direction). As highlighted in the thesis, the advantage of using an LC as the cavity medium is that structure and polarizability depend on applied electric fields and polarization, respectively. As a consequence, SOC effects are greatly enhanced and can be tuned by modifying the LC alignment, e.g., by varying the cavity thickness or applying an external electric field.

Thesis structure and main results. The thesis is motivated in Chap. 1, whereas Chap. 2 provides background knowledge about LCs, polarization states, cavities, polaritons, BEC, and Rashba-Dresselhaus SOC in uniform LCMC. Chapter 3 introduces the materials and experimental methods used in the thesis. This chapter reveals a strong technical expertise in measuring reflection, transmission, and emission spectra. The ability to resolve photon energies E as a function of the in-plane wavevector components k_x and k_y is remarkable as it provides compelling visualization of the phenomena emerging in LCMCs [e.g., volume plots in Figs 2.8a and 8.6].

Chapter 4 demonstrates lasing from a uniform LCMC due to the weak interaction of cavity modes with photoluminescent pyrromethene molecules. The LC birefringence splits the modes into doublets with orthogonal polarizations, which can be tuned by applying a voltage to enable RDSOC. Lasing reflects the LC anisotropy in the xy-plane of the cavity surface, where y is the LC tilt direction. Cavity modes hybridized by the RDSOC show two degenerate minima (valleys) along the k_y direction and one minimum along the

k_x direction. The degenerate minima are equally spaced from k_y = 0 and have orthogonal circular polarizations. Above the lasing threshold, these minima produce two coherent beams that propagate along the *y*-axis and interfere with each other creating a characteristic stripe pattern with a modulated intensity alternating between orthogonal polarizations. This pattern is the photonic analogue of the persistence spin helix (PSH) in solid-state systems, a unique platform for spin manipulation in spintronics that allows coherent spin transport over extended distances.

Chapter 5 builds on the results of Chap. 4 to achieve exciton-polariton BEC. Excitons are provided by the photoluminescent polymer MeLLLP deposited on the cavity surfaces. Strong light-matter coupling leads to polariton generation, as verified by measuring the increase in effective mass, i.e., the concavity of the lower polariton branch of the dispersion curve. Far from the RDSOC regime, BEC was confirmed using a femtosecond pulsed laser that revealed a high spatial and temporal coherence of light emission in single-shot measurements, as well as a very low threshold. Due to LC tunability, BEC can be obtained at different energies and different polarizations by matching the MeLLLP emission band. In the RSOC regime, the lower polariton inherits the double-valley structure discussed in Chap. 3, so that BEC produces the PSH stripe pattern. Single-shot experiments also reveal that the phase of the condensate wavefunction is either randomly distributed across the stripes or forms phase vortices. These observations indicate that polariton BEC in a uniform LCMC produces a superfluid state, i.e., a quantum fluid of weakly interacting polaritons, with a PSH stripe-like modulation of polariton density. This polariton condensate effectively is a “supersolid” state of the type expected at low temperature for massive quantum particles.

Chapter 6 explores the consequence of a 1D periodic modulation of the RDSOC potential parallel to the cavity. The modulation is obtained in a chiral nematic (cholesteric) under hometropic anchoring conditions, such that a uniform lying helix (ULH) is created. The helix lies in *xy*-plane with uniform period (pitch) and direction (y-axis). Depending on the sample, the LC director tilts along the perpendicular direction *x* (type-A ULH) or in a direction of the *xy*-plane at a slight angle from *x* (type-B ULH). As expected, the periodic modulation of the RDSOC potential opens one or more band gaps in the energy dispersion curve as a function of the wavevector k_y. Under normal incidence and zero applied voltage, only the extraordinary polarization (*x*-axis) experiences the modulation of refractive index in type-A samples, whereas the ordinary polarization is not affected. As a result, band gaps appear only for the extraordinary polarization. The type-B helix, on the other hand, mixes the two polarizations. Interference and hybridization between bands with the same parity and different polarization produce a characteristic quadrupolar band structure that breaks the inversion symmetry along the *y*-axis. Namely, two distinct pairs of bands appear, alternating between circular polarization and wavevectors k_y with the same magnitude but opposite signs. The good agreement between the data and simulations based on symmetry arguments and hybridization methods (tight binding and semi-analytical model in Fig. 6.9-6.10) is worth noticing. Lasing from the type-B ULH preserves the quadrupolar band structure. Notably, the quadrupolar band structure produces an interference stripe pattern resembling the PSH.

Chapter 7 focuses on the 2D modulation of the RDSOC potential created by torons in a cholesteric LC under homeotropic anchoring conditions. Both isolated and hexagonally close-packed torons were considered. An isolated toron acts as an axially symmetric photon-trapping potential. Resonance modes reflected this symmetry in the intensity distribution as a function of the k_x and k_y wavevectors, but show a quadrupolar alternance of polarization deriving from the chiral nature of the torons. Notably, the lowest-energy and first excited modes have zero and maximum intensity for k_x, k_y = 0, respectively. Lasing from isolated torons reproduces these characteristics. Notably, it creates a vortex beam carrying non-zero angular momentum from the lowest-energy mode. Honeycomb lattices of close-packed torons show additional features such as a six-fold symmetry of resonance mode and band gaps in the dispersion curve. These features deserve a more detailed investigation.

The last chapter is devoted to the blue phase I (BP-I) found in chiral LCs. Blue phases provide a 3d periodic modulation of refractive index and produce characteristic optical resonances known as Kossel patterns, even in the absence of a cavity. In Chap. 8, Kossel patterns are beautifully captured by the 3d tomographic reconstruction of photon energies as a function of the in-plane wavevectors. The resonances are carefully identified in terms of 3d lattice parameters and aligned relative to an excitation laser to stimulate coherent emission. It was found that lasing can be obtained with both right- and left-handed circular polarization, but only the polarization matching the LC chirality responds to a reorientation of the BP-I lattice relative to the excitation laser. In a microcavity, the periodic chiral structure of the BP-I phase affects the resonance modes. However, the effect is very similar to that of 1d periodic cholesteric and, notably, does not involve Kossel lines.

Evaluation. Apart from the minor revisions listed below, the thesis work demonstrates an outstanding scientific capacity and maturity. The dissertation is well written and clearly organized. In each chapter, the state of the art and perspectives are discussed concisely but inclusively, revealing a solid background in condensed matter physics and photonics, and an awareness of the research frontier that is uncommon among PhD students. The experimental data are presented in a clear and consistent way throughout the dissertation. In each chapter, the main findings are highlighted and explained qualitatively utilizing general concepts, before providing theoretical details or quantitative analysis. The findings are discussed in light of previous works by providing an accurate, updated, and inclusive bibliography. Contributions from other group members or other research groups are duly acknowledged, particularly for sample fabrication (MUT, IBM Zurich) and theoretical analysis (CNRS).

The ideas developed in the thesis are original, and I am truly impressed by the scope and depth of the PhD work. Clearly, the student did not leave any stone unturned in his research for emergent optical phenomena in LCMCs. Although the quality of scientific research is high throughout the thesis, I believe that Chap. 5 unearthed a veritable scientific gem deserving immediate publication: a supersolid polariton Bose-Einstein condensate at room temperature.

For all these reasons, I sincerely recommend M. Muszyńksi for a **distinction**.

Sincerely,

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Minor revisions

(a) The permittivity model for the LC-pyrromethene mixture introduced on p. 60 seems to neglect anisotropy in molecular polarizability and emissivity. Polarization anisotropy is considered for the non-resonant part of the permittivity, but the strength and lifetime of the resonant oscillators do not appear to depend on polarization. Anisomeric dyes such as pyrromethene typically show polarization-dependent polarizability and tend to align with the LC director. Polarized fluorescence confocal microscopy (PCFM), for instance, utilizes this property to obtain a 3d reconstruction of the director alignment across thin LC films [e.g., see Lavrentovich, O.D. (2012). *Confocal fluorescence microscopy*. In Characterization of Materials, E.N. Kaufmann (Ed.). https://doi.org/10.1002/0471266965.com127]. As noted in the thesis, lasing from the LC-pyrromethene occurs only for the *H*-polarization (Fig. 4.6), most likely because the dye molecules are anisomeric and align with the local director. This can be directly verified using PCFM and implemented in the dielectric model by allowing the oscillators to have different strengths and lifetimes for the extraordinary and ordinary polarization.

(b) In Chap. 6, the director of the type-A helix is normal to the helix axis (*y*-axis in Fig. 6.5a). Therefore, the *y*-polarization (V-polarization in Fig. 6.5) corresponds to the ordinary polarization under normal incidence ($k_y = 0$), and the 1d periodic director structure has little or no effect on the dispersion curve (no bandgap in the inset of Fig. 6.5a). When a voltage is applied, the director structure remains qualitatively the same, i.e., periodic, chiral, and normal to the helix. The only difference from the zero-voltage case is that the director rotates with a non-uniform rate along the helix (Fig. 6.5b). Yet, a bandgap opens up for the *y*-polarization under an applied voltage (Inset of Fig. 6.5b). I do not understand why the non-uniform rotation rate should produce this bangdap. Instead, I think that the director structure changes qualitatively under an applied voltage, e.g., it becomes similar to that proposed in Fig. 6.10a. Observations with a polarizing microscope should be sufficient to clarify this point, e.g., type-A sample could become similar to type-B samples.

(c) In a cholesteric film with homeotropic anchoring conditions, the dark areas surrounding torons and ULH domains are known to be nematic, i.e., chirality is suppressed. It is therefore incorrect to assume that these regions are in the isotropic phase, as stated in the legend of Fig. 6.18d.

(d) In Chap. 7, the energy difference between the first two bound states changes sign as the toron diameter decreases (Fig. 7.8). This finding is interpreted from the optical point of view. However, it is also possible that the defect structure changes in response to changes of LC film thickness (e.g., see Ackerman et al, *Phys. Rev. E* 90, 012505 (2014)]. This possibility could be ruled out or verified using polarizing microscopy or PCFM. Also, torons and other solitons in homeotropically confined cholesteric appear very differently as a function of the depth of focus within the LC film [e.g., see Fig. 5 in Bruni et al, *Phys. Rev. Lett.* 134, 223804 (2025)]. For the optical characterization of torons, therefore, it is important to specify the criterion for setting the focus depth.