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Letter to Prof. W. Satula, Chairman of the Council, University of Warsaw

Report on the PhD work of Mr. Mateusz Król
(Institute of Experimental Physics, Faculty of Physics, University of Warsaw)

After studying of the thesis dissertation of Mr. Mateusz Król, I confirm the **positive** fulfilment of the requirements to achieve a doctoral degree:

(1) *the candidate has demonstrated both theoretical (deep exploitation of the Berreman model to reproduce experiments) and experimental expertise (implementation of cavities, monolithic and open, embedding liquid crystals and strongly coupled to different types of quantum materials) to conduct high quality scientific research and achieve unprecedented results.*

(2) *the originality of the experimental results and their applicability towards novel photonic devices* based on liquid crystals and emergent quantum materials is demonstrated.

Additionally, I think that the thesis deserves a **distinction**. The experimental and theoretical research work carried out by the PhD candidate, Mr. Mateusz Król, contains a high level of novelty and quality. The PhD book discusses a total of six research papers that the candidate has driven as a main contributor. These papers have been published in highly competitive journals, such as Optica (Chapter 3), Science (Chapter 2), Nanoscale and 2D Materials (Chapter 5) and Science Advances (Chapter 6).[1] Although this is not specified in the manuscript, all these works have been presented in many international conferences and, they have attracted the attention of the scientific community studying light matter coupling in novel materials. Additionally, all these works are recent publications, from 2019 to 2022.

In page 200, the candidate lists another 18 contributed scientific papers, where he holds 1st or 2nd authorship in 7 of these research works. This is an exceptional record, since it is unusual to finalise a PhD thesis with 24 scientific papers.

The first part of the thesis is focused on the light polarisation engineering in a planar microcavity via an embedded electrically driven liquid crystal (Chapters 2-4). The anisotropic control of the index of refraction in the planar cavity opens an interesting research avenue to explore the interaction of optically confined modes with different HV polarizations, such as the Rashba-Dresselhaus spin orbit coupling (Chapter 2), the implementation of skyrmion and meron textures in polarisation (Chapter 3) and the (newly demonstrated) control and annihilation of exceptional points in the photonic modes of the microcavity (Chapter 4).

As a natural continuation of the previous chapters, Chapter 6 shows the generation of exciton-polaritons formed by the strong coupling between the LC cavity photons and two-dimensional perovskite excitons (a material that has gained a great relevance in the recent years to achieve polariton lasing at room temperature an topological photonics). In this Chapter, the PhD

[1] The paper described in Chapter 4 has not been published yet, but it is expected to have a great impact as the work reported in Chapter 2, due to the novelty and interest of the experimental results (first demonstration of EPs annihilation originating from different momentum valleys in the LC cavity system).

candidate shows the formation of exciton-polaritons following a Rashba-Dresselhaus spin-orbit coupling dispersion relation, and a gap-opening (DA splitting) at the diabolical points allowing to control the Berry curvature via an external voltage (HV splitting).

Chapter 5 is slightly decoupled from the rest of the thesis, as it reports the implementation of a different cavity system, a cryogenic open cavity, strongly coupled to excitons in a monolayer of WSe_2. The experimental results show valley polarisation phenomenology of non-thermalised exciton-polaritons. The results are very well timed and perfectly aligned with the current interest on open cavity system and exploiting the polarisation properties of transition metal dichalcogenide monolayers.

It is worth to mention that all these research works have been implemented in collaboration with renowned international collaborators in the exciton-polariton community such as the groups of P. G. Lagoudakis, M. Potemski, T. Liew, and D. Sanvitto. Also, it is noticeable that the results in Chapter 6 have inspired others to follow such lines of investigation (group of Prof. T. Gao in Tianjin University, China).

An important characteristic of a good PhD book is (1) its capacity to help future students to understand fundamental concepts of physics and (2) provide detailed descriptions of the thesis methodology in a clear and didactic way. Particularly, in Chapters 1 and 2, Mr. Król has done a fantastic work to explain the fundamentals of light-matter coupling, polarisation, and topology in light (which is fundamental for the comprehension of Chapters 3-6). The Berreman model, taking a primary importance along the whole book (except in Chapter 5, where the model it is not required), is very well described in Chapter 2,[2] accompanied with clear equations and graphical representations. The candidate has given quite insightful descriptions on the experimental setups (such as the hyper spectral tomography, key tool in most analysis presented along the book), the configuration of the microcavities (either monolithic or open) and the different materials involved in the experiments (liquid crystals, transition metal dichalcogenides, perovskites).

In general, the PhD candidate has done a very good work to provide results very systematically, he has identified the relevant parameters in simulations/experiments and systematically studied their impact/role in specific experimental/theoretical problems. Additionally, Mr. Król has demonstrated outstanding scientific representations skills in every possible plot style (colour and vectorial maps, 3D graphs, etc) and a unique skill to create pedagogic scientific illustrations (such as Figs. 1.1, 2.8, 2.16, …, 5.3, 6.1, etc).

Maybe, given the complex representation of certain 2D/3D effects, it could have been helpful to provide video animations (for example, accessible via an external link) where one could see the evolution of the photonic (or polaritonic) modes as a function of other parameters (HV/DA splitting, cavity lengths, etc); this could have helped to better understand and appreciate the presence of diabolical and exceptional points, the Berry curvature of the modes, etc.

In the following, I provide a deeper description of the main Chapters of the thesis:

In **Chapter 2**, it is interesting to study the three regimes of degeneracy (N,N), (N+1,N), and (N+2,N) between x- and y-polarised photonic modes, respectively. The (N,N) regime shows the build-up of the degeneracy (diabolical) points in specific positions along the k_y axis (k_x=0) of the momentum space (depending on the TE-TM splitting of the system, tunable in the LC cavity via the LC molecules rotation), yielding two conical intersections.

The (N+1,N) regime, with different mode parity, shows the Rashba-Dresselhaus like spin orbit coupling due to the inclusion of the term $2\alpha \mathrm{k_y} \hat{\sigma}_z$ in the Hamiltonian, which yields a dispersion relation with two paraboloids separated along the ky axis. The coefficient α depends on the

[2] I encourage the candidate to provide open access to the Berreman simulation codes, since I imagine that it would be relevant for the community.

two involved modes numbers, the cavity length, the ordinary and extraordinary indexes of refraction and, of course, the orientation of the LC molecules.

The (N+2,N) regime, equal mode parity, similar to the (N+1,N) regime, considers anisotropic masses along kx and ky directions, and the Hamiltonian is characterised by the terms $\delta_{xy}k_x k_y\hat{\sigma}_y$ and $(\delta_x {k_x}^2-\delta_y {k_y}^2)\hat{\sigma}_x$. Above resonance, there are four diabolical points (with total topological charge of zero) appearing along the axis kx and ky and depending on the HV detuning and the terms δ_x and δ_y, respectively. Setting the HV detuning to zero, brings the four diabolical points to kx=ky=0.

Chapter 3 studies the different polarization classes (skyrmions, merons) that emerge from the regimes (N,N) and (N+2,N) in the LC cavity, the unique tunability of this system (HV splitting) offers the possibility to study novel polarisation states of light in the LC cavity (second order antimeron polarization texture). These polarisation states are characterised by the parameters: vorticity (v), phase (gamma) and the topological charge (Q). In the (N,N) regime, the linearly [circularly] polarised light transmitted through the LC cavity, the experiments show the formation of a meron/antimeron [second order meron] spatial polarization in real space. In the (N+2,N) regime, the real space polarization patterns shows antimerons instead of merons (case of (N,N)) and vice versa.

Chapter 4 is devoted to the study of non-Hermitian physics; in the first part, the candidate introduces a simple 2D Hamiltonian in the circular polarisation basis (containing a diabolical point in kx=ky=0), then including an additional non-Hermitian term ($+i\xi\hat{\sigma}_x$). The non-zero value of ξ induces a separation of two exceptional points (along kx, ky=0), linked via the Fermi arc (degeneracy between the upper and lower modes of the model), this arc in momentum space is characterised for having a non-defined phase. Additionally, studying the S3 value of the bottom and lower branches (for non-zero value of ξ) we see that the exceptional points are highly polarised with S3 values of -1 and +1, respectively.

In the (N,N) regime, the inclusion of the non-Hermitian term $+i\xi\hat{\sigma}_x$ in the Hamiltonian (out of resonance case for the two HV modes) induces the creation of four exceptional points linked via two Fermi arcs (the two initial diabolical points present a topological charge of +1/2).

Finally, as a natural extension, in the (N+2,N) regime when the HV modes are above resonance, the theoretical analysis of the non-Hermitian term shows how the 4 diabolical points (DPs) become 8 exceptional points (EPs), linked via 4 Fermi arcs. Remarkably, in this case, the two pairs of exceptional points have opposite topological charge of +/-1/2 (zero total topological charge).

The experimental implementation of this Hamiltonian was done via the inclusion of a very low concentration (0.2%) of pyromethene 580 organic dye in the liquid crystal. The experiments show 8 exceptional points and how they can be annihilated in a controlled manner via the HV detuning, yielding a ring in momentum space after their annihilation. The candidate indicates that “this is the first demonstration of annihilation of exceptional points originating from different valleys in momentum space”, however the paper is yet to be accepted for publication in the moment when this report has been finalised.

Chapter 5 takes a totally different path with respect to the rest of thesis chapters, and implements a cryogenic open cavity strongly coupled to a transition metal dichalcogenide (TMDC) monolayer of WSe2. After giving a very detailed explanation about the optical properties of TMDCs and their technological relevance, the candidate explains in a very didactical way how to implement a light-matter coupled system composed by an open cavity and a deposited monolayer of WSe2. Many different simulations show the optimal composition of mirrors to strongly couple with the monolayer excitons. Then, the experiments are focused to characterise the WSe2 properties of emission (photoluminescence and reflectance). In the last part, the strong coupling between the planar open cavity and the WSe2 is demonstrated (for different number of monolayers and with/without hBN encapsulation, values around 25-40 meV are observed); under quasi-resonant excitation conditions, the candidate observes the (weak) valley polarisation physics of the lower polariton mode. The candidate analyses the circular degree of polarisation in the emission under different polarisation excitations. The circular degree of polarisation and intensity is also studied for different cavity detuning (different polariton Hopfield coefficient). The biggest circular degree of polarisation is observed when polaritons are resonant with the trion transition.

Chapter 6 retakes the LC cavity, this time including (room-temperature, two-dimensional) perovskite-excitons to strongly couple to the LC cavity photons. The cavity is one step further engineered to achieve DA splitting: the rubbing directions of the mirrors are at 45 degrees. The photoluminescence properties of the perovskite-excitons are studied in detail, its refractive index is obtained via ellipsometry measurements. After knowing the relevant experimental parameters of the system, the Berreman model predicts the strong coupling versus the cavity distance for the two relevant polarisations (from ~120 to ~40 meV for increasing distances), and the coupling strengths of the different photonic H/V modes anti-crossing the exciton mode. Experiments under non-resonant excitation allow to compare and validate the predictions from the Berreman model.

The Berreman model also studies the impact of the DA splitting in the LC cavity ($\frac{1}{2}\Delta E_{DA}\hat{\sigma}_y$ term in the cavity Hamiltonian), where the Rashba-Dresselhaus regime (N+1,N) regime shows a Dirac-cone gap opening (as a function of the ratio *l/L,* Fig. 6.11, see also Fig. 6.9). Calculating the Berry curvature on the LC photonic system, the model shows the opposite curvature values of the valleys (Fig. 6.9).

With the inclusion of the 2D perovskite strongly coupled to the photonic modes in the LC cavity with 0 [30] degree rubbing alignment, the experiments show a crossing [anti-crossing] of the (N+1,N) modes (Fig. 6.13d [6.14d or 6.15d]). The experiments also show the gapped Dirac cones and the behaviour of the Stokes parameters and Berry curvature on the polariton bands. The Berry curvature in the gapped Dirac points is clear (unclear) for the lower (upper) polariton band, and it is studied for different HV splitting values, both experiments and theory show a great agreement.

Maybe, provided the amount of novelty in this thesis, in **Chapter 7** I would have expected a more specific projection of the work rather than a summary, addressing next potential experiments or challenges, such as for example the inclusion of LC cavities with patterned lattices in order to explore complex topology phenomena that go beyond the results of C. E. Whittaker, et al., Nat. Photonics **15**, 193 (2021) or quantum phenomena, such as those expected from the recent results of L. Ferrier et al., Phys. Rev. Lett. **129**, 083602 (2022).

In the following, I include some comments and questions, most of them motivated by scientific interest/curiosity on the PhD topic.

Chapter 2

- On the **rubbing** process to promote the alignment of the LC, how is this rubbing process done? Is it applied directly on the DBRs or in the glass substrate (where the DBR layers are deposited)? Does the rubbing affect the photonic losses? Could a circular and micro-controlled rubbing induce local twisting in the LC? I wonder how much the rubbing can be engineered to control the alignment of the LC in the mirrors interface. What is the accuracy in the relative angle orientation between the rubbing directions? Apart from the rubbing technique, can you think of a different technique, such as a transversal electric field to orient the molecules with the controlled angle in the xy plane?
- It seems that there is an incompatibility between Fig. 2.9 labels (on distances between optical elements) and the central equation described in 2.32: y2=p2 and x1=p2-p1-y1, is this possible?
- In page 40, the liquid crystal photonic cavity is presented, I thought that at some point along the manuscript the PhD candidate would discuss the dynamic behaviour of the crystal, under a time-varying electric signal drive. It would have been interesting to know the timescales of such driving and the intrinsic limitations of the microcavity (mostly 2D size and cavity distance), acting as a capacitor.
- On the properties of liquid crystals, following results from other groups (for example, Appl. Phys. Lett. **94**, 071112, 2009), I wonder whether it is possible to push the boundaries to operate them at more extreme temperatures, or even whether other types of materials with electrically tuneable birefringence could offer similar or even bigger effects than the chosen liquid crystal of this thesis.
- In Fig. 2.18 it is not evident why the different H modes cross the V modes at different molecule rotation angles, this is due to which reason? Also, Fig. 2.2 seems to have a problem with the caption of the figure (15 degrees in bold, then indicated normal incidence, it seems an unedited caption from Fig. 2.18).
- Eq. 2.40 could have a typo in the denominator n_e is not squared, is this correct?
- The dispersion relation representation in Fig. 2.23 is in the same conditions (RD resonance) as the case represented in Fig. 2.24c (resonance for 2.46 V): why the dispersion relation represented in Fig. 2.26d does not resemble the one in the previous Fig. 2.23? Also, the bottom branch in Fig. 2.23b,c, for negative ky (kx=0), the branch should be sigma- (negative S3), then in Fig. 2.26f, supposedly representing the same conditions, the bottom branch has red colour, why? Also, from Fig. 2.23c, when representing E-<E> versus in-plane momentum, I would expect to see E-<E>=0, a whole line of degeneracy, along the kx axis, at ky=0, this is not the case in Fig. 2.26e, why?

Chapter 3

- Fig. 2.22a and Fig. 3.4a do not show the same type of dispersion relation since the first case consider isotropic masses in kx and ky directions and the second case takes anisotropic masses (Eq. 3.7), what would be the representation of the two H and V modes in 3D (energy, versus kx-ky)? I am trying to reconcile the plot in Fig. 3.4b (an ellipse at constant energy) with Fig. 3.8a (two concentric circles).
- Figure 3.5 presents some inconsistency in the legend, the birefringence increases from -0.4 to 0.4 in panels a-e, g-k and m-q, according to the text.
- The dependence between the TE-TM splitting and the birefringence in Fig. 3.6 is very interesting: could the candidate explain why the TE-TM splitting amplitude (|TE-TM

splitting|) is bigger for negative birefringence? I would like to know whether there is any intuitive explanation.

- In page 77 there might be some wrong references: Fig. 3.14a <-> Fig. 3.14d referring to experiments and, maybe, Eq. 3.1 <-> 3.7.
- From Fig. 3.14 it is difficult to imagine how big is the spot resulting from the cavity transmission, what is the intensity distribution? Also, what is the degree of depolarisation (sqrt(S1^2+S2^2+S3^2)<1) map? It would be interesting to see this map in Fig. 3.14 and Fig. 3.22.

Chapter 4

- In the introduction to non-Hermitian physics, the candidate describes that EPs can be used are sensors. This indeed seems an interesting proposal. Could the candidate describe how his LC cavity device could operate as a sensor or how it could exploit any other functionality in a straightforward way? Following this question, a recent work (Phys. Rev. Lett. 129, 083602 (2022)) proposes to use EPs in other to enhance the spontaneous emission of emitters, this system is composed by a grating on top of a Bragg mirror. Could the candidate observe any similar phenomenon? In the conclusions of this paper, the Authors write: "the effect of EPs on spontaneous emission of single quantum emitters is a salient perspective to explore the new regime of cavity-quantum electrodynamics for novel single photon sources.", could the candidate give his opinion on the matter?
- I understand that the non-Hermiticity in the system (terms in Eq. 4.21) is induced via the inclusion 0.2% of pyromethene 580 organic dye. Could the candidate describe what was the criteria to estimate the percentage of dye (translated into number of losses)?
- The criteria for estimating the position of the EPs in momentum space in Fig. 4.7a seems a bit unclear, could the candidate explain how their coordinates and their corresponding error were determined?
- The text indicates that two samples (with different cavity thicknesses) were implemented for these experiments, showing very similar dependence of the Fermi arc versus degree of non-Hermiticity (Fig. 4.7i). Was the organic dye concentration the same for both samples? Was this concentration value critical for changing radically the experimental results?

Chapter 5

- I wonder whether the liquid crystal could be interfaced with an open cavity, maybe an approach could be enclosing a double cavity, coupled to different materials (TMDCs, perovskites or other alternatives).
- In the open cavity sketch of Fig. 5.3, it is not possible to observe whether a mesa has been used in one of the mirrors to be able to approach the mirrors to closer distances. How big are these mirrors?
- About the open cavity system: the Oxford Instruments MicrostatHe-R cryostat used for the experiments is a vacuum cryostat: even if Fig. 5.12 shows a detail of the cavity holder, it is not clear how one can spatially scan to locate the monolayer between the mirrors, it is not clear either what is the optical access to the microcavity (apparently not visible in the figure, where a lateral view is shown). Is this cavity configuration constraining the maximum NA accessible for experiments? How were the DBRs pasted to assemble the cavity? In page 134, you indicate that the DBRs are grown in VIGO, could you give some more details about this?

- In general, it is difficult to fit the oscillations in the reflectivity appearing on the sides of the microcavity stopband (see Fig. 5.13a,b), this is something that happens in most cases: do you know what is the origin and how one could get a proper fitting of these oscillations too?
- Have you characterised the cavity length stability over time? Have you quantified its fluctuation due to vibrations (He flow, etc).
- In Fig. 5.26, the experimental data points in the dependence of Rabi splitting versus number of monolayers does not follow the square root projection, can you comment?
- In Fig. 5.28 is not clear whether in that case the monolayer is non-encapsulated.
- Is there any possible model to reproduce the experimental results in Fig. 5.32 (this is degree of circular polarisation versus energy, for several polariton detunings)?
- In the last part of the chapter, I was missing some discussion on the input-output curve of the system, it was not possible to increase the pump power and try to observe a change in the thermal distribution of polaritons?

Chapter 6

- Do the perovskite properties suffer any modification due to the application of a strong electric field (polarization, lifetime, other properties)?
- In Fig. 6.4, the 2D perovskite is represented on the surface of one of the mirrors, then in Fig. 6.10 the twisted nematic cavity represents the two mirrors with rubbing directions at 45 degrees. I wonder whether the 2D perovskite presence on the surface of one of the mirrors degrades the rubbing effects, could the candidate discuss?
- In the simulations with the twisted nematic LC, in the regions with thickness *l*, how is the rotation angle varying along z, linearly? This question applies to both theory simulations and the experimental device (if known).
- In page 182, it is mentioned that three samples are implemented with different rubbing angle orientation (30, 45 deg and parallel), from this, I understand that the reproducibility on the cavity thickness is extremely good, can the candidate comment on this? Where these samples requiring many trials until achieving the good ones? What is the typical duration of such samples (days, weeks)?
- In Eqs. 6.4 and 6.5, the theoretical analysis of the different coupling strengths of the H and V modes follow an unexpected trend, for example: in the V mode, the coupling strength shows modes (1) < (2) > (3), in the H mode, the coupling strength shows modes (1) > (2) > (3), why the coupling strength does not decrease in the vertical case? It also seems that the coupling strength is bigger with modes anti-crossing at higher k-vector, why? Finally, in Eq. 6.6 [6,7] (V [H] mode), we see that the experimental coupling strengths of the modes are (1) < (2) [(1) < (2) < (3)] follow a different trend for the modes, why? Additionally, what is the fitting error of these quantities?
- From the hyper spectral tomography, you can reconstruct the two dispersion bands, and an additional polarization tomography determines the total polarisation state of the bands. Given the spectral width of the bands, how is it the data processing done to associate the given degree of polarisation (DOP) to the band (see pseudo color scale S3 in Fig. 6.16a).
- Since the calculation of the Berry curvature imply derivates on the DOP maps, is it relevant to discuss the numerical errors in the momentum space distribution of the Berry curvature? There are points in momentum space where S1/S2 is zero/nonzero and S2, this makes the function Psi (Eq. 6.15) infinite, how is the derivative numerically calculated in that point? Is this creating any problem?
- How is the experimental Berry curvature affected by unpolarised light? In Fig. 6.18, first row on experimental results, it would be interesting to plot sqrt(S1^2+S2^2+S3^2).

- It remains a bit unclear why the observation of the berry curvature in the upper branch is so unclearly observed, the Stokes parameter maps are reasonably well following the theory predictions, but Fig. 6.19d clearly deviates from the two theory approaches. The candidate associates this experimental Berry curvature problem to the higher wave vectors of the gapped Dirac cones and the limit of numerical aperture, however, not even the sign of the Berry curvature is following the theory prediction, and same problem should hold for the lower branch.
- The Berry curvature units are wrong in several graphs (6.19, 6.20l, 6.22m,n), this should be microns^2, and the units are absent in the colour scale of Fig. 6.20a, 6.22f.
- Is there any intuitive explanation to understand the clear asymmetry of the Berry curvature centre of mass versus detuning HV for low DA splitting (Fig. 6.20k)?
- From the previous question on the numerical errors from the experimental calculation of the Berry Curvature, the candidate could discuss the (absent) error bars in the Fig. 6.22n, blue circles.
- The last two points in Fig. 6.22n disagree with theory since (following the book) the points of the gapped Dirac cones lie (partially) outside the angular collection of the objective. This is visible in the panels a-f, where the red curvature area on the right seems to be cut by the rim of a circle. This is not the case of the blue curvature area, which does not seem cut by the maximum angular collection of the objective. Why are the grey circle (depicting the maximum angular collection of the objective) and the clipping of the red curvature area not coinciding? Is this clipping arising from a different reason?
- In Fig. 6.22 we see the maximum berry curvature for different theory cases a smaller AD splitting increases the curvature value. Beyond the evident demonstration of a nonzero Berry curvature, maybe the candidate can discuss what value of maximum Berry curvature is optimum for some application? It does not seem clear.
- Following the previous comment, maybe the candidate can comment on experiments where one could measure the Berry phase from the Berry curvature. How would be this potential experiment?

Chapter 7

- Provided the degree of novelty shown in the different experiments, are you participating in the writing of any patent derived from your research? Related to this, do you think that the results reported in your thesis have the potential to create a start-up on photonic devices based on liquid crystal cavities and room temperature emitters?

In Madrid, 3rd of November of 2022.

Firmado por Carlos Anton Solanas el día 03/11/2022 con un certificado emitido por AC FNMT Usuarios

Carlos Antón-Solanas