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2022

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The interaction between wave and matter:
twisted light interacting with nanophotonics &
surface acoustic wave interacting with polymer and nanoparticles

By

Qingjun Wang

A dissertation submitted in partial satisfaction of the requirements for the degree of

Doctor of Philosophy

in

Engineering – Materials Science and Engineering

in the

Graduate Division

of the

University of the California, Berkeley

Committee in charge:

Professor Jie Yao, Chair
Professor Junqiao Wu
Professor Boubacar Kanté

Fall 2022

The interaction between wave and matter:
twisted light interacting with nanophotonics &
surface acoustic wave interacting with polymer and nanoparticles

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By
Qingjun Wang

Abstract

The interaction between wave and matter:

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Doctor of Philosophy in Engineering – Materials Science and Engineering

University of California, Berkeley

Professor Jie Yao, Chair

Light can carry spin angular momentum (SAM) and orbital angular momentum (OAM). OAM, as a new degree of freedom, has played an important role in lots of fields and leads to promising applications. Understanding how the SAM and OAM interact and convert to each other can pave the road of OAM application. To understand the mechanism, I explored spin to orbital angular momentum conversion in:

1. self-assembled concentric ring of gold nanoparticles and block co-polymer system, where the circularly polarized light is converted to light carry OAM by the plasmonic effect of Au.
2. the photonic crystal mimicking hexagonal structure system, where we try to understand the spin-orbit interaction from K space and real space point of view.
3. valleytronic system to see whether the involvement of OAM will change the selection rule.
4. twisted waveguide nanowires generating OAM.

These fundamental research projects will help with a better understanding about the new twist degree of freedom OAM in the nano-photonics system and will help pave the road of OAM applications.

Besides the optical wave, I also explored the acoustic wave in one of my projects by using interdigital transducer generated surface acoustic wave. Surface acoustic wave has similarity with respect to surface plasmonic polariton. And in this project, I explore how the surface acoustic wave can interact with matter and mediate the assembly of nanoparticles and polymer system. We found the surface acoustic wave can help form micro-sized structure and help with the formation of hierarchical structure. The research project reveals the potential power of using surface acoustic wave to control assembly and to explore the fundamental material science topics.

To my family.
And to the road I walked along.

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Acknowledgement

Five-years is like a blink for me. I still remember when I first came to UC Berkeley, I was very curious and excited about my future academic life here. And all sudden, it's near the end of my PhD.

I would like to first thank my advisor Professor Jie Yao who offers me the opportunity to study and do research in his group. His sound advice, careful guidance, and the support are invaluable to me during the whole process of conducting my research and writing this dissertation. And there are tons of memories flying by my mind when I write this: Prof. Yao is not only a good academic advisor but also provide a lot of warm support and help for the living of the students. The PhD degree in Berkeley is hard, especially for me who changed a major from EE to MSE. It's his support which encourages me to break through barriers by barriers to reach the destination.

And I hope to thank Prof. Junqiao Wu and Prof. Boubacar Kanté for serving in my committee and thank their generous help during my study here.

My academic life would not be complete without my colleagues and friends in the Yao Group and in UC Berkeley. And I cannot thank all my collaborators and friends enough. Without their help and inspiration, I cannot finish these exciting projects. And I feel very lucky to know these amazing people and to survive and thrive with them. And it's my proudness and happiness that we can work and have fun together. Although my study at UC Berkeley comes to an end, the friendships will last forever.

And I hope to thank my fiancé Zhaoyu who is always by my side for my PhD years, giving me lots of support and love, and help me become a better version of myself. He is an invaluable gift I received in Berkeley.

Finally, let me thank my parents and my grandparents for their unconditional support, not only for the last five years but for my entire life. They are and will always be my role model, my inspiration, my courage, and one of the most important reasons why I will always hope to go beyond myself to achieve something big.

Introduction

Throughout my Ph.D. research, I have had the chance to investigate many wave-matter-interaction-related topics in the photonic and acoustic systems. In this dissertation, I will break down different aspects of my research and show the SAM and OAM interaction (SOI) when light is interacting with different photonic systems, and the surface geometry manipulation when acoustic wave interacts with matter. The dissertation will include 5 chapters, covers the 5 projects. The order of them is a little bit different from the abstract and here I ordered them according to their completeness level. For each chapter, I will include some background information to prepare the readers for understanding the main results, and for example, some very important topics like OAM and SOI are included in Chapter 1 and Chapter 2 background sections correspondingly.

Chapter 1 is about the OAM generated from a bottom-up fabricated self-assembled concentric-rings structure. The mechanism is that the circularly polarized light can be divided into radially polarized light and azimuthally polarized light, each carrying OAM. When circularly polarized light shines on the concentric metal ring, the azimuthally polarized light will excite the plasmonic mode and be reflected and only radially polarized light can transmit. And this chapter will cover the fabrication method and the characterization of the device, and covers the optical setup used to characterize the device and the measurement results.

Chapter 2 is about the OAM generated from honeycomb photonic crystal structure. The previous research experimentally shows that the OAM is linked to pseudospin (which is the sublattice degree of freedom). My research is to verify that the generation of OAM is in fact a SOI phenomenon. This SOI phenomenon is mediated by pseudospin in real space and mediated by local berry curvature in reciprocal space. And this chapter will cover the design process, the fabrication, and the characterization of the photonic crystal device, and covers the optical setup used to characterize the transmission and band structure and the near-field optical setup to characterize the eigenmode.

Chapter 3 is about the surface acoustic wave interacting with polymer and nanoparticles. The surface acoustic wave can be formed by pairs of interdigital transducers. In the middle region of the pairs of interdigital transducers, there will be standing wave formed due to interference. And the standing wave will form potential nodes and antinode, which gives the force to drag the polymer and nanoparticles to the node's region. I successfully formed microstructure and nanostructure of polymer by using surface acoustic wave. This reveals the potential in acoustic wave aided bottom-up fabrication. This chapter will include the working mechanism of device, the characterization of the device, the experimental setup, characterization of formed structures.

Chapter 4 is about the OAM excite exciton. In this chapter, I explore whether OAM can influence the selection rule of exciton excitation. The valleytronic experimental results of OAM interacting with transition metal dichalcogenide monolayers (TMDC) shows that OAM generated from spatial light modulator barely will influence the exciton selection rule. And by analyzing, I believe this is caused by the size mismatch between the exciton wavefunction and the OAM diameter size. And

by using Cu_2O , which has huge exciton size, and surface plasmonic device to generate nano-sized OAM (below diffraction limit), theoretically the selection rule will be changed by OAM.

Chapter 5 is about the OAM generation from twisted nanowire. OAM mode can be generated when light injected and interact with the twisted nanowire. And this chapter will cover the mechanism equations, the simulation results, and preliminary experiments.

For chapter 1-3, the projects have been finished. And for chapter 4-5, I am halfway in the research and proposed the future direction. I hope my finding covered in chapter 1-3 can help readers gain some new knowledge in this field and chapter 4-5 will pave the road for the future discovery.

I love all these projects and they all made me super excited when I moved forward step by step. They are the invaluable markers of my days and nights from 2017 to 2022.

Chapter 1 Orbital angular momentum from self-assembled concentric nanoparticle rings

1.1 Background

1.1.1 OAM

Beam carrying OAM has the intensity property and phase property as shown in Figure 1-1. If a charge-coupled device (CCD) is put perpendicularly in front of the beam propagation direction, a donut intensity profile can be observed for the cross-sectional view of the beam. That's because of the phase singularity in the center of the beam caused by the helical wavefronts. It's similar to that on the north pole of the earth, time cannot be defined. And in the cross-sectional view of the beam, there is an azimuthal phase term $e^{i\ell\phi}$ which indicates the phase increment around the vortex core.

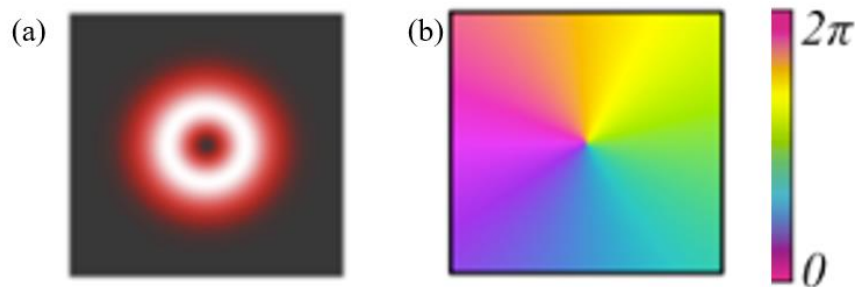


Figure 1-1 Cross-sectional view of a beam carries OAM. (a) intensity profile (b) phase distribution

Since the discovery of OAM around 30 years ago, OAM has been used in a lot of promising applications¹, from optical communication (OAM Multiplexing)², Manipulate particle in optical tweezer³, astronomy^{4,5}, OAM in chemical syntheses⁶, imaging⁷, to quantum entanglement⁸⁻¹⁰. Figure 1-2 shows some of these applications. For the application of optical communication, beams carrying OAM can dramatically increase the data transporting speed by using orbital angular momentum degree of freedom (ℓ different channels multiplexing). And this has not only been experimentally verified in laboratory², but also been tested in real world. Therefore, the applications of OAM are a flourishing field, and we can predict it will be applied in our life someday not too far away.

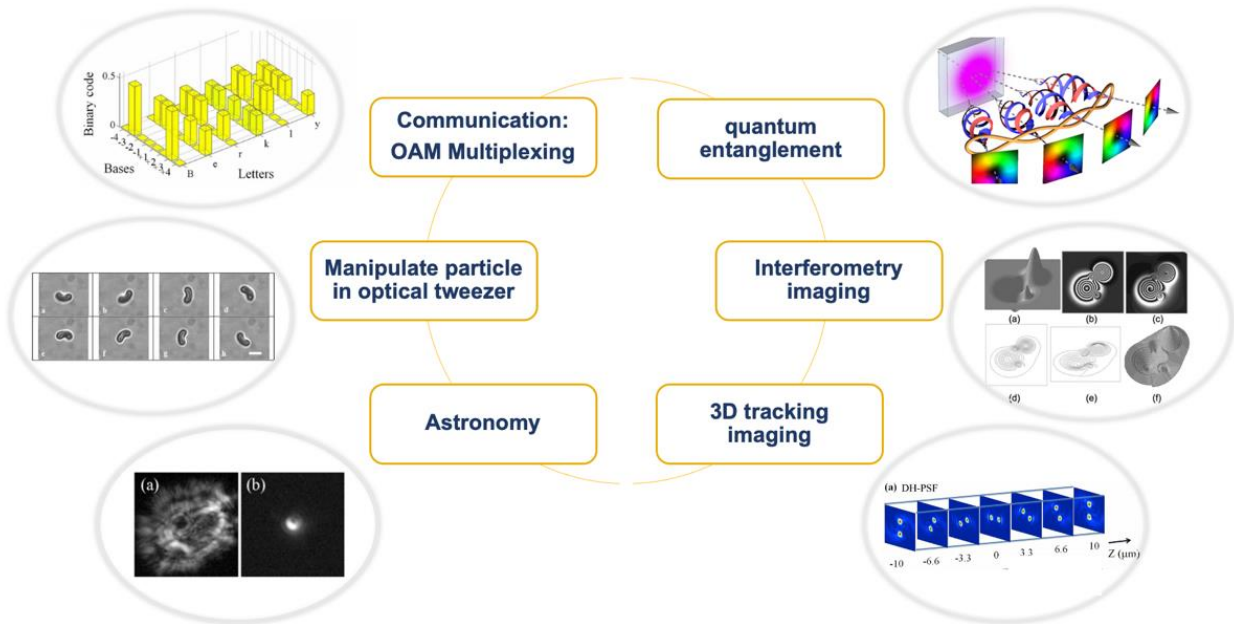


Figure 1-2 application of OAM

1.1.2 Wire-grid polarizer (WGP) by plasmonic mode

Wire-grid polarizer is one of the simplest linear polarizers. It consists of many fine parallel metallic wires placed in a plane. The separation between wires must be less than the wavelength of the incident radiation(subwavelength). In addition, the width of each wire should be small compared to the spacing between wires. WGPs mostly reflect the non-transmitted polarization and can thus be used as polarizing beam splitters as shown in Figure1-3¹¹.

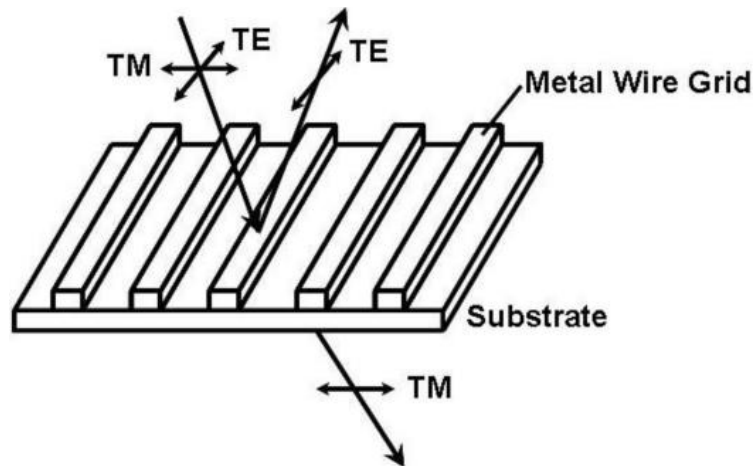


Figure 1-3 schematic image of wire grid polarizer

Metal wire grid grating can be designed to function as polarizer because of the existence surface plasmons (SPs) which consists of surface plasmon polariton (SPP) and localized surface plasmon

(LSP). Only the component of the laser polarization perpendicular to the grooves couples to surface plasmons propagating along the grating^{12–14}.

Electromagnetic waves that have a component of their electric fields aligned parallel to the wires will induce the movement of electrons along the length of the wires, couple to surface plasmons (SPs). Since the electrons are free to move in this direction, the polarizer behaves in a similar manner to the surface of a metal when reflecting light, and the wave is reflected backwards along the incident beam (minus a small amount of energy lost to Joule heating of the wire).

For waves with electric fields perpendicular to the wires, it can only excite the localized surface plasmon (LSP), and the electrons cannot move very far across the width of each wire. Therefore, little energy is reflected, and the incident wave can pass through the grid. In this case the grid behaves like a dielectric material.

1.2 Abstract

Ring-shaped nanostructures can focus, filter, and manipulate electromagnetic waves, but are challenging to incorporate into devices using standard nanofabrication techniques. Directed self-assembly (DSA) of block copolymers (BCPs) on lithographically-patterned templates has successfully been used to fabricate concentric rings and spirals as etching masks. However, this method is limited by BCP phase behavior and material selection. Here, we demonstrate a straightforward approach to generate ring-shaped nanoparticle assemblies in thin films of supramolecular nanocomposites. DSA is used to guide the formation of concentric rings with radii spanning 150-1150 nm and ring widths spanning 30-60 nm. When plasmonic nanoparticles are used, ring nanodevice arrays can be fabricated in one step, and the completed devices produce high-quality orbital angular momentum (OAM). Nanocomposite DSA simplifies and streamlines nanofabrication by producing metal structures without etching or deposition steps; it also introduces inter-particle coupling as a new design axis. Detailed analysis of the nanoparticle ring assemblies confirms that the supramolecular system self-regulates the spatial distribution of its components, and thus exhibits a degree of flexibility absent in DSA of BCPs alone, where structures are determined by polymer-pattern incommensurability. The present studies also provide guidelines for developing self-regulating DSA as an alternative to incommensurability-driven methods.

1.3 Introduction

“Top-down” nanofabrication has historically driven progress in nanodevice design and application, but it is limited by intrinsic machine and material constraints¹⁵. Some of these limitations—in particular, fabrication of small features and complex geometries—have been addressed with a hybrid approach, where simple lithographic templates guide the self-assembly of finer features^{15–19}. Directed self-assembly (DSA) of block copolymers (BCPs) has successfully produced a variety of functional nanodevices, including capacitors, memory storage, and photonic crystals^{20–22}. DSA of curved features, however, has not achieved the same level of success, despite the technological importance of ring and concentric ring patterns. Ring-based devices include zone plates, nanolenses, and optical ring resonators; certain memory storage devices, transistors, and sensors

also use nanoscale ring features^{23–29}. Compared to assembly along linear patterns, DSA of curved nanostructures has distinct requirements. In order for a lamellar or cylindrical BCP microdomain to form a ring, the polymer chains need to compress along the microdomain's inner edge and stretch along its outer edge^{30,31}. This asymmetric distortion is energetically unfavorable. In solution, spontaneous formation of rings or toroids requires that the BCP blocks be chemically modified to experience self-attraction^{32,33}. BCP rings and spirals have been observed within cylindrical confinement, though only a narrow set of BCP block ratios have been studied^{34–37}. The confined BCP assemblies often exhibit structural degeneracy, typical of a system trapped far away from its free energy minimum state.

Polymer nanostructures typically serve as etching masks for inorganic materials with the requisite electronic, magnetic, or optical properties. Alternatively, BCPs and inorganic nanoparticles (NPs) can be blended into nanocomposites, which offer additional advantages, including single-step fabrication and nanoparticle size- and arrangement-dependent properties^{38–42}. Indeed, ordered nanocomposites with long-range alignment across one, two, and three dimensions have been obtained with DSA. However, curved features have been notably absent. Incorporated particles increase the bending moduli of their host microdomains, and thus increase the energy penalty associated with curved features^{43,44}. To assemble successfully, the polymer matrix must accommodate both the particles and the template curvature. BCP-based supramolecules, which consist of small molecules non-covalently bonded to BCP chains, have demonstrated unparalleled flexibility in navigating interfacial interactions, NP incorporation, and geometrical confinement^{42,45–51}. Though supramolecular microstructures are visually similar to pure BCPs, small molecule redistribution makes the supramolecule sensitive to assembly kinetics. The microdomain morphology, periodicity, and orientation can all be selected by tuning the system's assembly kinetics and pathway; the system is agnostic to commensurability criteria⁵².

Here, we investigate DSA of supramolecular nanocomposites within shallow (50 nm) concentric ring templates (Error! Reference source not found. **A, B**). We hypothesize that self-regulation of the small molecule distribution will enable the supramolecular matrix to accommodate unfavorable curvature, confinement, and NP fillers. Indeed, the system successfully assembled across a variety of patterns and conditions. Using a one-step assembly process, a 10x10 array of nanoparticle rings was fabricated and prepared for integration into optical devices, all within minutes. The rings produce orbital angular momentum (OAM) effectively, while offering more versatility than nanodevices fabricated via a top-down milling technique⁵³. We note that the ring-shaped nanoparticle assemblies are insensitive to the pattern feature widths, molecular weight, and NP loading, but quite sensitive to the film thicknesses and NP size. A simple model of the curved microdomains confirms that small molecule redistribution can substantially decrease the energy of bending, and should be further explored to establish supramolecular nanocomposites as a rapid and flexible DSA platform.

1.4 Results and discussion

The supramolecule was constructed using a BCP, polystyrene-*b*-poly(4-vinylpyridine) (PS-*b*-P4VP), and the small molecule 3-pentadecylphenol (PDP), which hydrogen-bonds with the 4VP

monomer (schematically shown in Figure 1-4 A)⁵⁴. The PDP:4VP ratio was 2, so at least half of the small molecules were unbonded at any time. The PS-*b*-P4VP(PDP) supramolecules were blended with 3 vol% alkyl-passivated NPs, which have favorable interactions with the P4VP(PDP) microdomains. Two nanocomposites were studied, forming cylindrical PS microdomains with equilibrium periodicities of 31 nm and 57 nm, respectively (**Table 1-1** Compositions of supramolecular nanocomposites NC1 and NC2.). The first nanocomposite composition, denoted as NC1, consisted of a 19 kDa PS block and a 5.2 kDa P4VP block. The second nanocomposite (NC2) was constructed from a BCP containing 50 kDa PS and 17 kDa P4VP. The NP core chemistry has no effect on the self-assembly process, so iron oxide nanoparticles were used for the systematic DSA studies, and gold nanoparticles were used to fabricate the optical devices⁵⁰. The ring templates have three trenches and two mesas, with feature widths from 100-200 nm and an etch depth of 50 nm (**Table 1-2** Dimensions of lithographically-patterned templates P100, P150, and P200.).

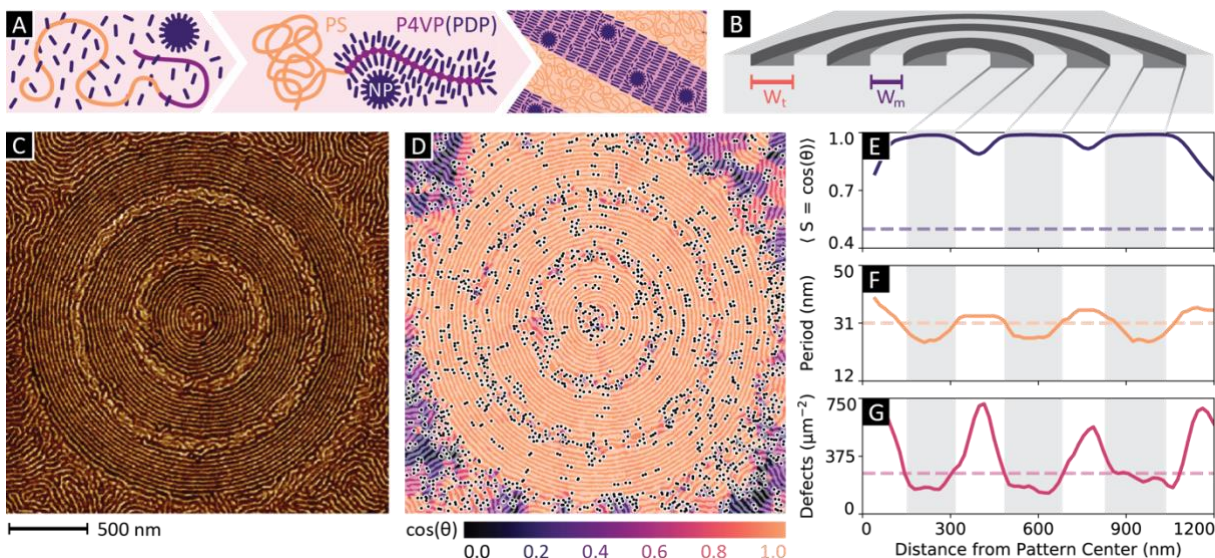


Figure 1-4 DSA of supramolecular nanocomposites within shallow (50 nm) concentric ring templates. [A] Schematic of the supramolecular nanocomposite system, including PS-*b*-P4VP BCP, hydrogen-bonded PDP small molecules, and nanoparticles with PDP-compatible ligands. The assembled structure is shown to the right [B] Schematic of the concentric ring template, with a trench width (w_t) of 200 nm, mesa width (w_m) of 150 nm, and a 50 nm etch depth. [C] AFM micrograph of the nanocomposite self-assembled on the ring-patterned substrate. Lighter microdomains are PS; darker microdomains are P4VP(PDP)+NPs. [D] The same micrograph shown in false color, with microdomains colored by the order parameter $S = \cos(\theta)$. Defects are labelled with black dots. [E] Order parameter, [F] microdomain period, and [G] defect density values are plotted radially from the pattern's center. The dashed lines show control values from an unpatterned sample; the shaded bars give the trench positions.

1.4.1 Characterization of assembly quality

We first examined the DSA of one supramolecular nanocomposite within one concentric ring template. The template, selected to be compatible with the requirements of OAM, consists of three 200 nm trenches separated by two 150 nm mesas (Error! Reference source not found. B)⁵³. The supramolecular nanocomposite samples were prepared as described previously⁵⁰. Briefly, after spin-coating the composite onto the patterned templates, films ~60 nm in thickness were annealed under chloroform vapor to a solvent fraction of 35%. The annealing process took less than 10 minutes. During annealing, the PS cylinders curved into toroids, with near-perfect alignment within the three trench regions (Figure 1-4 C-E). The microdomains on the raised mesas also became aligned, albeit to a lesser degree, with a high density of defects and a slightly larger periodicity. To quantify local microdomain alignment in the annealed samples, we define the order parameter $S = \cos\theta$, where θ is the angle between a polymer microdomain and the orientation of the underlying template. Quantitative image analysis of an atomic force microscopy (AFM) micrograph supports our qualitative observations: the radially-averaged S is above 0.96 within the three trenches, very close to a perfect value of 1.0, and above 0.88 within the two mesas. Past the outer trench, the aligning effect of the pattern decayed gradually, and the features became randomly oriented ($S \approx 0.5$) within five microdomain periods.

On a flat substrate, the supramolecular nanocomposite forms isotropically-oriented cylindrical microdomains with a period of 31 nm (**Figure 1-8**). When the patterned substrate was used, however, the microdomain periodicity was modulated by the underlying ring pattern. Within the trenches, the periodicity decreased up to 19% to form cylinders with a period of 25 nm (Figure 1-4 F). Complementary microdomain swelling is observed on the mesas and surrounding substrate, where the periodicity expanded up to 13% to form microdomains with a period of 35 nm. Periodicity changes within 10% of the equilibrium are routinely observed in BCP DSA whenever polymer chains must stretch or compress to achieve commensurability with the template^{30,55}. However, oscillations between increased and decreased periodicity, as shown in Figure 1-4 F, are rather rare. In templated BCP systems, microdomain stretching or compression starts at the pattern's edge and propagates inward¹⁶. Here, the increased periodicity past the template's edge suggests that the supramolecular system achieves commensurability through a different mechanism. We hypothesize that, instead of relying on entropically-unfavorable polymer chain deformation, free PDP is expelled from the trenches and collects on the mesas. Regions with lower concentrations of PDP have smaller equilibrium periodicities and lower bending moduli, so the confined supramolecules can potentially expel PDP until they fit neatly within the trenches. This global self-regulation process is consistent with previous results observed using linear trench templates^{41,52}. These observations are also consistent with studies of homopolymer-BCP blends, where homopolymers were shown to distribute nonuniformly within the BCP to reduce the entropic penalties associated with domain curvature and cross-sectional mismatch⁵⁶⁻⁵⁸.

Device-driven applications of DSA require high-quality pattern transfer with low defect densities^{18,59,60}. Within cylindrical morphologies, defects are defined as microdomain branches or terminal points⁶⁰. For this specific system, the unpatterned control sample has an average of 262 defects per μm^2 , distributed randomly across the sample surface (Figure 1-8). The templated

sample, conversely, has a spatially-patterned defect distribution. Just as the microdomain period oscillates across the trenches and mesas, the defect density decreased by more than 30% within the trenches, then surpassed the equilibrium defect density on the mesas and surrounding substrate (Figure 1-4 D, G). The increased defectivity is not due to poor ordering, because the order parameter S remains above 0.78 across the entire pattern. Within the regions of high defectivity, ie. the mesas, the microdomain periodicity is increased, and the aspect ratio of the PS cylinders is substantially reduced. These results suggest that the increased P4VP(PDP) fraction on the mesas caused a cylinder-to-sphere morphological transition. This morphological transition may thus be used to infer the spatial distribution of PDP (Figure 1-5).

1.4.2 Optical properties of the templated concentric rings

DSA of the supramolecular nanocomposite is not limited to a single pattern and can be readily scaled up. Here, we fabricated a 10x10 array of patterns on one wafer. After assembly, the concentric rings of the supramolecular nanocomposite were exfoliated from the silicon substrate (**Figure 1-5A**). The exfoliated films can subsequently be transferred anywhere, such as a fiber tip, demonstrating the versatility of the fabrication process. Here, the exfoliated film modulated circularly-polarized light to produce orbital angular momentum (OAM). Optical modes carrying OAM, typically represented by a helical wavefront and a phase singularity, have a broad range of applications, including communication, particle manipulation, astronomy, imaging, and quantum optics^{2-4,8-10,53,61,62}. To measure optical transmission through the patterned NP assembly and the OAM order of the transmitted light, a high-magnification microscopy setup was designed (Figure 1-5D). The set-up includes two magnification paths: a 25x magnification path to find the sample and a 250x magnification path to observe the OAM. Images of the sample under the two optical paths are shown in Figure 1-5B and 2C. When circularly-polarized light was transmitted through the nanocomposite with 3 vol% gold NPs, the assembly successfully produced light with OAM, identifiable by its donut-shaped intensity distribution and its good agreement with full-wave simulations, as discussed below. The clear array of donut shapes in Figure 1-5C suggests that OAM generation with these DSA devices is a novel approach with improved resolution and flexibility compared to previous, top-down-fabricated nanodevices⁵³.

In order to understand the origin of OAM, a circularly-polarized beam of light may be represented as a linear superposition of radially-polarized and azimuthally-polarized vortex beams. In a cylindrical coordinate system, a left-hand or right-hand circularly-polarized beam (LHC or RHC) with a topological charge of 1 may be expressed as:

$$\begin{aligned}\vec{E}_{LHC} &= e^{j\varphi} (\vec{e}_r + j\vec{e}_\varphi) \\ \vec{E}_{RHC} &= e^{-j\varphi} (\vec{e}_r - j\vec{e}_\varphi)\end{aligned}$$

where e_r and e_φ are the amplitudes of unit vectors in the radial and azimuthal directions, respectively. The gold NP concentric ring structures cause higher transmittance of the radially-polarized component, so light leaves the nanocomposite device as a radially-polarized vortex beam.

A finite element method (FEM) simulation of the device was used to further study the generation of OAM. The intensity and phase distributions of the electric field in the transmission region are shown in Figure 1-5F and 2G. The donut-shaped intensity and the vortex phase distribution again confirm the generation of Laguerre-Gaussian modes with OAM. Since the incident light was circularly-polarized light with no vortex, the donut-shaped intensity distribution on the transmission side indicates phase singularity and the generation of OAM with a topological charge of ± 1 , depending on the handedness of the incident circularly-polarized light. Simulation details are provided in the Experimental section.

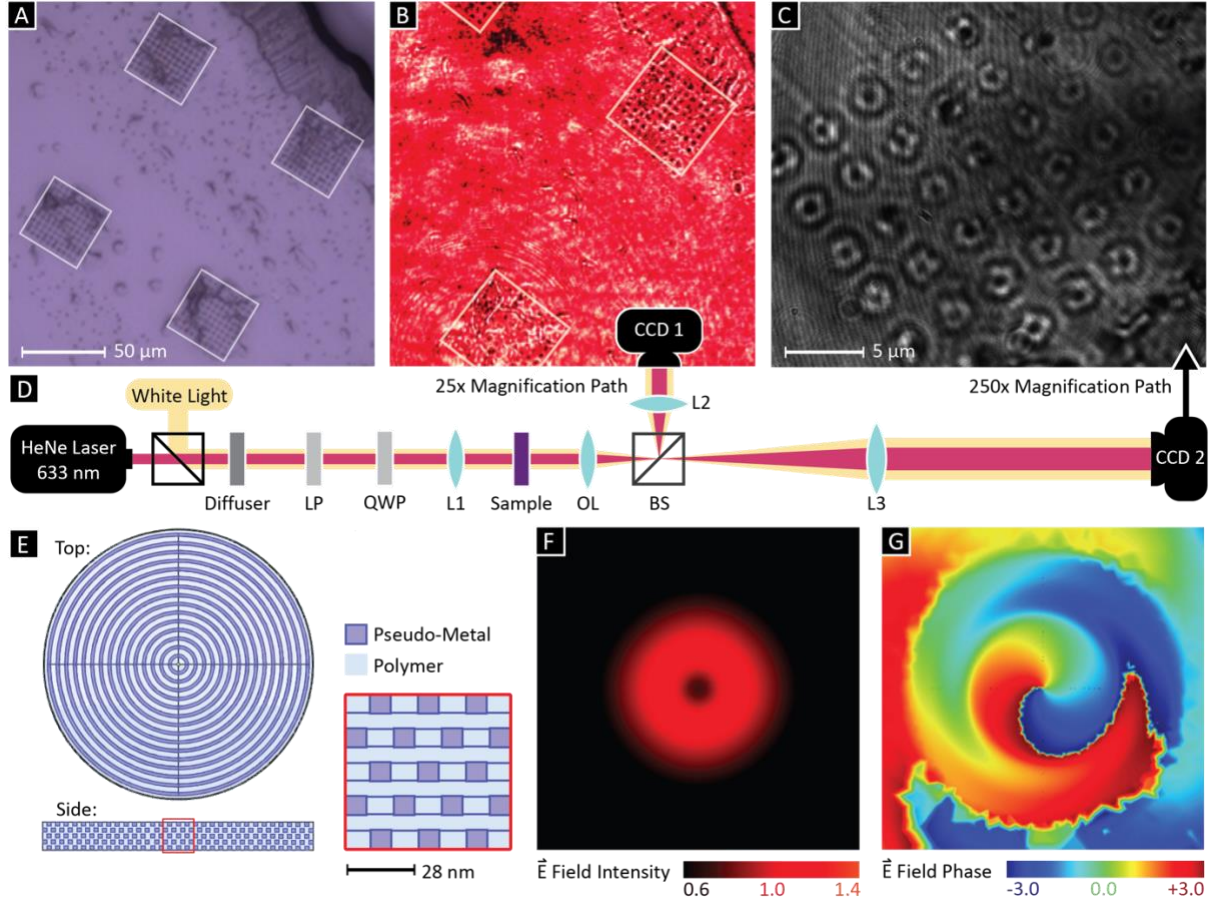


Figure 1-5 Optical Properties of the Templated Concentric Rings. [A] Optical microscope image of the exfoliated sample. Device arrays are boxed in white. [B] Image captured by CCD1, with a magnification of 25x. [C] Image captured by CCD2, with a magnification of 250x. The characteristic donut-shaped patterns of OAM are clearly seen. [D] Schematic of the optical setup used to test the devices' interactions with transmitted light. Light passes through a diffuser, a linear polarizer (LP), a quarter wave plate (QWP), and a first lens (L1) before it reaches the sample. Beyond the sample, light is focused with an optical lens (OL) before being split by a beam splitter (BS). The first optical path uses a second lens (L2, $f = 100$ mm) to achieve 25x magnification. The second optical path passes through a different lens (L3, $f = 1000$ mm) to achieve 250x magnification. [E] Top and side view of the staggered five-layer concentric ring model used for

FEM simulation. Regions with NPs were treated as pseudo-metallic elements. [F] FEM simulation result showing the intensity distribution of the transmitted electric field. [G] FEM simulation result showing the phase distribution of the transmitted electric field.

1.4.3 Design flexibility of supramolecular DSA

In comparison to DSA of BCP-only systems, where the template dimensions dictate the microdomain periodicity, the supramolecular nanocomposite system is not constrained by pattern-polymer commensurability. To this end, the same nanocomposite composition was used for DSA within two additional ring patterns: P150 ($w_t = 150$ nm and $w_m = 200$ nm) and P100 ($w_t = 100$ nm and $w_m = 100$ nm). As shown in **Figure 1-6**, the supramolecular nanocomposite assembled successfully in both P150 and P100. Compared to the original ring pattern, P200, the new patterns have thinner trenches with tighter radii of curvature. The periodicity oscillations noted within P200 are observed in both smaller patterns, consistent with the proposed PDP redistribution mechanism. In the area surrounding P100, the periodicity converged to the system's equilibrium value within 200 nm, or about 6 microdomain periods. Unlike the assemblies on the two larger patterns, the defect density of the nanocomposite guided by P100 was effectively constant across the patterned area. The cylindrical microdomains are aligned across the trenches and mesas, forming a continuous series of rings and spirals. The presence of periodicity oscillations suggests that the supramolecule was still incommensurate with the underlying trench pattern. However, for this pattern, much of the excess PDP appears to have been ejected from the pattern entirely, reducing the PDP accumulation on the mesas. This suggests self-regulation across an astonishing distance of 500 nm, or 20 microdomain periodicities.

The ring templates were used to guide another nanocomposite system, NC2, with an equilibrium periodicity of 57 nm. Due to its longer polymer chains, NC2 is more prone to chain entanglement than the first nanocomposite system, NC1. Nevertheless, the larger microdomains were successfully guided by all three ring patterns (Figure 1-6K, 1-10, 1-11). NC2 is more sensitive to pattern curvature than NC1, and the quality of the assembly decreased both across the set of patterns, from the largest (P200) to the smallest (P100), and within each pattern, from the outermost trench to the innermost. The NC2 nanocomposite film on P200 had the same general trends—oscillating periodicity, defect density, and order parameter—observed in the series of NC1 samples. Within the more tightly-curved P150, the periodicity decrease within each trench was not balanced by a periodicity increase atop the mesas, and the defectivity increased overall (Figure 1-10). By P100, the microdomains were well-ordered only within the outermost trench, and the defectivity was high across the full pattern (Figure 1-10). These results suggest that, while the supramolecular nanocomposite can accommodate a variety of confinement widths, it remains sensitive to feature curvature.

In addition to tight pattern curvatures, the supramolecular nanocomposite cannot accommodate nanoparticles larger than 10 nm or film thicknesses less than 50 nm (Figure 1-11). The NP size constraint is driven by the unfavorable polymer chain stretching that increases with particle size^{40,43}. At a certain NP size, the NP-related chain stretching is expected to dominate the template-related chain stretching, and the supramolecular system accommodates the NPs but ignores the

underlying pattern. This is the flip side of the system's robustness: it almost always achieves an ordered state, but may do so at the cost of the applied constraints. The film thickness constraint provides another insight into the system's assembly pathway. The system only exhibits global PDP redistribution when the film thickness is slightly greater than the trench depth and stays intact across the mesa features. Films thinner than 50 nm have relatively poor ordering because the supramolecules are fully isolated within the trenches, and cannot use the unconfined mesa regions as PDP reservoirs (Figure 1-11). In this case, the blended system resembles a pure BCP, and would require more annealing to achieve alignment with the underlying pattern³⁰.

1.4.4 Local and global self-regulation in supramolecular nanocomposites

The energetics of bending microdomains are further explored with a simple physical model. For an ideal solid, the elastic strain energy of small deformations is proportional to the squared volume change, $|\Delta V/V_0|^2$. A model is constructed for one repeat unit of the nanocomposite, including two PS block regions, two P4VP(PDP)+NP regions, and two block-block interfaces (**Figure 1-7A**). To simplify the volume change calculation, the morphology is approximated as lamellae rather than cylinders; this approximation will strictly underestimate the bending energy⁶³. Upon microdomain bending, both block interfaces are defined as neutral axes with fixed lengths. The supramolecules are pinned at the interface, and the inner block is compressed while the outer block is stretched. The volume change of each block is calculated as a function of the curvature radius and microdomain widths. The resulting strain describes a system with no volume redistribution, equivalent to a BCP system. Model details are provided in the Supplementary Information.

Next, two possibilities for volume redistribution are considered. In the first, excess PDP from the compressed P4VP(PDP) block is used to fill the neighboring stretched P4VP(PDP) block. The net volume change decreases, particularly at tight curvatures. In the second case, excess PDP is allowed to relieve the P4VP(PDP) and PS microdomains, further decreasing the strain. The three PDP redistribution modes are plotted together in Figure 1-7B. The presence of PDP in the PS domains has been shown experimentally, though this simple model does not account for the different mixing enthalpies of PDP with the two blocks. In reality, PDP redistribution is determined by many competing energies: the polymer chain conformational entropy, the PDP mixing entropy, the block-block and block-NP interfacial enthalpies, and any enthalpies associated with PDP-polymer mixing. The model presented here only includes one of these terms, but it still captures two important trends. First, the demand for PDP redistribution increases with curvature and with microdomain width, ie. molecular weight (Figure 1-7B-C). Second, local PDP redistribution cannot entirely alleviate the bending energy. This suggests that the system will rely on global PDP redistribution—redistribution well beyond the scale of individual microdomains—to further decrease the energy cost of curved features. This is consistent with experimental observations: the sizes of the PDP-rich and PDP-poor domains are set by the template feature size, not the microdomain periodicity (Figure 1-9D-E).

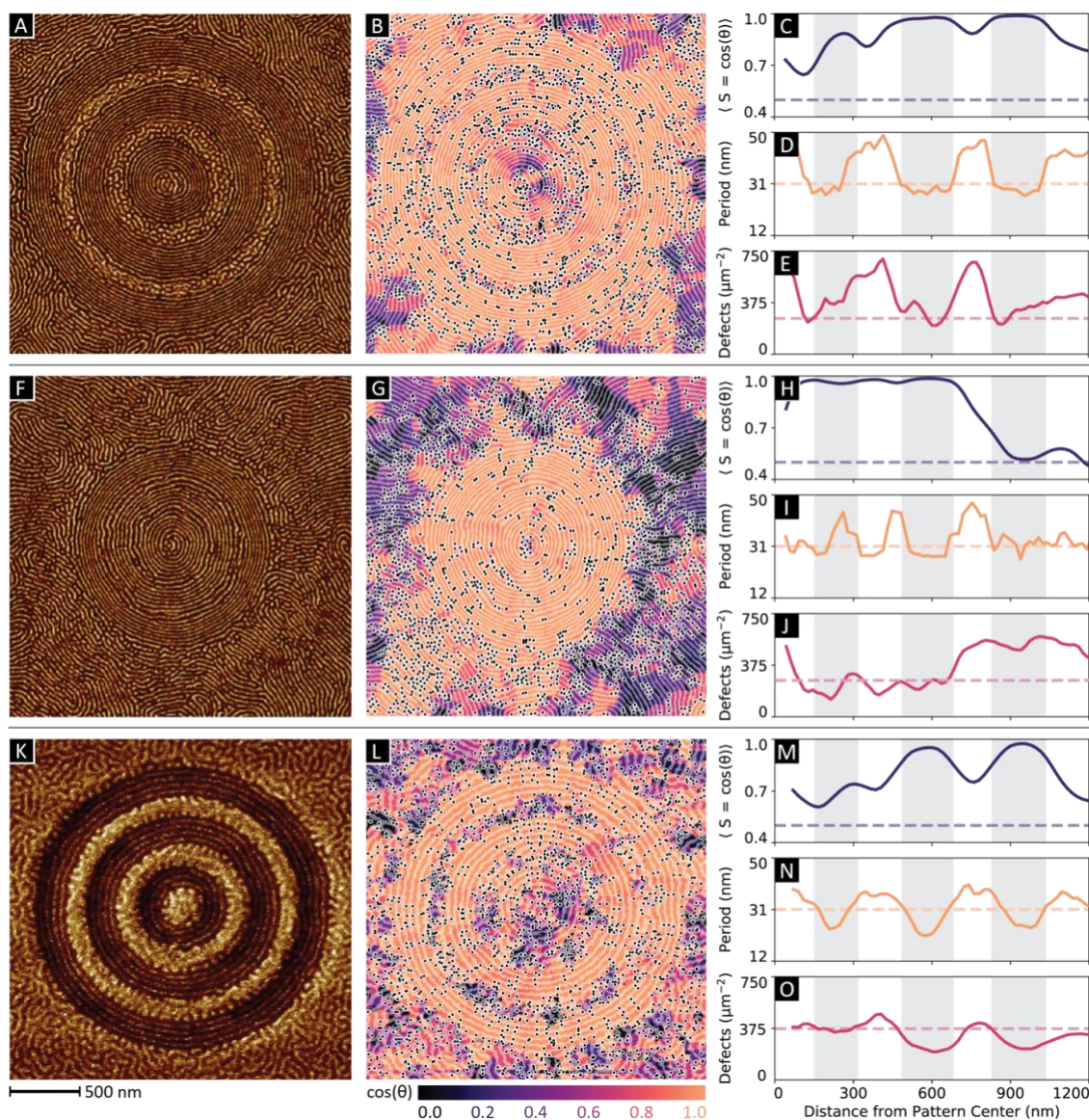


Figure 1-6 [A] AFM micrograph of the nanocomposite self-assembled on P150. [B] The same micrograph shown in false color, with microdomains colored by the order parameter $S = \cos\theta$. Defects are labelled with black dots. [C] Order parameter, [D] microdomain period, and [E] defect density, plotted radially from the pattern's center. The dashed lines show control values from a non-patterned sample; the shaded bars give the trench positions. [F-J] The same measurements and calculations are repeated for a nanocomposite sample assembled on P100. [K-O] The same measurements and calculations are repeated for the higher-molecular weight nanocomposite NC2, assembled on P200.

The supramolecules' propensity for small molecule redistribution leads to a set of behaviors not typically seen with BCPs. As described in the previous sections, the supramolecular

nanocomposites exhibit rapid assembly kinetics, large (~15%) periodicity changes, and spatially-patterned morphological shifts. They also respond differently to pattern-polymer incommensurability. In BCP DSA, the number of microdomains within a trench increases with a characteristic “stair-step” pattern as the trench width is continuously increased^{19,30,55}. When BCPs are assembled within an incommensurate template, the microdomains reliably stretch or compress into the state nearest to the equilibrium periodicity. Degeneracy is only observed along the step verticals, where stretched states of n microdomains are energetically similar to compressed states of $n+1$ microdomains. The supramolecular nanocomposite deviates significantly from this BCP behavior. As a visual comparison, Figure 1-7D-E shows all observed states of nanocomposites NC1 and NC2 within patterns P100, P150, and P200, plotted with the commensurability-defined step pattern. The supramolecular assemblies have a high degree of structural degeneracy: for every trench width, 3 to 5 different states were observed. For example, a 140 nm trench, which is 4.5 equilibrium periods wide, was filled by 4, 5, or 6 cylinders. This behavior suggests that the energy penalty from distorting the microdomains is too small to drive the system into one equilibrium-like state. We posit that the free energy landscape of the supramolecular nanocomposite is not funnel-shaped, but is instead fairly flat with multiple local minima. Energy landscapes of this sort can be navigated by tuning the system’s assembly kinetics, and allow access to a diverse range of non-equilibrium states⁶⁴. Importantly, the flat energy landscape effect is not localized to the trenches, and extends across the whole area affected by PDP redistribution. As a result, the unconfined mesas exhibit the same degree of structural degeneracy as the trenches (Figure 1-14).

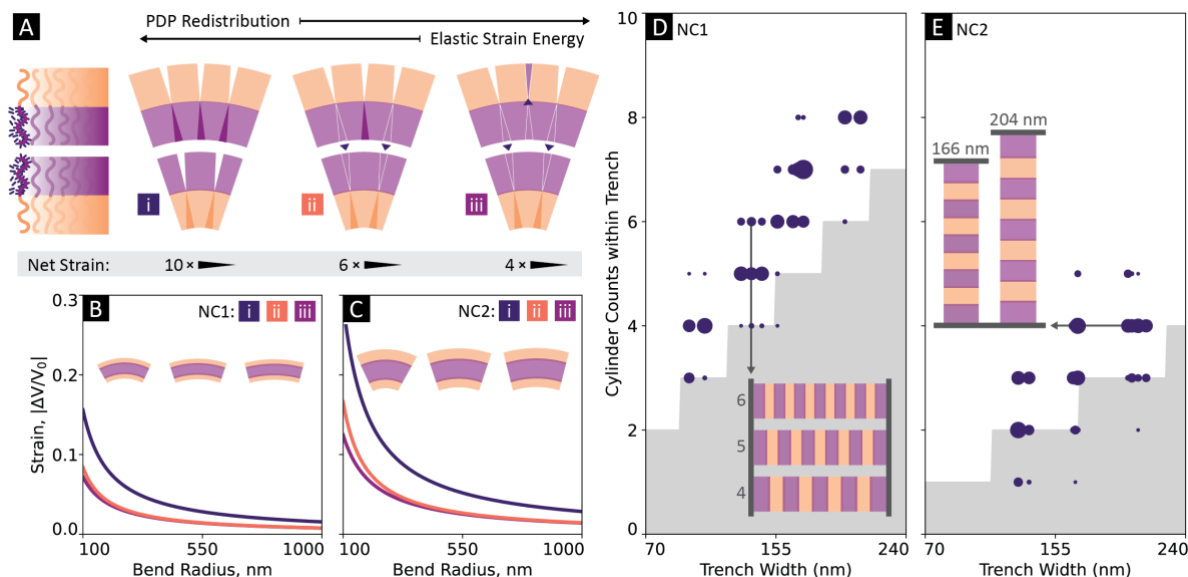


Figure 1-7 [A] Schematic diagram of microdomain bending and three possible response modes: i, no PDP redistribution; ii, PDP redistributes within neighboring P4VP(PDP) microdomains; iii, PDP redistributes within all neighboring microdomains. PS domains are drawn in orange, P4VP(PDP) domains are drawn in purple. [B-C] Strain vs. curvature radius for assemblies of NC1 and NC2, respectively, under redistribution modes i, ii, and iii. [D-E] All observed states of the nanocomposites NC1 and NC2, respectively, on patterns P100, P150, and P200. The gray stair-step pattern shows the commensurate state closest to the equilibrium periodicity for all trench

widths. The marker size is scaled by the number of times a state was observed; ie., the most common states are drawn with the largest markers.

1.5 Conclusion

DSA of a supramolecular nanocomposite is a straightforward method for self-assembly of concentric NP rings within an organic matrix. Oscillations in the microdomain order parameter, periodicity, and defect density indicate that the supramolecule responds to incommensurability through small molecule rearrangement rather than polymer chain distortion. These versatile thin film devices are not limited by supramolecule periodicity or trench width due to the cooperative nature of the self-assembly process. Additionally, the use of small, discrete NPs instead of solid features presents new possibilities for incorporating interparticle coupling interactions into device designs. High-magnification microscopy and an FEM simulation were used to confirm that the patterned NPs successfully produce OAM. Successful DSA within a variety of curved patterns suggests that the same technique can be transferred to other geometries. The observed structural degeneracy is consistent with the hypothesis of a relatively flat free energy landscape, which is particularly suitable for kinetic control. Overall, these studies introduce a robust method for fabricating thin film devices for advanced optical applications.

1.6 Experimental section

Materials

PS(19 kDa)-b-P4VP(5.2 kDa) (PDI = 1.06) and PS(50 kDa)-b-P4VP(17 kDa) (PDI = 1.15) were purchased from Polymer Source, Inc. 3-Pentadecylphenol (90%–95%) was purchased from ACROS Organics. Chloroform (Amylene as preservative) was purchased from Fisher Scientific. Hydrogen tetrachloroaurate (III) trihydrate ($\text{HAuCl}_4 \cdot 3\text{H}_2\text{O}$, Sigma-Aldrich, 99.995%, trace metals basis), oleylamine (Sigma-Aldrich, technical grade, 70%), tert-butylamine-borane (TBAB) complex (Aldrich, 97%), 1,2,3,4-tetrahydronaphthalene (tetralin, anhydrous, 99%), 1-dodecanethiol (Sigma-Aldrich, $\geq 98\%$) methanol (Fisher Scientific, analytical reagent grade), chloroform (Sigma-Aldrich, contains 100-200 ppm amylenes as stabilizer, $\geq 99.5\%$) and acetone (Sigma-Aldrich) were used to synthesize the 5 nm gold NPs, using previously published methods. 5 nm iron oxide NPs dispersed in chloroform were purchased from Ocean Nanotech.

Sample Preparation

Supramolecular solutions were prepared by dissolving the polymer and the small molecule in chloroform and stirring for 72 h. The two supramolecule compositions used in this work are given below. Thin films with film thicknesses of approximately 55 nm were prepared via spin-coating and solvent vapor annealing, using previously-published methods⁵². Gold NPs were used for the optical measurements, and iron oxide NPs were used for the AFM studies. It has been established that the composition of the NPs does not affect their assembly behavior, as long as their ligand chemistries are consistent⁵⁰.

Table 1-1 Compositions of supramolecular nanocomposites NC1 and NC2.

Supramolecular nanocomposite	PS molecular weight [kDa]	P4VP molecular weight [kDa]	4VP:PDP ratio	Equilibrium periodicity [nm]	5 nm NP loading [vol%]
NC1	19	5.2	2	31	3
NC2	50	17	2	57	3

Substrate Patterning

The circular trenches were fabricated using electron beam (e-beam) lithography. A variety of patterns were fabricated, with intended dimensions given below. Due to the limitations of e-beam lithography, the true trench and mesa widths deviate slightly from these values. The shaded bars in Figures 1-4, 1-6, and 1-7 use dimensions measured from AFM images of the bare substrates (Figure 1-12).

Table 1-2 Dimensions of lithographically-patterned templates P100, P150, and P200.

Template	Trench width [nm]	Mesa width [nm]	Trench depth [nm]
P100	100	100	50
P150	150	200	50
P200	200	150	50

Atomic Force Microscopy (AFM)

AFM was performed on a Bruker Dimension Edge instrument. The spring constant of the cantilever was 5–10 N/m with a resonant frequency in the range 90–210 kHz. The set point for the auto tune was ~6.0 V. The set point amplitude was 80–90% of the free vibration value.

Image Analysis

Image analysis was performed in Python using methods inspired by the ADABlock application written by Murphy et al⁶⁵. Nanocomposite periodicities were determined via fast Fourier transform (FFT). Local microdomain orientations were determined by a sliding window FFT technique. For defect analysis, AFM phase images were binarized and skeletonized. Defects were found by counting the nearest neighbors of every skeleton pixel; points with extra neighbors are junctions and points missing neighbors are ends. Defects are paired across the PS and P4VP(PDP)+NP phases—a terminal point in one requires a branch in the other—so only defects in the latter phase were counted. Radial values (periodicity, order parameter, and defect density) were calculated using a series of overlapping donut-like masks aligned with the lithographic pattern.

Sample Exfoliation

Krazy Glue KG92548R was used in the exfoliation process. First, the glue was used to bond the sample substrate to a first glass slide. Then, glue was dropped onto a second glass slide. The sample was immediately placed onto the glue droplet, with the sample surface facing toward the glue. The glue was allowed to completely dry, then the two glass slides were separated. After separation, the

sample was exfoliated to the second glass slide. Figure 1-13 shows the exfoliation process schematically.

Optical Setup for Transmission Measurements

The optical setup is shown in Figure 1-5d. A HeNe laser (MWK Industries P171NS12 Plasma Tube Laser Head) was used. The 25X magnification path was a 4f system composed of an objective lens, OL, (LEICA 566074, N Plan EPI 50X NA = 0.75, tube length = 200 mm) and a quartz lens, L2, with f=100mm. The 250X magnification path was a 4f system composed of the OL and a quartz lens, L3, with f=1000mm. A linear polarizer (Thorlabs, GTH10) and a quarter wave plate (Thorlabs, FR600QM) were used to generate circularly-polarized incident light. A quartz lens, L1, with focal length of 38mm was used to focus the laser on the sample in order to provide sufficient light intensity. Two charge-coupled devices (CCD), CCD1, (Thorlabs, DCC1645C-HQ), and CCD2, (Thorlabs, DCC3260M, HD-Resolution, Low-Noise USB 3.0 CMOS Cameras with Global Shutter) were used to capture the images. A transmissive laser speckle reducer (Optotune, LSR-3010) was used to reduce the laser speckle noise on CCD1, but was removed before capturing images on CCD2 to maintain the incident gaussian beam profile.

Finite Element Simulation

The finite element method (FEM) simulation of the sample was performed using COMSOL Multiphysics. In the simulation, a staggered five-layer concentric ring model was built to represent the sample (Figure 1-5E). The cross-sections of the pseudo-metal regions are 10 nm by 10 nm; regions within the same layer are separated by 17 nm. The polymer layers are 9 nm thick. For the pseudo-metal P4VP(PDP)-Au regions, the material refractive index is calculated according to Maxwell Garnett Effective Medium Approximation:

$$\epsilon_{MG} = \epsilon_m + 3f\epsilon_m \frac{\epsilon_{Au} - \epsilon_m}{\epsilon_{Au} + 2\epsilon_m - f(\epsilon_{Au} - \epsilon_m)} \quad (3)$$

where f is the volume fraction of the nanoparticles within the pseudo-metal regions (40%), ϵ_{Au} is the complex permittivity of gold, and ϵ_m is the complex permittivity of the polymer matrix. The refractive index of the polymer regions is defined as the average of the refractive indices of PS and P4VP. All optical properties are calculated at a wavelength of 633 nm.

Table 1-3 Optical constants used in the FEM simulation. The functions Re and Im denote taking only the real or imaginary part, respectively, of a complex-valued quantity.

Material	Refractive Index $n = \text{Re}(\sqrt{\epsilon})$	Refractive Index $k = \text{Im}(\sqrt{\epsilon})$	Permittivity, $\epsilon' = \text{Re}(\epsilon)$	Permittivity, $\epsilon'' = \text{Im}(\epsilon)$
PS	1.59	0	2.53	0
P4VP	1.53	0	2.34	0
Au	0.183	3.43	-11.73	1.26
Pseudo-metal	4.94	1.21	22.94	11.95

1.7 Supporting information

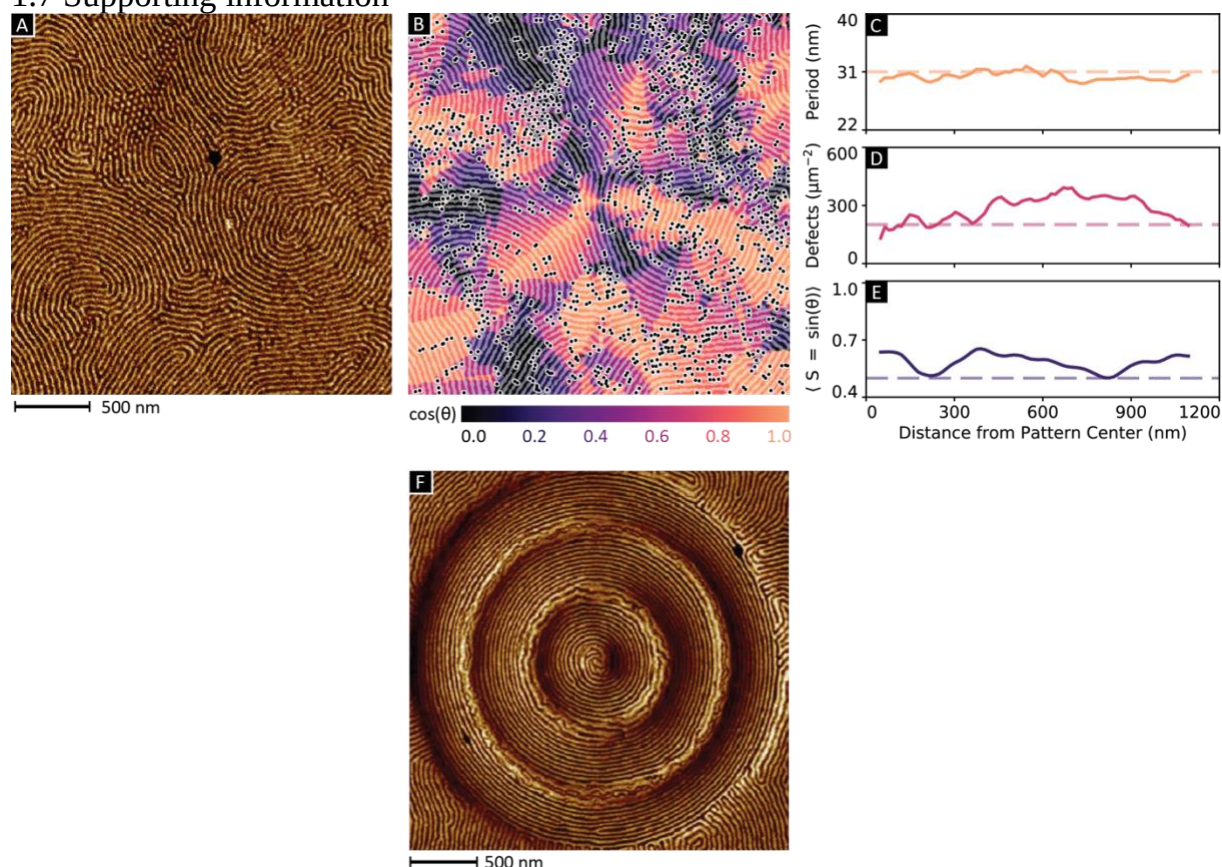


Figure 1-8 [A] AFM micrograph of the nanocomposite self-assembled on a flat, unpatterned substrate. [B] The same micrograph shown in false color, with microdomains colored by the order parameter $S = \cos\theta$. Defects are labelled with black dots. [C] Order parameter, [D] microdomain period, and [E] defect density, plotted radially from the micrograph's center. The dashed lines are averages over the full control image. [F] AFM micrograph of the 19-*b*-5.2 kDa PS-*b*-P4VP(PDP) supramolecule without nanoparticles self-assembled on P200. Under the same annealing conditions as NC1, a similar concentric ring morphology was observed.

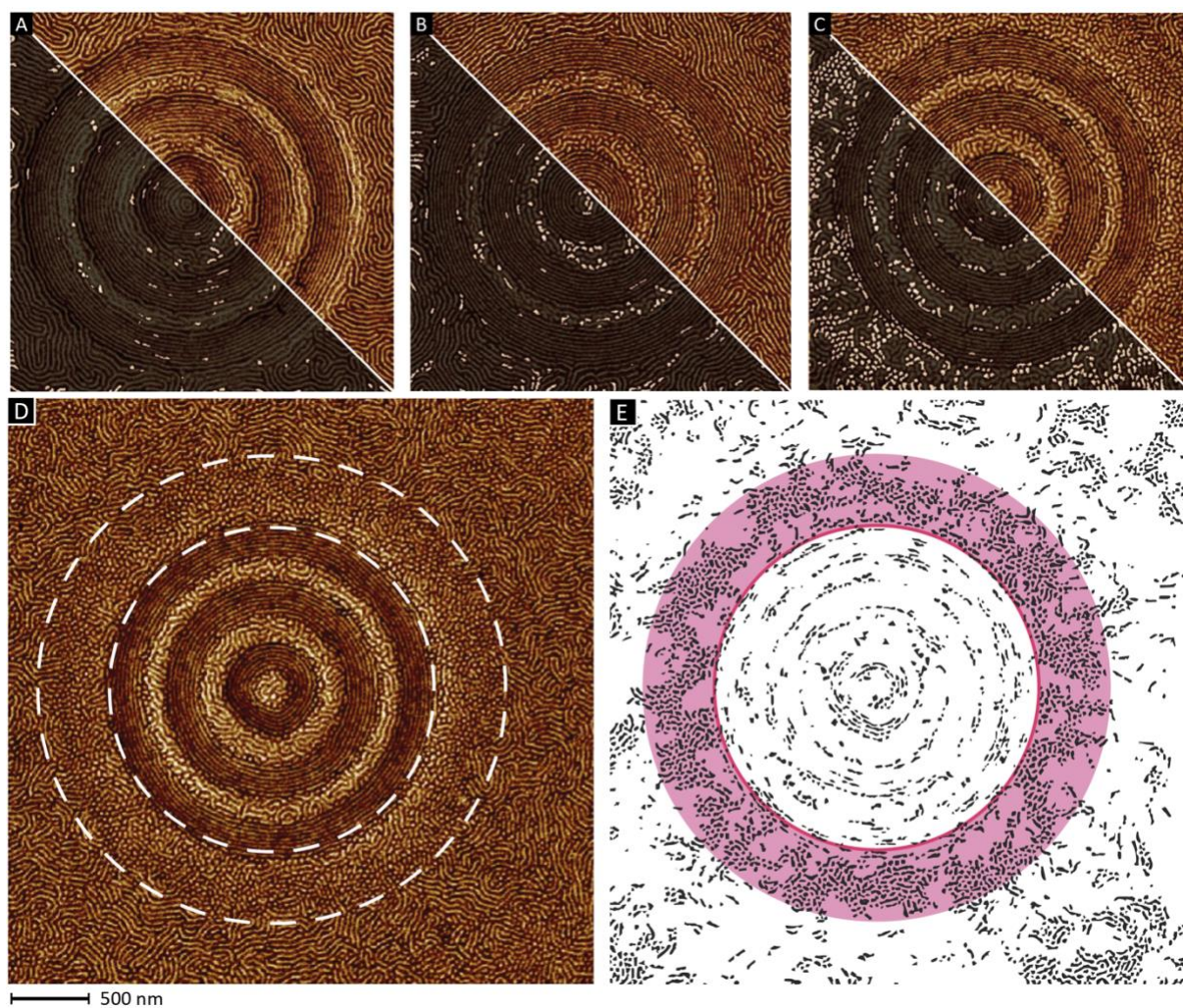


Figure 1-9 AFM micrograph of the nanocomposite NC1 with different nanoparticle loading. [A] AFM micrograph of the nanocomposite NC1 with a 1 vol% nanoparticle loading. The morphology is almost entirely cylinders with a small number of spheres, highlighted in white on the darker overlay. [B-C] AFM micrographs of the nanocomposite with 3 vol% and 5 vol% nanoparticle loadings, respectively. The area covered by spheres increases with NP loading, consistent with a cylinder-to-spheres morphological transition as a result of PDP redistribution. Each scan is 2.4 μm wide. [D] Lower-magnification AFM micrograph of the sample with 5v% particles and [E] false-color image with low-aspect-ratio microdomains (spheres) colored black. The morphological transition forms a ring (colored pink) that extends well beyond the outer edge of the template. This is consistent with global PDP redistribution: the system's self-adjustment occurs on the length-scale of the template, an order of magnitude larger than the characteristic microdomain size.

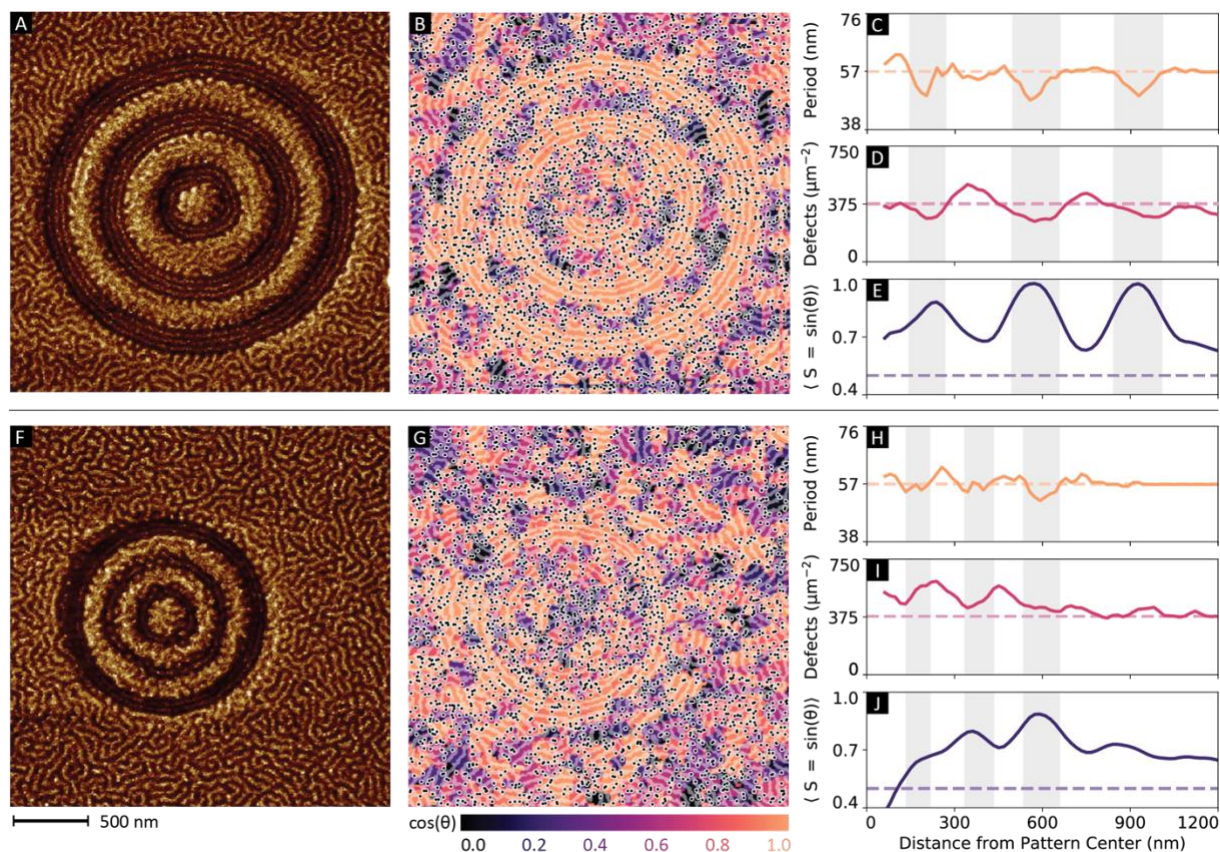


Figure 1-10 AFM micrograph of the nanocomposite NC2 self-assembled on different template. [A] AFM micrograph of the nanocomposite NC2 self-assembled on P150. [B] The same micrograph shown in false color, with microdomains colored by the order parameter $S = \cos\theta$. Defects are labelled with black dots. [C] Order parameter, [D] microdomain period, and [E] defect density, plotted radially from the pattern's center. The dashed lines show control values from a non-patterned sample; the shaded bars give the trench positions. [F-J] The same measurements and calculations are repeated for an NC2 sample assembled on P100.

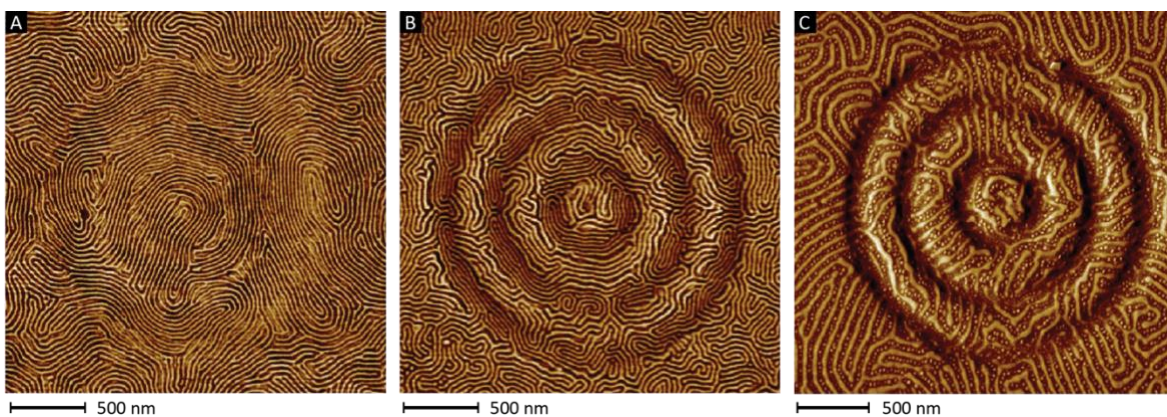


Figure 1-11 AFM micrograph of NC1 cast. [A] AFM micrograph of NC1 cast with a film thickness of 70 nm. The sample preparation was otherwise the same as the successfully-patterned samples, but the confinement does not substantially affect the top layer of this thicker film. [B] AFM micrograph of NC1 cast with a film thickness of 50 nm. The sample preparation was otherwise the same as the successfully-patterned samples, but the film was not able to self-adjust across the raised mesas. There is no significant difference in periodicity, alignment, or morphology between the trenches and mesas. [C] AFM micrograph of NC2 with 3 vol% 15 nm particles. The microdomains of NC2 are large enough to accommodate the particles, and the unpatterned regions show relatively low defect densities and large grain sizes. However, the microdomains are not guided by the underlying template.

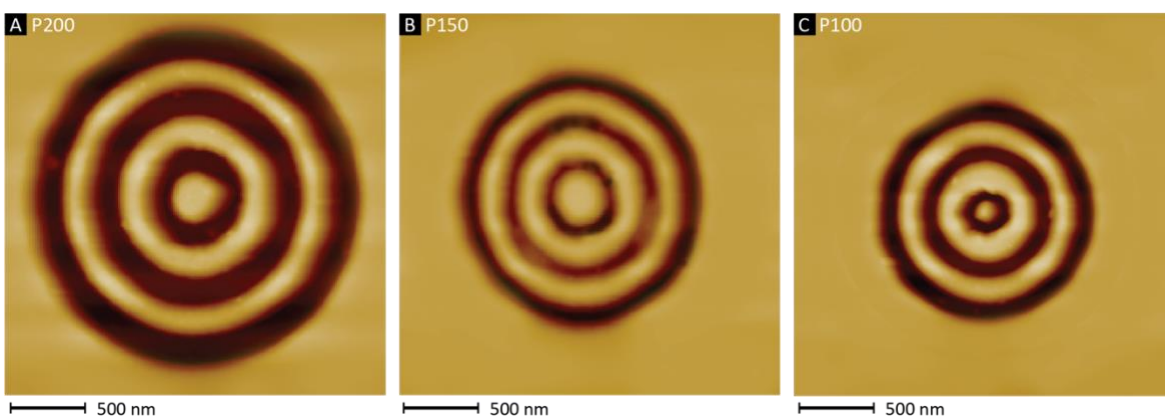


Figure 1-12 AFM micrographs of the lithographically-patterned templates. [A] P200, [B] P150, and [C] P100. A small degree of irregularity and eccentricity is observed, due to the limitations of e-beam lithography.



Figure 1-13 Schematic of the film exfoliation process. [A] Krazy Glue KG92548R is used to bond the silicon substrate to glass slide #1, which is later used as a holder. [B] Next, a drop of glue is applied to glass slide #2. The sample is immediately face-down onto the glue droplet. [C] After the glue has completely dried, the glass slides are separated. The nanocomposite sample is exfoliated to glass slide #2. The patterned substrate on slide #1 may be reused.

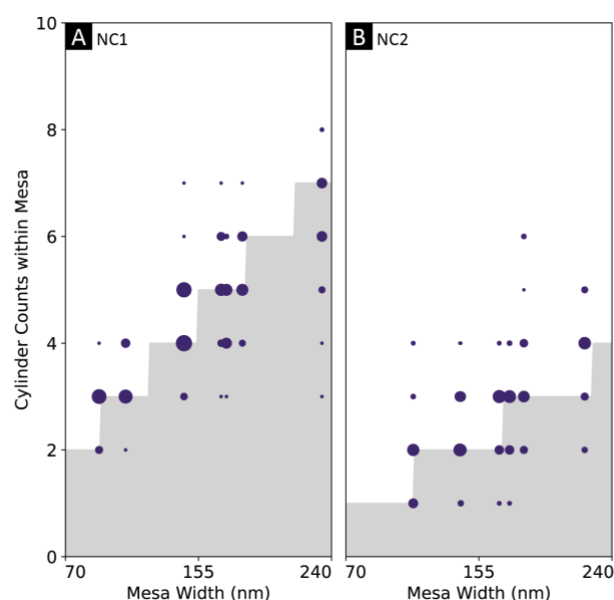


Figure 1-14 [A-B] Complementary to Figure 1-7D-E, these plots show all observed states of the nanocomposites NC1 and NC2, respectively, on the mesas of P100, P150, and P200. The gray stair-step pattern shows the commensurate state closest to the equilibrium periodicity for all mesa widths. The marker size is scaled by the number of times a state was observed; ie., the most common states are drawn with the largest markers. Though the nanocomposite is not confined, structural degeneracy is still observed.

Strain Energy Calculations

This strain energy model assumes that the deformations are purely elastic. This small-strain assumption is often made in BCP microdomain-bending models, in large part because plastic deformation is challenging to model in inhomogeneous systems^{28,66}. The energy-strain proportionality is derived by substituting the elastic stress-strain relationship,

$$\sigma = E\epsilon \quad (\text{S1})$$

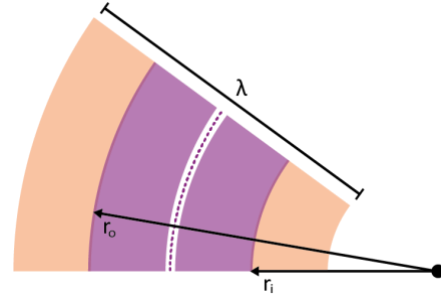
for stress σ , strain ϵ , and elastic modulus E , into the definition of elastic strain energy per unit volume, U ,

$$U = \frac{1}{2}\sigma\epsilon \quad (\text{S2})$$

for a final relationship of

$$U = \frac{1}{2}E\epsilon^2 \propto \epsilon^2 \quad (\text{S3})$$

The modulus E is not easily calculated, because of the inhomogeneous nature of the nanostructured system. We assume, however, that its value is constant for the system. As a result, the squared change in volume (which is easily calculated) is directly proportional to the elastic strain energy. The strain energy calculations are performed on a single curved microdomain. The cylindrical morphology is approximated as lamellar so that the system may be reduced to a simple 2D geometry (above). An inner radius (r_i) and an outer radius (r_o) are defined at the neighboring block interfaces. Both interfaces are



defined as neutral axes, which separate the outer stretched region from the inner compressed region, and do not change arc length upon domain bending. The intrablock interfaces, however, (dashed line) will change length, and the domain areas will change accordingly. For example, the stretched and compressed P4VP domains undergo the following area changes:

$$\Delta A_{P4VP,i} = \pi r_i^2 - \pi \left(r_i + \frac{\lambda}{4} \right)^2 - \frac{\lambda}{4} \cdot 2\pi r_i \quad (\text{S4})$$

$$\Delta A_{P4VP,o} = \pi r_o^2 - \pi \left(r_o - \frac{\lambda}{4} \right)^2 - \frac{\lambda}{4} \cdot 2\pi r_o \quad (\text{S5})$$

The stretched and compressed PS domains also change area:

$$\Delta A_{PS,i} = \pi r_i^2 - \pi \left(r_i - \frac{\lambda}{4} \right)^2 - \frac{\lambda}{4} \cdot 2\pi r_i \quad (\text{S6})$$

$$\Delta A_{PS,o} = \pi r_o^2 - \pi \left(r_o + \frac{\lambda}{4} \right)^2 - \frac{\lambda}{4} \cdot 2\pi r_o \quad (\text{S7})$$

So long as the strains are relatively small and the material responds elastically, the area change $|\Delta A|/A_0$ will be proportional to the strain energy. When PDP redistribution is allowed, area loss in the P4VP areas can be used to compensate for area gain in adjacent areas before the absolute value $|\Delta A|$ is calculated.

Chapter 2 Berry curvature mediated spin-orbit interaction in honeycomb photonic crystal

2.1 Background

2.1.1 Photonic crystal

To find the material which can control light propagation, we rely on the analogy with the successful electronic materials⁶⁷. The theory of quantum mechanics explained why electrons can avoid scattering from the constituents when propagates through the crystal with periodic arrangement of atoms or molecules.

The optical analogue of these “crystal” is the photonic crystal, in which the atoms or molecules are replaced by macroscopic media with differing dielectric constants and the periodic potential is replaced by a periodic index of refraction. If the dielectric constants of the materials in the crystal are sufficiently different and the absorption is minimal, then the refraction and reflections of light from all the various interfaces can produce many of the same phenomena for photons that the atomic potential produce for electrons.

To study the propagation of light in a photonic crystal, we begin with the Maxwell equation. Casting the Maxwell equations as a linear Hermitian eigenvalue problem will show that the electromagnetic problem is a close analogy with the Schrodinger equation. This not only allows us to take advantage of some well-established results from the quantum-mechanical to study photonic crystal, but also give us the chance to take advantage of photonic crystal to mimic and study some condensed matter physics system. On way in which the electromagnetic case differs from the quantum-mechanical case is that photonic crystal does not generally have a fundamental scale. This makes photonic crystal scalable in a way the traditional crystal is not, which means we can shift the wavelength of the photonic crystal eigenmode easily and this can help me better access the experimental resources to do the study.

Under approximations and derivations, the Maxwell equations become **master equation**:

$$\nabla \times \left(\frac{1}{\varepsilon(\mathbf{r})} \nabla \times \mathbf{H}(\mathbf{r}) \right) = \left(\frac{\omega}{c} \right)^2 \mathbf{H}(\mathbf{r})$$

And once solve the eigenmode of $\mathbf{H}(\mathbf{r})$, we can recover $\mathbf{E}(\mathbf{r})$.

We can see the master equation has a very similar format as the Schrodinger equation, shown in Figure 2-1⁶⁷.

	<i>Quantum Mechanics</i>	<i>Electrodynamics</i>
Field	$\Psi(\mathbf{r}, t) = \Psi(\mathbf{r})e^{-iEt/\hbar}$	$\mathbf{H}(\mathbf{r}, t) = \mathbf{H}(\mathbf{r})e^{-i\omega t}$
Eigenvalue problem	$\hat{H}\Psi = E\Psi$	$\hat{\Theta}\mathbf{H} = \left(\frac{\omega}{c}\right)^2 \mathbf{H}$
Hermitian operator	$\hat{H} = -\frac{\hbar^2}{2m}\nabla^2 + V(\mathbf{r})$	$\hat{\Theta} = \nabla \times \frac{1}{\epsilon(\mathbf{r})} \nabla \times$

Figure 2-1 comparison of master equation and Schrodinger equation. Copy right⁶⁷.

Here I make a Figure 2-2 to further compare the difference between crystal and photonic crystal

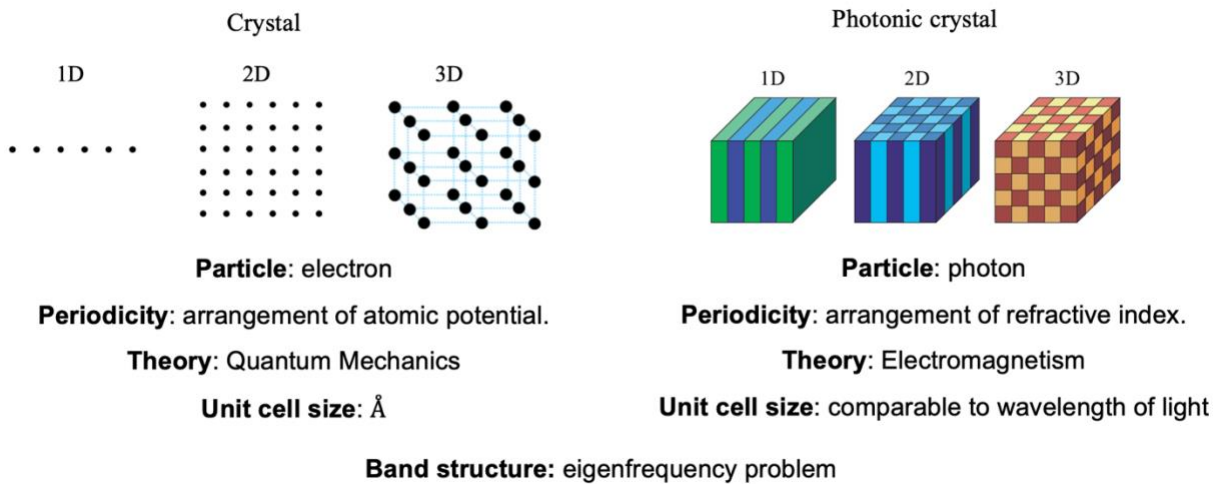


Figure 2-2 comparison between crystal and photonic crystal

2.1.2 Spin-orbit interaction

Light carries both spin angular momentum (SAM) and orbital angular momentum (OAM)⁶⁸. These properties are determined by the polarization and spatial degrees of freedom of light. Spin-orbit interaction (SOI) is the phenomenon when spin and orbital properties become strongly coupled. And it can be further categorized into three different types as shown in Figure 2-3, where intrinsic orbital AM (IOAM) represents optical vortex beams with helical phase fronts, and extrinsic orbital AM (EOAM) represents beams propagating at some distance from the coordinate origin.

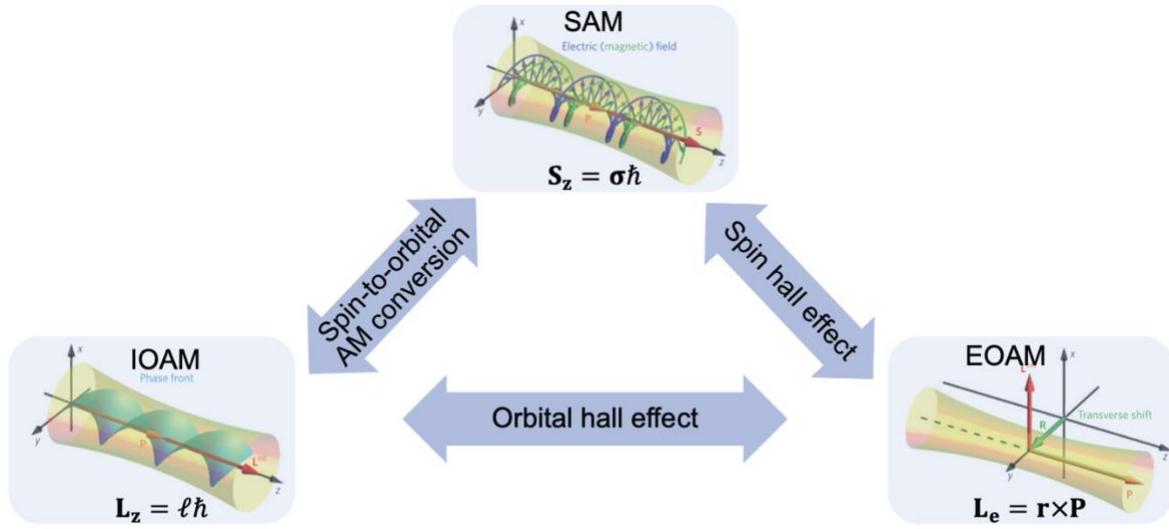


Figure 2-3 types of SOI.

Spin-orbit interaction (SOI) is a wavelength scale phenomenon. The traditional geometrical optics ignore the SOI phenomena by treating the spatial and polarization properties of light separately according to paraxial approximation. These degrees of freedom can be independently manipulated: by lenses or spatial light modulator, on the one hand, and polarizers or waveplates, on the other.

Nano-optics, photonics and plasmonic analyze the phenomenon at the subwavelength scales. At this wavelength, the geometrical optics (based on the properties of scalar waves) does not work and should be substituted by the full-vector analysis of Maxwell waves. In this non-paraxial range, spin and orbital properties become strongly coupled with each other and the SOI phenomena become significant. Spin-orbit interactions are inherent in all basic optical processes, and that they play a crucial role in modern optics.

The SOI of light has both fundamental and applied importance for physics. On the one hand, these phenomena allow the direct observation of fundamental spin-induced effects in the dynamics of relativistic spinning particles (photons). Measurements of similar effects, for example, for Dirac electrons or analogous condensed-matter quasiparticles, are far beyond current capabilities. On the other hand, akin to the significant enhancement of electron SOI in solid-state crystals, the SOI of light are considerably enhanced by material anisotropies and can be artificially designed in optical nanostructures, including metamaterials. This paves the way to spin-optics: an optical counterpart of electron spintronics in solids. Introducing additional spin degrees of freedom for the smart control of light promises important applications in photonics, optical communications, metrology, and quantum information processing. In this manner, the SOI of light conform to the most important trends in modern engineering: miniaturization of devices down to subwavelength scales and increasing the amount of information available through additional internal degrees of freedom.

Because of the similarity between Maxwell equation (write in master equation format) and Schrödinger equation⁶⁷, SOI in the optical systems^{68–70} are analogous to the SOI in relativistic

quantum particles^{71,72} and electrons^{73,74} in solids. The SOI of light has both fundamental and applied importance for nano-optics^{68,75}. First, the SOI phenomenon must be taken into account in the analysis of any nano-optical system, and this is absolutely essential in the conception and design of modern optical devices⁶⁸. Second, introducing SOI for the smart control of light promises important applications to optical nano-devices, such as far-field optical nanoprobng⁷⁶, optical metasurface⁷⁷, photonic spin-Hall shift⁷⁸, light OAM generation⁷⁹, optical spintronics, etc.⁸⁰. Third, these phenomena allow the direct observation of fundamental spin-induced effects in the dynamics of relativistic spinning particles (photons). As mentioned before, the measurements of similar effects, for example, for Dirac electrons or analogous condensed-matter quasiparticles, are far beyond current capabilities. Thus, in the new reality of nano-optics, studies and applications of optical spin-orbit phenomena are urgently and strongly needed.

2.1.3 Pseudospin

It's a concept in graphene structure. It can be simply understood as graphene sublattice degree of freedom. It is introduced initially due to the mathematical analogy between the graphene sublattice degree of freedom and the electron spin in the original Dirac equation. It has certainly turned into one of the paradigms of graphene physics⁸¹⁻⁸⁴.

2.1.4 Geometric phase or Berry phase

In 2016, the Nobel prize of physics was awarded to three scientists for their contributions to the topological theory in solids⁸⁵. The discovery of the topological insulator has been one of the biggest topics in the last decade⁸⁶. The root of the topological phenomena, however, was discussed decades ago when people were trying to understand the abnormal electron velocity in the ferromagnetic materials⁸⁷. The geometric phase, later referred to as the Berry phase, was proposed as an extra phase beside the propagation phase. People at that time did not realize that this concept would completely change the landscape of physics, materials science, even photonics in the twenty-first century⁸⁸. The mathematic expression of Berry phase will be discussed in 2.1.5.

2.1.5 Berry connection and Berry curvature

In physics, Berry connection and Berry curvature are related concepts which can be viewed, respectively, as a local gauge potential and gauge field associated with the Berry phase or geometric phase. The Berry connection is gauge dependent. Hence the local Berry connection can never be physically observable. Berry curvature is gauge invariant local manifestation of the geometric properties of the wavefunctions in the parameter space and has proven to be an essential physical ingredient for understanding a variety of electronic properties^{89,90}.

The closed-path Berry phase can be expressed as $\gamma_n = \oint_C d\mathbf{R} \cdot \mathcal{A}_n(\mathbf{R})$, where $\mathcal{A}_n(\mathbf{R}) = i\langle n(\mathbf{R}) | \nabla_{\mathbf{R}} | n(\mathbf{R}) \rangle$ is a vector-valued function known as the Berry connection (or Berry potential), and $n(\mathbf{R})$ is the eigenstate of the Hamiltonian $H(\mathbf{R})$.

Berry curvature can be derived from the berry connection via

$$\Omega_n(\mathbf{R}) = \nabla_{\mathbf{R}} \times \mathcal{A}_n(\mathbf{R})$$

For a closed path $\mathcal{C}$ that forms the boundary of a surface $\mathcal{S}$, the closed-path Berry phase can be rewritten using Stokes' theorem as

$$\gamma_n = \int_{\mathcal{S}} d\mathbf{S} \cdot \boldsymbol{\Omega}_n(\mathbf{R})$$

2.2 Abstract

Global topological invariant emerges from momentum-space integrals of the Berry curvature. It is becoming increasingly evident that the local (in momentum space) quantum geometry can have dramatic physical consequences as well. Here we utilize a honeycomb photonic crystal to mimic the condensed matter physics system. With inversion symmetry breaking, the Dirac cone degeneracy is lifted. Using this system, we proved the generation of orbital angular momentum (OAM) in different pseudospins is a local berry curvature mediated spin-orbit interaction (SOI) phenomenon. These fundamental finding not only helps us understanding the SOI in photonics system but also deepens our understanding of the SOI in condensed matter counterpart like BN structure. Besides, by linking spin to pseudospin and berry curvature, not only we proved the circular dichroism (CD) as an effective way to characterize local berry curvature, but also the photonic crystal can work as a chiral and OAM generation device.

2.3 Introduction

Light carries both spin and orbital angular momentum. The Spin-Orbit Interactions (SOI) phenomenon in optical system describes how spin (circular polarization) angular momentum affect and control orbital (spatial) angular momentum of light ⁶⁸ Because of the similarity between Maxwell equation (write in master equation format) and Schrödinger equation⁶⁷, SOI in the optical systems ^{68–70} are analogous to the SOI in relativistic quantum particles^{71,72} and electrons^{73,74} in solids. Understanding the spin-orbit interactions (SOI) in photonic lattices is essential in fundamental understanding of light-matter interactions, and it can also bring novel optical functionalities and allow information to be encoded and retrieved using polarization degrees of freedom.

In periodic solids, a global topological invariant is defined by momentum-space integrals of the Berry curvature, which is the foundation of topological insulators (TIs) and superconductors.⁹¹ However, independently of the global topology, it is becoming increasingly evident that the local (in momentum space) quantum geometry can have dramatic physical consequences as well, for example, Fermi surface for Fermi-liquid transport⁹², the fractional quantum Hall effect⁹³, the superfluidity in flat-band systems⁹⁴, the strongly anisotropic high-harmonic generation signals from hexagonal boron nitride (hBN) or transition-metal dichalcogenides (TMDC)⁹⁵, the valley Hall effect⁹⁶, the nonlinear Hall effect^{97,98} in topologically trivial systems etc.

There is a theory study⁹⁹ shows the possibility of mapping out the signatures of the momentum-resolved Berry curvature in two-dimensional materials by exploiting its intimate connection to the orbital polarization. Here in our study, by experimentally reveal the circular dichroism angle-resolved transmission spectroscopy, we linked the SAM-LBC-OAM relationship and proved that the generation of OAM in different pseudospins is a local berry curvature mediated SOI

phenomenon, and that the circular dichroism indeed will provide a powerful tool that can resolve the topological and quantum-geometrical character in momentum space.

For the hexagonal photonic crystal structure, there are some literatures¹⁰⁰ discussing the generation of OAM mode, but they only linked it with spin. In fact, it's an SOI effect, the pseudospin mediated the spin to orbital angular momentum transition. And there are also some theoretical discussions of hBN system local berry curvature characterization using circular dichroism, but since the difficulty of experiments, there is no direct evidence to support it. Here our study will fill the gaps and experimentally verify the mechanism behind the SOI phenomenon in hexagonal structure from both the real space and reciprocal space point of view.

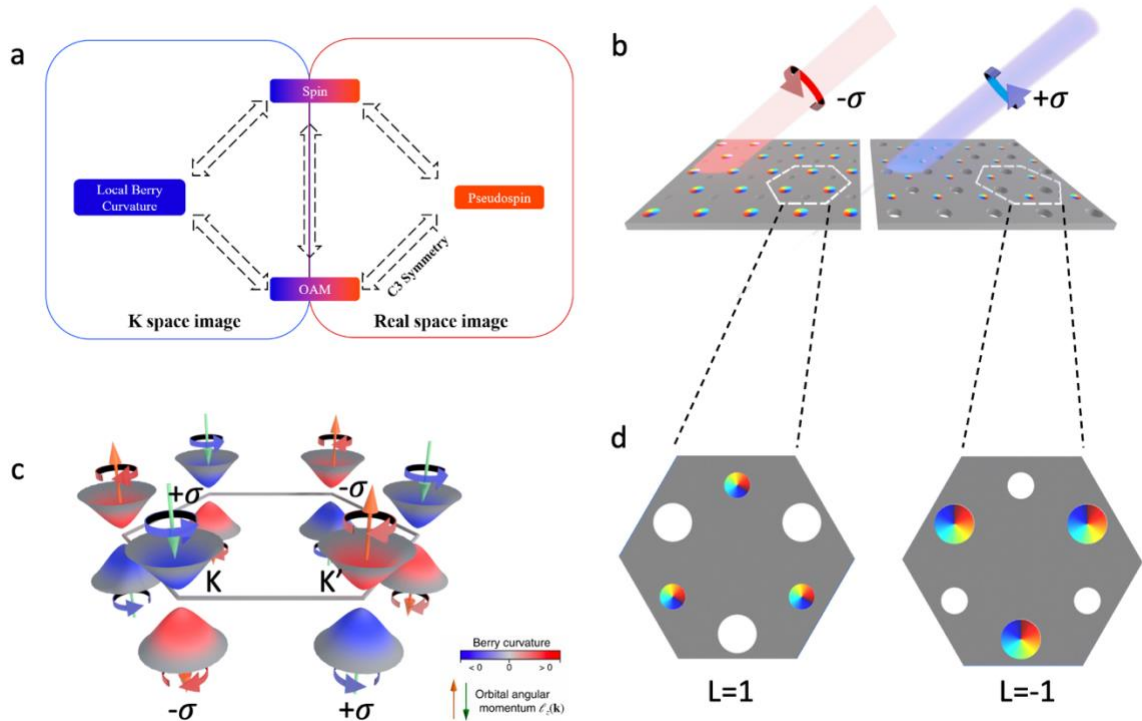


Figure 2-4 (a) the principle of SOI in hexagonal structure (b) real space schematic image: spin will excite different pseudospin mode which corresponds different OAM (c) K space schematic image: the local berry curvature in K space and its spin correlation (d) the schematic zoom-in image of OAM phase distribution

Figure 2-4a shows the schematic principle of SOI. The link between pseudospin and OAM is based on the symmetry of the eigenmodes. And the spin and pseudospin connection are caused by SOI. And for the K space valley selection rule of SOI phenomenon, the mechanism is the berry curvature connection. There is theoretical study showing that the berry curvature coming from the unbalanced OAM, and because of the SOI, the orbital polarization will be linked to local berry curvature and the circular dichroism will be a direct way to probe the berry curvature.

2.3 Results

In the schematic image, Figure 2-4b shows that spin will excite different pseudospin mode which corresponds different OAM shown in Figure 2-4d. And Figure 2-4c shows the local berry curvature in K space. And for K and K' point, the upper band and lower band will exchange the signs of local berry curvature, which connects different spin-OAM conversion.

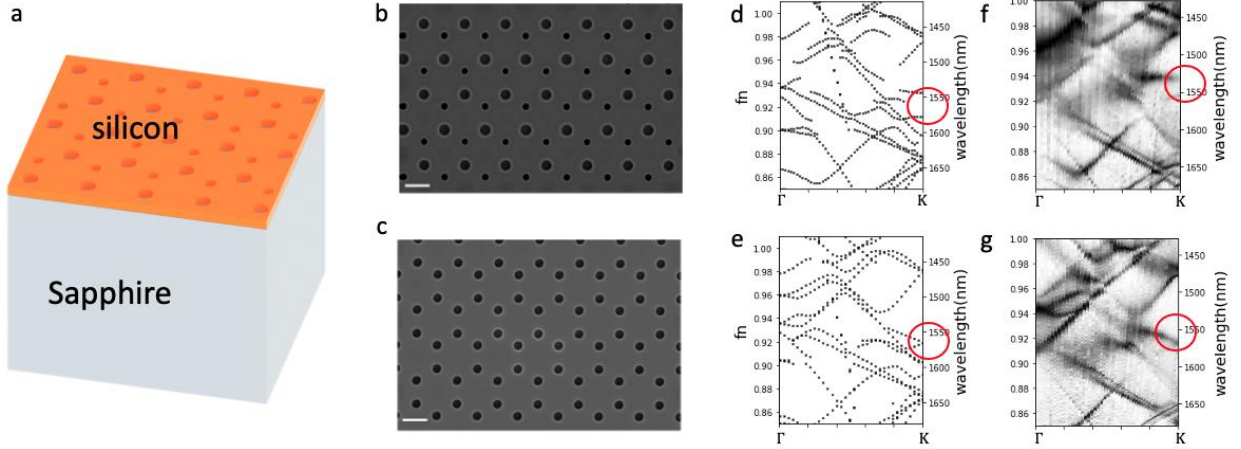


Figure 2-5 (a) 3D image of the sample (b) SEM image of the sample with inversion symmetry breaking, scale bar represent 1μm (c) SEM image of the sample without inversion symmetry breaking, scale bar represent 1μm (d) The COMSOL simulated band-structure of the sample with inversion symmetry breaking (e) The COMSOL simulated band-structure of the sample without inversion symmetry breaking (f) band-structure measured by experiments of the sample with inversion symmetry breaking (g) band-structure measured by experiments of the sample without inversion symmetry breaking

Figure 2-5a shows the 3D image of the sample. The sample is 230nm thickness silicon on sapphire substrate. And as shown by the SEM images in Figure 2-5b and Figure 2-5c (scale bar is 1μm), hexagonal photonic crystal structure with inversion symmetry breaking and without symmetry breaking is fabricated, and the diameter error is less than $\pm 10\text{nm}$. From Figure 2-5d and Figure 2e, the simulated band structure is shown, and the fn is the normalization number of frequency over $2\pi c/a$ in which a is the smallest distance between two holes in the same sublattice. The Dirac point is pointed out by the red circles. Figure 2d, the Dirac cone degeneracy is lifted, and Figure 2e shows there are two modes degenerated. And the Figure 2f and g shows the corresponding experimentally measured band structure, which match the simulation quite well.

Figure 2-6a shows the experimental setup for far-field measurements. Figure 2-6b shows the angle-wavelength transmission map from 0 degree to 60 degrees. Figure c and d shows the blue and purple cross-sectional transmission intensity distribution marked in Figure 2-6b. The Red and Black color shows the transmission of RCP and LCP incident light. And the transmission lines verified that the two modes at the Dirac point from K point are excited by RCP and LCP incident

light respectively. Figure e and Figure f shows the CD response of c and d respectively. The CD is defined as $\pm 2(\frac{\sigma^+ - \sigma^-}{\sigma^+ + \sigma^-})$. The strong CD response experimentally verified the SOI phenomenon.

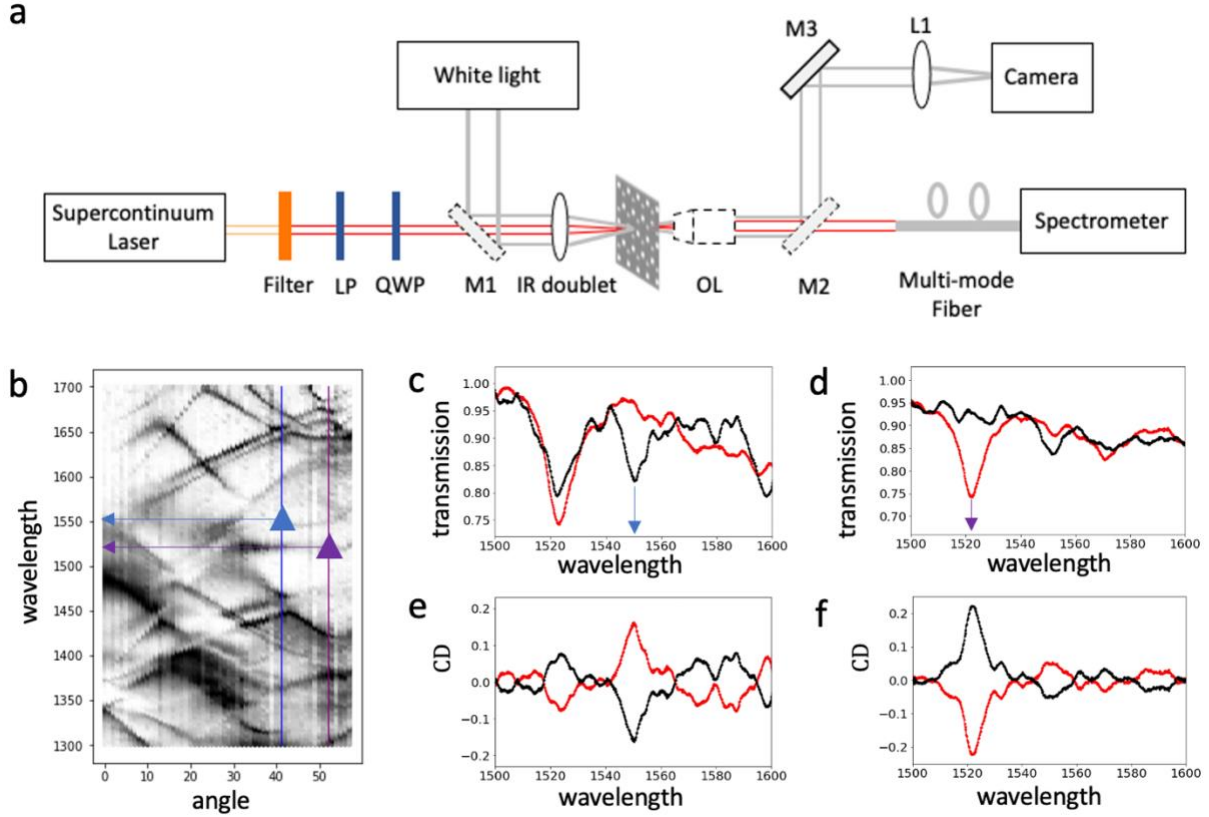


Figure 2-6 (a) the experimental setup for far-field measurement (b) the angle-wavelength transmission map from 0 degree to 60 degrees (c) blue cross-sectional transmission intensity distribution from Figure 2-6b, the Red and Black color shows the transmission of RCP and LCP incident light (d) purple cross-sectional transmission intensity distribution from Figure 2-6b, the Red and Black color shows the transmission of RCP and LCP incident light (e) CD response of Figure c (f) CD response of Figure d.

Figure 2-7a shows the reciprocal space of the hexagonal photonic crystal. And Figure 2-7b shows the calculation results of the berry curvature along edge of Brillouin zone. The results are for BN⁹⁹, which share the same symmetry with our photonics sample. The calculation results for the strict photonics crystal are under review and please refer to the formal publication for more details. And Figure 2-7c shows the CD band-structure around the K' point along Ky direction (along red line in Figure a). And Figure 2-7d shows the CD band-structure from the K' point to Γ point to K point along Kx direction (along blue-orange line in Figure a). And Figure 2-7e shows the CD band-structure around the K point along Ky direction (along green line in Figure a). The blue circles in

Figure 2-7c, d, and e circled out the two bands interested. These results match the local berry curvature distribution in Figure 2-4c and calculation in Figure 2-7b quite well.

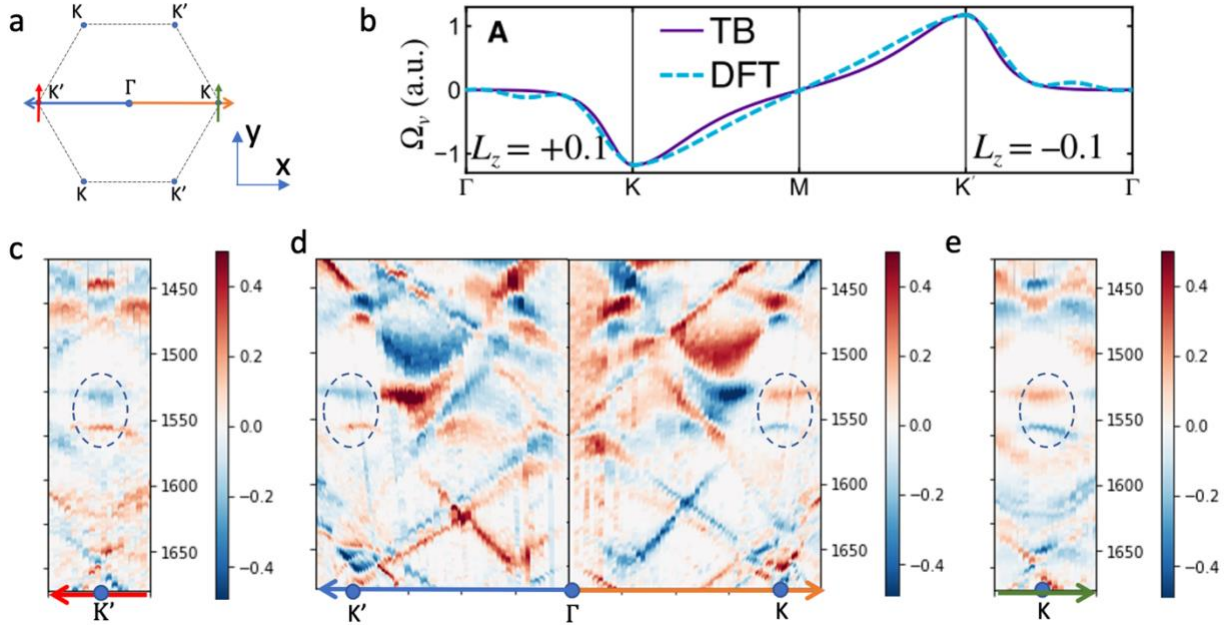


Figure 2-7 (a) the reciprocal space of the hexagonal photonic crystal, and the red/blue/orange/green arrows show different path in K space (b) calculated berry curvature results using DFT⁹⁹ (c) CD band-structure around the K' point along Ky direction, along red line in Figure a (d) CD band-structure from the K' point to Γ point to K point along Kx direction, along blue-orange line in Figure a (e) CD band-structure around the K point along Ky direction, along green line in Figure a.

2.4 Conclusion

We fabricated high quality photonic crystal to explore SOI effect. From the transmission results, we verify two eigenmodes in Dirac cone carrying opposite OAM at different pseudospin will be excited with different spin. And from reciprocal space point of view, we calculated the local berry curvature in different valley and use band structure CD experiments to verify this SOI is valley-linked and local berry curvature mediated phenomenon and proved the CD can effectively resolve the berry curvature. In short, we wrap up the SOI mechanism in hexagonal photonic crystal from real space and reciprocal space point of view as shown in Figure 2-4a. And the device can function as periodic near filed OAM source and work as a dichroism device. And our results also shine lights on its condensed matter physics counterpart and prove CD can be used for berry curvature detection where the corresponding experiments is hard to be carried. And we anticipate that with the rising of berry curvature related phenomena, the finding from work will be helpful to pave the road to the new discoveries.

2.5 Supplementary information

Photonic crystal design fabrication and characterization

By engineering the geometric and material properties of photonic crystals using the COMSOL simulation software, we designed an optical graphene system with tunable staggering potential, and the designed structure and the design process is shown below.

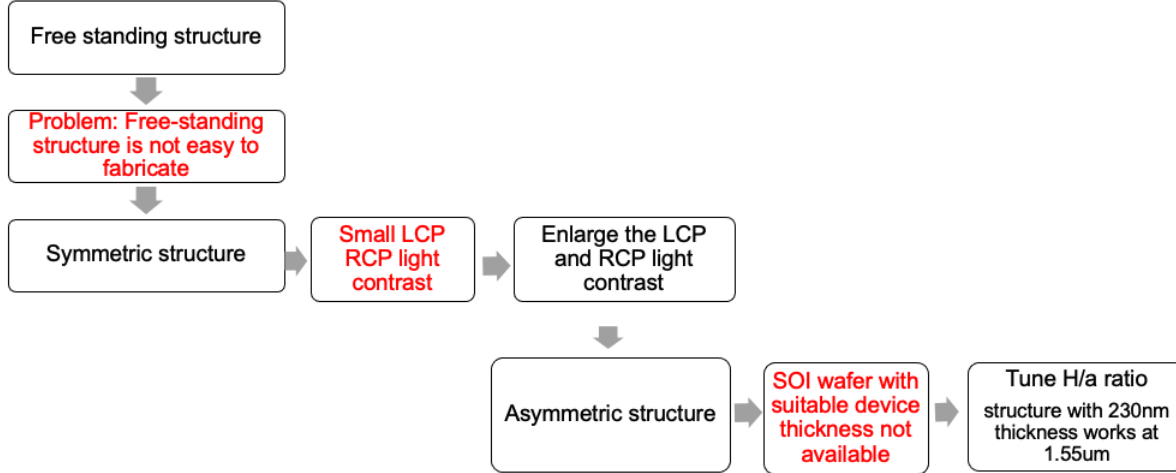


Figure 2-8 Photonic crystal design

The photonic crystal pattern is defined by EBL. And TP-RIE is used to etch silicon and high-power oxygen RIE is used to strip the photoresist. Zeiss-SEM is the characterization tool to get high-quality image. The overall size of the sample region is about 500um*500um with 429nm and 286nm diameter hole features, error within ± 10 nm. The SEM and FIB characterizing the fabricated nanostructure in-plane size uniformity and the wall straightness.

NSOM measurement

Near-field scanning optical microscope (NSOM) facility is used to characterize the near-field distribution of the mode. The source light will be incident from the bottom side of the sample, and the near field signal will be collected through a fiber tip coated by metal (with a hole aperture area uncoated). The results are still in process and please refer to the formal publication.

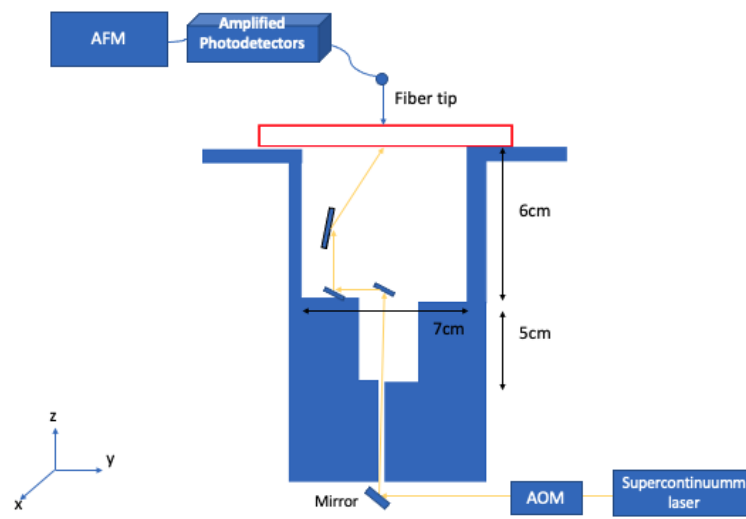


Figure 2-9 optical setup for NSOM measurement

Chapter 3 Optical grating from surface acoustic wave guided self-assembled polymer structure

3.1 Background

3.1.1 Interdigital transducer

An interdigital transducer (IDT) is a device that consists of two interlocking comb-shaped arrays of metallic electrodes (in the fashion of a zipper). These metallic electrodes are deposited on the surface of a piezoelectric substrate to form a periodic structure, which helps to transform electric signals to surface acoustic wave (SAW). When AC voltage is applied, the substrate surface will move up and down due to Inverse Piezoelectric effect and generate SAW, as shown in Figure 3-1¹⁰¹.

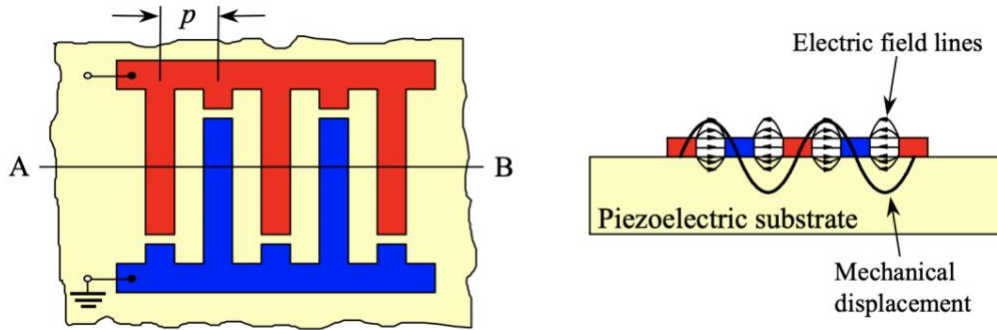


Figure 3-1 Working mechanism of IDT

3.1.2 Resonant frequency

The total output of the entire IDT transducer is the vector sum of the output of each IDT electrode:

$$E = E_0 e^{j\omega t} [1 - e^{j\Delta\theta} + e^{j2\Delta\theta} - \dots (-1)^{N-1} e^{j(N-1)\Delta\theta}]$$

where N is the number of IDT electrodes and E_0 is the amplitude of excited waves of each electrode. $\Delta\theta$ is the phase difference of the SAW of the adjacent IDT electrode and can be expressed as:

$$\Delta\theta = \omega \lambda_{SAW} / 2v_s$$

where v_s is the acoustic velocity in substrate.

When $\Delta\theta = \pi$, the maximum total output is achieved and the corresponding frequency is called synchronization frequency or resonant frequency, which is what we apply in experiment to get the strongest respond.

$$f_r = v_s / \lambda_{SAW}$$

It can be further understood in this way. For a particular substrate, acoustic velocity is a constant and the acoustic wavelength λ is determined by the applied f . The resonant f corresponds to the strongest SAW that meets the condition $\lambda = \lambda_{SAW}$, as shown in Figure 3-2. The x-axis is the value of λ_{SAW} / λ . The y-axis is the main item of transfer admittance, which is the value of I_{output} / V_{input} . It basically shows the shape of sinc function. When $\lambda = \lambda_{SAW}$, the respond is the strongest¹⁰².

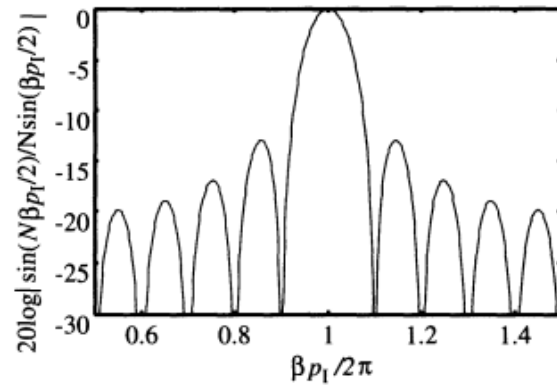


Figure 3-2 transfer admittance.

3.1.3 Surface acoustic waves

SAWs are sound waves that are generated and propagate along the surface of elastic materials. The most common SAW is Rayleigh wave, which confines most of the energy to the surface and decay exponentially with the depth of the substrate.

3.1.4 Theory of fluid main equations

The general governing equations for a fluid are the continuity and the Navier-Stokes equations¹⁰³, respectively,

$$\begin{aligned} \frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{v}) &= 0, \\ \rho \frac{\partial \mathbf{v}}{\partial t} &= -\nabla p - \rho(\mathbf{v} \cdot \nabla) \mathbf{v} + \mu \nabla^2 \mathbf{v} + \left(\frac{1}{3} \mu + \mu_B \right) \nabla(\nabla \cdot \mathbf{v}) \end{aligned}$$

where ρ , $\mathbf{v}$, p , μ and μ_B are the density, velocity, pressure, dynamic viscosity and bulk viscosity. By linearizing this equation using a perturbation approximation, we can obtain the first-order equations for the acoustic field:

$$\frac{\partial \rho_1}{\partial t} + \rho_0 \nabla \cdot (\mathbf{v}_1) = 0,$$

$$\rho_0 \frac{\partial v_1}{\partial t} = -c_0^2 \nabla \rho_1 + \mu \nabla^2 v_1 + \left(\frac{1}{3} \mu + \mu_B \right) \nabla (\nabla \cdot v_1)$$

The parameters with subscript 1 denote the first-order terms, whereas the subscript 0 indicates constant properties of a quiescent fluid. c_0 is the speed of sound in fluid. By assuming a single harmonic time dependence of $e^{i\omega t}$ and time-averaging first-order equations, we derive the second-order equations which describe the streaming field:

$$\begin{aligned} \rho_0 \nabla \cdot \langle v_2 \rangle &= -\nabla \cdot \langle \rho_1 v_1 \rangle, \\ -\nabla p_2 + \mu \nabla^2 \langle v_2 \rangle + \left(\frac{1}{3} \mu + \mu_B \right) \nabla (\nabla \cdot v_2) &= \rho_0 \langle \partial_t v_1 \rangle + \rho_0 \langle (v_1 \cdot \nabla) v_1 \rangle \end{aligned}$$

Similarly, the parameters with subscript 2 denote the second-order terms.

3.1.5 Forces

In the microfluidic chamber, the acoustic radiation force is the main forces driving the movement of polymer in this system. The acoustic radiation force can be derived from the gradient of Gorkov potential, which is the acoustic pressure field induced by SAWs at the substrate/fluid interface¹⁰³. **The net effect of acoustic radiation force is to push polymer from pressure antinodes to pressure nodes**, and its equation is given by

$$F_{\text{rad}} = -\nabla \left\{ V_p \left[\frac{f_1}{2\rho_0 c^2} \langle p_1^2 \rangle - \frac{3\rho_0 f_2}{4} \langle v_1 \cdot v_1 \rangle \right] \right\},$$

with

$$\begin{aligned} f_1 &= 1 - \frac{\rho_0 c^2}{\rho_p c_p^2}, \\ f_2 &= \frac{2(\rho_p - \rho_0)}{2\rho_p + \rho_0}, \end{aligned}$$

where V_p is the volume of the sphere particle, ρ_p and c_p are its density and acoustic phase velocity, and p_1 and v_1 are the acoustic pressure and acoustic velocity derived from the first-order equations.

An intuitive description of the acoustic pressure and the radiation force¹⁰⁴ can be found in Figure 3-3. The acoustic radiation forces drive material to nodes or antinodes in the standing pressure wave, depending on the sign of the acoustophoretic contrast factor A .

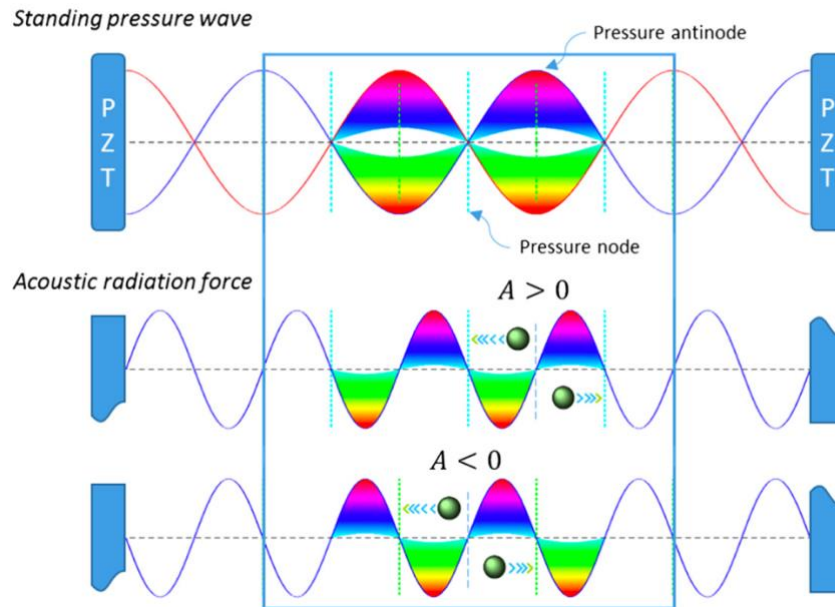


Figure 3-3 Standing pressure wave and acoustic radiation force

3.1.6 Rayleigh angle

When a travelling SAW contacts a liquid, the viscosity difference causes part of the SAW to refract into the liquid as a longitudinal wave. The refracted wave moves along the direction given by a refraction angle known as Rayleigh angle:

$$\theta_R = \arcsin(c_l/c_s)$$

And the schematic image of Rayleigh angle is shown in Figure 3-4¹⁰⁵:

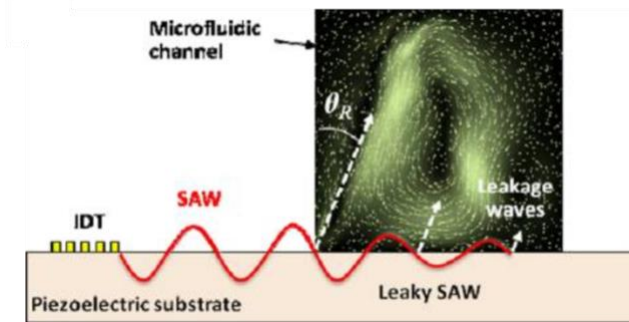


Figure 3-4 Illustration of SAW attenuation when encountering liquid.

3.1.7 Loss

In the research, we can mainly focus on the dissipation caused by liquid and ignore the influence of air. It can be illustrated by a simple calculation. The attenuation of SAW energy into the liquid/air domain is mainly contributed by a transverse wave, whose attenuation coefficient is

$$\alpha = \frac{\rho v}{\rho_s v_s} \frac{1}{\lambda}$$

where ρ , v , ρ_s and v_s are the density of the liquid/air, the sound velocity in liquid/air, the density of the substrate, the SAW velocity in the substrate. Using values in Table 3-1, the attenuation coefficient caused by air is 0.3793 (1/m), which is several orders of magnitude smaller than that caused by water.

Table 3-1 Density and acoustic velocity of different materials.

	ρ (kg/m ³)	v (m/s)
Water	997	1480
Air	1.225	343
LiNbO ₃	4650	3970

3.2 Abstract

Polymer-based microstructure are crucial for many optical applications. Here we found a brand-new method to fabricate microstructure of polymer by using surface acoustic wave (SAW). The generated acoustic pressure field from SAW device can excite micro-patterned structures on the surface. We simply encapsulate the SAW device and polymer solvent using a chamber to elongate the interaction time between the polymer and the surface acoustic wave, and as the solvent gradually evaporate, the patterned microstructures can be successfully fabricated. And we characterized the grating efficiency and found a 20% power in 1st order. This new fabrication method is simple, cheap, and fast. The studies open the door of developing a brand-new polymer fabrication method.

3.3 Introduction

“Top-down” nanofabrication has historically driven progress in nanodevice design and application, but it is limited by intrinsic machine and material constraints⁵⁹. “Bottom-up” fabrication of microstructure is a way to bypass the constraints, but it lacks the flexibility of pattern fine tuning and is hard to design. The integration of the sophisticated techniques of the top-down active device for bottom-up approaches is an exciting development, which is offering a unique opportunity to fabricate complex structured with high degrees of control and significantly expanding the possibilities of fabrication and functions.

Polymer-based microstructures have been widely utilized in optical device¹⁰⁶, tissue engineering¹⁰⁷, biomimetic smart surfaces¹⁰⁸, and organs-on-chips¹⁰⁹. However, the fabrication approaches of polymer microstructure are limited into two categories: transfer method^{110,111} and additive manufacturing, such as 3D printing¹¹². Among these fabrication methods, physical or chemical

molds and templates are usually required, and additional fabrication processes and procedures are involved, which lead to a long fabrication period and high cost. And once the mold is made, the fabrication doesn't have any turnability and flexibility. We developed a novel method using interdigital transducer (IDT) pair generated surface acoustic wave (SAW) to guide the bottom-up fabrication of the polymer microstructure patterns. This fabrication method has several advantages: a rapid fabrication period (approximately 5 minutes), a relatively low cost, and most importantly, the active device will offer the flexibility to fine-tune the period of the pattern and can be used to fabricate the pattern with more complicated structure, like beating pattern, Moiré pattern, polymer lens etc.

3.4 Main results

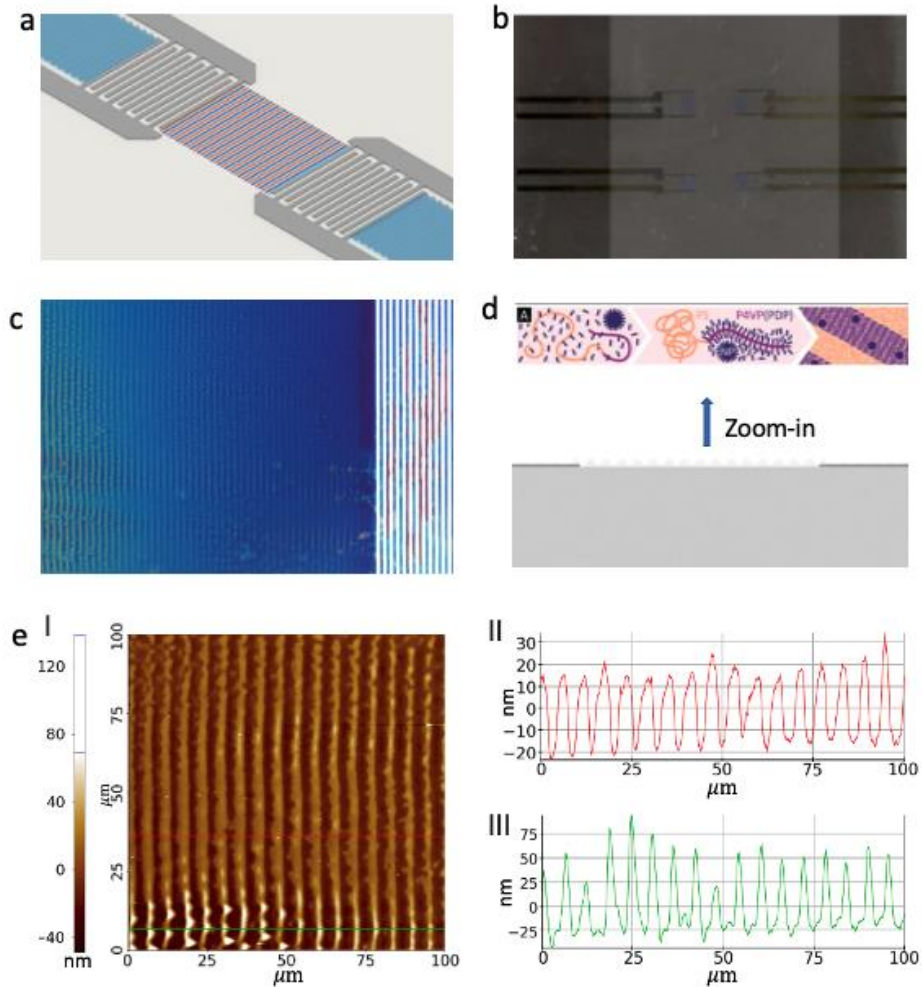


Figure 3-5 (a) the schematic interdigital transducer (IDT) device and the potential field distribution. (b) the image of the device (c) the microscopy image of the generated microstructure (g) schematical side-view of the pattern (e) The left panel I is the AFM image of the generated

microstructure. The right panels II and III are the cross-sectional view of the red and green lines in panel I.

Figure 3-5a schematically shows the interdigital transducer (IDT) device and the potential field distribution. There are two IDTs: one is at left, and the other is at right of the chip. And in the middle of the two IDTs, the surface acoustic wave generated from the IDTs will form the interference patterns and potential fields of acoustic wave. And the pattern shows the acoustic potential field distribution, there are nodes and antinodes formed. The image of the real device is in Figure 3-5b, and the pitch of the electrode is 12 μ m. The polymer and nanoparticle material will be dragged to the low acoustic potential region, resulting in the formation of microstructure shown by the microscope image in Figure 3-5c. And the schematic side view of the pattern is shown in Figure 3-5d, and the top image in Figure 3-5d shows the zoom in structure of the pattern, which is the nm sized nanoparticles and polymer. And the AFM image of the microstructure is shown in Figure 3-5e left panel, and the cross-sectional view of the red and green lines are shown in the right panel.

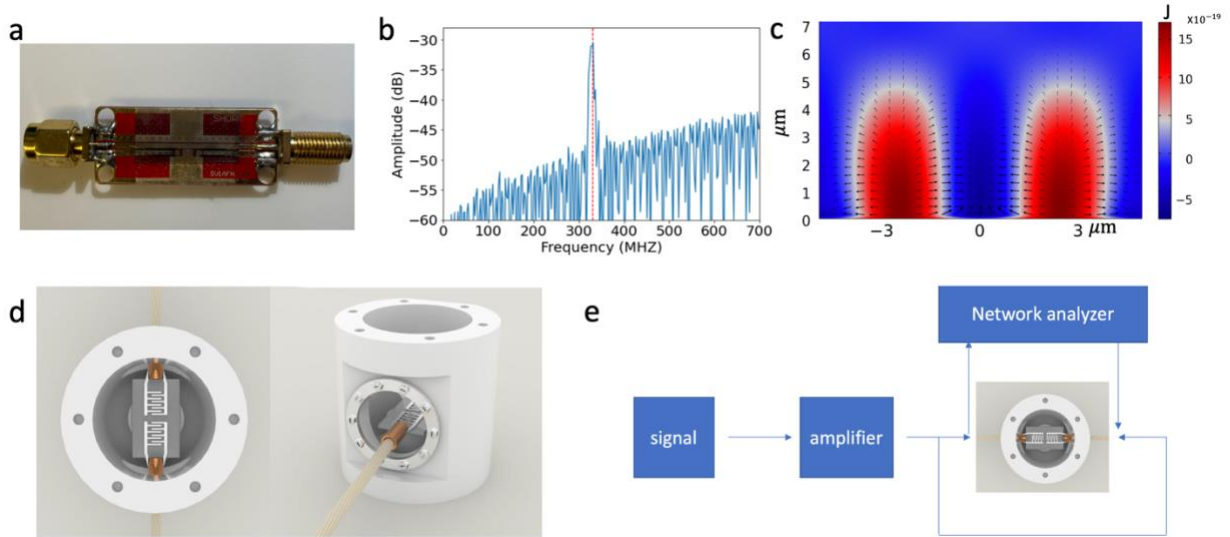


Figure 3-6 (a) the packaged device (b) network analyzer response image (c) Gorkov potential field distribution on the device from COMSOL simulation (d) the chamber used to encapsulate the sample and provide the vapor environment (e) the experimental setup

To input high frequency RF power to the device, I used a customized PCB board to package the sample, which is shown in Figure 3-6a. Given $v_s = 3970$ m/s and $p = 6$ μ m under our experimental condition, the resonant frequency is 330MHz (330MHz is marked as the dashed red line in Figure3-6b). The Figure 3-6b shows network analyzer response image, from the image, we can see the resonance peaks are in 330MHz, which matches the estimation. The Gorkov potential field distribution from COMSOL simulation results of the IDT components is shown in Figure 3-6c. For the polymer and nanoparticles, we must resolve them into the highly evaporate solvent,

and to increase the interaction time between the solution and the acoustic wave before the solvent evaporate, we need to use the chamber create a vapor environment. We encapsulate the device into a chamber shown in Figure 3-6d. The chamber is made by Teflon. It has a lid which is not shown in the image. And Figure 3-6e shows the experimental setup. The electrical signal is generated by the signal generator and the signal is then transmitted through the RF amplifier and be amplified. And the device is bonded using wire bonding to a customized PCB board and it is placed inside the chamber, where the solvent vapor environment is formed and prevent the solvent to evaporate immediately, so the liquid of polymer and nanoparticles still has reasonable viscosity and can interact with the acoustic wave and get moved and shaped to the line patterns.

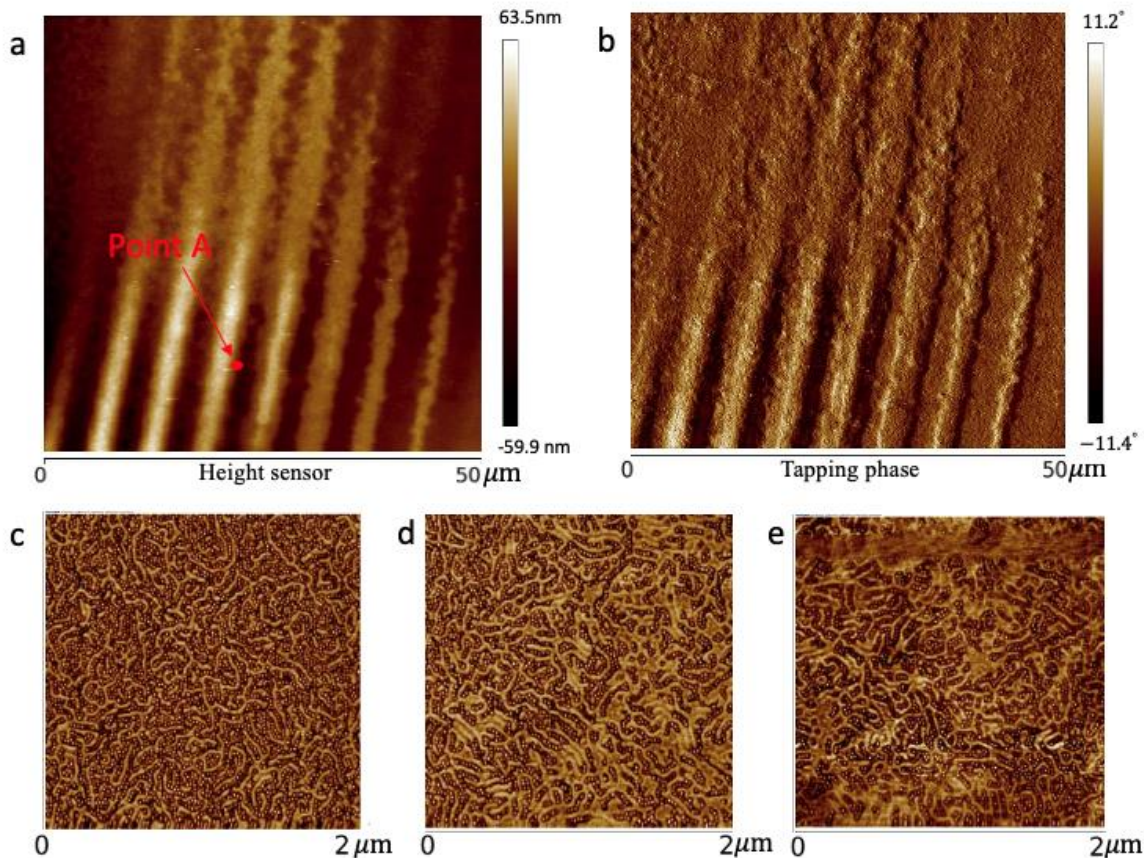


Figure 3-7 a show the AFM image of the microstructure of formed patterns in height sensor, and the Figure 3-7b shows the AFM image of the microstructure of formed patterns in Phase sensor. We hope to see the nanopattern formed within the microstructure near the edge of the peak in image a (the edge of the trench is represented using the red dot point A). Figure c shows the zoom-in nanostructure at the left-hand side of point A. Figure d shows the zoom in nanostructure around

the point A. And the Figure shows the zoom-in nanostructure at the right-hand side of point A. From Figure c d e, we know the nanoparticles are concentrated in the high peak region.

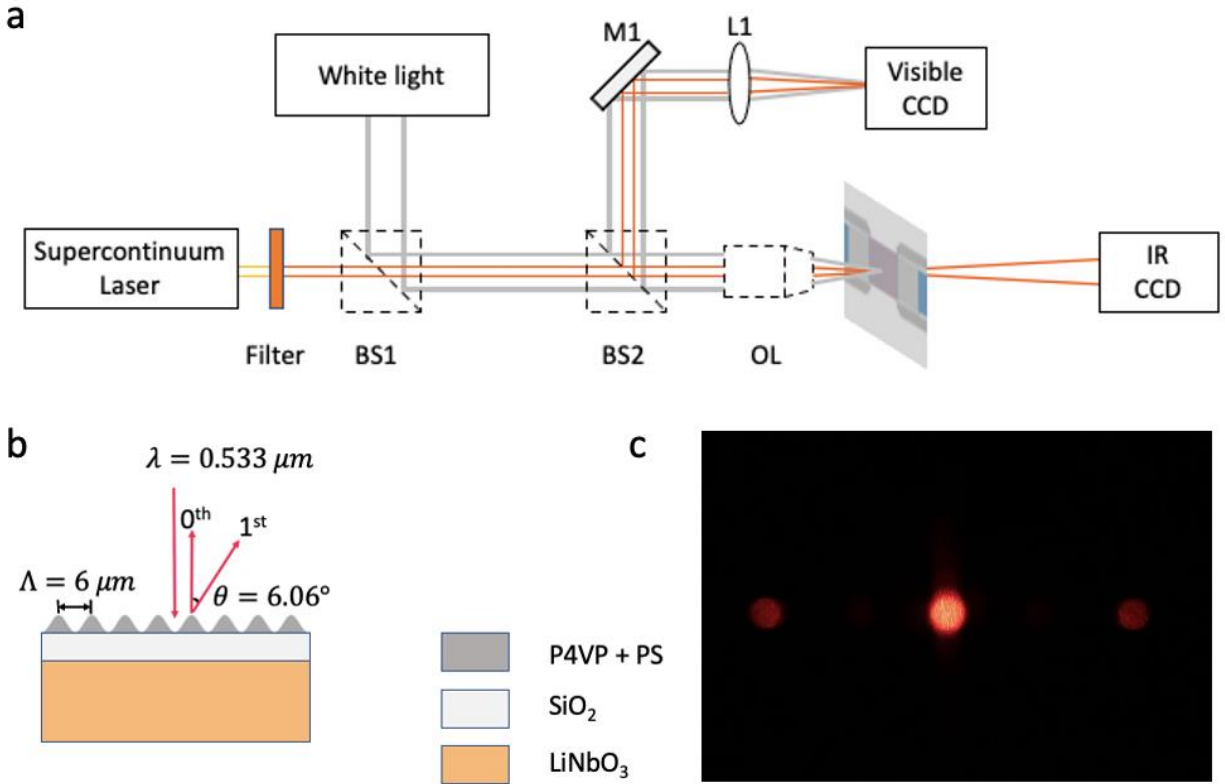


Figure 3-8 (a) optical setup to characterize the grating formed by the microstructures. (b) the schematic image of the sample (c) Experimental transmission results through pattern region, the image is captured by the CCD.

And Figure 3-8a showed the optical setup to character the optical property of the sample. And Figure 3-8b showed the sample schematic image. According to both calculation and experimental results, the 1st order in reflection should along around 6 degrees direction. Figure 3-8c shows both the 0 order and the $\pm 1^{\text{st}}$ order. The experimental results show that our surface acoustic wave generated grating can functional well with an efficiency around 20%.

3.5 Conclusion

Here we used the IDT pair generated surface acoustic wave to successfully fabricate the polymer microstructure. And we have characterized the microstructure as optical grating and characterized the grating efficiency and found a 20% power in 1st order. This is a brand-new bottom-up fabrication method using active device for polymer microstructure fabrication. This method is simple, cheap, and fast. And it also contains the flexibility of top-down fabricated active device.

We anticipate using this method, we can fine-tune the structure by shifting the frequency and by this method, we can fabricate beating pattern, moiré pattern, polymer lens etc.

Chapter 4 Orbital angular momentum changing selection rule of excitons

4.1 Background

4.1.1 Valleytronics selection rule

A local minimum in the conduction band or local maximum in the valence band is referred to as a valley. In addition to charge and spin, an electron is also endowed with a valley degree of freedom, which specifies the valley that the electron occupies. The possibility of using the valley degree of freedom to store and carry information (similar to spin in spintronics) leads to conceptual electronic applications known as valleytronic^{113–116}.

In systems that have strong spin–orbit coupling (for example, TMDs), the time-reversal symmetry dictates the spin splitting to have opposite signs at the +K and –K valleys, giving rise to an effective coupling between spin and valley pseudospin. The binary valley pseudospin can be optically addressed (that is, excited and read out) with circularly polarized light. The broken inversion symmetry of TMD systems gives rise to a valley-dependent optical selection rule, in which right circularly polarized light couples to interband transitions in the +K valley and left circularly polarized light couples to interband transitions in the –K valley.

This valley polarization, η , can be experimentally read out by measuring the circularly polarized components of the emitted photoluminescence intensity^{117–120}, PL ($\sigma \pm$), and can be expressed as

$$\eta = \frac{\text{PL}(\sigma+) - \text{PL}(\sigma-)}{\text{PL}(\sigma+) + \text{PL}(\sigma-)}$$

Valley excitons (bound valley electron–hole pairs) dominate the optical response of monolayer TMDs and will have an important role in optically driven valleytronic devices based on these materials. In principle, a valleytronic circuit based on optically injected valley excitons can be designed, in which information is stored in the exciton valley pseudospin.

For the case of monolayer MoS₂ as an example, the states at the top of the valence bands involve only $d_{x^2-y^2}$ and d_{xy} on Mo, and p_x and p_y states on S. At +K, the d-states on Mo hybridize as $1/\sqrt{2}(d_{x^2-y^2} + id_{xy})$ ($l = +2$) to interact with as $1/\sqrt{2}(p_x + ip_y)$ ($l = +1$). At –K, the d-states on Mo hybridize as $1/\sqrt{2}(d_{x^2-y^2} - id_{xy})$ ($l = -2$) to interact with $1/\sqrt{2}(p_x - ip_y)$ ($l = -1$). At the bottom of the conduction band, mainly d_{z^2} state on Mo ($l = 0$) is involved with a minor

contribution from the p-orbitals of S. These atomic orbitals form such linear combinations in accordance with the D3h point group symmetry and the lattice translational symmetry.

The optical selection rule is rooted in the phase winding of the Bloch states under rotational symmetry, threefold rotation ($\widehat{C}_3$) for TMDCs¹¹⁸. Given the symmetry-adapted linear combinations of the atomic orbitals, the azimuthal phase associated with the threefold rotation at $\mathbf{K}_{\pm}$ is readily calculable,

$$\widehat{C}_3|v(\mathbf{K}_{\pm})\rangle = |v(\mathbf{K}_{\pm})\rangle$$

And

$$\widehat{C}_3|c(\mathbf{K}_{\pm})\rangle = e^{\mp i2\pi/3}|c(\mathbf{K}_{\pm})\rangle$$

where v and c correspond to the valence and conduction band extrema, respectively.

Now the chiral optical selectivity of the valleys can be deduced. The bottom of the conduction bands at the valleys, dominated by the $l = 0$ d -states on Mo, bears an overall azimuthal quantum number $m_{\pm} = \pm 1$, at $\mathbf{K}_{\pm}$. At the top of the valence bands, $m = 0$. Then for an optical transition at $\mathbf{K}_{\pm}$, the angular momentum selection rule indicates that $\Delta m_{\pm} = \pm 1$, corresponding to the absorption of left- and right-handed photons.

4.1.2 Giant Rydberg excitons in the copper oxide Cu₂O

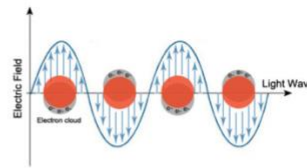
The Coulomb attraction between a negatively charged electron in the conduction band and a positively charged hole in the valence band leads to the formation of bound exciton states¹²¹, which are essential to the optical properties of semiconductors. The exciton in bulk crystals has strong similarities with the hydrogen atom, which have been substantiated by the observation of exciton states with binding energies as $E_{n,l}$ (shown by equation) below the bandgap. Here Ry is the Rydberg energy, n is an integer that is analogous to the principal quantum number n of hydrogen, and $\delta_{n,l}$ is the quantum defect induced by the non-parabolic hole dispersions¹²². The observation of highly excited excitons is typically prevented by a small Rydberg energy of a few millielectronvolts (for example, 4.2 meV in the prototypical semiconductor GaAs with $n = 3$ as its highest observed state), which is about three orders of magnitude smaller than in hydrogen owing to the small, reduced mass of the electron and hole and dielectric screening from the many-body surroundings. Therefore, higher exciton states are energetically spaced too closely to each other and to the ionization continuum to be resolvable.

$$E_{n,l} = \frac{Ry}{(n - \delta_{n,l})^2}$$

Semiconductor copper oxide (Cu₂O), facilitated by the comparatively large Rydberg energy of around 100 meV . Cu₂O has a direct bandgap. The highest valence and the lowest conduction bands are formed from Cu states, the 3d and 4s orbitals, respectively. The excitons associated with these two bands form the so-called yellow series with energies around 2.1 eV , corresponding

to a wavelength of 590 nm for excitation by light. Both bands have the same parity. Beyond the valence and conduction band symmetry, the symmetry of the exciton envelope wave function also must be considered. With photon carries one $\hbar$ of angular momentum (right and left circularly polarized light) only P excitons ($\Delta l = 1$) can be excited. For s- and d-type valence and conduction bands ($\Delta l = 2$ from the $3d$ valence to the $4s$ conduction band), the excitation is forbidden¹²³.

4.1.3 Localized surface plasmon resonance (LSPR)



Light incident on a metal nanoparticle causes the conduction band electrons to oscillate. This is the localized surface plasmon.

- LSP has two important effects: electric field near surface are greatly enhanced and the particle's optical absorption has a maximum at the plasmon resonance frequency.
- Characterized by dark field image and scattered spectra
- Spectra sensitive to the geometries, for tunability; Spectra sensitive to environment, for sensor

$d \ll \lambda$, quasi-static approximation; ideal dipole for each nanoparticles

As d is comparable to λ , mie theory; multipole (higher-order) plasmon modes can be excited due to phase retardation of the field inside the particle

4.1.4 Plasmonic bright modes and dark modes

Here I summarize some comparison between these two modes^{124,125} (For Dark mode: an efficient scheme to excite multipole plasmon resonances directly and selectively is to use size matched OAM mode^{126,127})

Bright modes: Modes with a strong dipole moment; couple very efficiently to the far field; decay radiatively; enhance both the near- and the far-field responses.

Dark modes (breathing mode): Modes exhibiting a weak dipole moment; radiative decay channel is suppressed; dark modes have a longer lifetime; representing resonators of a high quality-factor and therefore providing an extremely large near-field enhancement; pure near-field modes.

There are two types of dark mode: multipole plasmon resonances; interaction of the dipole modes in the coupled particles (Nanoparticle oligomers). Applications of dark mode: plasmon lasing, plasmon-enhanced luminescence, Fano resonance (where dark modes interfere with dipolar modes, provided that a spectral overlap exists), electromagnetically induced transparency in metamaterial. Limitations of dark mode: Associated with the weak coupling to free space, it is difficult to excite the dark modes by light in free space.

- Some dark modes can be excited by inducing Fano resonances *via* oblique incident angles or heterostructures, or breaking the symmetry of the NPs
 - allows multipole plasmons to be excited. But these modes are usually excited through bright plasmonic modes
 - the intensity of an excited multipole resonance is weaker than that of the simultaneously excited dipole resonance
- Excite by electron energy loss spectroscopy (EELS), dependent on the electron beam in extremely expensive instruments

4.2 Introduction

Optical selection rules fundamentally determine the optical transitions between energy states in a variety of physical systems, from hydrogen atoms to bulk crystals such as gallium arsenide. These rules are important for optoelectronic applications. Spin angular momentum plays an important role in exciton selection rule. According to Fermi-golden rule, OAM should also play a role for the selection rule.

4.3 Experiments on OAM interacting with transition metal dichalcogenide monolayers

The motivation of this project is the theoretical paper¹²⁸ talking about OAM changing the selection rule of valleytronic. In this paper, they predict OAM will make some of the excitations become forbidden as shown in Figure 4-1.

l		-4	-3	-2	-1	0	+1	+2	+3	+4
K	RCP	×	×	✓	×	×	✓	×	×	✓
	LCP	×	✓	×	×	✓	×	×	✓	×
-K	RCP	×	✓	×	×	✓	×	×	✓	×
	LCP	✓	×	×	✓	×	×	✓	×	×

Figure 4-1 OAM optical selection rule

To experimentally check it, I first carry out the valleytronic experiments in our lab. The optical setup is as Figure 4-2. The OL represents objective lens. HWP represents half waveplate. QWP represent quarter wave plate. LP represents linear polarizer. M represents mirror. And we keep the temperature of the sample at 4K.

And in the experiments, I prepared sample of MoS2 on top of BN shown in Figure 4-3, and the red circled area is the light incident spot. The sample image is shown LCP polarized light is incident, and the LCP and RCP photoluminescence is measured, which is marked as L-L and L-R, and then RCP polarized light is incident, and the RCP and LCP photoluminescence is measured, which is marked as R-R and R-L. The experimental results are shown in Figure 4-4. The results are as expected: RCP(LCP) incident light mainly excite RCP(LCP) photoluminescence, which is the traditional selection rule.

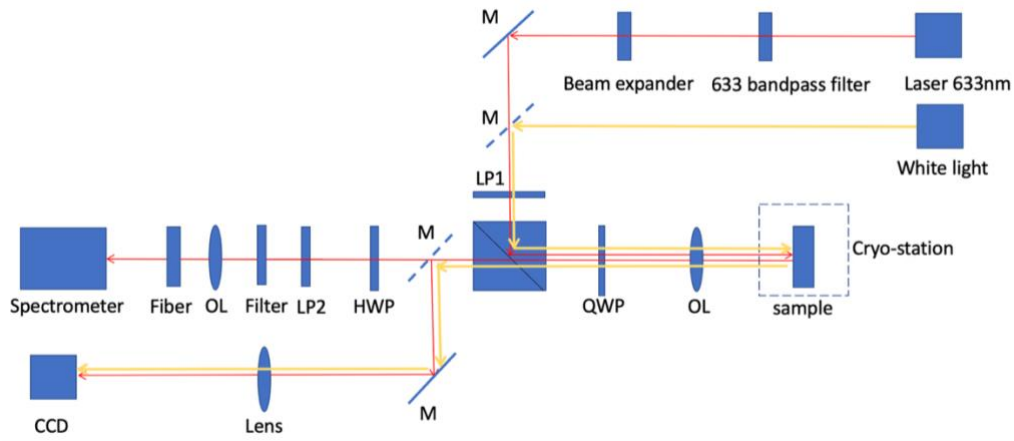


Figure 4-2 experimental setup for valleytronic measurement



Figure 4-3 image of sample, MoS2 on top of BN

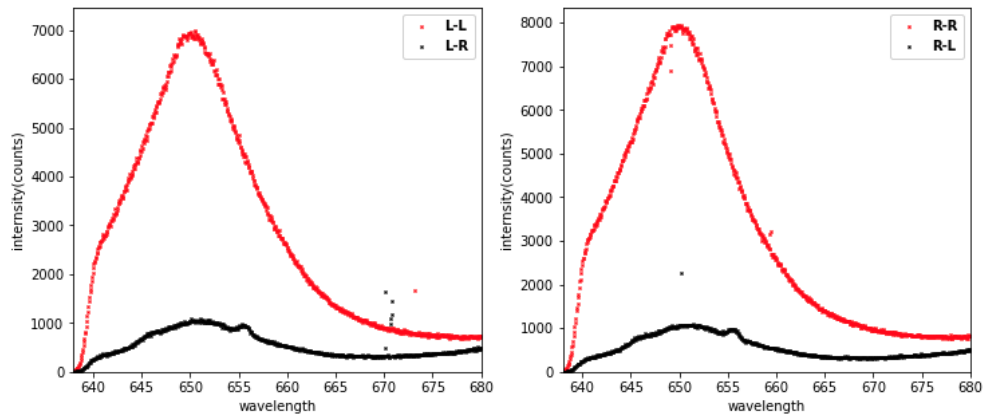


Figure 4-4 experimental results of valleytronics

To take OAM into consideration, I add spatial light modulator (SLM) into the optical path, and the optical setup is shown in Figure 4-5:

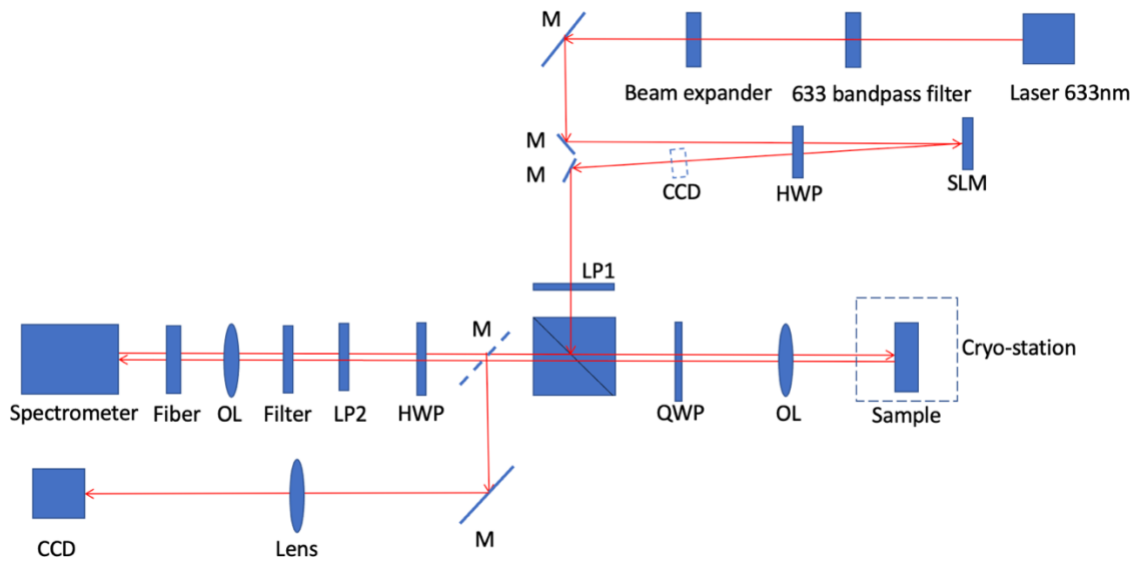


Figure 4-5 experimental setup for valleytronic measurement with OAM

And by using SLM, I tune the OAM from $l=1, 2, 3, 4$. And in the experiments, LCP polarized light is incident, and the LCP photoluminescence is measured, which is marked as L, and then RCP polarized light is incident, and the RCP photoluminescence is measured, which is marked as R. And the experimental results shown in Figure 4-6.

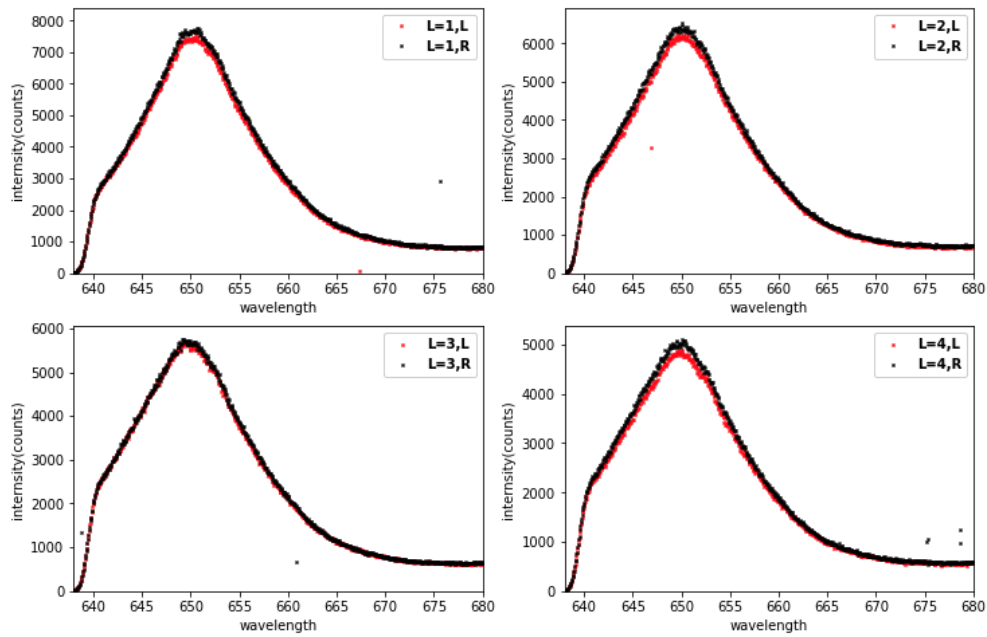


Figure 4-6 experimental results of valleytronics with OAM

From the results, we can see for $L=1$, LCP is still allowed, and for $L=4$ also LCP is still allowed. Based on the results, OAM doesn't have a strong influence on the valleytronic response. And we had some theoretical analysis and get some learnings.

Learning from the experiments: need size-matching.

The Conclusion is that the size of OAM doesn't match the size of the exciton wavefunction. Roughly speaking, for the exciton wavefunction, it has a spatial distribution of 10nm in diameter, but the OAM generated from SLM has ~ 10 micrometer in size. Therefore, for the exciton, it falls at the range of the dipole approximation, and what it sees is just locally circularly polarized response but it cannot see OAM^{123,129}. And for trapped ion it shows much more significant response¹³⁰. And this is also the same idea for so-called dark mode excitation in the surface plasmon resonance field, where the dark mode also needs the size-matching of the dark mode and the incident excitation^{126,127}.

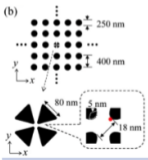
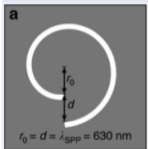
4.4 New experimental plans

Since the OAM is generated from SLM, the size of the wavefunction is limited by the pixel size of SLM. To match the size of OAM and the exciton wavefunction, we can

1. shrink OAM
2. choose a large exciton wavefunction

There are lot of plasmonic structure which can generate the focused OAM. There are two main categories: spiral structure^{131–135} and antenna structure¹³⁶. I did the summary of the difference between these two methods shown in Table 4-1.

Table 4-1 difference between two OAM focusing method

structure	Based on	tolerance against misalignment	Working spectrum range	Fab difficulty	Experimental result	Incident AM	Output AM	Consider E component	principle	Focus size
	LSPRs	400nm	30nm FWHM	The corner of the antenna	none	$S=1$ $L=1$	$j=2$	E _{xy}	focus	Antenna size (Related to incident frequency)
	surface plasmon polaritons (SPPs)	No need for alignment	None data found yet Shouldn't be too large (Based on interference)	λ_{spp} in geometry	Lots of	$S=1$ $L=m$	$j=m+s$	E _z	focus; Spin to OAM transition; Geometrical phase	$\sim \lambda_{spp}/2$ trade-off between confinement and loss demands

Choosing a large exciton wavefunction is also critical for size-matching. The transition metal dichalcogenide monolayers (TMDC) material typically have limited wavefunction size because of

the coulomb interaction is stronger for 2D material¹³⁷. And even modulating the environment dielectric screening can enlarge the exciton size, but the influence is still quite limited, and there is still a huge gap between OAM size and exciton size.

Some material like copper oxide (Cu₂O) has a giant Rydberg exciton, which is facilitated by the comparatively large Rydberg energy of around 100meV¹²¹, in which micro-meter size exciton can be observed. This is a very exciting material, which provides the potential opportunity to observe the interaction between the OAM and the exciton. With OAM, the previously forbidden S exciton potentially can be excited¹²³. And with the collaboration with Fangzhou Zhao, we calculated the exciton radius of P exciton and S exciton for different exciton level n shown in

Figure 4-7. And if you look at

Figure 4-7 and exciton energy difference in Table 4-2, you will realize there is a trade-off in experiment: as n(exciton level) increase, although we can get larger exciton size, but the energy difference between s and p exciton will become smaller, which require either tunable dye laser with ultra-thin line-width or a wavemeter with extremely high-resolution, which are very hard to be accessed by most labs. So, for experiments, a trade-off n is needed, and to combine OAM shrinking plasmonic device and a relatively high-resolution wavemeter.

n	P exciton	S exciton
1	N/A	1.665
2	5.55	6.66
3	13.875	14.985
4	25.53	26.64
5	40.515	41.625
6	58.83	59.94
7	80.475	81.585
8	105.45	106.56
9	133.755	134.865
10	165.39	166.5
11	200.355	201.465
12	238.65	239.76
13	280.275	281.385
14	325.23	326.34
15	373.515	374.625
16	425.13	426.24
17	480.075	481.185
18	538.35	539.46
19	599.955	601.065
20	664.89	666
21	733.155	734.265
22	804.75	805.86
23	879.675	880.785
24	957.93	959.04
25	1039.515	1040.625
26	1124.43	1125.54
27	1212.675	1213.785
28	1304.25	1305.36
29	1399.155	1400.265
30	1497.39	1498.5

Figure 4-7 exciton radius of P exciton and S exciton

Table 4-2 exciton energy difference and wavelength difference between P exciton and S exciton

p exciton	n	(meV)	wavelength(nm)	wl(n-1)- wl(n)	s exciton	n	(meV)	wavelength(nm)	wl(n-1)- wl(n)	delta wavelength(s-p) (nm)
	2	2147.3525	577.4552591			1	2030.3942			
						2	2139.5212	579.5689122		2.113653113
	3	2161.1317	573.7734619	3.681797143		3	2159.0792	574.3188958	5.250016344	0.545433912
	4	2166.0339	572.4748946	1.298567328		4	2165.234	572.6863749	1.632520932	0.211480308
	5	2168.2732	571.8836617	0.591232922		5	2167.8847	571.9861348	0.700240103	0.102473126
	6	2169.4715	571.567789	0.315872677		6	2169.252	571.6256034	0.360531416	0.057814387
	7	2170.1826	571.3804807	0.187308335		7	2170.0489	571.4156977	0.209905676	0.035217046
	8	2170.6389	571.2603892	0.120091466		8	2170.5516	571.2833436	0.132354155	0.022954357
	9	2170.9493	571.1787084	0.08168083		9	2170.8885	571.1947104	0.088633205	0.016001982
	10	2171.1689	571.1209373	0.057771114		10	2171.1254	571.1323661	0.062344235	0.011428861
	11	2171.3305	571.0784372	0.042500081		11	2171.2981	571.0869351	0.045431011	0.008497932
	12	2171.4528	571.0462678	0.032169392		12	2171.4281	571.0527553	0.034179823	0.0064875
	13	2171.5474	571.0213779	0.024889868		13	2171.528	571.0264851	0.026270192	0.005107177
	14	2171.6222	571.0017279	0.019650024		14	2171.6067	571.005789	0.020696096	0.004061104
	15	2171.6822	570.9859415	0.015786354		15	2171.6698	570.9891973	0.01659166	0.003255798
	16	2171.7312	570.9730665	0.012875066		16	2171.721	570.9757382	0.013459145	0.002671719
	17	2171.7716	570.9624345	0.010631943		17	2171.7632	570.9646505	0.011087655	0.002216007
	18	2171.8055	570.9535328	0.008901713		18	2171.7983	570.9554081	0.009242494	0.001875225
	19	2171.834	570.9460326	0.007500253		19	2171.828	570.9476096	0.007798417	0.001577061
	20	2171.8583	570.9396445	0.006388073		20	2171.8532	570.9409928	0.006616815	0.001348318
	21	2171.8792	570.9341503	0.005494154		21	2171.8748	570.9353104	0.0056824	0.001160073
	22	2171.8973	570.9293976	0.004752752		22	2171.8934	570.9304118	0.004898666	0.001014159
	23	2171.913	570.9252653	0.004132306		23	2171.9096	570.9261512	0.004260601	0.000885865
	24	2171.9268	570.9216377	0.003627548		24	2171.9238	570.9224227	0.003728499	0.000784913
	25	2171.9389	570.9184492	0.003188524		25	2171.9363	570.9191355	0.003287106	0.000686331

4.5 Conclusion

Since the limitation of my PhD time, I cannot see the exciting results by myself before my graduation. But I treasure this process that along the way as I read more, I have a progressive and iterative understanding for this problem, and that for the experiments, I become more and more clear about the plan. I look forward to seeing whoever follows the road can finish the experiments and prove the OAM interaction with exciton.

Chapter 5 Exploring the OAM generation from the twisted nanowire

5.1 Introduction

Paraxial optical beam can carry spin angular momentum (SAM) and orbital angular momentum (OAM). These dynamical properties are determined by the polarization and spatial degrees of freedom of light. Spin-orbit interactions not only generates OAM but also play critical rules in designing optical devices^{138,139}.

Since the discovery of OAM in 1992, it has been used in plenty of promising applications¹⁴⁰, from optical communication (OAM Multiplexing)², particle manipulation in optical tweezer³, astronomy^{141,142}, chemical syntheses¹⁴³, imaging¹⁴⁴, to quantum entanglement^{10,145,146}. For telecommunication applications, beams carrying OAM can dramatically increase the data transporting speed by using orbital angular momentum as another degree of freedom (l different channels multiplexing).

While emission and manipulation of twisted light in free space has been widely investigated, ways of generating OAM light in the integrated photonic systems are still limited¹³⁸. Inspired by the idea of OAM generation from twisted fiber and twisted photonic crystal fiber¹⁴⁷, we wonder whether it is possible to make another on-chip OAM generator using the novel twisted nanowire material synthesized in our group¹⁴⁸. Compared with the twisted fiber, this nanowire has advantages of scalable fabrication, smaller size, and larger tunability. Besides, this new type of waveguide may show some other novel optical phenomena, like the moiré pattern contributed exciton changing¹⁴⁹, which also deserves more exploration.

We aim to use EME method to simulate the spin to orbital angular momentum conversion in this twisted geometry.

5.2 Simulation method

Different from the standard FDTD methods, the Eigenmode Expansion method (EME) was used in our simulations. In this section, the fundamental physics of EME is demonstrated and the comparison between FDTD and EME is made.

5.2.1 Working principle of EME

The EME method has been well-established in photonic structure simulations for some time^{150,151}. EME is a frequency domain method to solve Maxwell's equation rigorously, and it is often used to simulate long-distance light propagation in structures like waveguides or Bragg gratings. Let's start with the definition of "Eigenmode" for waveguides, an Eigenmode is the electromagnetic

field distribution within the waveguide. Or, more strictly speaking, it is the solution to the waveguides Maxwell's equation below (assume we have harmonic time dependence)¹⁵²:

$$\nabla^2 E + \mu_0 \epsilon_0 E = 0 \quad (2.1.1)$$

Exact solution for the optical waveguide that does not vary in the z-direction would be in the form of¹⁵²:

$$E(x, y, z) = e_t(x, y) \cdot (C_1 \cdot e^{i\beta_{eff}z} + C_2 \cdot e^{-i\beta_{eff}z}) \quad (2.1.2)$$

Where $e_t(x, y)$ is the mode profile in the transverse plane and β_{eff} is the effective propagation constant for the mode, $e^{i\beta_{eff}z}$ and $e^{-i\beta_{eff}z}$ show the modes' bi-directional propagation nature within the waveguide. Mathematically speaking, $e_t(x, y)$ and β_{eff} can be viewed as the eigenfunction and eigenvalue solution for 2.1.1. For a waveguide, there exists several guided modes and infinite number of radiation modes. The combination of radiation modes and guided modes forms a complete basis set, in other words, any electromagnetic distribution within the waveguide can be expressed as the linear combination of modes within this set¹⁵²:

$$\begin{aligned} E(x, y, z) &= \sum_{m=1}^M e_m(x, y) \cdot (a_m \cdot e^{i\beta_m z} + b_m \cdot e^{-i\beta_m z}) \\ H(x, y, z) &= \sum_{m=1}^M h_m(x, y) \cdot (a_m \cdot e^{i\beta_m z} - b_m \cdot e^{-i\beta_m z}) \end{aligned} \quad (2.1.3)$$

With above analyses for the simple z-invariant structure, z-variant structure can also be analyzed. Let us consider the structure below, where a periodic refractive index change is introduced in the Z-direction:

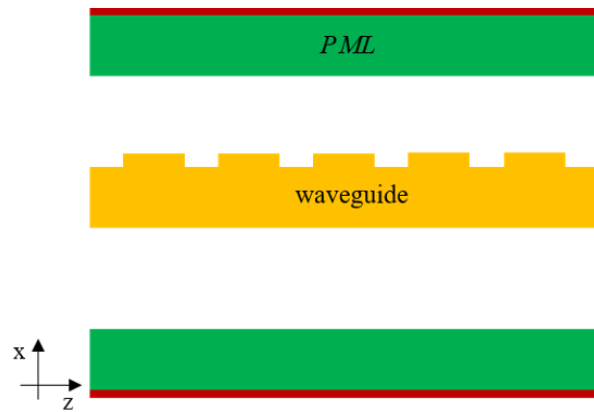


Figure 5-1 Graph for structure with periodic refractive index change in Z-direction

To perform accurate analysis, the structure is divided into many sub-waveguides, where each sub waveguide has z-invariant geometry:

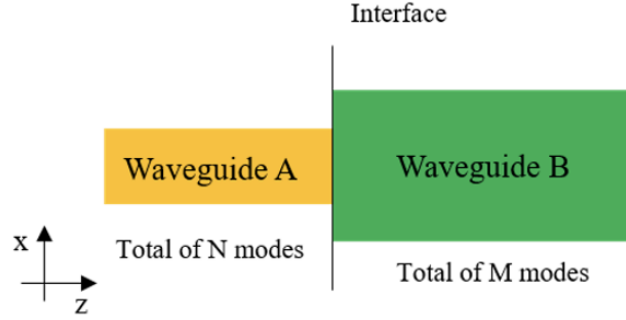


Figure 5-2 Two sub-waveguides and interface within the structure

Such simplification allows us to adopt the formulas of z-invariant structure for each sub-waveguide, and connects adjacent sub-waveguide's modes by applying Maxwell's boundary conditions (tangential electric field must be continuous on each side of the interface)¹⁵²:

$$\begin{aligned} \sum_{n=1}^N e_{n,t}(x, y) \cdot (a_n^{(+)} \cdot e^{i\beta_n z} + a_n^{(-)} \cdot e^{-i\beta_n z}) \\ = \sum_{m=1}^M e_{m,t}(x, y) \cdot (b_m^{(+)} \cdot e^{i\beta_m z} + b_m^{(-)} \cdot e^{-i\beta_m z}) \end{aligned} \quad (2.1.4)$$

This equation can also be expressed in the matrix form¹⁵²:

$$\begin{bmatrix} A_- \\ B_+ \end{bmatrix} = S \begin{bmatrix} A_+ \\ B_- \end{bmatrix} \quad (2.1.5)$$

Where: $A_+ = (a_1^+, a_2^+, \dots, a_N^+)$; $A_- = (a_1^-, a_2^-, \dots, a_N^-)$; $B_+ = (b_1^+, b_2^+, \dots, b_M^+)$; $B_- = (b_1^-, b_2^-, \dots, b_M^-)$.

S matrix is called scattering matrix with dimension $(N+M) \times (N+M)$, and it is the most important parameter we want to extract in the EME simulations, since it provides us with the transmission and reflection between adjacent subwaveguides.

After obtaining the single scattering matrix from the interface between adjacent subwaveguides, the total S matrix for the whole z-variant structure can be easily calculated by cascading all the individual S matrix from all the interfaces. This total S matrix provides the transmission and reflection from the input mode to the output mode.

5.2.2 Comparison between EME and FDTD

For smoothly varying geometries like the taper waveguide and long periodic structure like Bragg gratings, EME is the best method for simulation. Computation time is much shorter since the S

matrix technique provides solution for all input ports at the same time. In EME method, when one part of a device is altered, only the S-matrix of that part needs to be recomputed. FDTD method needs larger computation cost, but it provides more information, since it stores field profiles at each time step and each position. One drawback for the EME method is that different from FDTD's broad application range, EME can only be implemented in the waveguide-based systems.

5.3 General formulism of coupled-mode analysis for twisted nanowire structure

Coupled-mode theory is a great tool to get quantitative information like the amount of transmission and reflection for the periodic structures¹⁵³. There are other existed theories for the system, like: considering the twist as a small quantum perturbation to the cylindrically symmetric step index fiber¹⁵⁴. But here we consider only the coupled-mode theory since it is straightforward and intuitive.

5.3.1 Coupled-mode theory

In the ideal waveguide, where no z-dependent refractive index change is introduced, electric field can be expressed by superposition of the modes¹⁵³:

$$E_t(x, y, z) = \sum_j e_{jt}(x, y) \cdot (A_j(z) \cdot e^{i\beta_j z} + B_j(z) \cdot e^{-i\beta_j z}) \quad (3.1.1)$$

Where A_j and B_j are the varying amplitude of the j th mode in the +z and -z direction. In ideal waveguide, guided modes are orthogonal to each other, and hence they are not coupled. However, when some periodic modulation in the z-direction is introduced, such dielectric perturbation brings modes to be coupled. In the coupling case, the evolution of the amplitude along propagation direction can be expressed as¹⁵³:

$$\begin{aligned} \frac{dB_j}{dz} &= -i \sum_k A_k (K_{kj}^t - K_{kj}^z) \cdot e^{i(\beta_k + \beta_j)z} - i \sum_k B_k (K_{kj}^t + K_{kj}^z) \cdot e^{-i(\beta_k - \beta_j)z} \\ \frac{dA_j}{dz} &= i \sum_k A_k (K_{kj}^t + K_{kj}^z) \cdot e^{i(\beta_k - \beta_j)z} + i \sum_k B_k (K_{kj}^t - K_{kj}^z) \cdot e^{-i(\beta_k + \beta_j)z} \end{aligned} \quad (3.1.2)$$

K_{kj}^t is the transverse coupling coefficient between mode j and mode k , which can be calculated by the formula¹⁵³:

$$K_{kj}^t(z) = \frac{\omega}{4} \int \int dx dy \Delta\epsilon(x, y, z) e_{kt}(x, y) \cdot e_{jt}(x, y) \quad (3.1.3)$$

Where $\Delta\epsilon(x, y, z)$ is the dielectric perturbation to the system. Generally, the coupling in the transverse direction is way larger than the coupling in the propagation direction, so we can simply neglect the K_{kj}^z term in equation 3.1.2. Also, the majority of the waveguide systems only care about the coupling between two modes, which further simplifies the expression for amplitude evolution along the propagation direction.

5.3.2 Selection rules for twisted nanowire

According to the literature, the OAM generation is related to the mode conversion from core mode to cladding mode¹⁵⁵. Based on the coupled-mode theory in 5.3.1, two requirements are needed for the modes carrying different angular momentum to be coupled¹⁵⁵:

$$\begin{aligned}\beta_1 - \beta_2 + N\kappa_u &= 0 \\ M_1 - M_2 \mp N &= 0\end{aligned}\quad (3.2.1)$$

where $\beta_{1,2}$ and $M_{1,2}$ are the effective propagation constants and angular momentum charges of two eigenmodes that propagate independently in the reference waveguide, respectively. $k_u = \frac{2\pi}{\Lambda}$ is the twist ratio (Λ is the twist pitch of the N fold symmetry fiber). First equation is the phase matching condition. Second equation shows conversion of the Angular Momentum in mode coupling, where the interactions between OAM and SAM are involved.

The mode basis set is defined as follow¹⁵⁶:

$$\begin{aligned}HE_{mn}^{\pm} &= HE_{mn}^e \pm jHE_{mn}^o \approx F_{m-1,n}(\hat{x} \pm j\hat{y})e^{\pm j(m-1)\phi} \\ EH_{mn}^{\pm} &= EH_{mn}^e \pm jEH_{mn}^o \approx F_{m+1,n}(\hat{x} \mp j\hat{y})e^{\pm j(m+1)\phi}, \quad m \geq 1 \\ V_{0n}^{\pm} &= TM_{0n} \mp jTE_{0n} \approx F_{1n}(\hat{x} \pm j\hat{y})e^{\mp j\phi}\end{aligned}\quad (3.2.2)$$

And the specific coupling conditions derived from the selection rules 3.2.1 are listed in the table 5-1 below:

Table 5-1 specific coupling conditions¹⁵⁶

Fiber Mode 2 Excited Mode	AM-Matching Condition	Fiber Mode 1 Generated Mode
$HE_{mn}^+(s=1, l=m-1, j=m)$ $EH_{mn}^+(s=-1, l=m+1, j=m)$	$M_1 = \pm N + m \neq 0$	$HE_{ \pm N+m f}^{\sigma_1}(s=\sigma_1, l=\pm N+m-\sigma_1, j=\pm N+m),$ $EH_{ \pm N+m f}^{\sigma_1}(s=-\sigma_1, l=\pm N+m+\sigma_1, j=\pm N+m),$ $\sigma_1 = \text{sign}(\pm N + m)$
$HE_{mn}^-(s=-1, l=1-m, j=-m)$ $EH_{mn}^-(s=1, l=-1-m, j=-m)$	$M_1 = \pm N - m \neq 0$	$HE_{ \pm N+m f}^{\sigma_2}(s=\sigma_2, l=\pm N-m-\sigma_2, j=\pm N-m),$ $EH_{ \pm N-m f}^{\sigma_2}(s=-\sigma_2, l=\pm N-m+\sigma_2, j=\pm N-m),$ $\sigma_2 = \text{sign}(\pm N - m)$

Detailed definitions of the hybrid HE, EH, TE, TM modes; even modes, and odd modes are listed below¹⁵⁶:

$$\begin{cases} e_{zm}(r, \phi) = E_{zm}(r) \begin{cases} \cos(m\phi) \\ \sin(m\phi) \end{cases} \\ e_{rm}(r, \phi) = F_m^{HE}(r) \begin{cases} \cos(m\phi) \\ \sin(m\phi) \end{cases} \\ e_{\phi m}(r, \phi) = F_m^{HE}(r) \begin{cases} -\sin(m\phi) \\ \cos(m\phi) \end{cases} \end{cases} \quad \text{for } HE_{mn} \text{ modes}$$

(3.2.3)

$$\begin{cases} e_{zm}(r, \phi) = E_{zm}(r) \begin{cases} \cos(m\phi) \\ \sin(m\phi) \end{cases} \\ e_{rm}(r, \phi) = F_m^{EH}(r) \begin{cases} \cos(m\phi) \\ \sin(m\phi) \end{cases} \\ e_{\phi m}(r, \phi) = F_m^{EH}(r) \begin{cases} \sin(m\phi) \\ -\cos(m\phi) \end{cases} \end{cases} \quad \text{for } EH_{mn} \text{ modes}$$

5.4 Results and discussions

5.4.1 Geometric construction

A simple Lumerical script was used to build the twisted nanowire structure with rectangular cross section. The geometric twisted structure is shown in Figure 5-3a to mimic the nanowire.

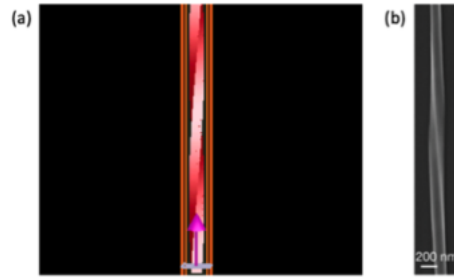


Figure 5-3 The geometry structure of the twisted wire in (a) simulation model, (b) SEM image of a twisted nanowire adhering to a substrate¹⁵⁷

5.4.2 Mode construction

Due to the limitations of EME setting in the commercial software, instead of directly solving the supported OAM modes in the waveguide, different OAM modes are constructed manually based on the following equation:

$$OAM_n^+ = HE_{n1}^{odd} + iHE_{1n}^{even} \quad (4.2.1)$$

The nth order OAM mode can be generated by applying a $\frac{\pi}{2}$ phase shift between the corresponding linear polarized even and odd mode in the untwisted reference waveguide.

In our system, it can be calculated from selection rules in Equation 3.2.1 that two coupled OAM modes should have the order difference of 2. The default mode solver in EME is used to extract

mode profiles together with their effective index, which is needed for the OAM mode construction later.

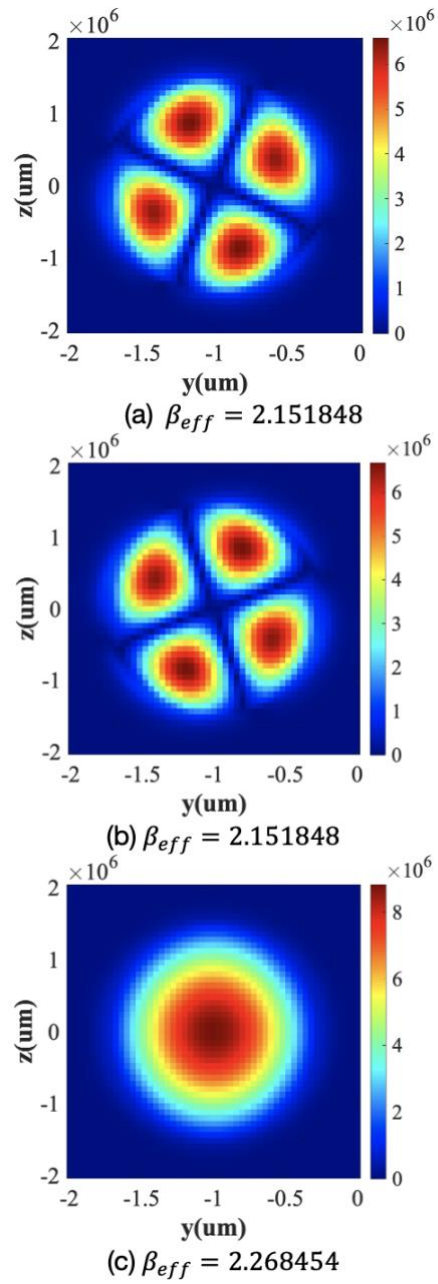


Figure 5-4 mode profile and effective index of (a) HE_{31}^{odd} , (b) HE_{31}^{even} , and (c) HE_{11}^{even} in FDE solver's

It can be seen easily from Equation 4.2.1 that HE_{11}^{even} and HE_{11}^{odd} is used to generate fundamental mode with OAM order equals to zero. HE_{31}^{even} and HE_{31}^{odd} is used to generate higher order mode with OAM order equals to two.

Mode profiles and phase information for these two modes are plotted in Figure 5-5 and Figure 5-6 (here in the simulation, the light is propagating along x direction). Only the phase distribution in the transverse plane is considered.

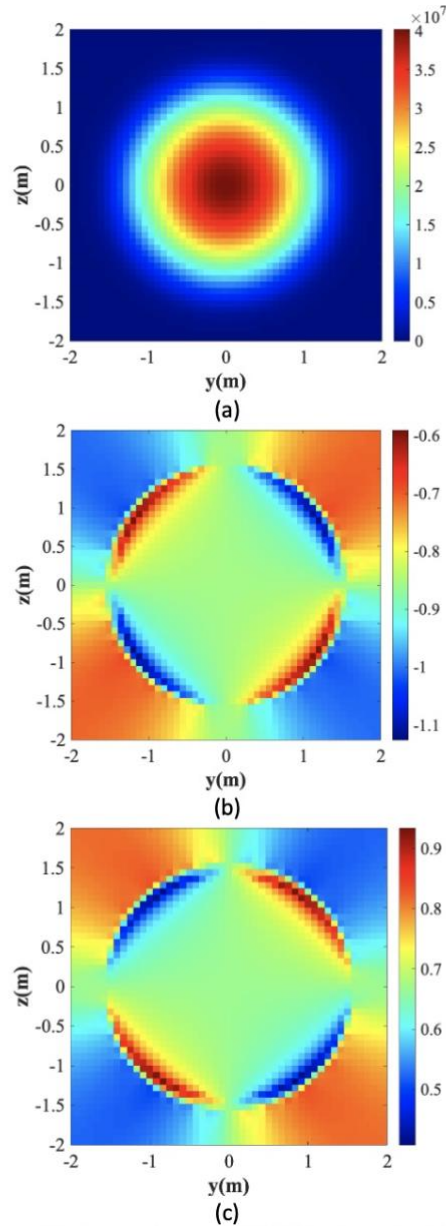


Figure 5-5 (a) mode profile for fundamental OAM = 0 mode (b) y-direction phase distribution for OAM =0 mode (c) z-direction phase distribution for OAM =0 mode

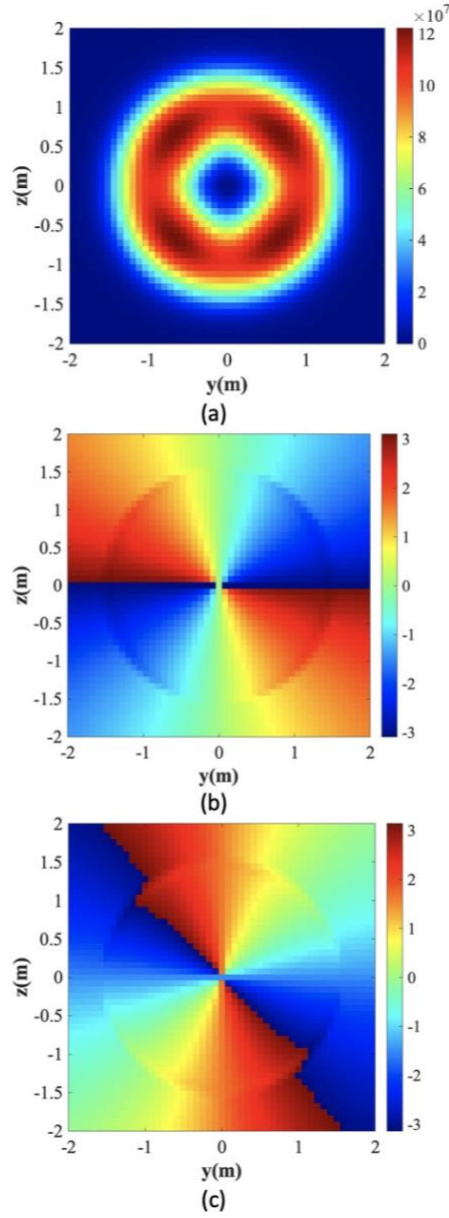


Figure 5-6 (a) mode profile for fundamental OAM = 2 mode (b) y-direction phase distribution for OAM =2 mode (c) z-direction phase distribution for OAM =2 mode

By comparing Figure 5-5 with Figure 5-6, An intensity hole can be observed in the Figure 5-6(a). And in the Figure 5-6(b) and (c), one can clearly see the phase distribution for the higher order OAM mode with 2 nodes in the azimuthal direction.

5.4.3 Mode coupling

Combining selection rule analysis, EME method, and mode construction formulas we have discussed above, we are able to simulate our twisted nanowire system. The final S matrix we get in the simulation confirms what we proposed: there is a large transmission from OAM = 0 mode to OAM = 2 mode in the twisted nanowire structure, while there is a near-zero transmission between the two modes in the reference untwisted nanowire system.

5.5 Experiments preparation

5.5.1 Nanowire fabrication

GeS nanowires are grown on top of the silicon and are tightly adhere to the substrate. To prepare the transfer of GeS nanowires, the bonding need to be removed. The PMMA are dropped on top of the substrate and encapsulates the nanowires. After that, Hydrogen fluoride (HF) is used to treat the substrate to release the PMMA solid carrying nanowires from the substrate.

Then, nanowires are released from PMMA. Putting the PMMA on the aimed substrate and immerse them in acetone liquid with heating. The nanowire will be released to the new substrate.



Figure 5-7 Nanowire on new substrate

Finally, we selectively transfer the nanowires with proper length and diameter to the edge of the substrate using PDMS. The nanowire is circled out by black circle in Figure 5-8. And its distance to the edge is ~500um, which is within the focal length range of objective lens.

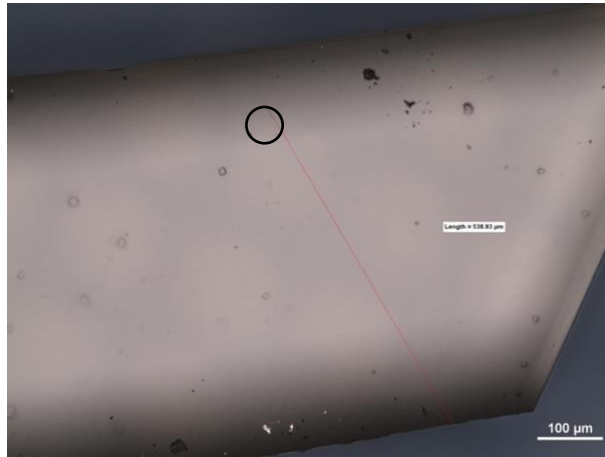


Figure 5-8 PDMS transferred nanowire on the edge of the substrate

5.5.2 Optical measurements

Since the transferred nanowires are on the edge of the substrate, it is much easier to couple light into the nanowire using objective lens. Quarter waveplate can generate circularly polarized light from NIR tunable laser source. And the light can be coupled into the nanowire waveguide by the edge coupling and the transmitted profile will be collected and imaged, and the transmission spectrum will also be detected by the spectrometer. We expect the donut pattern of OAM will be imaged and the spectrum results will show the wavelength selectivity.

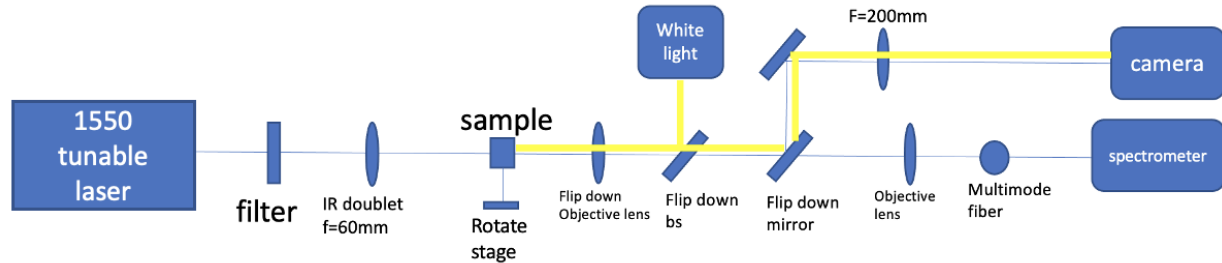


Figure 5-9 optical setup for transmission profile and spectrum analysis

5.6 Conclusion

In this project, I successfully demonstrate the spin-orbit interactions in the twisted nanowire system using EME simulation method. Coupled-mode theory is implemented to obtain the phase & angular selection rules for the OAM mode conversion. More specifically, fundamental OAM = 0 mode is coupled to higher order OAM = 2 mode in our nanowire. And I did some preliminary experiments preparation. For potential future works, investigating the effects of refractive index twist nanowire would be interesting. We would like to know whether this kind of index twist can generate similar OAM mode selection rules as the geometry twist or not. And future experimental verification will also be meaningful.

Chapter 6 Summary and Outlook

In this dissertation, I have presented my research on the interaction between wave and nano-material and nanostructure, including optical wave and acoustic wave.

For optical wave:

I explored OAM generation from nanostructure like concentric pseudo-metal rings, photonic crystal, twisted nanowire. The spin-orbit-interaction will help to convert SAM to OAM. And the twisted geometric in nanostructure will also contribute OAM. And for material valence to conduction band transition, OAM can potentially play a role to compensate the angular momentum requirement.

For acoustic wave:

The acoustic wave has a much larger force compared to optical wave, and I have explored using acoustic wave to help the assembly of polymer and nanoparticle.

Nano-devices open the doors for us to fully control the wave. This is a new field with lots of opportunities. The limitation is only our imagination of the nanostructure. And the fully controlled wave inversely will help us modify the material properties, and these modifications can then be iterated into the future nano-device design.

With the development of science and technology our understanding of nature is quickly expanding. Yet the more we learn, the more we learn how little we know. In the exploration of the vast unknown territory of science, we are touching a world at smaller, faster, and more complex scales. I am honored to be able to get involved in some of the most exciting discoveries. And I am sure there will be many more down the road and let's see what treasures are behind these opened doors.

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