Observation of photonic spin-momentum locking due to coupling of achiral metamaterials and quantum dots

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Abstract

Chiral interfaces provide a new platform to execute quantum control of light-matter interactions. One phenomenon which has emerged from engineering such nanophotonic interfaces is spin-momentum locking akin to similar reports in electronic topological materials and phases. While there are reports of spin-momentum locking with combination of chiral emitters and/or chiral metamaterials with directional far field excitation it is not readily observable with both achiral emitters and metamaterials. Here, we report the observation of photonic spin-momentum locking in the form of directional and chiral emission from achiral quantum dots (QDs) evanescently coupled to achiral hyperbolic metamaterials (HMM).

Efficient coupling between QDs and the metamaterial leads to emergence of these photonic topological modes which can be detected in the far field. We provide theoretical explanation for the emergence of spin-momentum locking through rigorous modeling based on photon Green’s function where pseudo spin of light arises from coupling of QDs to evanescent modes of HMM.

Keywords: topology, metamaterials, quantum dots, spin-momentum locking

Supplementary material for this article is available online

(Some figures may appear in colour only in the online journal)

1. Introduction

Topological photonics [1, 2] is an emerging area where various novel phenomenon like photonic spin Hall effect [3–6], photonic topological insulators [7–9], unidirectional propagation of light [10–14] etc have been reported using various degrees of freedom of light coupled with advanced design of metamaterial and metasurfaces [15, 16]. Spin-momentum locking, a key manifestation of topological phase, has been reported for photonic systems where extrinsic helicity of light or explicitly chiral emitters are coupled to surface plasmon polaritons or waveguide modes [13, 17, 18] in near field [19]. Various chiral structures have been used to demonstrate chirality in emission including nanofibers with trapped nanoparticles or atoms on it [10, 17, 20] and engineered photonic crystal waveguides having chiral modes [11, 12, 21, 22].
Another system which has been widely used to demonstrate photonic topological effects are metamaterials which have several fascinating properties [23–26, 26–28], etc. Demonstration of spin-momentum locking in metamaterials [9, 29] and metasurfaces [30, 31] have been mostly restricted to intrinsically chiral materials or in combination with chiral emitters and with directional far field excitation.

A special class of metamaterials which possesses a hyperbolic iso-frequency surface known as hyperbolic metamaterials (HMM) have a number of novel properties like anisotropy and large photonic density of states which can be used for a number of applications [32–38]. The high wavevectors (high-$k$) of an emitter are evanescent in conventional dielectric media but HMM supports propagation of high-$k$ wavevectors [39–41]. However, it has not yet been possible to utilize the properties of these modes by transporting them to the far-field. In general, it has been demonstrated that when chiral emitters couple with evanescent high-$k$ modes of HMMs unidirectional and chiral emission emerges in the near-field [19, 42]. HMM provides the extrinsic momentum to photon by making it directional which couples to intrinsic spin of photon (polarisation state of light) analogous to a spin–orbit interaction. However, transport of these photonic topological modes, especially with intrinsically achiral structures and normal incidence excitation, to the far field has not yet been reported.

Here, we report experimental observation and theoretical modeling of photonic topological effects in the form of directional and chiral far field emission from intrinsically achiral semiconductor quantum dots (QDs) coupled to a two dimensional (2D) achiral HMM. We observe clear signatures of directional and chiral emission from the hybrid system in wavevector resolved emission map in experiments. We report a new regime of spin-momentum locking phenomenon where extrinsic pseudo helicity of light is provided by coupling quantum emitters to evanescent high-$k$ modes of HMM without the need for directional excitation using standard normal incidence illumination. The observed directional emission, in experiments, coincides with the appearance of strong splitting of the photoluminescence (PL) spectra from the QDs on HMM [43] [supplementary information (https://stacks.iop.org/JPCM/33/015701/mmedia) (SI)]. The experimental results are explained reasonably well using rigorous numerical scheme to evaluate electric field distribution using method of photon Green’s function for geometry similar to that used in experiment. Significantly, we also demonstrate the ability to detect the photonic topological modes generated due to the high-$k$ evanescent HMM modes to the far-field which, coupled with normal incidence illumination, generalises the scope for observation of spin-momentum locking phenomenon with emitters and metamaterials. Note that this locking refers to the locking of the propagation direction to the spin or the polarization of the photons and has been the subject of extensive discussion in literature [4, 6]. Overall, our study provides a methodology to obtain far field photonic spin-momentum locking without having to use chiral emitters or chiral metamaterials.

2. Results and discussion

The experimental results are based on fabricated 2D HMM templates consisting of hexagonal arrays of silver nanowires (length 800 nm) embedded in alumina matrix prepared using methods described earlier [44]. To fabricate the HMM, nano-porous alumina membrane was prepared by two step-anodization of high purity aluminium metal. Porous alumina membrane contains the ordered array of pore with diameter 50 nm and inter-pore distance 110 nm. Silver nano-wires were grown into porous alumina membrane using electrochemical deposition process as detailed elsewhere (SI). Fabricated HMM was a structure with ordered array of silver nanowire (length 800 nm) embedded in alumina matrix. A thin polymer layer of polystyrene was spin coated on top surface of HMM as a spacer layer to control the distance between QD monolayer and avoid the quenching caused by metal. The purpose of the polymer spacer layer in experiments is critical since it controls the effective near field coupling strength of the QDs to the HMM evanescent modes. Another important aspect is the refractive index matching between the compact QD layer and the intervening layer between the HMM and QD. The refractive index is reasonably well matched for the QD and polymer leading to reduced scattering loss of the QD emission which excites the HMM modes. Thickness of polymer layer was controlled by concentration of polymer. We used the cadmium selenide (CdSe) QD as an achiral and isotropic emitter with mean PL at 660 nm to ensure that the emission couples and excites the hyperbolic modes of the template (SI, figure S1). A compact monolayer of CdSe QDs, synthesised by well known methods [43, 45, 46] (SI), was placed on top of the metamaterial using the well known Langmuir–Blodgett technique as discussed earlier [47, 48] as well as in SI. The metamaterials prepared undergo a wavelength dependent topological transition to a phase having hyperbolic optical dispersion hence called HMM depending on silver nanowire filling fraction ($f = 0.15$ in our study) (SI). The peak emission wavelength of the QDs was chosen such that it lies well inside HMM dispersion spectral regime (SI). Atomic force microscopy (AFM) and scanning electron microscopy (SEM) images of the top surface of HMM is shown in figures 1(a) and (b), respectively, revealing the ordered structure of the array as well as the lattice spacing. AFM image of the same template after transfer of a compact QD monolayer on top of a polymer spacer is shown in figure 1(c) with the thickness of the film (7 nm) being revealed from the typical height profile in figure 1(d) which roughly corresponds to the diameter of the QDs used. For back focal plane, Fourier imaging the QD monolayers on HMM templates were excited by focusing a 633 nm continuous wave (CW) laser using 100× objective with numerical aperture (NA) being 0.95. Emission was collected by the same objective [49] (SI). Polarization of incident excitation is controlled by half wave plate in front of laser. Circular polarization (CP) excitation is generated by a combination of half wave plate and quarter wave plate. The helicity of CP incident light was switched from left-handed circular polarization (LCP) to right-handed circular polarization (RCP) by changing the angular orientation of quarter wave plate with
Figure 1. (a) AFM image of top surface of HMM template before transfer of QD monolayer. (b) SEM image of top surface of HMM template showing silver nanowire diameter of $\sim 50$ nm and inter-wire separation of $\sim 110$ nm (inset shows fast Fourier transformation of SEM image). (c) Shows the AFM image of HMM after transfer of compact QD monolayer with the corresponding line profile along the red line shown in (d).

Figure 2. (a) Schematic of the experimental setup while the inset shows a typical back focal plane Fourier image pattern. Fourier space emission profiles with unpolarised excitation for (b) QDs on HMM corresponding to $d = 10$ nm and (c) for QD on glass. (d) QD emission intensity as a function of in-plane wave vector $k_x/k_0$ for the patterns shown in (b) and (c).

respect to incident polarization direction (SI). For PL spectroscopy measurements excitation was performed with 514 nm laser as described earlier [43] and in SI.

Figure 2(a) shows schematic of experimental system which demonstrates the directional and chiral emission from QD monolayer coupled to HMM. In figures 2(b) and (c), we have shown the Fourier images for QDs on HMM and QD on glass, respectively for spacer layer thickness, $d = 10$ nm. A clear signature of directional emission was observed for QDs placed in the near-field of HMM and emission is azimuthally symmetric and wavevector preferential. Azimuthally symmetric rings of maximum intensity are observed for QDs placed in
near-field of HMM at a separation of 10 nm, which is shown in figure 2(b). We also performed a reference measurement for the same QD monolayer on glass as shown in figure 2(c). Figure 2(c) shows a completely isotropic and homogeneous emission from the same QD monolayer on glass. To quantify this data further, we extracted the line profile of emission intensity for a band of $k_y/k_0 = 0 \pm 0.12$ along $k_x$ as indicated in figures 2(b) and (c) by a pair of lines. Emission intensity is integrated over a band of $k_y/k_0 = 0 \pm 0.12$ to avoid the spatial resolution limited fluctuations in emission intensity. These integrated intensity profiles when plotted as a function of $k_x/k_0$ in figure 2(d) reveal two symmetric peaks of maximum emission intensity $k_x/k_0 = \pm 0.70$ for $d = 10$ nm, whereas for the reference sample (QD on glass) it does not show any wavevector preferential emission. In summary, figure 2 manifests that the emergence of directional emission from QDs on HMM is due to the coupling of excitons to high-$k$ modes of HMM.

After confirmation of the connection of directional emission with high-$k$ modes, we explore the study of the chiral nature of these modes. In general, these high-$k$ modes are evanescent in nature. To study this, we excited our system with LCP and RCP polarised 633 nm CW laser and emission was collected in the Fourier image. Figures 3(a) and (b) show LCP and RCP excitation dependent Fourier images for QDs on HMM and QDs on glass systems. It should be noted that we have eliminated the central region ($NA = 0$ to $\pm 0.24$) of the Fourier image which has emitted signal of the uncoupled QDs with high-$k$ modes of HMM. The resultant Fourier image of LCP and RCP excitation shows a circular ring of maximum intensity followed by a minima located at mutually opposite azimuthal angles ($\phi$) as visible in figures 3(a) and (b), which shows the helicity dependent emission. The radius of the ring gives us the directions of emission which are characterized by $k_x$ and $k_y$. To extract the emission intensity line profiles from the Fourier images, a pair of white dotted lines are drawn in a fixed direction. Extracted emission intensity profile is normalised with maximum intensity of extracted line profile and plotted in figure 3(c). Normalised emission intensity profile shows antisymmetric intensity peak for LCP and RCP excitations. For RCP excitation, maximum intensity peak occurs at $k_x > k_0$ and the case is reverse for LCP excitation. After confirming the chiral nature of emission from HMM based system, we calculated chirality ($C$) using following expression:

$$C = \frac{I_m(P_+) - I_m(P_-)}{I_m(P_+) + I_m(P_-)}, \quad (1)$$

where $I_m(P_+)$ and $I_m(P_-)$ are local maximum intensity for $k_x > k_0$ for LCP and RCP incident polarizations and we have $0 \leq C \leq 1$. The value $C = 0$ stands for the excitation by using a linear polarization, while $C = 1$ implies a truly unidirectional excitation of the field. The chirality parameter, $C$ was found to be 0.35, from above-mentioned analysis of Fourier image as indicated in figure 3(c). We also performed LCP and RCP excitation dependent Fourier imaging for QDs on glass as reference measurement as shown in figures 3(d) and (e), respectively. Emission intensity line profiles for QDs on glass are plotted in figure 3(f) which clearly shows no chiral

![Figure 3](image-url)
pattern. Thus, our experimental observations suggest the 
ability of the QDs to couple HMM high \( k \) modes leading to 
observation of spin-momentum locking detectable in far-field. 
Interestingly, this phenomenon disappears for larger value of 
spacertickness, \( d \), (SI, figure S4), suggesting an intricate 
connection between the emergence of spin-momentum lock-
and the observation of splitting of PL spectra for QDs on 
HMM templates as shown in detail by us earlier [43] and in 
(SI, figure S3).

3. Theoretical modeling

We next develop a model for the observations. We use rigorous 
solutions of Maxwell equations, i.e., the development is similar 
to a recent one [50] but is generalized to the present experi-
mental geometry. Unlike the layered medium in the previous 
work, the HMM in the present work is different and is char-
acterized by complex effective dielectric tensor. The detailed 
modeling is given in SI. We can numerically investigate the 
emission characteristics radiating by a two-dimensional dipole 
on the nanowire HMMs, where the unit dipole moment of 
the dipole is \( \hat{P}_x = \hat{p}_x \hat{x} + \hat{p}_z \hat{z} \) with \( \hat{p}_x^2 + \hat{p}_z^2 = 1 \). We select 
the related parameters corresponding to the experiment as: 
\( \epsilon_1 = \epsilon_4 = 1 \), \( \epsilon_2 = 2.5 \), \( d = 10 \) nm, and \( l = 800 \) nm. In order 
to make the coupling mechanism clear, we first consider the case 
of \( k_y = 0 \). In this case, the s-polarized waves have no contribu-
tion to the electric field intensity from the equation (8) in SI.

Then the electric field intensity \( I \) of the p-polarized waves in 
the Fourier transformed space can be described as [51]

\[
P = \frac{|p|^2(k_x^2 + |k_{1z}|^2)}{k_0^2} \cdot |p_x + p_zk_{1z}/k_{1z}|^2,
\]

where \( k_{1z} = \sqrt{k_0^2 - k_x^2} \) and \( k_0 = \omega/c \). As discussed in SI, 
the reflection \( |r|^2 \) is symmetric along the \( k_x = 0 \). Therefore, 
the equation (2) can be seen as \( I \propto |p_x + p_zk_{1z}/k_{1z}|^2 \) 
for evanescent waves with \( |k_x| > k_0 \), the term \( k_{1z} \) is a com-
plex number and then \( I \propto |p_x - ip_zk_{1z}/|k_{1z}|^2 |^2 \). If we let \( p_x \) 
and \( p_z \) be real numbers without a \( \pi/2 \) phase difference used 
for the unpolarized case, the electric field intensity can be 
written as \( I \propto |p_x|^2 + |p_zk_{1z}/k_{1z}|^2 \), which is a linear super-
position of the contributions from the horizontal and vertical 
oriented components of the dipole. No interference pheno-
menon occurs in this case. However, the case is different 
when \( p_x \) and \( p_z \) have a \( \pi/2 \) phase difference. For instance, 
for the right-circularly polarized dipoles, we have \( p_x = \frac{1}{\sqrt{2}} \) 
and \( p_z = \frac{1}{\sqrt{2}} \). Consequently, the electric field intensity can be 
reduced to \( I \propto |1 + k_{1z}/k_{1z}|^2 / 2 \). The horizontal and vertical 
oriented spectral amplitudes for \( k_x > k_0 \) add constructively, 
whereas for \( k_x < k_0 \) destructive interference occurs. There-
fore, we emphasize that chiral emission occurs when a circu-
larly polarized dipole excites the multilayered nanostructure.

More generally, elliptic polarization with a phase difference 
of \( \pi/2 \) between the \( x \) and \( z \) components is required. These 
very general findings on the expected chiral behaviour are 
in agreement with the experimental results in figure (3). We 
also discussed the case without absorption in silver, which is given 
in SI (SI, figure S7).

In the following, we give a quantitative analysis about the 
electric field intensity \( I \) for different dipolar polarization 
states. The unit vector of the horizontally (vertically) polar- 
ized dipole is \( p_x = \hat{x} \) (\( p_z = \hat{z} \)). For the circularly polarized 
dipoles, the unit polarization vectors are \( p_x = \frac{1}{\sqrt{2}}(\hat{x} \pm \hat{z}) \).

The dielectric constant tensor for the HMMs can be 
described by the effective medium theory, as given in SI, by 
\( \epsilon_1 = 3.79 + 0.0088i \) and \( \epsilon_2 = -0.843 + 0.0672i \) for 
the wavelength \( \lambda = 660 \) nm. In figure 4(a), we present the 
electric field intensity map induced by the unpolarized dipole with 
\( p_x = 1/\sqrt{2} \) and \( p_z = 1/\sqrt{2} \), which can match well with that 
in figures 2(b) and (d). Figure 4(b) shows the normalized 
emission intensity profile along the \( k_x \) direction with \( k_x = 0 \).

It has two symmetric peaks with approximately equal intensity 
at \( k_x/k_0 = \pm 1.085 \), which denotes that there is no chiral 
behaviour but only directional characteristics.

The intensity maps containing information for both TE 
and TM modes for different dipolar polarizations are shown 
in figures 5(a–b) and (d–e), which give a circular ring of 
maximum intensity followed by a minima and show direc-
tional emission. The figure 5 demonstrates clearly the spin 
momentum locking for evanescent waves. This locking refers
to the locking of the propagation direction to the spin or the polarization of the photons and has been the subject of extensive discussion in literature [4, 6]. Moreover, chiral emission for the cases of circularly polarized dipoles with a forward-propagating mode for \( k_x > k_0 \), and backward propagation is suppressed for \( k_x < -k_0 \). However, the case for a left-circularly polarized dipole \( (P_-) \) is converse. For the right-circularly polarized dipole, the position of peak appears at \( k_x = 1.08k_0 \) and the peak for \( k_x < -k_0 \) appears as a relatively broad shoulder. Even though, emission intensity line profiles still show chiral behaviour for circularly polarized dipoles with \( C = 0.682 \) at \( k_x = 1.08k_0 \). This theoretical value of \( C \) is higher than the experimental value. This is because the theoretical result is calculated from the peak value, whereas the experimental data is averaged over a range of \( k_x \) vectors. This clearly will bring down the observed value of \( C \). Instead, for linearly polarized dipoles, the excited field is bidirectional propagation, as shown in figure 5(f). The forward-propagating and backward-propagating modes are symmetrical modal distribution. Electric field intensity of the high-\( k \) propagating mode induced by a \( z \)-polarized dipole is larger than that of an \( x \)-polarized dipole. We can find that even though there is no preference in the \( k_x \) direction, the directional emission can be realized by using the circularly polarized dipoles. In the experimental data, the measured wave vectors are in free space whereas in theoretical modeling \( k_x \) and \( k_y \) are in glass objective which has refractive index 1.45. A closer look at figure 3(a) shows for example that the emission max-min corresponds to \( \kappa \) about 0.7 which translates to \( \kappa \) in glass as 1.015 which matches well with the maximum in the figure 5(c) obtained in the electromagnetic calculations.

The polarization degree of freedom provides a new approach for studying a variety of novel optical effects. Finally, we show how the dielectric constant tensor \( \epsilon \) and the intensity ratio between the left-circularly \( (P_-) \) and right-circularly \( (P_+) \) polarized components is defined as

\[
D_1 = \frac{P^x(P_-)}{I^x(P_+)} = \left( \gamma - \sqrt{\gamma^2 - 1} \right)^4, \quad (3)
\]

where \( \gamma = k_x/k_0 \). The chiral parameter \( C \) can be given by \( C = (1 - D_1)/(1 + D_1) \). We can also define the intensity ratio induced by the horizontally and vertically polarized dipoles as

\[
D_2 = \frac{I^y(P_+)}{I^y(P_+)} = 1 - 1/\gamma^2. \quad (4)
\]

In figure 6, we present the intensity ratios \( (D_1 \) and \( D_2) \) and chiral parameter \( C \) as a function of the wave vector component \( k_x \) for \( k_x > k_0 \) and \( k_x = 0 \). When increasing the value of \( k_x \), the intensity ratio \( D_1 \) goes down, while the chiral parameter \( C \) and intensity ratio \( D_2 \) grow up. Therefore, we can obtain larger

![Figure 5](image_url)

**Figure 5.** Electric field intensity maps when considering the absorption in silver with different polarization, (a) \( P_x = \hat{P}_x = \hat{P}_z = \hat{x} \), and (c) \( P_+ = \hat{z} \). Intensity profile induced by circularly polarized (c) and linearly polarized (f) dipoles as a function of \( k_x \) are plotted for \( k_0 = 0 \). The electric field \( E_x \) used for normalization corresponds to the larger peak value induced by a circularly polarized dipole in (c). In figure 5(c) the respective scales for solid and dashed curves are shown on top and bottom.
components of the dipolar QDs in evanescent high-interference between the longitudinal and transverse polarized explained. The origin of chirality is attributed to the near-field

tonic spin-momentum locking in experiments, quantitatively,

HMMs are presented theoretically and the origin of pho-

ral emission from a circularly polarized dipole coupled with

Based on the method of photon Green’s function, the chi-

momentum locking in the form of directional and chiral emis-

we observe in our experiments.

In conclusion, we have demonstrated photonic spin-momentum locking in the form of directional and chiral emission from excitons in compact monolayers of QDs efficiently
coupled to evanescent metamaterial modes in a 2D HMM. Based on the method of photon Green’s function, the chiral emission from a circularly polarized dipole coupled with HMMs are presented theoretically and the origin of pho-

tonic spin-momentum locking in experiments, quantitatively, explained. The origin of chirality is attributed to the near-field interference between the longitudinal and transverse polarized components of the dipolar QDs in evanescent high-k modes of the HMM. Equivalently, the directional emission can be observed by exciting an elliptically polarized dipole to break the inversion symmetry, similar to the spin Hall effect. Our study thus provides a generalized method to obtain chiral and directional photonic effects in the far field without having to use chiral emitters or chiral metamaterials and without directional far field excitation.

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