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OPTICAL DISPERSION RELATIONS FROM THREE-DIMENSIONAL CHIRAL GOLD NANOCUBES IN PERIODIC ARRAYS

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Recommended Citation

Boora, Manpreet, "OPTICAL DISPERSION RELATIONS FROM THREE-DIMENSIONAL CHIRAL GOLD NANOCUBES IN PERIODIC ARRAYS", Open Access Master's Thesis, Michigan Technological University, 2020.

<https://doi.org/10.37099/mtu.dc.etdr/1081>

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OPTICAL DISPERSION RELATIONS FROM THREE-DIMENSIONAL CHIRAL
GOLD NANOCUBES IN PERIODIC ARRAYS

By

Manpreet Boora

A THESIS

Submitted in partial fulfillment of the requirements for the degree of

MASTER OF SCIENCE

In Applied Physics

MICHIGAN TECHNOLOGICAL UNIVERSITY

2020

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This thesis has been approved in partial fulfillment of the requirements for the Degree of MASTER OF SCIENCE in Applied Physics.

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Author Contribution Statement

This dissertation is submitted in partial fulfillment for the Degree of Master of Science in Applied Physics. Analysis of angle-resolved optical transmission measurements on chiral gold nanostructures has been done to study the dependence of surface lattice resonance on the chirality of the structures. To analyze their energy-momentum dispersions, wavelength-incident angle relations are obtained. The angle-resolved transmission spectra are acquired by a custom-built setup, which consists of motor controlled rotational stage, polarizer, lens, white-light source, collimator and spectrometer. The samples are periodic arrays of chiral gold nanocubes, fabricated and provided by Prof. Ki Tae Nam at Seoul National University (South Korea). The periodic PDMS (polydimethylsiloxane) templates were fabricated and provided by Prof. Min Hyung Lee at Kyung Hee University (South Korea).

Acknowledgement

Foremost I would like to express my gratitude to my advisor Dr. Jae Yong Suh for his continuous guidance, support and motivation during my research. I would also like to sincerely thank Dr. Chito Kendrick and Dr. Miguel Levy for their valuable time and insightful advices. A much of my sincere thanks goes to my research group members and friends. Furthermore, I would like to thank my family: my parents and my brother for supporting me emotionally throughout writing this thesis and my life in general. They were a constant source of my strength and motivation during this time. Lastly, I am thankful to Neerav for being the best companion, best teacher and the best friend I could have.

List of abbreviations

1. SP: Surface Plasmon
2. SPP: Surface Plasmon Polariton
3. LSP: Localized Surface Plasmon
4. LSPR: Localized Surface Plasmon Resonance
5. SEM: Scanning Electron Microscope
6. SLR: Surface Lattice Resonance
7. LCP: Left Circularly Polarized (light)
8. RCP: Right Circularly Polarized (light)
9. CD: Circular Dichroism
10. OA: Optical Activity
11. ORD: Optical Rotatory Dispersion
12. RA: Rayleigh Anomaly

Abstract

Chiral metamaterials are artificially tailored materials that are composed of nanoparticles that lack in-plane mirror symmetry. Application of chirality to plasmonic nanostructures can lead to chiro-optical effects such as asymmetric transmission of circularly polarized light, circular dichroism and optical activity. When light is incident on metallic nanoparticles, conduction electrons undergo collective oscillations within the nanoparticles, resulting in localized surface plasmon (LSP) resonance. Surface lattice resonances (SLRs) are hybrid plasmonic-photonic modes arising from the radiative coupling between LSPs and electromagnetic waves diffracted in the plane of the periodic array. Because of the collective dipole interactions, SLRs have less radiative losses compared to LSP resonances. In this thesis, SLRs in chiral metamaterials are studied and discussed. Three-dimensional chiral gold plasmonic nanocubes were fabricated and transferred to periodic arrays of hexagonal and square lattices. The components of chirality tensor are obtained from the asymmetric transmission dispersions of SLR modes. The strength of the asymmetry in dispersion relation is scaled as the optical activity that originates from the broken space inversion symmetries of the nanocubes in the arrays.

Chapter 1

1.1 Introduction

Plasmonics is a bridge between nanophotonics and electromagnetics. Metallic nanostructures supporting surface plasmons can concentrate light in nanoscale regime. As the length scale for plasmonics lies in between photonics, where device feature sizes are usually above 1000 nm, and electronics, where fabricated circuit sizes are nearly 10 nm [1], nano structures supporting surface plasmons offer a solution to the size compatibility limitation. Usually noble metals like gold and silver are used in plasmonics because of their high bulk plasma frequency $\omega_p = \frac{n_e e^2}{m_{eff} \epsilon_0}$ where n_e , e , m_{eff} and ϵ_0 are the electron density, electron charge, effective electron mass and the permittivity of the free space respectively, which makes them support surface plasmons in visible or near infrared range [5].

Also, the small ohmic losses of noble metals make them good plasmonic materials. Applications of plasmonics can be seen in many fields especially in optical imaging, nanophotonics, bio-chemical sensors, and photovoltaics [2].

Surface plasmons (SPs) are electromagnetic waves that propagate along the surface of conductors, usually noble metals [3]. The optical response of metal to external electromagnetic field can be understood by Drude's formula:

$$\epsilon_m(\omega) = 1 - \frac{\omega_p^2}{\omega^2 + i\omega\gamma} \quad (1.1.1)$$

where ϵ_m , ω_p , ω and γ are the frequency-dependent dielectric function of metal, plasma frequency, frequency of incident field and inverse electron momentum relaxation time (i.e. damping factor) respectively [5]. When photon couples with an SP, the resulting hybridized excitation is called a Surface Plasmon Polariton (SPP). SPP is a propagating mode along the interface of a metal, and it loses energy via scattering or absorption. Surface plasmons also exist within metallic nanoparticles, called Localized Surface Plasmons (LSPs). Surface plasmons helps in exploiting the unique properties of light-matter interactions below diffraction limit, which is discussed in detail in section 1.2. At optical frequencies, the oscillations of conduction electrons in metal can be seen as a harmonic oscillator with the eigen resonance frequency as the plasma frequency of the conduction electrons.

In Section 1.3, another type of surface plasmons is discussed. When light is incident on nanoparticles, conduction electrons can oscillate coherently without significant phase retardation. Since the surface plasmons are strongly confined to nanoparticles at the resonant frequency, this mode is called localized surface plasmon resonances (LSPRs). LSPRs have few limitations. These exhibit broad linewidth (i.e. low quality factors) because of large radiative damping. Thus, the potential applications of LSPs in optical and electronic system is limited. A way to mitigate the losses in LSPs is also discussed in this section. The metallic nanoparticles arranged in an array can support collective resonances, called surface lattice resonance (SLRs). Because of the limitations of LSPRs, research interest grew more in SLRs which are delocalized modes resulting from the coupling of LSPs with diffractive states. SLRs become stronger when nanoparticles are arranged in such a way that the distance between them is an integer multiple of the wavelength of incident light. SLRs are of interest because their q factors are significantly higher than that of LSPRs. Applications of SLRs can be extended to metamaterials, enhancement of weak magneto-optical Kerr effects in magnetic nanoparticles, surface enhanced Raman scatterings and refractive index sensing [10].

Chiral metamaterials are discussed in section 1.4. Chiral materials are composed of nanoparticles that lack mirror symmetry in their structures, and thus can have chiro-optical responses. The chirality in material can manifest as different optical response to left and right circularly polarized light or as the rotation of the incident plane of linearly polarized light. Chiro-optical responses like optical activity (OA) and circular dichroism (CD) can be enhanced with the optical response of chiral nanostructures. For instance, Optical Rotatory Dispersion (ORD) and CD can be tuned by the geometry of the nanostructures [11]. Chiral nanoparticles larger in size can support strong dipole moments and higher-order modes which leads to strong chiral optical responses [14]. SLRs excited in periodic arrays of three-dimensional chiral gold nanocubes are to be measured and analyzed. The effect of broken space inversion symmetry of these chiral nanostructures on the SLRs is revealed in their dispersion relations. SLRs are shown to have dependence on periodicity, shape and size of nanoparticles, chirality, and dielectric environment surrounding these nanoparticles. From dispersion relations of surface lattice modes, the asymmetry appeared in their positive and negative in-plane momenta regions is measured to extract the components of the chirality tensor. The delocalized nature of the SLR where the radiative fields of individual particles extend over several unit cells of the structure beyond the nearest-neighbor coupling regime may give its potential uses in topological plasmonics [8].

1.2 Dispersion Relations

SPPs are collective excitation of surface electrons at dielectric-metal interface. The electric field in the direction perpendicular to the interface decays exponentially with distance from the surface. The field in the perpendicular direction is said to be evanescent or near field, and is a consequence of the bound, non-radiative nature of SPs [2].

To understand the nature of SPPs, energy-momentum or frequency- wavevector dispersion relations are studied. For a free space light , dispersion relation is linear, and simply given by $\omega = k_0 c$ where ω is the frequency, $k_0 = 2\pi/\lambda$ is the wavevector and c is the speed of light in free space. The slope of a dispersion curve gives a group velocity. To understand the propagation properties of SPPs, we begin with a derivation of the dispersion relation.

Consider an electromagnetic wave propagating along x direction as shown in Figure 1.

$$E(r, t) = E(r)e^{-i\omega t} \quad (1.2.1)$$

Figure 1 : Surface plasmon propagating along x direction. Evanescent field is decaying in z -direction perpendicular to metal-dielectric interface. Here ϵ_1 and ϵ_2 are relative permittivities of metal and dielectric respectively [6].

At $z = 0$ interface, the electric field is given in the following.

$$E(x, y, z) = E(z)e^{i\beta x} \quad (1.2.2)$$

Here k_o is the wave vector of the propagating wave in vacuum and $\beta (= k_x)$ is the propagation constant. Substituting this plane wave solution in the Helmholtz equation

$$\nabla^2 E + k_o^2 E = 0 \quad (1.2.3)$$

we get the following form

$$\frac{d^2 E(z)}{dz^2} + (k_o^2 \varepsilon - \beta^2) E = 0 \quad (1.2.4)$$

As there is no translation symmetry in y -direction, relative permittivity $\varepsilon = \varepsilon(z)$. Using the Maxwell curl equations, we can get six field components for both E and H fields.

$$\begin{aligned} \frac{\partial E_x}{\partial z} - \frac{\partial E_z}{\partial x} &= i\omega\mu_o H_y, & \frac{\partial E_y}{\partial x} - \frac{\partial E_x}{\partial y} &= i\omega\mu_o H_z, & \frac{\partial E_z}{\partial y} - \frac{\partial E_y}{\partial z} &= i\omega\mu_o H_x \\ \frac{\partial H_x}{\partial z} - \frac{\partial H_z}{\partial x} &= -i\omega\varepsilon_o \varepsilon E_y, & \frac{\partial H_y}{\partial x} - \frac{\partial H_x}{\partial y} &= -i\omega\varepsilon_o \varepsilon E_z, & \frac{\partial H_z}{\partial y} - \frac{\partial H_y}{\partial z} &= -i\omega\varepsilon_o \varepsilon E_x \end{aligned}$$

Using the propagation of the field along x direction $\left(\frac{\partial}{\partial x} = i\beta\right)$, spatial translation symmetry along y direction $\left(\frac{\partial}{\partial y} = 0\right)$ and harmonic time-dependence $\left(\frac{\partial}{\partial t} = i\omega\right)$ in the above equations, we get

$$\frac{\partial E_x}{\partial z} - i\beta z = i\omega\mu_o H_y, \quad i\beta E_y = i\omega\mu_o H_z, \quad \frac{\partial E_y}{\partial z} = -i\omega\mu_o H_x$$

$$\frac{\partial H_x}{\partial z} - i\beta H_z = -i\omega\varepsilon_0\varepsilon E_y, \quad i\beta H_y = -i\omega\varepsilon_0\varepsilon E_z, \quad \frac{\partial H_y}{\partial z} = -i\omega\varepsilon_0\varepsilon E_x$$

There exist two sets of self-consistent solutions with the different polarization properties of the propagating waves [5]. Only the TM (transverse magnetic) solution with nonzero field components (E_x, E_z, H_y) can excite SPPs, and only those are used hereafter. In a TM mode, E_x and E_z can be expressed in terms of H_y

$$E_x = \frac{-i}{\omega\varepsilon_0\varepsilon} \frac{\partial H_y}{\partial z}, \quad E_z = -\frac{\beta}{\omega\varepsilon_0\varepsilon} H_y \quad (1.2.5)$$

After putting TM modes in Helmholtz wave equation (1.2.3), we get

$$\frac{\partial^2 H_y}{\partial z^2} + (k_o^2\varepsilon - \beta^2)H_y = 0 \quad (1.2.6)$$

Solving the above equation, we get the solutions for both halves

For $z < 0$:

$$H_y(z) = A_1 e^{i\beta x} e^{k_1 z},$$

$$E_x(z) = -iA_1 \frac{1}{\omega\varepsilon_0\varepsilon_1} k_1 e^{i\beta x} e^{k_1 z}, \quad E_y(z) = -iA_1 \frac{\beta}{\omega\varepsilon_0\varepsilon_1} k_2 e^{i\beta x} e^{k_1 z}$$

For $z > 0$:

$$H_y(z) = A_2 e^{-i\beta x} e^{-k_2 z},$$

$$E_x(z) = iA_2 \frac{1}{\omega \varepsilon_0 \varepsilon_2} k_2 e^{-i\beta x} e^{-k_2 z}, E_y(z) = -iA_1 \frac{\beta}{\omega \varepsilon_0 \varepsilon_2} k_2 e^{-i\beta x} e^{-k_2 z}$$

The continuity of the field components ($H_y, \varepsilon_1 E_z, \varepsilon_2 E_z$) at the interface requires the condition that $A_1 = A_2$ and $\frac{k_2}{k_1} = -\frac{\varepsilon_2}{\varepsilon_1}$. For SPP to exist at the interface, we need the opposite signs of dielectric functions for metal and surrounding dielectric medium. As we know for metals, $Re(\varepsilon_1) < 0$, ε_2 is positive. After putting H_y in (1.2.6), we get

$$k_1^2 = \beta^2 - k_0^2 \varepsilon_1, \quad k_2^2 = \beta^2 - k_0^2 \varepsilon_2 \quad (1.2.7)$$

Combining all the equations above, we finally get the dispersion relation as follows

$$\beta = k_0 \sqrt{\frac{\varepsilon_1 \varepsilon_2}{\varepsilon_1 + \varepsilon_2}} \quad (1.2.8)$$

From Equation (1.2.8), we can see that the propagation constant for SPPs depends on the permittivities of the metal and the surrounding dielectric medium. The permittivity of metal is a complex function of frequency. Using eqn. (1.1.1) with $\gamma = 0$ and $\varepsilon_m(\omega) = -1$, the maximum frequency of surface plasmon at large values of β can be written as

$$\omega_{SP} = \frac{\omega_p}{\sqrt{2}} \quad (1.2.9)$$

where ω_{SP} is the frequency of surface plasmon [5]. It can also be seen from dispersion relation that SPPs only exist for TM polarization. At higher frequency, wavevector of SPPs lies inside the light cone and thus the metal behaves as a dielectric. Exponential

decaying fields transform to propagating fields and its physical interpretation would be the Brewster effect. When SPP dispersion curve lies to the right of the light line of the dielectric, it is not possible to excite SPPs with free space light because the in-plane component momentum of SPPs is larger than the momentum of light. The mismatch must be compensated if light is to be used to generate SPs. The light is coupled with SPPs at a certain angle of incidence θ when the in-plane component of wavevector of free space light ($k_o \sin \theta$) equals the propagation constant β of SPPs. There are a few techniques to excite SPPs using prism coupling such as in Otto configuration or Kretschmann configuration. Periodic gratings or periodic array of nanoparticles on metallic surface can compensate the momentum mismatch between light and SPs. The gratings satisfy momentum conservation by providing an additional in-plane momentum to the photon such that the additional momentum is equal to the integer number of the reciprocal lattice vectors $G = \frac{2\pi}{p}$ where p is the period of grating. When propagating on periodic structures, SPPs behave like Bloch waves and thus follow the Bloch theorem which states that the amplitude of a wave depends on the periodicity of the medium. These Bloch waves, which are similar to surface electromagnetic waves, will have wavefunction (φ) in the following general form

$$\varphi = e^{ik \cdot r} \cdot v(r) \quad (1.2.10)$$

where k is the wavevector and $v(r) = v(r + p)$ is the periodic amplitude modulation. The SPPs satisfying the Bloch waves condition can be excited, and thus they propagate on such a periodic surface [7]. Because of the translation symmetry, SPP-Bloch wave modes can be generated under the Bloch coupling condition

$$k_{spp} = k_o \sin \theta \pm m G_x \pm n G_y$$

$$|k_{spp}| = |k_o| \sqrt{\frac{\epsilon_m \epsilon_d}{\epsilon_m + \epsilon_d}} = |k_{\parallel} + G_{mn}| \quad (1.2.11)$$

where k is the wavevector of light incident at a polar angle θ , $k_{\parallel}$ is in-plane parallel component of wavevector of light, $G_m = G_n = \frac{2\pi}{p}$ is the magnitude of the reciprocal

vectors of the periodic arrays and m and n are integers associated with the mode of the SPP wave vector k_{spp} . Moreover, the propagation of SPPs on periodic structures modifies dispersion relation. Because of the Bragg's scattering from periodic boundaries, two standing waves with different energies can make photonic bandgap appear. This bandgap appears at the brillouin zone boundaries in the reciprocal space because of the coupling of forward and backward propagating modes, and therefore the dispersion curves are folded into the first brillouin zone boundaries [5].

1.3 Surface Lattice Resonances (SLRs)

The second type of surface plasmons are non-propagating and so-called localized surface plasmons (LSPs). The oscillation of the surface charge within metallic nanoparticles is constrained by size, geometry and dielectric environment of nanoparticles [5]. The optical response of the LSP in a metallic spherical particle of radius a can be described by the particle polarizability α that relates the incoming electric field E_0 to the electric dipole moment P as follows

$$P = \alpha E_0 \quad (1.3.1)$$

where

$$\alpha = \frac{\epsilon_{metal} - \epsilon_{medium}}{\epsilon_{metal} + 2\epsilon_{medium}} 4\pi\epsilon_0 a^3 \quad (1.3.2)$$

In the above equation, ϵ_{metal} and ϵ_{medium} are the dielectric functions of metal nanoparticle and the medium surrounding the nanoparticle, respectively. The LSPR occurs when the denominator in (1.3.2) vanishes ($\epsilon_{metal} + 2\epsilon_{medium} = 0$). When this condition is met, LSP characterized by the polarizability is enhanced relative to the incident field. LSPs can be used to enhance and control light-matter interactions nearby light emitters and absorbers on subwavelength scale [4]. The localized surface plasmons in neighboring particles may mutually couple to each other, and, in general, their resonances are severely broadened due to the strong radiative damping. Hence, LSPRs exhibit low quality factors (Q) [5][17].

Periodic arrays of metal nanoparticles can support collective resonance modes. SLRs occur from constructive interference of scattered field arising from the nanoparticle

arrays [9]. When the distance between the neighboring particles is an integer multiple of the incident wavelength

$$p = \frac{m\lambda}{n} \quad (1.3.3)$$

where p is the periodicity of the lattice, m is the order of diffraction, n is the refractive index of the medium and λ is the resonance wavelength of a single nanoparticle [13]. The LSPs can couple strongly with one another in array. SLR arises from the diffractive coupling of LSPRs of individual particles, following the diffraction orders of a periodic structure. Hence, this coupling is mediated by Rayleigh anomaly (RA), which corresponds to the condition whereby a diffracted wave propagates in the plane of the array. At a Rayleigh wavelength, a diffracted wave propagates tangentially to the surface of the gratings. Classically, “SLRs can be described as driven damped coupled oscillators in which one oscillator has the natural frequency of the LSPR while the other has the frequency of the diffraction order. The spectrum of SLRs can be calculated from the dissipated power of this driven coupled oscillator system, in which one oscillator represents the LSPRs with a natural frequency independent of the wave vector and the other is dispersive” [20]. Like SPPs, RAs are also surface electromagnetic waves and follow Bragg’s scattering condition in periodic structures [4]

$$k_{RA} = \frac{\omega}{c} \sqrt{\epsilon_d} = k_0 \sin \theta (\cos \varphi + \sin \varphi) \pm mG_x \pm nG_y \quad (1.3.4)$$

which is related to ϵ_d and the grating pitch but not the grating material (i.e. no dependence on ϵ_m) [21][23]. Here $G_x = G_y = \frac{2\pi}{p}$ is the magnitude of the reciprocal vectors, p is the periodicity of array, and θ and φ are respectively the polarization and azimuthal angles of the incident light.

At normal incidence ($k_0 = 0$) in (1.3.4), the expression for frequency of SLRs can be written as

$$\omega_{SLR} = \frac{c}{\sqrt{\epsilon_d}} \sqrt{\frac{m^2 + n^2}{p^2}} \quad (1.3.5)$$

SLRs follow the dispersion of the RAs with a small shift emerging from the coupling with RAs [20]. SLRs are collective modes which arise from the constructive interference of the scattered lights from each nanoparticle in the plane of the lattice [22]. SLRs are dispersive modes as the spectral position is dependent on the incident angle. Furthermore, SLRs can have very narrow lineshapes because of the radiative coupling between LSPs. Thus, the q -factors can be an order of magnitude higher than their localized counterparts. Coherence length of SLRs is also higher due to their delocalized nature (tens to over 100 μm) as compared to that of SPPs (a few tens of microns) [4]. SLRs can also be easily tuned in visible and near infrared range of frequencies depending on the periodicity, shape of nanoparticle, lattice type, and polarization of the light [4][18].

1.4 Chiral Metamaterials

A metamaterial is an artificial composite structure engineered to have properties not present in nature. An object is called chiral if there is no translational or rotational transformation that allows its mirror image to be superimposed onto the original one. A chiral metamaterial is designed to have strong chiro-optic responses as compared to naturally occurring chiral materials. Because of the recent development of chiral metamaterials with exceptional light-manipulating capabilities such as polarization control, negative refractive index and chiral sensing, there is a growing interest in the field. Optical activity and circular dichroism are two major characteristic (but different) manifestations of chiral materials [16].

Chiral materials have broken space inversion symmetry which leads to the property of optical activity (OA). In OA, linearly polarized light is gradually rotated as it passes through the medium. Broken time-reversal symmetry in all materials under magnetic fields leads to the magneto-optical (MO) effect such as the Faraday effect [12].

Linearly polarized light is regarded as a coherent superposition of left and right-handed circularly polarized light. In circularly polarized light, the tip of the electric field sweeps out a helix in real space. OA is caused by electromagnetic induction in the chiral structures. As a result, the OA is reciprocal when the rotation direction of light polarization is dependent on the light propagation direction [12].

As mentioned previously, frequency-dependent optical responses of a medium can be studied by dispersion relations. A medium with no broken symmetries has degenerate and symmetric dispersion relation as

$$\omega = \omega_{L,R}(-\vec{k}) \quad (1.4.1)$$

where L and R are left and right circularly polarized light, respectively. A diagram of dispersion relation for materials with space inversion and time reversal symmetry is shown in Figure 2. It can be seen that the material has a degenerate and symmetric dispersion relation. Also, asymmetric dispersion relation for materials with broken space inversion symmetry is shown in Figure 3.

Figure 2 : Degenerate and Symmetric dispersion relation for materials with space inversion and time reversal symmetry [12].

Figure 3 : Asymmetric dispersion relations in materials with broken space inversion symmetry. Here, L and R represents LCP and RCP waves [12].

Medium preserving space inversion symmetry would have the following dispersion relations

$$\omega_L(\vec{k}) = \omega_L(-\vec{k}) \quad (1.4.2.A)$$

$$\omega_R(\vec{k}) = \omega_R(-\vec{k}) \quad (1.4.2.B)$$

Hence, a system with broken space inversion symmetry has inequality between the positive and negative momentum regions in the dispersion relations above. In Figure 3, the optical response of medium is different for LCP and RCP waves. Chirality is a kind of structural asymmetry [12]. To understand optical activity, we parameterize the chirality that give rise to asymmetric dispersion relations.

Chiral media are a subset of bi-isotropic media where the electric and magnetic fields of light waves are coupled to each other [16]. Thus, the electromagnetic or optical responses of bi-isotropic media can be described by the following constitutive relations [15],

$$D(r, t) = \varepsilon_o \hat{\varepsilon} E(r, t) - i \frac{\hat{\xi}}{c} H(r, t) \quad (1.4.3.A)$$

$$B(r, t) = \mu_o \hat{\mu} H(r, t) + i \frac{\hat{\xi}}{c} E(r, t) \quad (1.4.3.B)$$

where $\hat{\epsilon}, \hat{\mu}, \epsilon_0, \mu_0$ are the relative permittivity of the medium, relative permeability of the medium, permittivity of vacuum and the permeability of vacuum, respectively. The chirality tensor $\hat{\xi}$ represents an effect of structural chirality that breaks the space-inversion symmetry. It also represents coupling strength between electric and magnetic fields because if $\hat{\xi}$ is zero, the aforementioned constitutive relations will have normal form as in case of normal isotropic materials [19]. For a cubic geometry and linear isotropic chiral medium, supposing that the chiral axis is parallel to the z -direction, we can consider only one diagonal element in the chirality tensor. The chirality tensor is thus written as

$$\hat{\xi} = \begin{pmatrix} \xi & 0 & 0 \\ 0 & \xi & 0 \\ 0 & 0 & \xi \end{pmatrix} \quad (1.4.4)$$

Using the plane wave solution for $E, H \sim e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)}$, Maxwell's curl equations are given by

$$\nabla \times E(r, t) = -\frac{\partial B(r, t)}{\partial t} \quad (1.4.5.A)$$

$$\nabla \times H(r, t) = -\frac{\partial D(r, t)}{\partial t} \quad (1.4.5.B)$$

where E, B, H, D are the electric field, magnetic flux density, magnetic field and the electric flux density respectively. After substituting the plane wave solutions, we get

$$k \times E(r) = \omega B(r) \quad (1.4.6.A)$$

$$k \times H(r) = -\omega D(r) \quad (1.4.6.B)$$

Using (1.4.3.A) and (1.4.3.B), we can write E and B in terms of H in a chiral material as follows

$$E = \frac{1}{\epsilon_0 \epsilon} \left[\frac{-k \times H}{\omega} + \frac{i \hat{\xi}}{c} H \right] \quad (1.4.7.A)$$

$$B = \hat{\mu} \mu H + \left[\frac{-1}{\omega} k \times H \right] \frac{i}{c \epsilon_0 \epsilon} \hat{\xi} \quad (1.4.7.B)$$

The wave equation can thus be written as

$$-k \times (k \times H) + i \frac{\omega}{c} k \times (\hat{\xi} H) + i \frac{\omega}{c} \hat{\xi} (k \times H) = \frac{\omega^2}{c^2} \hat{\xi} \mu \epsilon H - \frac{\omega^2}{c^2} \hat{\xi}^2 H \quad (1.4.8)$$

The dispersion relation can be obtained by separating the propagation direction in the transverse and longitudinal modes $k = (k_t, k_z)$. For linearly isotropic, reciprocal chiral media, simplifications can be done as follows

$$\frac{k_t^2}{(n_t^\pm)^2} + \frac{k_z^2}{(n_z^\pm)^2} = \frac{\omega^2}{c^2} \quad (1.4.9)$$

$$n_t^\pm = \sqrt{\epsilon \mu} \pm \frac{\xi}{2} \quad (1.4.10)$$

$$n_z^\pm = \sqrt{\epsilon \mu} \quad (1.4.11)$$

where $n_t^\pm, n_z^\pm$ are refractive indices and $\pm$ represent polarization states (i.e. Right Circularly Polarized (RCP) light and Left Circularly Polarized (LCP) light). From above dispersion relations (1.4.10), it can be seen easily that LCP and RCP waves have different propagation constants and refractive indices. Consequently, the unequal

refractive indices confirm that the difference in transmitted amplitude and phase between RCP and LCP waves results in the circular dichroism and optical activity.

1.5 Research Objectives

Chiral plasmonic lattices composed of three-dimensional gold nanostructures combine characteristics chirality with plasmonic systems having strong light matter interactions enhancing local field to tailor chiral optical responses. The presence of SLR modes of these chiral gold structures in the visible range has made them interesting for numerous reasons.

Gold nanocubes or nanodisks in arrays exhibit SLRs. Chiral nanocubes have cubic geometry with chirality present on each six face. The objective of transmission angle resolved measurements is to observe an optical effect of chirality present in individual nanocubes on their SLR dispersions. The chirality can be manifested as asymmetry in their dispersion relations, bolstering the fact that the optical response is different for incident LCP and RCP waves. The other physical phenomena related to chirality such as CD and ORD have been also confirmed in the chiral nanocubes. In this work, both circular dichroism and optical activity from these chiral nanostructures are discussed. Angle-resolved transmission spectra are taken and translated into the dispersion relations, which display the asymmetry of the SLR modes caused by the chirality of the structures. The knowledge of SLRs in chiral plasmonics may be used for making plasmon lattice lasing or topological nanophotonic devices where the delocalized nature and longer lifetimes of SLRs are utilized for creating Dirac materials that enable topologically protected edge states in analogy to the physics of topological insulators.

Chapter 2

2.1 Experimental Setup

A home-built setup was used to obtain angle-resolved transmission spectra of optically transparent samples. Collimated white light from a 20W halogen lamp is impinged on the sample surface. White light passes through an optical fiber into a series of collimating lenses, an aperture, and a linear polarizer. The measurement schematic is shown below in Figure 4.

Figure 4 : Optical measurement setup for taking angle-resolved transmission spectra. Incident angle θ is changed via a pair of motor-controlled rotational stages. Azimuthal angle ϕ is changed in clockwise direction.

Samples consisting of chiral nanostructures are illuminated by linearly polarized lights, and transmitted light is collected by a combination of collimating lens and optical fiber. The collimator in the setup minimizes the beam divergence. The transmitted lights are sent to a spectrometer (Ocean Optics, USB 2000), which records the intensity spectra in visible range.

Samples are rotated around the vertical axis where the incident light passes through. Azimuthal angle is defined by the angle between the lattice orientation of the samples and the polarization direction of the incident light (Figure 5). The angular resolution of all the measurements was set to be 0.4° .

Dispersion relations for any diffractive modes can be obtained by recording the transmission spectra (intensity versus wavelength) as a function of incident angle θ . Energy-momentum dispersion relation can be retrieved by wavelength-angle relation by the following conversion formula [6],

$$E = \frac{hc}{\lambda}, \quad \hbar\beta = \hbar k_0 \sin \theta \quad (2.1.1)$$

where $E, \hbar, \lambda, \beta$ are the energy, Planck's constant, the wavelength of the incident light and the propagation constant of the SLR mode, respectively.

Figure 5 : Real image of homebuilt optical setup consisting of computer controlled Rotational Stage which can be used for both reflection and transmission angle resolved spectra.

2.2 Sample description with SEM images

The sample consists of three-dimensional asymmetric chiral gold nanocubes. The gold nanocubes are synthesized using amino acid and peptide agents, which control the asymmetric growth of nanocubes. For synthesis of 432 helicoid III nanoparticles, colloidal seed growth method was used and templates of periodic nanocubes were fabricated by Dr. Nam's research group at Seoul National University. Further details of chemical synthesis are mentioned in [14]. These helicoid nanoparticles belong to special symmetry group 432 and have pinwheel-like structures, consisting of four highly curved arms of increasing width—on each of the six faces of the cubic geometry as shown in Figure 6. The chiral elements in 432 helicoid III are twisted with a larger curvature and the gaps between them are carved more deeply in the central direction which can be seen clearly in Figure 6b. Imaging of these samples after performing Ion Milling using helium-ion microscopy shows the curved surfaces located inside the gaps (Figure 7). The morphology of nanostructures is discussed in detail in [14].

The gold nanoparticles display strong chiral plasmonic optical activity (a dissymmetry of 0.2). The Kuhn's dis-symmetry factor (g -factor) is a dimensionless quantity that is useful for quantitative comparisons of chiro-optical properties among different systems [14]. Figure 8 shows that the chiral structures show strong CD response and ORD for individual nanocubes. CD means that the chiral medium responds differently to the absorbance of LCP and RCP light. CD can also be represented by a difference in the imaginary part of the refractive indices for LCP and RCP light. The magnitude of circular dichroism (CD) is given by [14],

$$CD \sim A_{LCP} - A_{RCP} \frac{\log(10)}{4} \text{ rad} \quad (2.2.1)$$

Figure 6 (A) SEM image of 432 helicoid III chiral nanoparticles in hexagonal array of pitch 400 nm. The scalebar is 3.00 μm . (B) The original pinwheel-like structure of 432 helicoid III is highlighted in yellow. 432 helicoid III chiral nanoparticles are synthesized using an octahedral seed. Figure adapted from [14].

Figure 7 : Helium-ion microscopy secondary electron images of 432 helicoid III during the He^+ ion milling process exposure to a He^+ ion beam allow visualization of the interior parts of the curved surfaces, as indicated by red arrows. Figure adapted from [14].

Figure 8 : Circular dichroism and optical rotatory dispersion spectra of 432 helicoid III chiral nanoparticles. Figure adapted from [14].

2.3 Results and Discussions

Dispersion measurements from the arrays of chiral nanocubes display the SLR modes. The refractive indices for left and right circularly polarized light are measured to be different, which confirms the circular dichroism in Figure 9-12. For chiral isotropic medium, the modified refractive indices and the chirality-dependence are given in the Equation (1.4.10)

$$n^{\pm} = \sqrt{\epsilon\mu} \pm \frac{\xi}{2} \quad (2.3.1)$$

where n_- and n_+ are refractive indices for left-handed and right-handed circularly polarized light and ξ is the chirality tensor component. A linearly polarized light can be described as the superposition of an LCP and an RCP light with identical amplitude; however, the equation above shows that light of opposite handedness (LCP and RCP) will obtain different accumulations of phases as they travel through a chiral material. The first term on the right-hand side corresponds to conventional symmetric dispersion relation without any spatial symmetry breakings. The second term has a directional dependence, and thus represents the structural optical activity as shown in Figure 8. Thus, the electric field direction of linearly polarized light can be rotated by a very short angle while passing through a chiral medium [12].

This difference in the refractive indices for two circular polarizations is called circular birefringence and leads to the rotation of linear polarization and circular dichroism. If chirality tensor is zero, there is no circular birefringence. Using the modified refractive indices for RCP and LCP waves, the chirality parameter can be expressed as

$$\xi = (n^+ - n^-) \quad (2.3.2)$$

A chiral medium is an optically active medium, and the optical rotation of linear polarization direction passing through chiral medium can be expressed by the angle of rotation ϕ as follows

$$\phi = \frac{(n_{RCP} - n_{LCP})\pi d}{\lambda} = \frac{\xi\pi d}{\lambda} \quad (2.3.3)$$

where d and λ are the width of the medium and the wavelength of the light. The angle of rotation ϕ is a dimensionless quantity.

Angle-dependent transmitted spectra are recorded with an exposure time of 100 ms while the sample is rotated from -50° to 50° in steps of 0.4° . By using (1.3.4), SLRs are calculated to appear near the diffractive orders of a nanocube array in the momentum space. The SLR modes for negative and positive range of wavevectors are plotted in Figure 9a for $\varphi = 25^\circ$ and in Figure 9b for $\varphi = 24^\circ$, respectively. Inspection of Figure 9c and 9d shows that as the incident angle of polarization θ increases, the degenerate SLR modes split into a combination of first order modes $(-1,0)$, $(0, -1)$, $(1,0)$ and $(-1,0)$. The dispersion curves for the first order modes are close to each other, and thus the energy difference between the two modes is very small. The energy difference between the dispersion curves of the SLR modes is smaller for positive range of wavevectors k_+ as compared to negative range of wavevectors k_- . Hence, the lattice plasmon modes respond selectively to the handedness of polarized light. The broad dispersionless resonance centered at 680 nm is the LSP mode.

The SLR modes for negative and positive wavevectors are plotted in Figures 10a for $\varphi = 54^\circ$ and 10b for $\varphi = 55^\circ$, respectively. At $k = 0$ (i.e. incident angle 0°), the degenerate SLR mode is a bright mode, as the intensity of transmission is measured to be maximum. The energy difference between dispersion curves for SLR modes in Figure 9 is smaller as compared to the energy difference between modes in Figure 10, demonstrating the dependence on azimuthal angle (Eq. 1.3.4). Figure 10c and 10d show that the energy of the SLR modes decreases and the gap between two dispersion curves increase as the incident angle increases.

Figure 9 : Transmission spectra for pitch 400 nm sample for negative and positive incident angles in the range from -50° to 50° . Azimuthal angle $\varphi = 25^\circ$ for RCP and $\varphi = 24^\circ$ for LCP. The solid blue lines are showing an overlay of SLR modes and the color bar represents the transmission intensity. The dispersion for negative and positive wavevectors are shown in (A) and (B) respectively. The corresponding SLR modes are plotted in (C) and (D).

Figure 10 : Transmission spectra for pitch 400 nm sample for negative and positive angles with azimuthal angle $\varphi = 54^\circ$ and $\varphi = 55^\circ$ for RCP and LCP. Note that the increase in the azimuthal angle, the gap between the SLR modes becomes broader. The solid lines are SLR modes and the color bar represents the transmission intensity. The dispersion for negative and positive wavevectors are shown in (A) and (B) respectively. The corresponding SLR modes are plotted in (C) and (D).

From Figures 9c and 9d and Figures 10c and 10d, the refractive indices for LCP and RCP are measured to be different as the following

$$n_{LCP} = 1.29$$

$$n_{RCP} = 1.32$$

To retrieve the chirality tensor, Equation (2.3.1) is used

$$\xi = 1.32 - 1.29 = 0.03 \quad (2.3.4)$$

The chirality tensor, which signifies the coupling strength of electric and magnetic field in chiral medium, is a dimensionless constant [19]. The value of the chirality tensor $\xi = 0.03$ is measured to be independent of the polarization state of incidence or the azimuthal angle. Because of the cubic geometry of the chiral helicoid, all the diagonal elements of chirality tensor are to be ξ .

The angle of rotation ϕ can also be calculated from the chirality parameter ξ by using Equation (2.3.3)

$$\phi = \frac{(n_{RCP} - n_{LCP})\pi d}{\lambda} = \frac{\xi\pi d}{\lambda} = 1.34^\circ \quad (2.3.5)$$

where d is 50 nm and $\lambda = 400$ nm. It can be seen that the chirality effectively changes (rotates) the angle ϕ (the equivalent effect of changing azimuthal angle). As shown in Figure 9 and Figure 10, the difference between azimuthal angles for LCP and RCP light is $\phi = 1^\circ$. This difference results in asymmetric behavior of dispersion curves of SLRs.

In Figures 11, the periodicity of the sample is 700 nm and the s-polarized light is incident onto it. The SLR modes are mapped in Figures 11a and 11b using the dispersion relation (1.3.4). The SLR modes for negative and positive wavevectors are plotted respectively in Figures 11a and 11b with $\varphi = 15^\circ$ and $\varphi = 15^\circ$. At normal incidence ($k = 0$) or $\sin\theta = 0^\circ$, there exists first and second order SLR modes. The SLR mode

at $k = 0$ is a bright mode, and therefore its intensity is high. The degenerate mode exists around wavelength 700 nm which can be found using (1.3.3).

Figure 11 : Transmission spectra for pitch 700 nm sample for negative and positive incident angles with azimuthal angle $\phi = 25^\circ$ of the incident light respectively. The solid lines are SLR modes and the color bar represents the transmission intensity. The dispersion for negative and positive wavevectors are shown in (A) and (B) respectively. The corresponding SLR modes are plotted in (C) and (D).

In Figures 11c and 11d, the refractive indices for LCP and RCP are found to be different.

$$n_{LCP} = 1.17$$

$$n_{RCP} = 1.14$$

$$n_+ - n_- = 0.003$$

To retrieve chirality tensor, Equation (2.3.1) is used

$$\xi = 1.17 - 1.14 = 0.03$$

Thus, from Figures 11, the chirality tensor is obtained as $\xi = 0.03$. Chirality is an intrinsic geometry property, and thus the chirality parameter is the same for pitch 400 nm and pitch 700 nm samples since the same 432 helicoid nanocubes are present in both cases.

Again, the angle of rotation θ can be calculated by using (2.3.3)

$$\phi = \frac{(n_{RCP} - n_{LCP})\pi d}{\lambda} = \frac{\xi\pi d}{\lambda} = 0.77^\circ$$

In Figure 11, the difference between azimuthal angles for LCP and RCP light is $\phi = 1^\circ$ and the asymmetry in the SLR dispersion curves can also be observed.

2.4 Conclusions

The spectral shifts of SLR modes are clearly observable as the refractive index of the material changes for RCP and LCP due to the chirality. The SLR modes redshift for RCP and blueshift for LCP because of the broken space inversion symmetry. Since SLR modes are coupled modes between LSPs and diffractive states, the SLR modes depend on the geometric parameters of the array and individual nanoparticles. SLRs depend on the interparticle distance and on the long-range order in the lattice [3]. When nanoparticles are arranged in a lattice with a spacing comparable to the optical wavelength, the resonances of the individual particles can couple strongly with those of neighboring particles via Rayleigh anomalies (RAs) [3]. For pitch 400 nm, only the first order RA modes are seen whereas the higher order modes are present for pitch 700 nm.

For pitch 400 nm and pitch 700 nm samples, we have observed the effect of chirality on the SLR modes appeared in their dispersion maps. The parameter chirality is calculated to be the difference in modified refractive indices obtained from fitted dispersion curves. We find in our experiments that the strength of chirality controls the degree of asymmetry for the given handedness of illuminating light. With retrieval of chirality parameter from measurements, angle of rotation ϕ can also be estimated. It is found that the effect of changing azimuthal angle in dispersion curves is equivalent to the effect of changing the angle of rotation for the same polarization state of incident light.

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Appendix A

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