Chapter 6

Conclusion

This chapter summarizes the overall conclusion of the research work as a part of this thesis.

This chapter also discusses the scope and future work to be carried out.

6.1 Conclusion

Light scattering is a fundamental phenomenon that comes into existence when an electromagnetic (EM) wave interacts with material, resulting in elastic scattering due to spatial fluctuations (such as variations in density, refractive index) and inelastic scattering due to temporal fluctuations (such as molecular motion or acoustic waves), causing light redirection and energy distribution. Research on the scattering of light by matter, at the nano scale, is not only foundational but also pivotal for advancing both scientific understanding and technological innovation. Structures like nanoparticles have ability to precisely manipulate the light scattering due to their unique plasmonic characteristics. By controlling the interaction between light and nanoparticle, we can tailor the scattering behaviour to achieve desired outcomes, such as improved resolution in imaging systems, and even more efficient energy transfer in photovoltaic devices. Nanoparticles can be designed to manipulate light's direction, intensity, and wavelength, unlocking a range of applications from advanced medical diagnostics to quantum computing. On the other hand, fine-tuning incident light properties such as polarization, wavelength, or angle of transmission or reflection during scattering, offers a paralleled control over light-matter interaction. This ability to structure light not only deepens our understanding of fundamental optical processes but also enables the development of innovative technologies in biosensors, spectroscopies, and imaging, etc.

This thesis emphasizes on the investigations on the scattering of a structured laser beam by a single nanoparticle. We have considered Mie scattering resulting from spatial fluctuations in the nanostructures. More specifically, tightly focused beams having high convergence angle and high numerical aperture (NA) are considered, and their effects on the plasmonic response are inspected.

We examined the intensity enhancement in the near-field of a nanoparticle influenced by the beam profile of an incident EM wave, and the numerical simulations are performed on the MATLAB platform. Mie scattering theory based on the multipole expansion approach is utilized to examine the scattering of a light beam through a nanoparticle. Plasmon assisted monometallic (uncoated) nanoparticles such as gold (Au), silver (Ag), and core-shell (coated) nanoparticles gold coated silver (Ag@Au), silver coated gold (Au@Ag) nanoparticles are considered for the study. We separated radial and tangential components of near-field intensity for more significant observations. We analyzed the effects on the plasmonic response of nanoparticles in different surrounding dielectric media air, BK7 (glass), and silicon for an incident beam of wavelength range from 300 nm to 900 nm.

A detailed examination is done on the scattering of a linearly polarized plane wave by a nanoparticle in Chapter 2. Results obtained on COMSOL are presented, and the preferred use of the MATLAB platform is highlighted. An algorithm based on the Mie scattering theory is developed to investigate the near-field intensity enhancement on the surface of a nanoparticle. Detailed investigations are carried out for varying radii of monometallic nanoparticles from 1 nm to 100 nm, and results are presented and analyzed. Near-field intensity enhancement for a core-shell nanoparticle in different media is examined. Tightly focused beams and their numerical representation using Richard and Wolf's formalism are presented in Chapter 3. We studied the interaction of a tightly focused linearly polarized beam with a monometallic and core-shell nanoparticle supporting plasmon resonance. Effects of a linear polarization in high NA focusing on intensity enhancement in the near-field are analyzed by developing a Mie scattering algorithm. Average intensity enhancement is observed by varying outer shell thickness of a core-shell nanoparticle, and results are presented.

In Chapter 4 we studied the scattering of a tightly focused radially polarized beam with a nanoparticle. Radial polarization impacts focal spot and it has significant role on the plasmonic response of nanoparticle. We developed a Mie scattering algorithm for investigating the interaction of tightly focused radially polarized beam with monometallic and core-shell nanoparticles. Intensity enhancement is observed for nanoparticles in the near-field for a range of incident beam wavelength ranging from 300 nm to 900 nm, in different dielectric media. Influence of a tightly focused radially polarized beam on plasmonic response of Au@Ag core-shell nanoparticle is examined by increasing outer shell thickness. The outcomes on near-field intensity enhancement for three different illuminations namely plane wave, tightly focused linearly polarized, and tightly focused radially polarized beams are categorized and compared. A detailed comparison is done for monometallic Au, and Ag nanoparticles and core-shell Ag@Au, and Au@Ag nanoparticles for different incident beams and dielectric media. Results showing more intensity enhancement for an incident tightly focused radially polarized beam are also discussed.

In Chapter 5, the presence of optical aberration during the interaction of a tightly focused beam with a nanoparticle is discussed, and its impact on the Mie scattering by a nanoparticle is discussed. We investigated the scattering of a tightly focused linearly polarized beam by a monometallic nanoparticle under spherical aberration. We developed the Mie scattering algorithm by utilizing Mie theory, Richard and Wolf's formalism, and Zernike polynomials to analyze the effects of spherical aberration on the plasmonic response of a nanoparticle. The involvement of modes is also discussed for different values of the coefficient of spherical aberration. A balancing condition is introduced by using defocusing to compensate for the effects of spherical aberration on the near-field intensity enhancement. The effects of low and high NA focusing are also analyzed by varying the convergence angle of the incident beam.

6.2 Future plan

The scattering of structured light by a nanoparticle represents a cutting-edge approach to control the light-matter interaction at the nanoscale. Structured light includes complex beam profiles with tailored phase and polarization, which are the additional degrees of freedom that can be exploited during scattering. When such light interacts with a nanoparticle, the scattering process becomes highly customizable, allowing for precise manipulation of light's direction, intensity, and angular momentum. This level of control has far-reaching implications for applications in the domains like super-resolution microscopy, optical trapping, and quantum information processing, etc. We have investigated the scattering of a tightly focused beam based on its polarization state. By exploring how nanoparticles scatter structured light for engineered polarization, phase, modes, and coherence, we can find more applications in enhancing the sensitivity of sensors, spectroscopy sensors, and imaging systems, etc. Some important topics which needs a special attention are

1. The scattering of tightly focused beams for other non-uniform polarization needs to be further explored and we are currently working on the examination of an azimuthally polarized beam interaction with a nanoparticle.
2. Illuminating nanostructures by cylindrical vector beams (CVBs) instead of planar beams, transverse unidirectional scattering can be used in completely new ways, offering useful solutions for measurements of nanoscale displacement. The unique amplitude and phase distribution of CVBs fields makes them commonly used in light-matter interaction. Some CVBs are Hermite–Gaussian (HG) beams, Laguerre–Gaussian (LG) beams, and Orbital angular momentum (OAM) beams, etc. We can continue our study for such incident structured beams interacting with nanoparticles.
3. Coherence of an EM wave plays an important role in understanding the properties of light and light-matter interaction. By controlling optical coherence, we can tailor the spatial, spectral & polarization distributions of propagating statistical beams. The

coherence of incident beam illuminating grating, nanostructures also affects their plasmonic response. For a fully coherent, a partially coherent and an incoherent beam different outcomes are observed when an interaction with plasmonic structures are considered, and a significant study can be performed.

4. In this thesis, scattering by a single nanoparticle is considered. We can extend our investigations for many nanoparticles, and multiple scattering.
5. Effects of aberration during interaction of a light with nanoparticles needs to be exploited more. We are also working on aberration caused due to refractive index mismatch while a tightly focused EM wave is propagating through multi-layer or stratified media.

1) Mie Scattering Algorithm determining average intensity enhancement for gold (Au) nanoparticle of radius $r = 50\ nm$, in air illuminated by a tightly focused linearly polarized beam, with convergence angle $\alpha = 60°$.

```
clc
clear all
close all

a=50;                    %radius of the particle(nm)
n_m=1.00;                %optical index of medium(glass)
N=15;                      %order of multipole
alpha=pi/3;

h1=[];
g1=[];

for n=1
   syms t;
   at=cos(t)^(1/2);
   Pn=legendreP(n,cos(t));
   dPnx=diff(Pn,t);
   Pnx=(-sin(t)).*dPnx;
   PnX=Pnx./sin(t);
   dPnX=diff(Pnx,t);
   P=PnX+dPnX;
   PL=at.*P.*sin(t);
   Palpha=int(PL,t,0, alpha);
   A=(-1i)^n.*((2*n)+1)./(2*n^2.*(n+1)^2).*Palpha;
   A1=abs(A)^2;
   A1=double(A1);
end

for Inter_lambda=300:1:900

    lambda=[301 311 320 331 342 354 368 381 397 413 430 451 471 496 521 548 582 616
              659 704 755 821 891];    %wavelength of incident light (nm)
   R=[1.53 1.53 1.54 1.48 1.48 1.50 1.48 1.46 1.47 1.46 1.45 1.38 1.31 1.04 0.62
       0.43 0.29 0.21 0.14 0.13 0.14 0.16 0.17];        %Real part of refractive index
                                                              of gold nanoparticle
    I=[1.889 1.893 1.898 1.883 1.871 1.866 1.895 1.933 1.952 1.958 1.948 1.914
        1.849 1.833 2.081 2.455 2.863 3.272 3.697 4.103 4.542 5.083 5.663];
                                   %Imaginary part of refractive index of gold nanoparticle

   Aur=interp1(lambda,R,Inter_lambda);
   Aui=interp1(lambda,I,Inter_lambda);
```

```
    Kralpha=0;
    Ktalpha=0;

      n_Au=Aur+(1i*Aui);          %optical index(complex) of particle(gold/silver)
      k=2*pi*n_m/Inter_lambda;    %wavenumber
     m=n_Au/n_m;                 %m=relative refractive index
     x=k*a;                      %x=size parameter
     z=m*x;

for n=1:N
   sx=sqrt(pi*x/2);
   sz=sqrt(pi*z/2);

   Phinx=sx.*besselj(n+0.5,x);
   Phin1x=sx.*besselj(n-0.5,x);
   Phinz=sz.*besselj(n+0.5,z);
   Phin1z=sz.*besselj(n-0.5,z);

   Xinx=sx.*((besselj(n+0.5,x))+1i*(bessely(n+0.5,x)));
   Xin1x=sx.*((besselj(n-0.5,x))+1i*(bessely(n-0.5,x)));

   Phindx=Phin1x-(n.*Phinx./x);
   Phindz=Phin1z-(n.*Phinz./z);
   Xindx=Xin1x-(n.*(Xinx)./x);

an=((m*Phinz.*Phindx)-(Phinx.*Phindz))./((m*Phinz.*Xindx)-(Xinx.*Phindz));
bn=((Phinz.*Phindx)-(m*Phinx.*Phindz))./((Phinz.*Xindx)-(m*Xinx.*Phindz));

W=((abs(an))^2.*(abs(Xinx))^2)+(abs(Phinx))^2-(2*real(an.*Xinx).*Phinx);
X=((abs(an)^2).*(abs(Xindx)^2))+((abs(bn)^2).*(abs(Xinx)^2))+(abs(Phinx)^2)+(abs(Phi
       ndx)^2)-(2*real(an.*Xindx).*Phindx)-(2*real(bn.*Xinx).*Phinx);
Y=(2*(n^3)*(n+1)^3)./((2*n)+1);
Z=(2*(n^2)*(n+1)^2)./((2*n)+1);


    syms t;
    at=cos(t)^(1/2);
    Pn=legendreP(n,cos(t));
    dPnx=diff(Pn,t);
    Pnx=(-sin(t)).*dPnx;
    PnX=Pnx./sin(t);
    dPnX=diff(Pnx,t);
    P=PnX+dPnX;
    PL=at.*P.*sin(t);
    Palpha=int(PL,t,[0 alpha]);
    B=(-1i)^n.*((2*n)+1)./(2*n^2.*(n+1)^2).*Palpha;
    An=abs(B)^2;
    An=double(An);

    An60=An./A1;
```

```
    Kra=An60.*Y.*W;
    Kralpha=Kra+Kralpha;

    Kta=An60.*Z.*X;
    Ktalpha=Kta+Ktalpha;

end

        Kr60=9.*Kralpha./(16.*x^4);
        Kt60=9.*Ktalpha./(16.*x^2);

        h1=[h1,Kr60];
        g1=[g1,Kt60];
end

Inter_lambda=300:1:900;

figure(1);plot(Inter_lambda,h1,'r',)
hold on
axis square
figure(2);plot(Inter_lambda,g1,'r')
hold on
axis square
```

2) Mie Scattering Algorithm determining average intensity enhancement for gold (Au) nanoparticle of radius $r = 50\ nm$, in BK7 (glass) illuminated by a spherically aberrated tightly focused linearly polarized beam, with convergence angle $\alpha = 60°$.

```
clc
close all
clear all


N=7;                    %order of multipole
AAn=[];

%ha=[];                 %aberration free
%ga=[];
% h=[];                  %aberattion with defocusing
% g=[];

hAs=[];               %aberattion without defocusing
gAs=[];

alpha=pi/3;   %%%%%% 60 degree

for Inter_lambda=300:1:900
    lambda=[301 311 320 331 342 354 368 381 397 413 430 451 471 496 521 548 582 616
               659 704 755 821 891];    %wavelength of incident light (nm)
   R=[1.53 1.53 1.54 1.48 1.48 1.50 1.48 1.46 1.47 1.46 1.45 1.38 1.31 1.04 0.62
        0.43 0.29 0.21 0.14 0.13 0.14 0.16 0.17];        %Real part of refractive index
                                                                         of gold nanoparticle
    I=[1.889 1.893 1.898 1.883 1.871 1.866 1.895 1.933 1.952 1.958 1.948 1.914
        1.849 1.833 2.081 2.455 2.863 3.272 3.697 4.103 4.542 5.083 5.663];
                                          %Imaginary part of refractive index of gold nanoparticle

   Aur=interp1(lambda,R,Inter_lambda);
   Aui=interp1(lambda,I,Inter_lambda);

   Bg1=1.0396;
   Bg2=2.3179*(10^(-1));
   Bg3=1.0104;
   Cg1=6.0069*(10^3);
   Cg2=2.0017*(10^4);
   Cg3=1.0356*(10^8);
   bg1=(Bg1*(Inter_lambda^2))/((Inter_lambda^2)-Cg1);
   bg2=(Bg2*(Inter_lambda^2))/((Inter_lambda^2)-Cg2);
   bg3=(Bg3*(Inter_lambda^2))/((Inter_lambda^2)-Cg3);
   BK7=sqrt(1+bg1+bg2+bg3);                  %%% Glass
```

```
% Input Parameters
   n_m=BK7;                  %optical index of medium(glass/air)
   a=50;                     %radius of the particle(nm)
   n_Au=Aur+(1i*Aui);         %optical index(complex) of particle(gold/silver)
   k=2*pi*n_m/Inter_lambda;         %wavenumber
   m=n_Au/n_m;               %m=relative refractive index
   x=k*a;                    %x=size parameter
   z=m*x;

   As=0.5*Inter_lambda;        %Spherical Aberration
%    Ad=-As;                   %Defocussing
   Ad=0.0;

    Kralpha=0;
    Ktalpha=0;

for n=1:N
   sx=sqrt(pi*x/2);
   sz=sqrt(pi*z/2);

   Phinx=sx.*besselj(n+0.5,x);
   Phin1x=sx.*besselj(n-0.5,x);
   Phinz=sz.*besselj(n+0.5,z);
   Phin1z=sz.*besselj(n-0.5,z);

   Xinx=sx.*((besselj(n+0.5,x))+1i*(bessely(n+0.5,x)));
   Xin1x=sx.*((besselj(n-0.5,x))+1i*(bessely(n-0.5,x)));

   Phindx=Phin1x-(n.*Phinx./x);
   Phindz=Phin1z-(n.*Phinz./z);
   Xindx=Xin1x-(n.*(Xinx)./x);

an=((m*Phinz.*Phindx)-(Phinx.*Phindz))./((m*Phinz.*Xindx)-(Xinx.*Phindz));
bn=((Phinz.*Phindx)-(m*Phinx.*Phindz))./((Phinz.*Xindx)-(m*Xinx.*Phindz));

W=((abs(an))^2.*(abs(Xinx))^2)+(abs(Phinx))^2-(2*real(an.*Xinx).*Phinx);
X=((abs(an)^2).*(abs(Xindx)^2))+((abs(bn)^2).*(abs(Xinx)^2))+(abs(Phinx)^2)+(abs(Phindx
)^2)-(2*real(an.*Xindx).*Phindx)-(2*real(bn.*Xinx).*Phinx);
Y=(2*(n^3)*(n+1)^3)./((2*n)+1);
Z=(2*(n^2)*(n+1)^2)./((2*n)+1);

    syms t;
    at=cos(t).^(1/2);
    rho=sin(t)./sin(alpha);
%     W_rho=As.*(rho.^4);
    W_rho=As*(rho.^4)+Ad*(rho.^2);
    A3=exp(1i.*2.*pi.*W_rho./Inter_lambda);
    Pn=legendreP(n,cos(t));
    dPnx=diff(Pn,t);
    Pnx=(-sin(t)).*dPnx;
```

```
    PnX=Pnx./sin(t);
    dPnX=diff(Pnx,t);
    P=PnX+dPnX;
    PL=A3.*at.*P.*sin(t);
    Palpha=int(PL,t,[0 alpha]);
    B=(-1i).^n.*((2*n)+1)./(2*n.^2.*(n+1).^2).*Palpha;
    An=abs(B).^2;
    An=double(An);
    AAn=[AAn,An];
    A1=AAn(1);
    An60=An./A1;

    Kra=An60.*Y.*W;
    Kralpha=Kra+Kralpha;
    Kta=An60.*Z.*X;
    Ktalpha=Kta+Ktalpha;
end

    Kr60=9.*Kralpha./(16.*x.^4);
    Kt60=9.*Ktalpha./(16.*x.^2);

ha=[ha,Kr60];
ga=[ga,Kt60];
end

Inter_lambda=300:1:900;

figure(1);plot(Inter_lambda,ha)
hold on
axis square

figure(2);plot(Inter_lambda,ga)
hold on
axis square
```