Bio inspired synthesis of silver nanoparticles and its applications to spin – orbit interactions of light

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Abstract

Spin–orbit interaction of light serves as an important property of light, which deals with the study of polarization and phase modulations in the light beam. These studies are essential and principal characteristics of light beam that have been used for most of the nanophotonics applications. Silver nanoparticles (Ag NPs) prepared via biosynthesis are used for one of such nanophotonics application in scattering via studying the light scattered through these nanoparticles. The silver nanoparticles Ag NPs were synthesized using green method, where reduction of silver ions to silver nanoparticles happen during the reaction of aqueous solution of Ag NO₃ with the biomolecules present in fresh leaf extract of Coleus amboinicus plant. The nanoparticles were characterized using UV-visible (UV–vis) spectroscopy, Transmission electron microscopy (TEM) and Fourier Transform Infrared (FTIR) spectroscopy. TEM analysis shows the wide size distribution of spherical shape nanoparticles with 80 nm average size. The study of polarization and phase changes in the scattered light field has been carried out using Stokes polarimetry in forward direction scattering. Under the preliminary measurements of Polarimetry, the modification in the polarization components was studied by demonstrating changes in the Stokes S₂, S₃ parameters, polarization orientation (ψ) and ellipticity angle (χ) using transverse magnetic (TM) polarized Gaussian light beam.

1. Introduction

Nanotechnology is a progressive research domain as it has large scope for modifications based on desired technological application which makes it ever thrusting area. Silver nanoparticles (Ag NPs) have received paramount attention due to their unique properties and utilized in various applications such as photonics [1, 2], catalysis [3, 4], Surface Enhanced Scattering [5, 6]. On the other hand, its natural antimicrobial properties lead to bio-medical applications [7–9]. As performance of nanoparticles depend on their size, shape and composition hence main focus is on synthesis methods. There are plenty of reports available on various chemical and physical methods of synthesis of Ag NPs and other metal nanoparticles [10–13]. As these methods of synthesis involve usage of hazardous chemicals which in turn threat to health and Environment and also tedious, time-consuming preparation methods. This leads the way to explore and investigate the simple, rapid, eco-friendly and cost-effective synthesis methods as an alternative. In all these synthesis methods, most successful is green synthesis which uses biological materials such as bacteria [14, 15], fungi [16, 17] and plant parts [18–23] as the reducing agent, among them widely reported is using plant parts. In the present study for the synthesis of Ag NPs, we have used the fresh leaf extract of the plant Coleus amboinicus, which is known as Indian Borage (family of lamiaceae). It is a medicinal plant containing phytochemicals such as carvacrol (monoterpenoid), caryophyllene (bicyclic sesquiterpene) and patchoulane and flavonoids. It is well established that phytochemicals and flavonoids present in the leaf extract are responsible for the reduction and stabilisation of the nanoparticles to a great extent.

In context to the application of these bio-inspired Ag NPs, large part of the reports is focused on their bio-medical applications [24–26]. In our earlier papers we demonstrated the non-linear optical properties of these bio-inspired Ag NPs [27, 28]. There is large scope for nanophotonics application where light interacts with the...
The leaves of Coleus amboinicus (Family, Lamiaceae) were collected from the home premises. Silver nitrate (AgNO₃) was purchased from Sigma Aldrich (India).

2.2. Preparation of leaf extract
The leaf extract was prepared with 20 g of fresh and healthy Coleus amboinicus leaves which were thoroughly rinsed with DDW (double distilled water) and chopped into small pieces. The chopped leaves were boiled in 100 ml of DDW for 5 min and filtered. The cooled extract was stored at 4 °C for further use for the synthesis purpose. The filtrate is used as reducing and stabilising agent for synthesis of Ag NPs.

2.3. Synthesis of Ag nanoparticles
The leaf extract of 5.0 ml was added to 50 ml of 10⁻³ M AgNO₃ aqueous solution and kept at room temperature. The experiment was done in triplicate to confirm the reproducibility. Reduction of silver ions present in the aqueous solution of 1 mM Ag NO₃ during the reaction with the biomolecules present in the Coleus amboinicus leaf extract is monitored through the UV–vis spectrum. As we mix the Coleus leaf extract to the aqueous silver ion, complex solution start changing from colourless to yellow colour indicating the formation of Ag NPs. The bio-reduced Ag NPs solution was collected and measured by using UV-visible spectra.
3. Structural and morphological properties of Ag NPs

The Spectroscopic techniques like UV-Visible and Fourier Transform Infrared (FTIR) were carried out to understand the successful generation of Ag NPs and to find out the presence of functional groups of biomolecules present in the leaf extract of Coleus amboinicus which are responsible for the reducing and stabilising the formation of Ag NPs respectively. The microscopic technique like Transmission Electron Microscopy (TEM) were carried out to study the morphological properties like size and shape of the prepared Ag NPs.

3.1. UV-visible spectroscopy

UV-visible spectroscopy analysis was carried out using a UV-Visible absorption spectrophotometer (JASCO V-670) with a resolution of 1 nm between 300 and 800 nm possessing a scanning speed of 300 nm min \(^{-1}\). The distilled water was used as a blank to adjust base line. Equivalent amounts of the Ag NPs solution (0.4 ml) were diluted in a constant volume of distilled water (4 ml) and subsequently analysed at room temperature. The progress of the reaction between metal ions and the leaf extracts were monitored by UV–Visible spectra of Ag NPs. The spectra were recorded at different time intervals of 0 min, 30 min and 60 min.

The progress of the reaction between metal ions and the Coleus amboinicus leaf extracts were monitored by UV-visible spectra of Ag nanoparticles in aqueous solution with different reaction times are shown in figure 1. It shows the absorption spectra of Ag NPs prepared from the leaf extract of Coleus amboinicus plant at different intervals of time. It was observed from the figure that at time \(t = 0\), there is no Surface Plasmon Resonance (SPR) peak, this indicates the absence of Ag NPs. With the increase in the reaction time, it is observed that the SPR peak at \(t = 30\) min is centred at 427 nm is red shifted to 435 nm at \(t = 60\) min. It is also observed that the SPR peak at 427 nm \((t = 30\) min) is slightly narrow indicates the formation of stable spherical shape nanoparticles, whereas the peak at 435 nm \((t = 60\) nm) has broad in nature indicating the varied size and shape of nanoparticles. These results imply that the reduction of silver ions to formation of stable Ag NPs within an hour, making this method one of the fastest bio-inspired methods [46].

3.2. FTIR spectroscopy

The FTIR spectrum was recorded using FTIR Spectrometer (Bruker Alpha II) at wave length ranging from 500–4000 cm \(^{-1}\). The spectrum was recorded with solution of Ag NPs and distilled water was used for the base line correction. The spectrum is recorded for both the fresh leaf extract (red plot) and the Ag NPs reduced from the leaf extract (black plot) in the range of 500–4000 cm \(^{-1}\), are shown in figure 2.

From the figure (red plot), three strong peaks and two weak peaks are noticed in the spectrum obtained from the leaf extract. The wavenumbers of these peaks are observed at 693, 1645, (strong peaks) and 3500 (broad peak) cm \(^{-1}\) correspond to C–Cl–, N–H bending (corresponds to amide I), −C=−C= aromatic and H bonded OH stretches, respectively. The weak peaks are observed at 2068 and 2534 cm \(^{-1}\) due to −C triple bond C stretching and S–H stretching thiol group. Figure 2 (black plot) shows the spectrum obtained from Ag NPs reduced from the leaf extract. It is observed from the spectrum that three strong peaks observed from the plot. The wave
numbers are 761, 1650 and broad 3580 cm\(^{-1}\) due to C= C bending, N—H bending, amine group and OH stretching alcohol group, respectively. These observations confirmed that the functional groups of phytochemicals which are present in the leaf extract are responsible for the reducing and stabilising of Ag NPs. The phenolic compounds which contains Hydroxyl and Carboxyl groups which possesses the ability to bind metals [46].

3.3. Transmission electron microscopy (TEM)
TEM technique was used to visualize the size and shape of Ag NPs. The 200 kV Ultra High-Resolution Transmission Electron Microscope (JEOL-2010) has been used. TEM grids were prepared by placing a drop of the particle solution on a carbon-coated copper grid and drying under lamp.

Figure 3(a) shows the typical bright-field TEM micrograph of the synthesized Ag NPs and it is observed that most of the them are spherical in shape. It is also observed that a few agglomerated Ag NPs in some places indicates possible sedimentation at a later time. Figure 3(b) shows the histogram taken from several such micrographs at different places in the grid. It is evident that the particle size varies from broad range from 10 nm to 250 nm and the average size estimated is around 80 nm.

The high-resolution lattice imaging was done with UHR pole piece (URP 22) of JEOL 2010 TEM operating 200 keV. This URP pole piece, with a spherical aberration coefficient of 0.5 mm, gives a lattice resolution of 0.005 nm easily. We have magnified (raw image) the region shown in yellow box in figure 3(c) and also shown enlarged figure 3(c). It is clearly seen that the particle single crystalline with an inter-planar spacing of 0.236 ± 0.006 nm. The error bar is obtained by fitting the lattice fringes to Gaussian peaks (several of them) and obtaining their peak position along with the mean value. The error bar is estimated with Average full width at half maximum of fitted Gaussian peak value and divided by the number of fringes taken care of.

Figure 3(d) shows the Selection Area Electron Diffraction (SAED) pattern of electrons associated with a polycrystalline nature, indicating that the Ag NPs synthesized from the leaf extract were randomly oriented. The diffraction rings, from inside out, can be indexed with planes (111), (200), (220), (311), (222), (331), (420) and (422) respectively corresponding to the Ag crystalline FCC (JCPDS file No 65 2871).

3.4. The measurements of Ag NPs concentration
The concentration of Ag NPs was determined using the method which has been previously reported in [47]. To determine the average number of atoms per nanoparticle

\[
N = \frac{\pi \rho D^3}{6M} N_A,
\]

where \(N\) is the number of atoms per nanoparticles, \(\rho\) is the density of face cantered cubic (fcc) silver (= 10.5 g cm\(^{-3}\)), \(D\) is the average diameter of nanoparticles (= 80 nm = 80 × 10\(^{-7}\) cm), \(M\) is the atomic mass of silver (= 107.868 g), \(N_A\) is the number of atoms per mole (Avogadro’s number) (= 6.023 × 10\(^{23}\)). Molar concentration, \(C\) of nanoparticles is determined by

\[
C = \frac{N_T}{NVN_A},
\]

where \(C\) is the molar concentration of nanoparticle solution, \(N_T\) is the total number of silver atoms added as AgNO\(_3\) = 1 M, ‘\(N\)’ is the number of atoms per nanoparticle (from equation (1) \(N\) is equal to 1.63 × 10\(^7\) nanoparticles), ‘\(V\)’ is the volume of the reaction solution in litres, \(N_A\) is the Avogadro’s number.
The concentration of nanoparticle is calculated from the equation (2) and it $\sim 61$ nm and the number of nanoparticles in 20 ul solution which is used to drop cast on the glass plate of $1 \times 1$ cm$^2$ is $\sim 6 \times 10^5$ nanoparticles.

Figure 3. (a) A typical bright field TEM image of bio-reduced Ag NPs (b) Histogram of the Ag NPs. (c) HRTEM image of single spherical nanoparticle and (d) SAED pattern of the Ag NPs with the rings arisen due to the reflections from $(111)$, $(200)$, $(220)$, $(311)$, $(222)$, $(331)$, $(420)$ and $(422)$. 
For the spin–orbit interaction measurements, we have prepared the substrate by cutting the microscopic glass slide with the dimension of $1 \times 1$ cm$^2$. For each measurement, 20 µl of this Ag NP solution were dropped onto the microscopic glass slide and dried naturally. A fixed volume micro-pipette with disposable tip was used to prevent contamination. In our tests the dropped solution soon spread over the whole glass slide. The number of Ag NPs under focused laser beam of size 2 µm are approximately 2000.

4. Application to nanophotonics and spin–orbit interaction

The average size of the Ag NPs is observed as 80 nm. They can be used as good scatterer candidate to study the spin–orbit interaction of light. They scatter the light beam upon interacting with them and thus changing the polarization which depends on the particle size, refractive index and angular spread of light beam. Angular spread of light beam and beam size can be controlled in accordance with particle size by using a microscope objective lens of desired Numerical Aperture (NA). High NA lens focuses the beam to a size comparable to that of particle size and the interaction between light and nano and micro particles can happen. As a result, the nanoparticle scatters the light beam and properties of Ag NPs changes, which is translated in the polarization behaviour of output light changes. The polarization change in comparison to input polarization can be measured via performing Stokes Polarimetry measurement on the output light beam to measure four Stokes parameters ($S_0, S_1, S_2, S_3, S_4$). Stokes parameters provide the complete information about the polarization, phase and amplitude of light beam. $S_0$ describes the total intensity of beam, $S_i$ describes the component of linear TM and TE polarizations, $S_4$ describes linear $\pm 45^\circ$ polarizations and $S_3$ describes the component of right and left circular polarizations in the light beam across the cross section.

4.1. Experimental measurement

The Experimental setup for polarimetry measurement to measure the Stokes parameter is shown in figure 4. We have used a collimated monochromatic light source of wavelength $\lambda = 632.8$ nm from He–Ne laser. Polarizer $P_1$ and $P_2$ is used to select the light in a required polarization state. In the experiment $P_1$ is kept at $0^\circ$ to get transverse magnetic (TM) polarized beam. The TM polarized light beam then focused via a microscope objective lens of NA = 0.4 and is allowed to fall on the glass plate containing Ag NPs. A drop of Ag NPs was put on the glass slide and then allowed to dry overnight before it was used for the experiment. The glass slide containing Ag NPs is kept in the focal plane of focused light beam where it interacts with the light and the scattering happens. With the objective lens of NA = 0.4, the light beam was focused to a spot size given by $0.61 \lambda/NA$, which is calculated approximately $1.8 \mu m$ [48]. While the average size of Ag NPs in 80 nm, the beam size is much bigger therefore many nanoparticles interact with different parts of beam cross section and changes the polarization of beam accordingly. From quick calculation it was observed that approximately 2000 Ag NPs were present in the focal spot of light beam to scatter the light. After the scattering, the light beam is collimated and collected by another objective lens of same NA (= 0.4). A Quarter-wave-plate (QWP) and a polarizer ($P_2$) is used to perform Stokes Polarimetry measurement. At the end of set up, a CCD camera is used to record the intensity images and then processed in Matlab for further analysis.

As a preliminary measurement the experiment was performed for TM polarized light beam by setting the polarizer $P_1$ at $0^\circ$. Six intensity images ($I(\phi, \theta)$) were recorded at different orientations of QWP ($\phi$) and polarizer $P_2$, ($\theta$), where $\phi$ is retardation angle of QWP and $\theta$ is orientation angle of polarizer. First four intensity images are recorded at $0^\circ$ retardation of QWP and $0^\circ$, $45^\circ$, $90^\circ$ and $-45^\circ$ orientation of Polarizer $P_2$, respectively. Last two images were recorded at QWP retardation of $90^\circ$ and polarizer orientation of $45^\circ$ and $-45^\circ$ respectively. From

![Figure 4. Experimental set up to measure the Stokes parameters. A He–Ne laser source is used for this purpose. $P_1$ and $P_2$ are Glan-Thompson polarizers, QWP is quarter-wave plate. Objective lens of NA = 0.4 is used for focusing and collecting the scattered light beam. CCD is a charged coupled device to record the intensity images.](image-url)
these intensity images, the Stokes parameters $S_0$, $S_1$, $S_2$ and $S_3$ are deduced as follows,

\[
S_0 = I(0^\circ, 0^\circ) + I(0^\circ, 90^\circ), \\
S_1 = I(0^\circ, 0^\circ) - I(0^\circ, 90^\circ), \\
S_2 = I(0^\circ, 45^\circ) - I(0^\circ, 135^\circ), \\
S_3 = I(90^\circ, 45^\circ) - I(90^\circ, 135^\circ). 
\]  

(3)

The ellipse orientation and ellipticity angle of polarization ellipse is given by below equations,

\[
\psi = \frac{1}{2} \tan^{-1} \left( \frac{S_2}{S_1} \right), \\
\chi = \frac{1}{2} \tan^{-1} \left( \frac{S_4}{S_0} \right). 
\]  

(4)

4.2. Results and discussion

It can be observed from the images of Stokes parameters shown in figure 5 that the beam has acquired an additional polarization component across the beam cross section upon scattering which is approximately uniform. The measurement of Stokes parameters was performed at two different positions of Ag NPs on the glass plate and shown in column 2 and 3 of figure and compared with the result without Ag NPs sample shown in column 1. From the distribution of $S_2$ and $S_3$ across the beam cross section, we can see that a uniform polarization accumulation happened in the cross section of beam, suggesting that the nanoparticles interact with the beam in a similar manner. Though the interaction of nanoparticle happened at different places throughout the beam cross section. It can be seen that one side of beam has acquired one polarization and other side has acquired the opposite polarization. Stokes parameter $S_2$ shows that the upper half of the beam cross section has acquired linear $45^\circ$ polarizations accumulation and lower half of the beam cross section has acquired $-45^\circ$ polarization accumulation. The image of $S_3$ shows that the upper cross section has acquired right circular polarization accumulation and lower part has acquired left circular polarization accumulation in the beam cross section. This suggest the splitting of beam into two opposite polarizations, typically separation of two spin states.

Figure 5. Stokes parameters are shown for glass plate without nanoparticle sample (first column) and with samples (second and third columns) at two different positions of glass plate. (a) Stokes parameter $S_2$ which shows the presence of components of linear $\pm 45^\circ$ polarizations in the beam cross section. (b) Stokes parameter $S_3$ which shows components of right and left circular polarization in the beam cross section.
in the linear polarized TM beam upon scattering by Ag NPs, that can be connected to giant spin-Hall effect of light [30, 41]. Spin-Hall effect of light is splitting of TM and TE polarized light into right and left circular polarizations. This splitting can be further increased by increasing the concentration of nanoparticles.

Ellipse orientation ($\psi$) and ellipticity angle ($\chi$) across the beam cross section is shown in figure 6. Ellipse orientation actually measures the orientation of major axis of polarization ellipse with respect to horizontal direction and related to describing the orientation of polarization components other than TM and TE. While ellipticity angle is angle between major and minor axis of polarization ellipse and related to the measure of circularity present in the beam. This is clearly presence of spin angular momentum in light beam. These two measurements again confirmed the presence of polarization modulation obtained upon scattering of TM polarized beam by Ag NPs. One can see that the upper half of the beam cross-section has acquired positive ellipse orientation (a) and ellipticity angle (b), while the lower half has acquired the negative ellipse orientation and ellipticity angle.

Figure 6. Polarization ellipse parameters (a) ellipse orientation and (b) ellipticity angle. First and second columns are result for two different positions of glass plate.

5. Conclusions

A simple, and economical biosynthesis of Ag NPs using fresh leaf extracts results in Ag NPs that are mostly spherical in shape and it is demonstrated that this method produces a broad distribution of Ag nanoparticles. We have also shown that the particles synthesized by this method can have potential application in spin–orbit interaction studies. We have demonstrated this by studying the Stokes parameters $S_2$ and $S_3$ and polarization ellipse parameters ellipse orientation ($\psi$) and ellipticity angle ($\chi$) of input TM polarized beam using Polarimetry measurement. After TM polarized beam interacted with the nanoparticles, we observed that it induces an additional polarization component in the beam cross section that can be evidenced from $S_2$, $S_3$, polarization orientation ($\psi$) and ellipticity angle ($\chi$) measurements. It is also observed that these additional polarization components are acquired uniformly across the beam cross section showing that nanoparticles scatter the light beam in a similar manner.

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Conflicts of Interest

There are no conflicts of interest.

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