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Theory of the Scattering Phase Matrix Determination for Atmospheric Aerosols

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ABSTRACT

A theoretical foundation is presented for the experimental determination of the scattering-phase matrix for spherical aerosols. In this paper we present a calculation of the angular distribution of light for the atmospheric aerosols. Scattered light patterns produced by spherical transparent particles of a wide range of diameters and for a useful range of forward scattering angles (0-360°) are calculated by using Lorenz-Mie theory . A detailed comparison of the results leads to a definitive assessment of the accuracy of Lorenz-Mie theory in the forward direction.

Keywords : The Lorenz-Mie Theory, Aerosol, Scattering of Light, Phase Matrix Elements, Refracting Index

I. INTRODUCTION

Aerosols are small solid or liquid particles of range of sub-microns to several microns, suspended in the atmosphere. Basically there are two types of Aerosols, Primary and Secondary. The Primary are directly emitted as particle in the atmosphere and the secondary are originate from chemical reactions [1]. Aerosols play multiple roles in the climate system. They affect the radiation budget directly by scattering and absorption of solar radiation and indirectly, by causing changes in the cloud albedo and lifetime. The polarization of a plane wave from spherical particles is an old object of research, but it is still maturing as new applications demand more detailed understanding. Its rigorous mathematical solution was obtained as the well-known infinite series of partial waves, generally known as the Mie solution. There is now considerable amount of interest in the polarization of radiation reflected from particles of small size in atmosphere. The four Stokes parameters are the key to understand the effect of particles in atmosphere. A good description of Mie theory is provided by Liou K. N [2] and the notation in his book is thus adopted. To compute parameters obtained by using Mie Theory programming language FORTRAN has been used. This code provides accurate results for small and large particles with size parameters.

In this paper, we present a theoretical foundation for determining experimentally the four-by-four scattering phase matrix for spherical aerosol. Moreover, the corresponding relationship of the approximate period of phase functions and the relative refractive index (m) is studied.

II. THEORETICAL BACKGROUND

2.1 Properties of Atmospheric Aerosols

Many types of particles like aerosols, water droplets, and ice crystals to raindrops, snowflakes, and hailstones are exist in earth’s atmosphere. All of these particles are produced by a number of physical and dynamic processes. Atmospheric aerosols reside mainly in the two lowest layers of the atmosphere: the troposphere and the stratosphere [3]. Primary aerosols are directly emitted into atmosphere. Sea spray, mineral dust, volcanic ash, plant and animal debris are the example of primary aerosol. Secondary aerosol are formed in the atmosphere by gas-to-particles conversion processes. SO_2, NO_2 and volatile organic compounds (VOC) are responsible for gas-to-particle conversion, thus producing secondary aerosols [4].

Size is the most fundamental property of an aerosol particle and there is always a distribution of differently sized particles in the atmosphere. When the size of aerosol is less than 2.5μm then it is known as fine mode, if it is greater than 2.5μm then it is known as coarse mode [5]. The fine mode can be further divided into a cluster mode (1–3 nm), a nucleation mode (3–25 nm), an aitken mode (25–100 nm), and an accumulation mode (100–1000 nm) [6].
The earth's radiation budget is the managed by incoming energy from the sun and outgoing energy from the earth. The earth will warm if the Earth and Earth's atmosphere absorb more solar energy than it radiates back to space. The earth will cool if the earth and the earth system radiate more energy to space than it receives from the sun, [7]. When the shortwave emitted from sun is reaches to the earth atmosphere, about one third of the shortwave radiation is reflected back to space by clouds, aerosols, atmospheric molecules and the surface. In addition, a fraction of the incoming solar radiation that is transferred by the atmosphere to the surface is absorbed by greenhouse gases present in the atmosphere. Therefore, only half of the direct shortwave radiation reaches the surface and is absorbed as heat. Due to the ability of greenhouse gases to absorb longwave radiation, the thermal radiation emitted by the surface in turn heats the atmosphere. The thermal radiation emitted by both the atmosphere and the surface is absorbed by clouds and aerosols, so the temperature of atmosphere should be increase [8].

Aerosols have short lifetime and variety of sources, so their effect on climate is much more complex than that of greenhouse gases. Their radiative effects are determined by their concentration, chemical composition, size and shape, are highly variable in both space and time. As a result aerosols affect both regional and global climate [9].

2.2 Interaction of Electromagnetic Radiation with Aerosols

Aerosols influence climate by absorbing and scattering of solar and thermal radiation. These two processes reduce the amount of shortwave radiation reaching the Earth's surface. By reflecting the solar radiation back to space, aerosols contribute to cooling the atmosphere and the surface, whereas absorbing processes lead to positive climate forcing [10].

Scattering and absorption properties of a particle are determined by its chemical composition, size, and the wavelength of the incident radiation. These processes are governed by two wavelength (λ) dependent parameters; the refractive index m and the dimensionless size parameter *x*:

$$m = m_r - im_i$$

$$x = \frac{2\pi}{\lambda} a$$

where the real part represents the scattering whereas the imaginary part represents absorption. The real part of the refractive index in the visible spectrum is not very sensitive to the chemical composition. The real refractive index is variable in the range between mr=1.3-1.65 for most aerosol types and relative humidity while the imaginary part of the index is much more variable, ranging from 1 x 10-9 (e.g water droplets) to more than 0.1 in very sooty or iron-rich dust aerosols [11]. In equation 1.2, λ is the wavelength of incident radiation and a is radius of aerosol.

The elastic scattering can be classified further into two: Rayleigh scattering and Mie scattering, depending on the size of the particle as compared to wavelength of the incident radiation. When the particle size is much smaller than the incident wavelength, the scattering is called Rayleigh scattering (x<<1). Blue sky is an excellent example of Rayleigh scattering. When the particle size is same as the incident wavelength, the scattering is called Mie scattering (x˜1). Except in Microwave region, all the particle size is larger than the solar radiation so the Rayleigh scattering theory is not applicable. When the particle size is larger than incident wavelength, the scattering is known as Geometric scattering [5]. Except in Microwave region, all the particle size is larger than the solar radiation so the Rayleigh scattering theory is not applicable [11].

2.3 Polarization

Light, as any other electromagnetic wave, nearly always propagates as a transverse wave, with both electric and magnetic fields oscillating perpendicularly to the direction of propagation. The direction of the electric field is called the polarization of the wave. In a linearly polarized plane wave, the electric field remains in the same direction as the wave propagates. Natural light as a mixture of waves with different polarizations is also called unpolarized light or, more precisely, randomly polarized light. According to the Poynting theorem, the energy flow (intensity of the light or illuminance, I) associated with plane electromagnetic waves is proportional to the square of the amplitude of the electric field. When light interacts with matter, its behaviour is modified, mainly its intensity and its velocity. Moreover, some materials are able to modify light differently in each spatial direction. Linear polarizers for example can convert unpolarized light into linearly polarized light. An ideal polarizer fully attenuates light polarized in one direction, and fully transmits light with the orthogonal polarization [12]. Consider a beam of linearly polarized light incident upon a linear polarizer. The amplitude of the electric field before the polarizer is E0 and its intensity is I0 ˜E_0^2. Let θ be the angle between the axis of the polarizer and the polarization of the incident light. The electric field that passes through the polarizer is the component in the direction of the axis, E = E0 cosθ. Therefore, the intensity of the light passing the polarizer is

I = I_0 cos2θ

This is the so-called Malus law, named after the French physicist Louis Malus, who discovered optical polarization [13].

2.4 Scattering Phase Matrix:

To specify the polarization configuration of a radiation beam, the Stokes parameters I, Q, U, V are required. The Stokes parameters are defined as follows

$$\begin{aligned} I &= E_{\parallel}E_{\parallel}^{*} + E_{\perp}E_{\perp}^{*}, \\ Q &= E_{\parallel}E_{\parallel}^{*} - E_{\perp}E_{\perp}^{*}, \\ U &= E_{\parallel}E_{\perp}^{*} + E_{\perp}E_{\parallel}^{*}, \\ V &= -i(E_{\parallel}E_{\perp}^{*} - E_{\perp}E_{\parallel}^{*}). \end{aligned} \qquad (1)$$

where an asterisk denotes the complex conjugate value [14].

Letting the subscript 0 denote the incident components, we show that

$$\begin{bmatrix} I \\ Q \\ U \\ V \end{bmatrix} = \frac{\mathbf{F}}{k^2r^2} \begin{bmatrix} I_0 \\ Q_0 \\ U_0 \\ V_0 \end{bmatrix} \qquad (2)$$

where the matrix is F is called the transformation matrix of light scattering of single sphere. In general, F has all 16 components. But for the sphere, they are reduce to four components.

$$\mathbf{F} = \begin{bmatrix} \frac{(M_2 + M_1)}{2} & \frac{(M_2 - M_1)}{2} & 0 & 0 \\ \frac{(M_2 - M_1)}{2} & \frac{(M_2 + M_1)}{2} & 0 & 0 \\ 0 & 0 & S_{21} & -D_{21} \\ 0 & 0 & D_{21} & S_{21} \end{bmatrix} \qquad (3)$$

where its components are defined by

$$M_1 = S_1(\theta)S_1^{*}(\theta),$$

$$M_2 = S_2(\theta)S_2^{*}(\theta),$$

$$S_{21} = [S_1(\theta)S_2^*(\theta) + S_2(\theta)S_1^*(\theta)]/2,$$

$$-D_{21} = [S_1(\theta)S_2^*(\theta) - S_2(\theta)S_1^*(\theta)]i/2 \qquad (4)$$

Where, $s_1(\theta)$ and $s_2(\theta)$ are scattering functions [15]

$$s_1(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [a_n \pi_n(\cos\theta) + b_n \tau_n(cos\theta)] \qquad (5)$$

$$s_2(\theta) = \sum_{n=1}^{\infty} \frac{2n+1}{n(n+1)} [b_n \pi_n(\cos\theta) + a_n \tau_n(cos\theta)], \qquad (6)$$

Where,

$$\pi_n(\cos\theta) = \frac{1}{sin\theta} P_n^m(cos\theta),$$

$$\tau_n(\cos\theta) = \frac{d}{d\theta} P_n^m(cos\theta).$$

Here, $P_n^m(\cos\theta)$ is associated Legendre polynomial of n degree and m order [16],

In eq.(5) and (6) a_n and b_n are known as Mie coefficients.

$$a_n = \frac{\psi_n'(y)\psi_n(x) - m\psi_n(y)\psi_n'(x)}{\psi_n'(y)\xi_n(x) - m\psi_n(y)\xi_n'(x)} \qquad (7)$$

$$b_n = \frac{m\psi_n'(y)\psi_n(x) - \psi_n(y)\psi_n'(x)}{m\psi_n'(y)\xi_n(x) - \psi_n(y)\xi_n'(x)} \qquad (8)$$

By using transformation matrix we may define a parameter referred to as the scattering phase matrix such that

$$\frac{\mathbf{F}(\theta)}{k^2r^2} = C\mathbf{P}(\theta). \qquad (7)$$

The coefficient C can be obtained from normalization of the matrix element in the form

$$\int_0^{2\pi}\int_0^{\pi} \frac{P_{11}(\theta)}{4\pi} \sin\theta \, d\theta \, d\phi = 1. \qquad (8)$$

From Eqs. (7) and (8), we obtain

$$C = \frac{1}{2k^2r^2}\int_0^{\pi} \frac{1}{2}[M_2(\theta) + M_1(\theta)]\sin\theta \, d\theta = \frac{1}{4k^2r^2}\int_0^{\pi} [i_1(\theta) + i_2(\theta)]\sin\theta \, d\theta. \qquad (9)$$

The coefficient C is also given by following equation [17].

$$C = \sigma_s/4\pi r^2 \, . \qquad (10)$$

Where, σ_s is scattering cross section.

$$\sigma_s = Q_s \pi^2$$

Where, Q_s is scattering efficiency.

$$Q_s = \frac{2}{x^2}\sum_{n=1}^{\infty}(2n+1)(|a_n|^2 + |b_n|^2). \qquad (11)$$

So we get,

$$\frac{P_{11}}{4\pi} = \frac{1}{2k^2\sigma_s}(i_1 + i_2), \qquad (12)$$

$$\frac{P_{12}}{4\pi} = \frac{1}{2k^2\sigma_s}(i_2 - i_1), \qquad (13)$$

$$\frac{P_{33}}{4\pi} = \frac{1}{2k^2\sigma_s}(i_3 + i_4), \qquad (14)$$

$$\frac{P_{34}}{4\pi} = -\frac{i}{2k^2\sigma_s}(i_4 + i_3), \qquad (15)$$

Where

$$i_j = S_j S_j^* = |S_j|^2, \qquad j = 1,2,$$

$$i_3 = S_2 S_1^*, \qquad i_4 = S_1 S_2^*$$

So the scattering phase matrix for a single homogeneous sphere is given by

$$\mathbf{P}=\begin{bmatrix} P_{11} & P_{12} & 0 & 0 \\ P_{12} & P_{11} & 0 & 0 \\ 0 & 0 & P_{33} & -P_{34} \\ 0 & 0 & P_{34} & P_{33} \end{bmatrix}. \qquad (16)$$

The phase matrix consists of 16 non zero elements if there is no assumption is made about the shape and position of the scatterer. But for a single sphere , the independent elements reduce to only four. Scattering phase matrix elements of P11, P12, P33 and P34 are known as phase function. The phase function represents the angular distribution of the scattered energy. It is the ratio of the scattered intensity at a specific direction to the integral of the scattered intensity at all directions [18]. The phase function depends on the orientation of the particle with respect to the direction of the incident radiation and on the particle characteristics [19].

III. RESULTS

To investigate in detail the effect of size of aerosol we select three different wavelength; λ= 500 nm, 700 nm, 900nm. Radius (*a*) of aerosol is taken equal to 5.30 μm. Through, the solution of a in the present study is arbitrary and represent only a test-case. Further, we have tested and analysed the result for two refractive indices;

Only with real part (mr).

Including imaginary refractive index (mi).

A complete computation is performed in DELL PRECISION 5810 work station on window platform, while FORTRAN-90 code is constructed to program calculations.

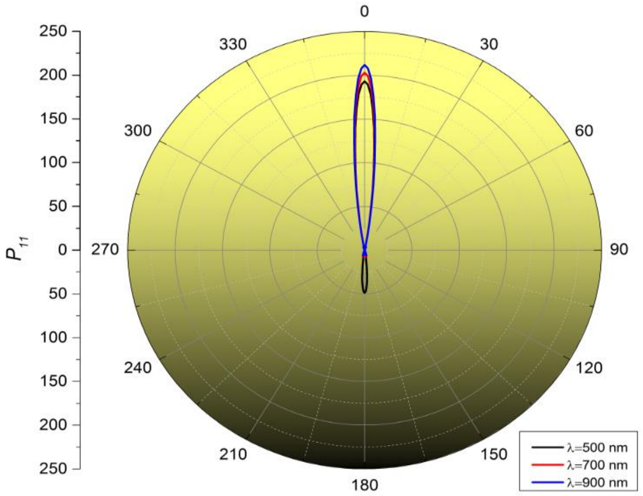

Figure 3.1: Scattering phase matrix element (P_{11}) as a function of scattering angle for m_r=1.5, m_i=0.0, λ=500 nm, 700 nm, 900 nm.

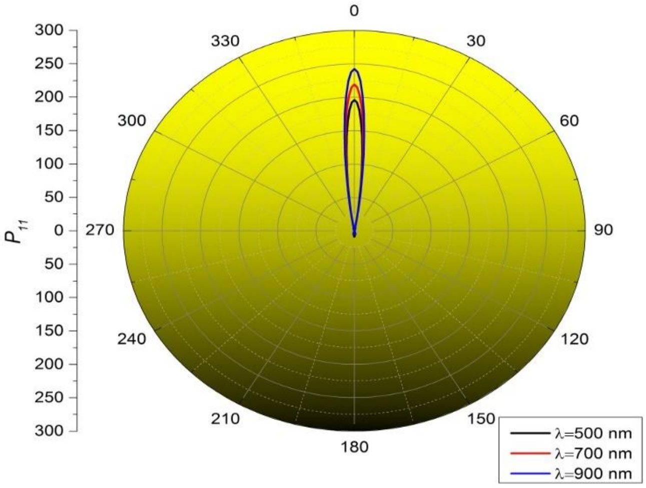

Figure 3.2: Scattering phase matrix element (P_{11}) as a function of scattering angle for m_r=1.5, m_i=0.01, λ=500 nm, 700 nm, 900 nm.

The Lorenz-Mie particles are characterized by strong forward scattering and backscattering. By using $\pi_n \cos\theta$ and $\tau_n \cos\theta$ we obtained the phase matrix elements. The phase matrix elements contain the full information about the scattering process. Characteristics of the normalized phase functions (P_{11}) of sphere are shown in above figs. 3.1-3.2.

When the wavelength incident unpolarised light is 500 nm, the intensity should be decrease up to 90° and then it should start to increase and maximum near back scattering direction. But When the wavelength is

700 nm, the intensity should be increase in forward scattering direction (0°< θ<20°) and then it decrease rapidly and then it became start to increase in back scattering direction. When the wavelength is 900 nm, it shows same behaviour as for 500 nm. The imaginary part Of refractive index is negligible in P_{11} .

There is enhanced intensity in back scattering direction (˜180˚) is known as 'glory'. This is due to spherical shape of scatterer which serves to focus certain rays at ˜180˚. The back scattering is smaller than forward scattering [20].

Here the scattering pattern consists of rapid fluctuation due to interference effects [21].

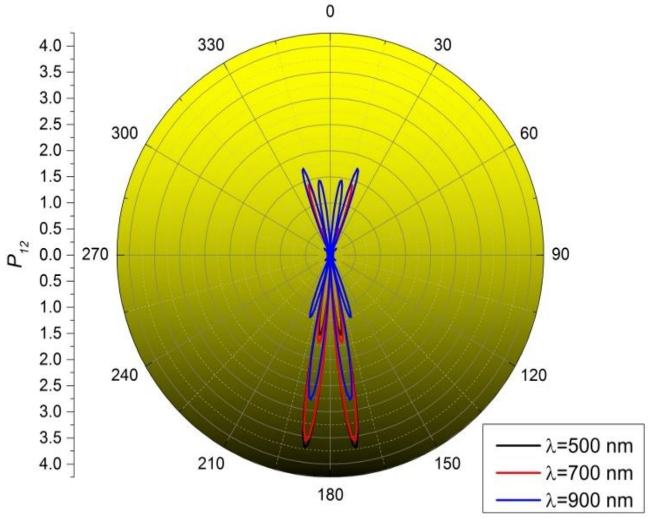

Figure 3.3: Scattering phase matrix element (P_{12}) as a function of scattering angle for m_r=1.5, m_i=0.0, λ=500 nm, 700 nm, 900 nm.

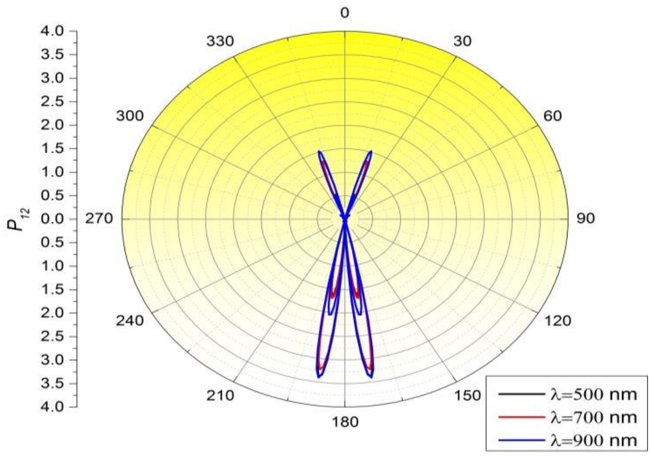

Figure 3.4: Scattering phase matrix element (P_{12}) as a function of scattering angle for m_r=1.5, m_i=0.01, λ=500 nm, 700 nm, 900 nm.

Figs. 3.3-3.4 show the angular distribution of scattering phase matrix element P_{12} as a function of scattering angle for three different wavelength in visible range. From figs. 3.3-3.4, we can say that the deviation in P_{12} is observed only in forward and back scattering direction[22].

When the refractive index has only real part then forward scattering is found negative for all three wavelength and the backward scattering positive [23]. But when the imaginary part is added in the refractive index the forward scattering and back scattering shows same behaviour as we discuss above but with decrease in magnitude. However, for 900 nm, we get reverse characteristics. This observation suggests that scattering phase matrix is a combined function of size parameter and wavelength both [24].

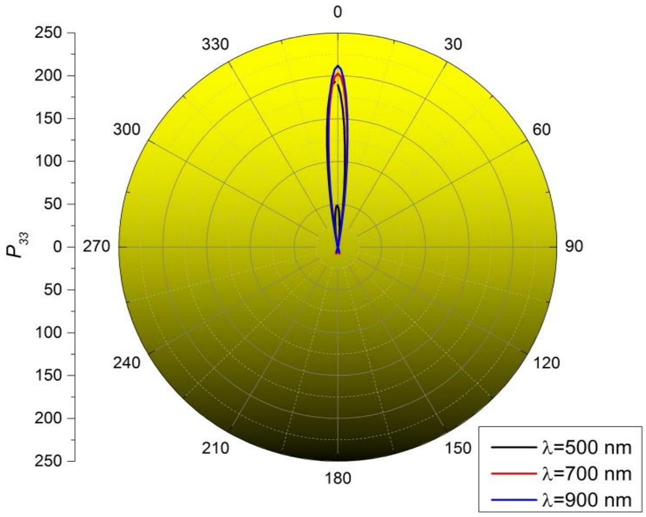

Figure 3.5: Scattering phase matrix element (P_{33}) as a function of scattering angle for m_r=1.5, m_i=0.0, λ=500 nm, 700 nm, 900 nm.

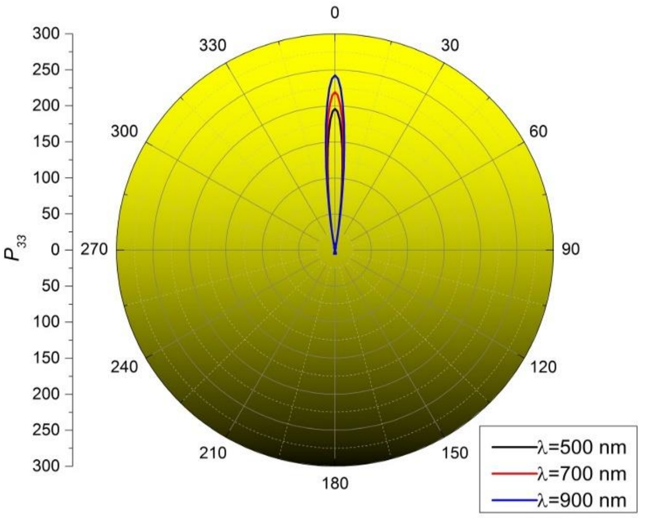

Figure 3.6: Scattering phase matrix element (P_{33}) as a function of scattering angle for $m_r=1.5$, $m_i=0.01$, $\lambda=500$ nm, 700 nm, 900 nm.

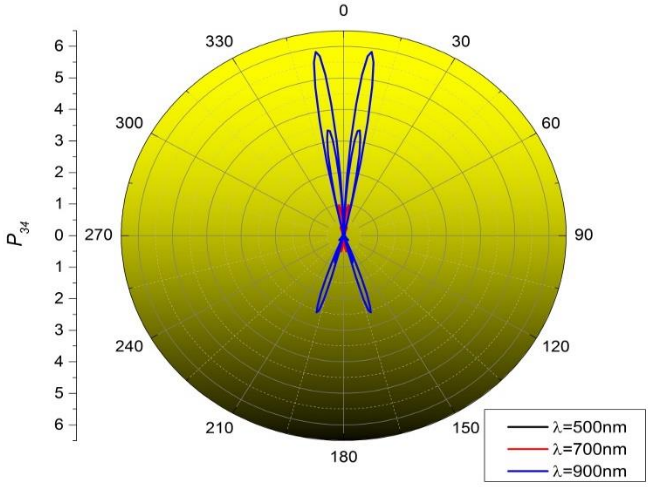

Figure 3.7: Scattering phase matrix element (P_{34}) as a function of scattering angle for $m_r=1.5$, $m_i=0.0$, $\lambda=500$ nm, 700 nm, 900 nm.

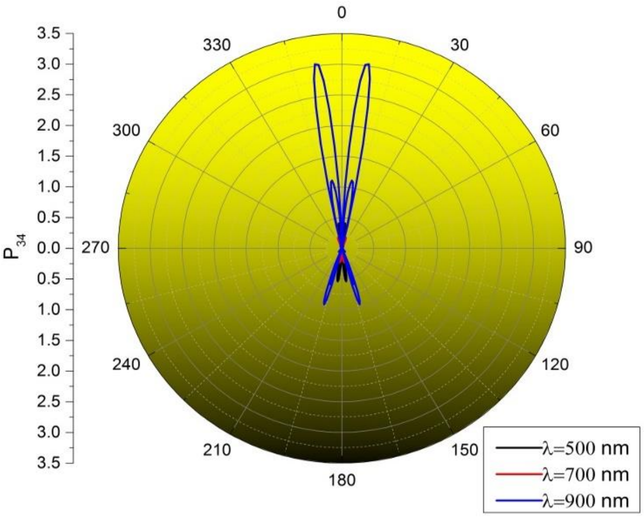

Figure 3.8: Scattering phase matrix element (P_{34}) as a function of scattering angle for $m_r=1.5$, $m_i=0.01$, $\lambda=500$ nm, 700 nm, 900 nm.

The phase matrix element P_{33} and P_{34} has same behaviour as P_{11} and P_{12} respectively [25].

IV. CONCLUSION

The results indicate that There is no much (non-noticable) deviation in P_{11} and P_{33} when the imaginary refractive indices increase from 0.00 to 0.05. The graph of P_{33} is similar to P_{11}. In all phase matrix elements, the maximum deviation occurs near the forward scattering direction and backward scattered direction, showing largest sensitivity and hence larger polarizability. This observation reveals importance of low-scattering angle scattering experiment. P_{12} and P_{34} also shows the negative values. By adding the imaginary part of refractive index, the change in the graph is quite noticeable; emphasising the fact that polarization is a sensitive function of refractive index.

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