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Spectral Dependence of Quasi-Rayleigh Polarization Leap of Nonspherical Particles：Polystyrene Beads Application

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Abstract Solid particles in Earth's atmosphere，such as polystyrene beads，are an important factor affecting the processes of absorption and scattering of light in the atmosphere. These processes affect on the solar energy transfer in the Earth's atmosphere，consequently they have influence on the regional and global climate changes and atmospheric visibility. In particular，great interest to study the scattering properties of small particles compared with wavelength，because of such particles experience low gravitational settlement and may have long time of life in the atmosphere. When scattering particle is much smaller than the wavelength of the scattered or absorbed light，this is the case of Rayleigh scattering. Scattering properties of these particles (such as intensity and the degree of linear polarization) at the Rayleigh scattering are simply derived from electromagnetic Maxwell's equations. But when the particles are large enough to be comparable with the wavelength，the deviations from Rayleigh scattering law are observed. One of the clear manifestations of such deviations is the recently discovered quasi-Rayleigh polarization leap of monodisperse spherical particles. This quasi-Rayleigh polarization leap allows remote sensing of the sizes of distant particles，based on the spectral position of quasi-Rayleigh polarization leap at different phase angles of observation. In this paper，we studied the effect of the non-sphericity of a scattering polystyrene particle on the magnitude and position of the quasi-Rayleigh polarization leap. It is established that the non-sphericity shifts the position of the quasi-Rayleigh polarization leap shorter wavelengths. It is shown that for non-sphericity of particles makes the quasi-Rayleigh polarization leap becomes less pronounced. Moreover，it was found，that increasing of the phase angle and degree of non-sphericity shift the quasi-Rayleigh polarization leap position to shorter wavelength. However，in the case of not very elongated particles，the quasi-Rayleigh polarization leap is quite well manifested. Therefore，this method is suitable for remote sensing not only the size，but also the degree of non-sphericity of the scattering particles. A simple formula has been obtained for polystyrene beads that relates the degree of non-sphericity of a particle with the wavelength and phase angles at which the quasi-Rayleigh polarization leap is observed.

Keywords Nanomaterials；Polarization；T-matrix；Quasi-Rayleigh polarization leap

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Introduction

The term《Rayleigh scattering》is commonly associated with light scattering by molecular and atomic gases and the explanation of《why the sky is blue?》[1]. In addition，Rayleigh scattering describes the scattering of light by particles much smaller than the wavelength of the light divided by the real part of the complex refractive index of scattering particle $m=n+ik$.

The scattering characteristics depend on the ratio between the size of scattering particle and wavelength of incident light. That is why the scattering particle is characterized by a size parameter $X=\frac{2\pi r}{\lambda}$，where λ is the wavelength of the incident light，and r is characteristic particle size，which in the case of non-spherical scattering particle can be

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treated as the radius of sphere of equivalent volume. For Rayleigh scattering, the following condition should be kept: $X|n| \ll 1$. Both intensity and another characteristics of scattered light depend on the phase angle α, i. e. on the angle between the direction lightsource—scatterer and scatterer—observer. The phase function of Rayleigh scattering depends on the phase angle as[2]:

$$I \sim \frac{1+(\cos\alpha)^2}{\lambda^4} \tag{1}$$

Even when scattering particle is illuminated by unpolarized light, scattered light will be partially polarized. For description of scattered light polarization state, the degree of linear polarization was introduced. For Rayleigh lightscattering, it can be described by the following equation[2]

$$P = -\frac{Q}{I} = \frac{I_\perp - I_\parallel}{I_\perp + I_\parallel} = \frac{1-(\cos\alpha)^2}{1+(\cos\alpha)^2} \tag{2}$$

Here, Q and I are the Stokes parameters[3], $I_\perp$ and $I_\parallel$ are the intensities of scattered light, that are polarized perpendicularly to the scattering plane and within it, correspondingly. Note that, in accordance with Eqn. (2), the degree of linear polarization of Rayleigh particles is always positive.

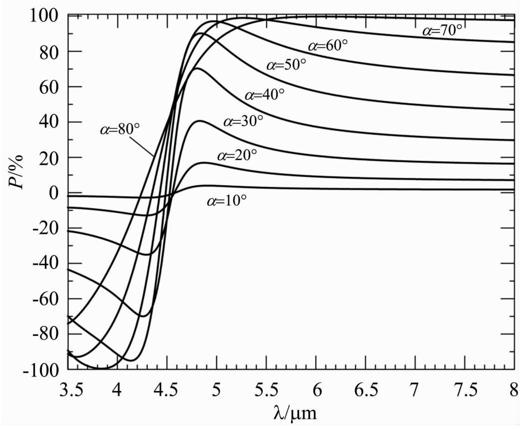

Fig. 1 Spectral dependence of the linear polarization degree for spherical polystyrene particle with size $R=1.227$ μm and $m=1.5722$ at different phase angles from 10° to 80°

The larger the particle size becomes, the more pronounced are the deviations from the Rayleigh scattering law. Most notably these deviations are manifested in the so-called quasi-Rayleigh polarization leap[4]. This phenomenon manifests itself precisely in the degree of linear polarization. It consists in a sharp drop in the degree of linear polarization from the values described by Eqn. (2) to negative values, when the wavelength is decreased. Figure 1 shows the quasi-Rayleigh polarization leap for spherical polystyrene particles with a refractive index[4] of $m=1.5722$ and a size[5] of 1.227 μm at different phase angles. It can be seen from the figure that the strongest quasi-Rayleigh polarization leap occurs at a phase angle of about 50 degrees.

1 Quasi-Rayleigh polarization leap of non-spherical particles calculation

Absolutely spherical particles are found in nature quite rarely. Even in the case of polystyrene particles exhibiting a fairly distinct Mie spectrum[6], there are some discrepancies in the determination of physical properties, which, according to the researchers, can be explained by deviations of the shape of the particles from the sphere[5]. The simplest way to take into account the non-sphericity of a particle is to consider the transformation of a sphere into a spheroid. A spheroid is a sphere elongated or compressed along one of the axes. In the first case, we can call it a prolate spheroid, and in the second, an oblate spheroid (see Fig. 2). Deviations from the sphericity of such particles can be characterized by the ratio of two mutually perpendicular axes a/b, where a is the length of the axis perpendicular to the rotation axis, and b is the length of the axis parallel to the rotation axis. In the case of a prolate spheroid $a/b<1$, as well as for an oblate spheroid $a/b>1$.

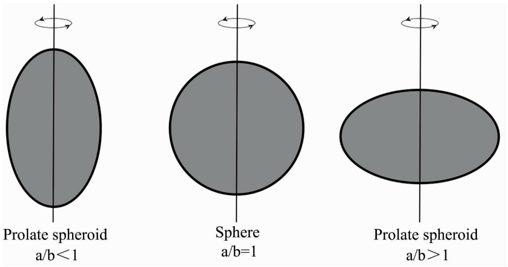

Fig. 2 An examples of spheroids with different axial ratio a/b

For calculation the degree of linear polarization of the spheroids, the T-matrix method[7-8] is widely used. This method based on the expansion of the incident and scattered electromagnetic fields in a series of vector spherical functions. The advantages of this method include fairly fast calculations, as well as the possibility of analytically averaging the characteristics of scattered light over the orientations of the scattering particle. Recently the modification of the T-matrix method was developed, namely so-called shape-matrix method or the Sh-matrix method[9], which allows factorization of the quantities depending on the particle shape. Although Sh-matrix method usually used for calculations of much more irregular shape particles[10], it is suitable for spheroids too[11]. For calculations we use the Sh-matrix method, controlling the accuracy of calculations using the well-known program realization of T-matrix method[12-13].

We performed a series of calculations of the degree of linear polarization for a set of axial ratios and phase angles. It should be noted that in all cases we kept the volume of the scattering particle equal to the volume of a spherical particle with a radius of 1.227 μm. Figure 3 shows calculated spectral dependence of the linear polarization degree for non-spherical polystyrene particles having different axial ratio at phase angle $\alpha=50°$. Upper panel (a) corresponds to the case of prolate spheroids; lower panel (b) shows dependence for oblate spheroids. As can be seen from the figure, the non-sphericity of the scattering particle has a significant effect on both the position and the magnitude of the quasi-Rayleigh polarization leap. Moreover, both the elongation and compression of the particle affect the quasi-Rayleigh polarization leap in a similar way, shifting its position to the region of shorter wavelengths. The non-sphericity also makes the decrease in the degree of polarization with the reduction of the wavelength less sharp, that is, it actually "erodes" the quasi-Rayleigh polarization leap. In view of this, it is interesting to study the effect of non-sphericity on particles of various refractive indices, not limited to the case of polystyrene particles alone. This is the subject of the next paper that will be published soon.

2 Main properties of quasi-Rayleigh polarization leap of non-spherical polystyrene particle

Figure 3 clearly shows that the rate of change in the degree of linear polarization depends on the wavelength. Accordingly, at a certain wavelength, this speed will be maximal at absolute value. This wavelength will be denoted by λ_{QRPL} and will be called the position of quasi-Rayleigh polarization leap.

Calculations show that there is a clear relationship between the position of the quasi-Rayleigh polarization leap, the phase angle, and the axial ratio of the non-spherical particle. Graphically, this relationship is shown in Figure 4. Each point on the surface corresponds to a specific set of wavelengths, phase angles, and axial ratios at which quasi-Rayleigh polarization leap is observed. The following features of the quasi-Rayleigh polarization leap of non-spherical polystyrene particles are visible from the figure:

(1) An increase in the phase angle leads to a decrease in λ_{QRPL};

(2) An increase in the degree of non-sphericity of the scattering particle (both in the case of an prolate and oblate particles) leads to a decrease in λ_{QRPL};

(3) Prolate particles have a much stronger effect on the λ_{QRPL} changing, than oblate ones;

(4) There is an ambiguity in the dependence of the axial ratio on λ_{QRPL} and phase angle. Those, each pair of λ_{QRPL} and α values will correspond to two a/b values.

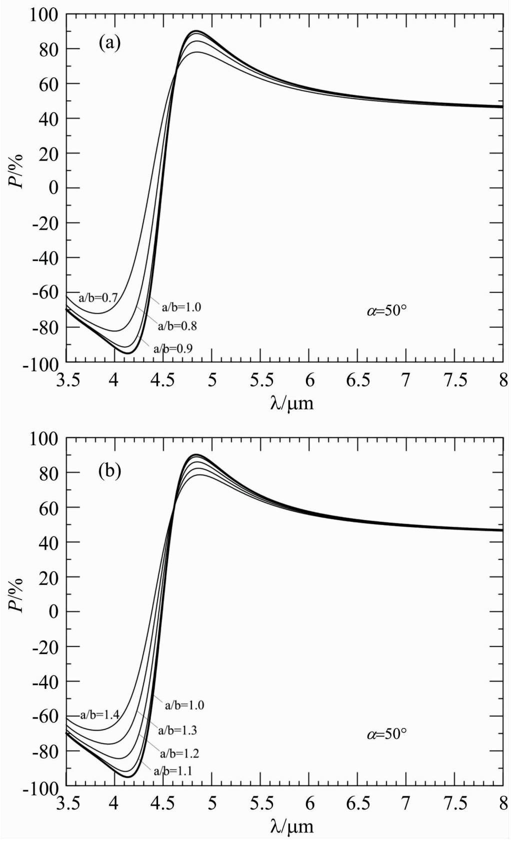

Fig. 3 Spectral dependence of the linear polarization degree for nonspherical polystyrene particles having different axial ratio at phase angle $\alpha=50°$

(a): Prolate spheroids with $a/b<1$;

(b): Oblate spheroids with $a/b>1$

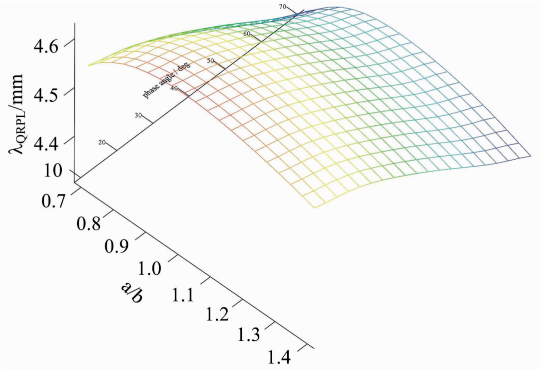

Fig. 4 3D surface, showing relation between the position of quasi-Rayleigh polarization leap λ_{QRPL}, phase angle α and axial ratio a/b

Fortunately, this dependence is quite smooth, and therefore the relation between these values can be approximated by a simple second-order equation

$$\lambda_{\mathrm{QRPL}}=4.0473+1.149\frac{a}{b}-0.5436\left(\frac{a}{b}\right)^2-2.2798\times$$

$$10^{-3}\alpha - 1.808\,1\times10^{-5}\alpha^{2} - 5.134\,2\times10^{-5}\alpha\,\frac{a}{b} \quad (3)$$

Here the phase angle α should be expressed in degrees, a/b is dimensionless value, and λ_{QRPL} will be in microns. Eqn. (3) valid for phase angles range from 10° to 70°, and a/b can change from 0.7 to 1.4. The maximum difference between Eqn. (3) and the exact meaning of λ_{QRPL} is 0.027 μm. For wavelengths of the order of 4.5 μm, this accuracy is very good.

3 Concluding remarks

In this paper, we investigated the influence of scattering particle non-sphericity on the phenomenon of a quick change of the degree of linear polarization, also called quasi-Rayleigh polarization leap. This feature appears with increasing of scattering particle size, when it becomes in several times higher than wavelength. The relations between main features that determine the quasi-Rayleigh polarization leap of polystyrene particles have been established.

It was found, that an increasing of the phase angle as well as degree of non-sphericity of the scattering particle shift the spectral position of quasi-Rayleigh polarization leap (λ_{QRPL}) to shorter wavelength. Moreover, a simple formula has been obtained for polystyrene beads. Eqn. (3) relates the degree of non-sphericity of a particle with the wavelength and phase angle at which the quasi-Rayleigh polarization leap is observed. So, the quasi-Rayleigh polarization leap can be used for remote sensing of distant non-spherical objects, in particular, polystyrene particles. Founded ambiguity in the dependence of the axial ratio on λ_{QRPL} and phase angle (when each pair of λ_{QRPL} and α values corresponds to two a/b values, for prolate and oblate spheroids, respectively) can be eliminated by a large number of measurements at different wavelengths and phase angles. Because of the established fact, that prolate particles have a much stronger effect on the quasi-Rayleigh polarization leap position, than oblate ones, a large set of measurements will make it possible to uniquely determine the degree of non-sphericity of the scattering particle.

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