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Theory of the scattering-phase-matrix determination for ice crystals

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A theoretical foundation is presented for the experimental determination of the scattering-phase matrix for nonspherical ice crystals. The scattering-phase matrix can be determined solely from four radiance measurements. Conditions for the phase matrix of a volume of ice crystals that are typical of those in cirrus clouds are presented.

Index Headings: Atmospheric optics; Scattering; Polarization; Crystals.

During the 60's, an enormous amount of effort was devoted to the electromagnetic scattering by water clouds consisting of spherical water drops, based on the exact Mie solution. With the growing capacity of the digital electronic computer, we are now in a position to evaluate the optical properties of water clouds for any wavelength in the solar and infrared spectra, provided that the cloud size distributions are given. Moreover, with the resulting comprehensive numerical tables[1] and the existing Mie computer programs, the problem of polarized light scattered by water spheres seems to have been completely solved.

However, ice clouds are composed of nonsymmetrical ice crystals whose shapes and sizes vary with atmospheric conditions. The determination of polarized light scattered by ice particles is made very difficult by their nonsphericity and the consequent problem of orientation. Knowledge of the scattering phase matrix of a volume of ice crystals is of vital importance for remote sensing of cloud compositions of the planet by means of radiance and polarization techniques,[2–6] as well as for radiation-budget studies in cloudy atmospheres.[7] In view of the great mathematical difficulties involved in deriving scattering solutions for nonspherical particles (e.g., ellipsoids and finite cylinders) whose sizes are much larger than the incident wavelength, it is unlikely that rigorous electromagnetic-wave solutions can be formulated and numerically computed for irregular ice crystals. Thus, information on the scattering of polarized light from ice crystals may best be obtained by the experimental approach.

Huffman and Thursby[8] reported angular scattering measurements for ice crystals, employing a visible wavelength of 0.5 μm without considering the polarization. Some aspects of measuring the polarized light scattered by atmospheric particles were discussed by Pritchard and Elliott,[9] who performed scattering experiments for a water fog, and by Holland and Gagne,[10] who measured the scattering of visible light by a sample of micrometer-size aerosols. In a classical paper, Jones[11] described a method for determining experimentally the two-by-two Jones matrix. Born and Wolf[12] also discussed procedures for producing different states of polarized light.

In this paper, we present a theoretical foundation for determining experimentally the four-by-four scattering phase matrix for nonspherical ice crystals typical of those in cirrus clouds.

I. STOKES PARAMETERS OF THE SCATTERED POLARIZED LIGHT

Consider a monochromatic light wave of circular frequency ω propagated in the positive z direction. Let

$$E_l = a_l e^{i\omega t}, \quad E_r = a_r e^{-i\delta} e^{i\omega t} \tag{1}$$

represent the components, at a point 0 of the electric vectors in two mutually orthogonal directions at right angles to the direction of propagation, where a_l, a_r are complex amplitudes and δ is the phase difference between two electric vectors.

Suppose that the r direction is subject to a retardation ϵ with respect to the l component, and consider the component of the electric vector in the direction that makes an angle ψ with the positive l direction, after the retardation ϵ has been introduced, is

$$E(t;\psi,\epsilon) = E_l\cos\psi + E_r e^{-i\epsilon}\sin\psi = a_l\cos\psi\, e^{i\omega t} + a_r\sin\psi\, e^{-i(\delta+\epsilon)} e^{i\omega t}. \tag{2}$$

The irradiance is therefore

$$I(\psi,\epsilon) = \langle E(t;\psi,\epsilon)E^*(t;\psi,\epsilon)\rangle, \tag{3}$$

where * is the conjugate, and $\langle\ \rangle$ represents a time average.

Substituting Eq. (2) into Eq. (3), we obtain

$$\begin{aligned} I(\psi,\epsilon) &= \langle a_l^2\cos^2\psi + a_r^2\sin^2\psi + 2a_l a_r\cos(\delta+\epsilon)\sin\psi\cos\psi\rangle \\ &= \langle a_l^2\rangle\cos^2\psi + \langle a_r^2\rangle\sin^2\psi + \tfrac{1}{2}\sin 2\psi(\langle 2a_l a_r \\ &\quad\times\cos\delta\rangle\cos\epsilon - \langle 2a_l a_r\sin\delta\rangle\sin\epsilon). \end{aligned} \tag{4}$$

The Stokes parameters of a monochromatic wave can be defined as

$$\begin{aligned} I &= \langle a_l^2\rangle + \langle a_r^2\rangle, \\ Q &= \langle a_l^2\rangle - \langle a_r^2\rangle, \\ U &= \langle 2a_l a_r\cos\delta\rangle, \\ V &= \langle 2a_l a_r\sin\delta\rangle. \end{aligned} \tag{5}$$

Thus, Eq. (4) becomes

$$I(\psi,\epsilon) = \tfrac{1}{2}[I + Q\cos 2\psi + (U\cos\epsilon - V\sin\epsilon)\sin 2\psi]. \tag{6}$$

To measure the Stokes parameters of the scattered light, we need only to obtain the irradiance for several different values of ψ (orientation of polarizer) and ϵ (delay introduced by the compensator). A set of irradiances may be measured at the convenient pairs of (ψ, ϵ); $(0°, 0)$, $(90°, 0)$, $(45°, 0)$, $(135°, 0)$ $(45°, \pi/2)$, $(135°, \pi/2)$. From these combinations, the Stokes parameters expressed in Eq. (5) are

$$\begin{aligned} I &= I(0°, 0) + I(90°, 0), \\ Q &= I(0°, 0) - I(90°, 0), \\ U &= I(45°, 0) - I(135°, 0), \\ V &= -[I(45°, \pi/2) - I(135°, \pi/2)]. \end{aligned} \tag{7}$$

From Eq. (7), it is evident that to obtain a complete set of Stokes parameters for the scattered polarized light, subsequent measurements of irradiance with a rotating polarizer that transmits components in the azimuths, ψ, of 0°, 90°, 45°, and 135° are required. A compensator that introduces a phase difference of a quarter period (e.g., a quarter-wave plate) between the components of 45° and 135° is also needed to obtain the V component. Note that the scattered irradiance can be determined from two measurements with a polarizer oriented in 0° and 90° directions.

II. MEANING OF THE PHASE MATRIX

First, we shall refer the electric vibrations to arbitrary but fixed rectangular axes. Because only linear processes are involved in optical instruments, the two orthogonal scattered electric vectors can be expressed as[13]

$$\begin{bmatrix} E_l \\ E_r \end{bmatrix} = \begin{bmatrix} A_2 & A_3 \\ A_4 & A_1 \end{bmatrix} \begin{bmatrix} E_{r0} \\ E_{l0} \end{bmatrix}, \tag{8}$$

where E_{l0} and E_{r0} are the incident electric vectors in two orthogonal directions, and

$$A_j = \frac{e^{-ikr}}{ikr} S_j, \quad j = 1, 2, 3, 4. \tag{9}$$

The S_j are nondimensional quantities dependent on the optical properties of the scattering medium; k and r denote the wave number and the distance, respectively.

In terms of the Stokes parameters, the scattered and incident waves for a fixed coordinate may be expressed as

$$\mathbf{I}(\Theta) = C\mathbf{M}(\Theta)\mathbf{F}_0, \tag{10}$$

where C is a constant of proportionality, $\mathbf{F}_0 = (F_0, Q_0, U_0, V_0)$ represents Stokes parameters of the incident wave in units of flux density, $\mathbf{I}$ is the scattered radiance in units of flux density per solid angle, the subscript 0 denotes Stokes parameters for the incident wave, Θ is the scattering angle, and the 4×4 nondimensional, normalized phase matrix $\mathbf{M}(\Theta)$ is defined in such a way that

$$2\pi \int_0^{\pi} M_{ij}(\Theta) \sin\Theta \, d\Theta = 1, \quad i, j = 1, 2, 3, 4. \tag{11}$$

M_{ij} represents the element in the ith row and jth column; its value can be obtained from the four combinations of $S_i S_j^*$. To measure these elements, it is not necessary to express them in terms of $S_i S_j^*$.

The reference planes of the incident and scattered electric vectors are normally related to different coordinate systems. Thus, in order to evaluate E_l and E_r correctly, a proper coordinate transformation must be carried out. For either the incident or the scattered electric vector, we may take new rectangular axes $0l'$ and $0r'$ such that $0l'$ makes an angle ϕ with $0l$. Then the components of the electric vector with reference to the new axes are

$$\begin{bmatrix} E_l' \\ E_r' \end{bmatrix} = \begin{bmatrix} \cos\phi & \sin\phi \\ -\sin\phi & \cos\phi \end{bmatrix} \begin{bmatrix} E_l \\ E_r \end{bmatrix}. \tag{12}$$

Hence, the Stokes parameters with respect to the new axes can be shown as

$$\mathbf{I}' = \mathbf{L}(\phi)\mathbf{I}, \tag{13}$$

with the angular transformation matrix

$$\mathbf{L}(\phi) = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\phi & \sin 2\phi & 0 \\ 0 & -\sin 2\phi & \cos 2\phi & 0 \\ 1 & 0 & 0 & 1 \end{bmatrix}. \tag{14}$$

It follows that the Stokes parameters of the scattered light expressed in Eq. (10) have to be modified to yield

$$\mathbf{I}(\Theta, \phi_1, \phi_2) = C\mathbf{L}(\phi_2)\mathbf{M}(\Theta)\mathbf{L}(\phi_1)\mathbf{F}_0, \tag{15}$$

where angles Θ, ϕ_1, and ϕ_2 can be expressed as functions of local zenith angles θ and θ' and azimuthal angles φ and φ' (see, e.g., Chandrasekhar[14]). The general expression of the phase matrix is then

$$\mathbf{P}(\theta, \theta'; \varphi, \varphi') = \mathbf{L}(\phi_2)\mathbf{M}(\Theta)\mathbf{L}(\phi_1). \tag{16}$$

However, in laboratory experiments, we may arrange a fixed coordinate system such that the polarizers in both the transmitter and receiver are in the same directions with respect to a fixed plane, say the optical table, so that $\mathbf{L}(\phi_1) = \mathbf{L}(\phi_2) = 1$, and $\mathbf{P}(\theta, \theta'; \varphi, \varphi') \equiv \mathbf{M}(\Theta)$. We may then simply employ Eq. (10) for the determination of the phase matrix. Furthermore, because each element of M_{ij} is normalized to unity, we may write symbolically that

$$\mathbf{M}(\Theta) \sim \frac{\mathbf{I}(\Theta)}{2\pi\int_0^{\pi}\mathbf{I}(\Theta)\sin\Theta\, d\Theta} \equiv \mathbf{I}'(\Theta). \tag{17}$$

This means that the phase matrix $\mathbf{M}(\Theta)$ can be determined solely from the measured Stokes parameters, without knowledge of C and $\mathbf{F}_0$.

III. DETERMINATION OF THE PHASE MATRIX

It is clear from Eqs. (10) and (17) that four measurements for the Stokes parameters of the scattered, partially polarized light are needed to determine the 16 elements of the phase matrix. The experimental determination of each set of Stokes parameters of the scattered light was described in Sec. I.

At time t_1, let an unpolarized incident flux density be $(F_{01}, 0, 0, 0)$. We use the subscript i in the Stokes parameters to refer to time t_i, where $i = 1, 2, 3, 4$. The four elements in the first row of the phase matrix can be first determined by

$$M_{11}=I_1', \quad M_{12}=Q_1', \quad M_{13}=U_1', \quad M_{14}=V_1' . \tag{18}$$

At time t_2, let a 100% linearly polarized light be incident, with the electric vector vibrating parallel to the plane of the optical table. In this case, the Stokes parameters of the incident light are $(F_{02}, Q_{02}, 0, 0)$. From the measured irradiances, the elements in the second row of the phase matrix are

$$M_{21}=I_2'-I_1', \quad M_{22}=Q_2'-Q_1', \quad M_{23}=U_2'-U_1', \quad M_{24}=V_2'-V_1'. \tag{19}$$

At time t_3, use a 100% linearly polarized light with the electric vector vibrating 45° to the plane of the optical table. Because $E_l=E_r$ and $\delta=0$, the Stokes parameters by definition are $(F_{03}, 0, U_{03}, 0)$. The third row of the phase-matrix elements may then be obtained from

$$M_{31}=I_3'-I_1', \quad M_{32}=Q_3'-Q_1', \quad M_{33}=U_3'-U_1', \quad M_{34}=V_3'-V_1' . \tag{20}$$

Finally, at time t_4, we employ the same linearly polarized light as at time t_3, but add a quarter-wave plate in front of the polarizer to produce a phase shift δ of $\pi/2$ between l and r components of the electric field. Thus, the Stokes parameters become $(F_{04}, 0, 0, V_{04})$. In a way similar to that used for Eq. (20), we derive the last row of the phase-matrix elements,

$$M_{41}=I_4'-I_1', \quad M_{42}=Q_4'-Q_1', \quad M_{43}=U_4'-U_1', \quad M_{44}=V_4'-V_1' . \tag{21}$$

The determination of the 16 phase-matrix elements of a scattering medium is thus completed.

IV. PHASE MATRIX OF A VOLUME OF ICE CRYSTALS

On the basis of an observation by Weickmann,[15] it is known that the ice particles in cirrus clouds are hexagonal columns whose lengths and radii are about 200 and 30 μm, respectively. The concentration generally varies from 0.1 to 1 particles per cm^3; crystals of nearly uniform size are usually observed. In a recent investigation, Heymsfield and Knollenberg[16] reported that cirrus generating cells are composed primarily of hexagonal columns having a mean length of ~600–1000 μm. Their concentration is found to be ~0.1–0.05 cm^{-3}. Generally, if no assumptions are made concerning the physical positions of the nonsymmetrical ice crystals in space, the phase matrix contains 16 independent parameters,

$$\begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{21} & M_{22} & M_{23} & M_{24} \\ M_{31} & M_{32} & M_{33} & M_{24} \\ M_{41} & M_{42} & M_{43} & M_{44} \end{bmatrix} . \tag{22}$$

Jayaweera and Mason[17] studied the behavior of freely falling cylinders in a viscous fluid. They found that if the ratio of diameter to length is less than unity, then cylinders fall with their long axes horizontal. Observations by Ono[18] also indicated that columnar crystals tend to fall with their major axes parallel to the ground. Plates fall with their major axes also horizontal. Hexagonal cylinders probably orient randomly in a horizontal plane, i.e., every possible position is equally probable in this plane. Under this condition, and if clouds contain cylinders, all of nearly equal size, then the law of reciprocity may be applied. We may reverse the directions of the incident and scattered polarized beams; the final results will be equal. Perrin[19] proved that the 16 phase-matrix elements must obey six conditions of symmetry or antisymmetry, and that they are reduced to 10 independent parameters. In an elegant way, Van de Hulst[13] also illustrated this by indicating that the scattering functions (S_3, S_4) are equivalent to $(-S_4, -S_3)$. The resulting phase matrix is given by

$$\begin{bmatrix} M_{11} & M_{12} & M_{13} & M_{14} \\ M_{12} & M_{22} & M_{23} & M_{24} \\ -M_{13} & -M_{23} & M_{33} & M_{34} \\ M_{14} & M_{24} & -M_{34} & M_{44} \end{bmatrix} . \tag{23}$$

It is clear from Eq. (23) that the ith row and ith column ($i \neq 3$) in the matrix are equivalent, except for a minus-sign difference in the third element.

Occasionally, however, cylinders may be oriented randomly in space owing, for example, to turbulence. If a sample of cylinders is indeed randomly oriented in space, then these cylinders are symmetrical with respect to the incident polarized beam, regardless of its direction. Consequently, the reference plane of the incident beam may be used as a plane of symmetry. Perrin[19] and Van de Hulst[13] showed that, under this condition, the number of independent phase-matrix elements is only eight. By use of this with the previous assumption, we have randomly oriented cylinders that have a plane of symmetry, these eight parameters may be further reduced to only six independent elements,

$$\begin{bmatrix} M_{11} & M_{12} & 0 & 0 \\ M_{12} & M_{22} & 0 & 0 \\ 0 & 0 & M_{33} & M_{34} \\ 0 & 0 & -M_{34} & M_{44} \end{bmatrix} . \tag{24}$$

Note that if a polarized beam is incident normal to a plane in which the cylinders of a sample are randomly oriented, then Eq. (23) should be reduced to Eq. (24), because the reference plane of the incident beam may be considered the plane of symmetry. The three forms in Eqs. (22), (23), and (24) are the possible phase matrices of a volume of ice crystals that are typical of those in cirrus clouds.

We illustrate in this paper that four convenient sets of radiance measurements can be employed to determine the four-by-four phase matrix for nonspherical ice crystals. In a controlled environment, such as a cold room, we may simulate the conditions for random orientation and preferred orientation in a horizontal plane and verify the zero and symmetrical elements in Eqs. (23) and (24).

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