MANIFESTATION OF LIGHT SCATTERING ANISOTROPY IN THE STRATOSPHERIC AEROSOL LAYERS

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Backscattering phase matrices (BPM) have been measured by a polarization lidar with controllable polarization of output laser radiation for measuring all the Stokes parameters at $\lambda = 532$ nm. The degree of orientation and the preferred orientation of particles are determined. To this end, the properties of BPM for the model of axisymmetric prolate particle (APP) ensemble are used. It has been suggested that scattering anisotropy of an aerosol layer, whose BPM is not described by the APP model, is caused by birefringence.

In our previous experiments[1] we have demonstrated the feasibility of determination of the preferred orientation of crystal cloud particles based on lidar measurements of the backscattering phase matrices (BPM's). In this paper we discuss some results of BPM measurements which provide reasons to suggest that in some cases polarized lidar measurements are capable to detect stratospheric crystal non–aqueous particles.

Identification of different types of particles is based on an analysis of the relations among the BPM elements. These elements, in their turn, are determined in terms of the elements S_{ij} of the amplitude conversion matrix (ACM) via the matrix equation[2]:

$$\mathbf{M} = \mathbf{U}(\mathbf{S} \times \mathbf{S}^{*})\mathbf{U}^{-1}, \tag{1}$$

where **M** is the 4×4 scattering matrix for the intensities (Müller's matrix), **S** is the 2×2 conversion matrix for the electromagnetic field amplitudes, and **U** is the 4×4 unitary matrix[1,2]. The symbol × denotes the Kronecker product of matrices.

The explicit expressions for the elements of the Mü ller matrix (in terms of the BPM elements) can be found, for instance, in Ref. 3 where it was also reported that the condition $S_{12} + S_{21} = 0$ is always valid for backscattering, and therefore the relations

$$M_{ij} = M_{ji}, \text{ if } i \neq 3 \text{ or } j \neq 3,$$

$$M_{ij} = -M_{ji}, \text{ if } i = 3 \text{ or } j = 3$$

are generally fulfilled for nondiagonal elements of the BPM.

In the analysis of the experimental BPM presented below it is advisable to present the form of the BPM for axisymmetric particles whose plane of mirror symmetry is perpendicular to the symmetry axis. The most widespread crystal ice particles in the form of needles and hexagonal columns belong to this type of axisymmetric prolate particles (APP). In addition axisymmetric plates also belong to this APP type.

When describing light scattering by particles of this type in a system of coordinate affixed to a particle in such a way that the axis of symmetry lies in the reference plane, all the nondiagonal elements of the BPM vanish (*xoz* plane is the reference plane with the *oz* axis in the direction of the wave vector of the incident electromagnetic wave). As a result, all the elements M_{ij} containing the factors S_{12} or S_{21} from the ACM vanish, and the BPM assumes the form

$$\mathbf{M}(0) = \begin{pmatrix} a & b & 0 & 0 \\ b & a & 0 & 0 \\ 0 & 0 & c & -d \\ 0 & 0 & d & c \end{pmatrix}, \tag{2}$$

where $a = (A_2 A_2^* + A_1 A_1^*) / 2$, $b = (A_2 A_2^* - A_1 A_1^*)/2$, $c = (A_2 A_1^* + A_1 A_2^*) / 2$, and $d = i(A_2 A_1^* - A_1 A_2^*)/2$.

Here and below we use the following designations for the ACM elements: $S_{11} = A_2$, $S_{22} = A_1$, $S_{13} = A_3$, and $S_{21} = A_4$. These designations were used in Ref. 3.

In the coordinate system rotated through the angle α about the direction of propagation the BPM can be obtained by transformation

$$\mathbf{M}(\alpha) = \mathbf{R}(\alpha)\mathbf{M}(0)\mathbf{R}(\alpha), \tag{3}$$

where $\mathbf{R}(\alpha)$ is the rotation operator. The explicit expression for this operator was given by us in Ref. 1.

In Ref. 4 BPM for polydisperse APP ensemble with preferred orientation being symmetrically distributed around the mode α_0 was given. This matrix can be written as follows:

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$$\mathbf{M} = \begin{pmatrix} \bar{a} & k_1\bar{b}\cos 2\alpha_0 & -k_1\bar{b}\sin 2\alpha_0 & 0 \\ k_1\bar{b}\cos 2\alpha_0 & \frac{\bar{a}-\bar{c}}{2} + k_2\frac{\bar{a}+\bar{c}}{2}\cos 4\alpha_0 & -k_2\frac{\bar{a}+\bar{c}}{2}\sin 4\alpha_0 & k_1\bar{d}\sin 2\alpha_0 \\ k_1\bar{b}\sin 2\alpha_0 & k_2\frac{\bar{a}+\bar{c}}{2}\sin 4\alpha_0 & \frac{\bar{c}-\bar{a}}{2} + k_2\frac{\bar{a}+\bar{c}}{2}\cos 4\alpha_0 & -k_1\bar{d}\cos 2\alpha_0 \\ 0 & k_1\bar{d}\sin 2\alpha_0 & k_1\bar{d}\cos 2\alpha_0 & \bar{c} \end{pmatrix}. \quad (4)$$

Here $\bar{a}$, $\bar{b}$, $\bar{c}$, and $\bar{d}$ represent the corresponding quantities from expression (2) averaged over polydisperse ensemble, k_1 and k_2 are factors depending on the orientation angle distribution function. For uniform distribution these factors vanish and matrix (4) becomes diagonal. When $\alpha_0 = 0$ or $\pi/2$, matrix (4) transforms into matrix (2).

Let us use the known properties of the BPM, including the above-stated ones, to interpret one of the previous results.

The vertical profile of a ratio of the total backscattering to the molecular backscattering measured in October of 1991 is shown on the left–hand side of Fig. 1. This profile obtained on July 11, 1991 when we first detected the layer of enhanced scattering at altitudes between 13 and 16 km, which was stably manifested in our subsequent measurements (on July 16, July 24, August 27, September 26, and September 30), is shown on the right–hand side of Fig. 1. The polarization characteristics of light backscattered by this layer were described by us earlier.[5] The normalized BPM of the layer is diagonal and absolute values of its elements are close to unity. This demonstrates the water droplet structure of particle ensemble.

Generally speaking, this behavior of the BPM can be observed in axisymmetric plate ensemble when the normals to these plates are oriented in the sounding direction. This assertion is almost evident since it is clear from symmetry considerations that the nondiagonal elements of the BPM for these particles are equal to zero. At the same time, from the general expression for the BPM of an ensemble containing n particles it follows that

$$M_{11} - M_{22} = \sum_{i=1}^{n} (A_3 A_3^*)_i. \quad (5)$$

The quadratic form on the right side of Eq. (5) will be equal to zero, if each term of this sum equals zero. Such is indeed the case in which all the normals to the particles are oriented strictly in the sounding direction. But natural flatter breaks the orientation and for the plates, whose normals lie neither in the reference plane nor in the perpendicular plane, the element A_3 will differ from zero.

Therefore from Eq. (5) it follows that

$$M_{11} > M_{22} \quad (6)$$

and matrix of absolute values $|M_{ii}|$ will differ from the unit matrix.

A less evident result was obtained in Ref. 6. It was demonstrated that radiation scattered by cylindrical particles of 10 μm radius, with lengths being uniformly distributed in the range 30 – 50 μm and cylinder axes lying in the horizontal plane and being oriented about a certain chosen horizontal direction, has the same polarization as radiation scattered by spheres. On account of Eq. (5) and expression for the BPM of cylindrical particles derived in Ref. 6, it may be concluded that for objects under consideration the condition

$$A_1 = -A_2 \quad (7)$$

is valid at least approximately for arbitrary angle of rotation about the sounding direction.

Probably, this condition violates for cylinder axis oriented at an angle with respect to the horizontal plane, because the calculations performed in Ref. 6 showed qualitatively different behavior of the Stokes parameters of an ensemble of particles of the same size whose axes are tilted at an angle of 45°. It is natural to assume that the tilt angles in real ensembles of prolate particles are distributed around the horizontal plane and relation (6) is also valid. Based on the aforesaid, the water–droplet structure of the above–discussed layer may be argued with reasonable confidence. The period of observation of this layer suggests its volcanic origin (Pinatubo volcano).

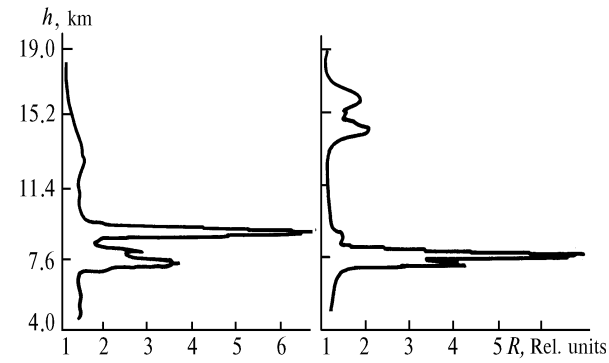

FIG. 1

Now we analyze the aerosol situation shown in Fig. 1, on the left–hand side. One can see two sharply pronounced layers whose maxima are at altitudes of 7.6 and 9.3 km and one weakly pronounced layer ($R = 1.5$) within the 10 – 16 km altitude range. The normalized BPM of the latter is diagonal and has the following values of its elements: $m_{11} = 1$, $m_{22} = 0.97$, and $m_{33} = m_{44} = -0.97$. The estimated absolute value of the error in measuring these elements was ±0.04. This form of the matrix and the altitude of the layer suggest that this is the same water – droplet layer of possible volcanic origin. The normalized BPM's for two other layers have the following values:

$$m(h = 7.6\ \text{km}) = \begin{pmatrix} 1 & 0.39 & 0 & 0 \\ 0.39 & 0.81 & 0 & 0 \\ 0 & 0 & -0.60 & -0.31 \\ 0 & 0 & 0.31 & -0.38 \end{pmatrix}, \quad (8)$$

$$m(h = 9.3\ \text{km}) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & 0.62 & 0 & 0.125 \\ 0 & 0 & -0.55 & -0.32 \\ 0 & 0.125 & 0.32 & -0.15 \end{pmatrix}. \quad (9)$$

The matrix of the 7.6 km layer can be described by APP model (2) and (4). It follows from Eq. (4) that

$$\alpha_0 = \frac{1}{2}\,\text{arccot}\,\frac{m_{21}}{m_{31}} = \frac{1}{2}\,\text{arccot}\,\frac{m_{43}}{m_{42}}, \quad (10)$$

from which we obtain two principal values $\alpha_0 = 0$ and $\alpha_0 = \pi/2$. From the results reported in Refs. 4 and 6 it follows that for cylinders the quantities b and d entering into equations for the elements of BPM (2) and (4) are negative when the cylinder axis lies in the reference plane and positive when the cylinder axis is perpendicular to this plane. Matrix (8) corresponds to the latter case,therefore $\alpha_0 = \pi/2$.

Generalizing this results to other APP, we may conclude that the axis of the particles in the layer being observed had preferred orientation in the direction perpendicular to the x axis of the lidar polarization basis, because in Eq. (8) $b > 0$ and $d > 0$. This approximately corresponds to the east–west direction. In addition, as was shown in Ref. 4, it is possible to obtain the parameter k_2 that characterizes the degree of particle orientation changing from absence of any preferred orientation ($k_2 = 0$) to the complete orientation ($k_2 = 1$).

From the relation

$$k_2 = \frac{m_{22} + m_{33}}{(m_{11} + m_{44})\cos 4\alpha_0} \quad (11)$$

we obtain $k_2 = 0.33$ for matrix (8).

Now we analyze matrix (9). One can see from Eq. (4) that if m_{12} and m_{13} equal zero simultaneously, they have zero values for any location of the reference plane (lidar polarization basis). This is a general property of the BPM, it is inherent not only to the APP model. Within the scope of the APP model, the zero values of m_{12} and m_{13} elements may result from $k_1 = 0$ or/and $b = 0$. The zero value of the parameter k_1 means chaotic orientation and this leads, within the scope of the APP model, to the diagonal BPM. However, nonzero values of m_{24} and m_{34} are indicative of the preferred orientation with $\alpha_0 = 10.5°$ and $k_2 = 0.11$. The rotation of lidar polarization basis about the sounding direction through this angle is to result in zero values of m_{24} and m_{42} elements, whereas the absolute values of m_{34} and m_{43} elements become maximum.

It follows from the above–discussed that within the scope of the APP model, for interpretation of matrix (9) we should assume $\overline{b} = 0$. The quantity $\overline{b}$ represents the sum

$$\overline{b} = \frac{1}{2}\sum_{i=1}^{n}(A_2A_2^* - A_1A_1^*)_i, \quad (12)$$

where n is the particle number density, A_1 and A_2 are the elements of the ACM written down for each particle in the coordinate system affixed to it in such a way that $A_3^i = A_4^i = 0$. The sum (12) may be equal to zero in particular when every term of the sum equals zero. In this connection condition (7) for large cylindrical particles comes to mind. But in this case the BPM must be diagonal with absolute values of the elements being close to unity. It is inconsistent with the measured matrix. As we have mentioned above, condition (7) is also valid for axisymmetric plates whose normals are oriented in the sounding direction. It can be shown that in the presence of random flatter the BPM remains diagonal and preferred tilt of the axes to the horizontal plane results in nonzero values of m_{12} and m_{13}. However, this is beyond the scope of our paper. Having rejected condition (7) and symmetric plate hypothesis, the sum (12) has to be regarded within the scope of this model as being equal to zero, on the average. Such an APP ensemble seems rather strange, since it should include both the particles for which $|A_1| < |A_2|$ in the coordinate system affixed to the particles and particles for which $|A_1| > |A_2|$. At the same time if we accept that $\alpha_0 = 10.5°$ is associated with the orientation of particle axes, the condition $|A_1| < |A_2|$ would be typically satisfied.

Since the above described contradictions in interpretation of the experimental BPM within the scope of the APP or symmetrical plate model exist, we risk to suppose that the preferred orientation determined by the relation between the elements m_{42} and m_{43} is primarily connected with optical anisotropy of particles rather than their geometry. This assumption is confirmed by the possible relation between genesis of the layer centered at an altitude of 9.3 km and the above layer of probably volcanic origin, as can be seen from Fig. 1.

Long–term observations of this volcanic layer in the form of a water–droplet layer indicate that it contains sulfuric acid and ammonia which can produce ammonium sulfate. It is well known that ammonium sulfate is a ferroelectric material at a temperature below –49°C, while acid ammonium sulfate already becomes ferroelectric at a temperature below –3°C. If we assume that the layer centered at an altitude of 9.3 km consists of the ammonium sulfate particles, the specific direction may be associated with the preferred orientation of the particle dipole moment in the Earth's electric field, for some reason exhibiting potential gradient in this direction. Then the zero values of m_{12} and m_{13} elements follow from particle isometry, whereas the nonzero values of m_{42} and m_{43} elements can be explained by the phase shift between the components of the electric field due to birefringence.

Such an interpretation of the experimental data opens the possibility of lidar investigation into the processes of transformation of gaseous pollution to the aerosol phase. But we are not sure that this interpretation is unique. In particular, in the review of our paper it was pointed out that there is another possibility to interpret matrix (9) within the scope of the APP model if we assume that the values of $|m_{21}|$ and $|m_{31}|$ differ from zero and this difference is within the limits of experimental error. We hope that further investigation will clarify this question.

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