# Symmetry of backscattering phase matrices as related to orientation of non-spherical aerosol particles

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It is shown that if there are forces in the atmosphere that may cause orientation, at least in part, of an ensemble of non-spherical aerosol particles, the direction of these forces action can be found from measurements of the backscattering phase matrices (BSPM). Moreover, some parameter  $\chi$  is being determined that characterize the degree of the orienting action of the force field on the aerosol ensemble.

Lidar measurements of the backscattering phase matrices (BSPM) have shown that deviations from random orientation of particles are often observed in crystal clouds. 1 Moreover, it is not only the action of aerodynamic forces on the falling down particles that may favor the orientation of particles, but also the forces of other origin can cause a preferred orientation about some azimuth direction. Possible reasons for that may be the wind shears and electric fields. Obviously, the direction of the preferred orientation is related to the direction of the action of these forces. The approach developed by the authors of Ref. 2 to determination of the parameters of orientation of a polydisperse ensemble of some axially symmetric particles was used in Ref. 1 determination of the direction of preferred orientation at interpretation of the experimentally measured BSPMs. It is not only the direction of preferred orientation but also the Mises distribution parameter,<sup>3</sup> which is the measure of grouping of the particle axes about the mode of the distribution, i.e., around the direction of preferred orientation, was determined for ensembles of such particles.

However, the representation of actual crystal clouds using the aforementioned ensembles is, of course, idealized. Numerous papers devoted microphysical investigations reveal a wide variety of the particle shapes, including asymmetric ones, for which it is difficult to construct a criterion of the preferred orientation, because no symmetry axes or planes can be isolated in this case. Therefore, it is expedient to consider a more general formulation of the question on what could be the reason for the BSPM invariance relative to rotation of the coordinate system, or to the rotation of a cloud as a whole, if that would suit understanding better.

The invariance is considered here as follows. Let M be the BSPM determined at a certain orientation of the coordinate system rigidly tied to the experimental setup, then, if turning the latter by an angle  $\Phi$  around the direction of the wave vector of the incident radiation (z-axis of the xoz coordinate system), the matrix M is transformed according to the following law

$$\mathbf{M}' = \Re(\Phi) \ \mathbf{M}\Re(\Phi), \tag{1}$$

where  $\Re(\Phi)$  is the operator of rotation

$$\Re(\Phi) = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\Phi & \sin 2\Phi & 0 \\ 0 & -\sin 2\Phi & \cos 2\Phi & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}.$$
 (2)

The requirement of invariance is expressed by the equality

$$\mathbf{M'} = \mathbf{M}.\tag{3}$$

Obviously, this requirement should hold for the BSPM of an isotropic ensemble, at least relative to the rotation around the z-axis.

Direct calculations of the matrix  $\mathbf{M'}$  at arbitrary elements  $M_{ii}$  of the initial matrix and comparison of  $M'_{ij}$  with  $M_{ij}$  show that the condition (3) is fulfilled only for the matrix of a certain form, which is as follows:

$$\mathbf{M} = \begin{pmatrix} M_{11} & 0 & 0 & M_{14} \\ 0 & M_{22} & 0 & 0 \\ 0 & 0 & M_{33} & 0 \\ M_{41} & 0 & 0 & M_{44} \end{pmatrix} \tag{4}$$

under the additional condition that

$$M_{22} = -M_{33}. (5)$$

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Besides,  $M_{14} = M_{41}$  is always fulfilled for the BSPM of the form (4), and, as a particular case, these elements can be equal to zero.

Matrices of the form (4) are determined<sup>4</sup> as the BSPM of ensembles of asymmetric particles of the same kind either chaotically oriented in space, or oriented so that the rotation symmetry relative to the wave vector direction is kept. The elements  $M_{14}$  and  $M_{41}$  vanish if the particles have such symmetry that they coincide with their mirror reflections, or asymmetric but the ensemble of such particles has mirror symmetry relative to any plane containing the *z*-axis.

Obviously, the requirement that particles have to be of the same kind is not obligatory. Owing to additivity of the scattering phase matrices, an ensemble composed of sub-ensembles of particles of different kinds, but so that each of them has the BSPM of the form (4), will have the total BSPM of the same form or its particular case at  $M_{14} = M_{41} = 0$ .

If the optical axis of a lidar is directed toward the zenith and there are no another orienting factors except for gravity, it is reasonable to expect the BSPM of the form (4). This is true because the particles are either truly randomly oriented in space or have some preferred orientation at a zenith angles  $\theta,$  while, at the same time, being randomly oriented over the azimuth  $\Phi.$  Let us call this state 2D random orientation. Superposition of 2D and 3D randomly oriented sub-ensembles is also possible. As it will be clear from the below, the presence of a sub-ensemble with 2D random orientation can be revealed by measuring BSPM at a slant position of the lidar optical axis.

Let us emphasize that the matrix (4) was obtained without any suppositions about the particle shape. It is the invariant of the transformation (1) and means the absence of any preferred azimuth direction. So, the BSPM of an isotropic, on the average, ensemble relative to the z-axis ought to have the form (4), and one can interpret any deviation from this form to be caused by the presence of asymmetry or a finite-order symmetry relative to the rotation about the z-axis.

Naturally, one can assume the violation of such symmetry in crystal clouds to be caused by the action of some factor that orients the particles, with the vector of the action not coincident with the vertical direction. As a result, one can isolate a plane  $P_0$  that contains both of these directions.

It is reasonable to assume that the vector field of orienting forces of non-gravitational origin is homogeneous, at least within the limits of the volume illuminated by a laser beam. Undoubtedly, the gravity is such a field. Then, if the orienting effect of these forces has led to an ordering of particles orientation along some direction more probable, this probability does not depend on which side from the plane  $P_0$  a particle is situated. Besides, the mirror reflected position of the initial particle relative to  $P_0$  has the same probability, because it is impossible in the field of uniform forces to point out a feature that would make one of these positions preferred. For example, the number of particles contributing to the lidar backscatter from crystal clouds at a time is about 104-10<sup>6</sup>. Therefore, one may expect that the mirror symmetry relative to  $P_0$  occurs, on the average, in such an ensemble and fluctuating deviations from it are insignificant. One can reach the coincidence of the reference plane xoz with the symmetry plane  $P_0$  by turning the coordinate system relative to the z-axis. The BSPM in this particularly chosen system of coordinates has a certain symmetry that follows from the symmetry of amplitude scattering phase matrices. 4,5 The frameworks of the paper do not allow to expound in detail the proofs, so let us only present the BSMP in this coordinate system:

$$\mathbf{M}_{0} = \begin{pmatrix} M_{11}^{0} & M_{12}^{0} & 0 & 0 \\ M_{21}^{0} & M_{22}^{0} & 0 & 0 \\ 0 & 0 & M_{33}^{0} & M_{34}^{0} \\ 0 & 0 & M_{43}^{0} & M_{44}^{0} \end{pmatrix}.$$
(6)

The zero superscript of the elements means that the matrix is determined at coincidence of the reference plane and plane of mirror symmetry of the ensemble of particles. The following condition is fulfilled as for any BSMP:

$$M_{12}^0 = M_{21}^0$$
 and  $M_{34}^0 = -M_{43}^0$ .

The condition (5) may not hold for this matrix. But, due to the known general property of BSPMs, according to which

$$M_{11} - M_{22} + M_{33} - M_{44} = 0, (7)$$

one can write

$$M_{11}^0 - M_{44}^0 = M_{22}^0 - M_{33}^0. (8)$$

Since the elements  $M_{11}$  and  $M_{44}$  of any BSPM are invariant at rotation, the right-hand side of the equality (8) is also invariant.

For the reasons, which will be clear from the below discussion, let us write the following identities:

$$\begin{split} M_{22}^0 &= \frac{M_{22}^0 - M_{33}^0}{2} + \frac{M_{22}^0 + M_{33}^0}{2} = E + F \ , \\ M_{33}^0 &= -\frac{M_{22}^0 - M_{33}^0}{2} + \frac{M_{22}^0 + M_{33}^0}{2} = -E + F \ . \end{split} \tag{9}$$

Let us also introduce the following notations:

$$M_{11}^0 = A, \quad M_{12}^0 = M_{21}^0 = B,$$
  
 $M_{34}^0 = D = -M_{43}^0, \quad M_{44}^0 = C.$  (10)

Let the BSPM which has the form (6) in a selected coordinate system related to the plane  $P_0$  be measured now in the coordinate system turned relative to the plane  $P_0$  by the angle  $-\Phi$  around the wave vector, i.e., clockwise if looking towards to the incoming scattered radiation. The elements of the BSPM in this new coordinate system are expressed through the elements of the matrix (6) by use of the transformation

$$\mathbf{M}_{-\Phi} = \Re(-\Phi) \ \mathbf{M}_0 \Re(-\Phi). \tag{11}$$

Formally one can present the matrix  $\mathbf{M}_0$  as a sum of diagonal matrix  $\mathbf{M}_0'$  with the elements A, E, -E and C (see notations (10)) and the matrix

$$\mathbf{M}_{0}'' = \begin{pmatrix} 0 & B & 0 & 0 \\ B & F & 0 & 0 \\ 0 & 0 & F & D \\ 0 & 0 & -D & 0 \end{pmatrix}, \tag{12}$$

and write (11) in the following form:

$$\mathbf{M}_{-\Phi} = \Re(-\Phi) \left(\mathbf{M}_0' + \mathbf{M}_0''\right) \Re(-\Phi).$$
 (13)

But  $\mathbf{M}_0'$  is composed from invariant of rotation and does not change at transformation (11), and (12) is transformed to the matrix

$$\mathbf{M}''_{-\Phi} = \begin{pmatrix} 0 & B\cos 2\Phi & -B\sin 2\Phi & 0 \\ B\cos 2\Phi & F\cos 4\Phi & -F\sin 4\Phi & -D\sin 2\Phi \\ B\sin 2\Phi & F\sin 4\Phi & F\cos 4\Phi & D\cos 2\Phi \\ 0 & -D\sin 2\Phi & -D\cos 2\Phi & 0 \end{pmatrix}$$
(14)

The matrix (14) is obtained by turning the coordinate system by the angle  $-\Phi$  relative to the plane  $P_0$ , the position of which is assumed to be preset. On the contrary, in experiments, the position of the plane xoz related to the lidar is known, and one needs to find the position of the plane  $P_0$ . Obviously, this problem is reduced to determination of the angle  $\Phi$  such that

$$\Re(\Phi) \ \mathbf{M}''_{-\Phi} \ \Re(\Phi) = \mathbf{M}''_{0}. \tag{15}$$

It can be ready seen that  $\Phi$  is determined by the ratio of the elements of matrix  $\mathbf{M}''_{-\Phi}$ , for example,

$$\mathbf{M}_{31}''/\mathbf{M}_{21}'' = \tan 2\Phi.$$
 (16)

It is easy to see that the direction  $\Phi$  is determined independently of the presence of particles, which do not undergo the orienting effect due to any cause, and compose the randomly oriented sub-ensemble.

Let us denote the BSPM of such a sub-ensemble by  $M_{\rm x}$ . It is the matrix of the form (4).

Let us represent the matrix of the whole ensemble as a sum of the matrices  $\mathbf{M}_x$ ,  $\mathbf{M}_0'$  and  $\mathbf{M}_0''$  and write the transformation

$$\mathbf{M}_{-\Phi} = \Re(-\Phi) (\mathbf{M}_{x} + \mathbf{M}'_{0} + \mathbf{M}''_{0}) \Re(-\Phi).$$

But the matrices  $\mathbf{M}_x$  and  $\mathbf{M}_0'$  are invariant relative to rotation, and the problem is reduced to transformation of the matrix  $\mathbf{M}_0''$  only related to subensemble of oriented particles. As before, the determination of  $\Phi$  is done by use of Eq. (16).

Let the experimentally determined matrix  $\mathbf{M}$  with the elements  $\mathbf{M}_{ij}$  correspond to  $\mathbf{M}_{-\Phi}$ . It can contain 10 different parameters. It follows from the property of amplitude scattering phase matrix

$$A_{12} + A_{21} = 0,$$

the consequence of which are the relationships for the off-diagonal elements of the BSPM

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

It is easy to see that these relationships hold for the matrix (14). The elements  $M_{11} = A$ ,  $M_{44} = C$ ,  $M_{14} = M_{41} = H$  are invariant of rotation. Besides, one more invariant is determined

$$E = (M_{11} - M_{44})/2 = (M_{22} - M_{33})/2.$$

These parameters are also related to BSPM of the whole ensemble and determine its invariant component. The parameters of the non-invariant component  $\mathbf{M}_0''$ : B, D, F will be found after determination of  $\Phi$  by Eq. (16). As was shown above that the transformation (15) reduces the non-invariant component of the BSPM to the form (12). Applying this transformation to the elements of the experimental BSPM  $M_{ij}$ , we obtain

$$B = M_{12} \cos 2\Phi - M_{13} \sin 2\Phi;$$

$$D = M_{34} \cos 2\Phi - M_{24} \sin 2\Phi;$$

$$F = \cos 4\Phi (M_{22} + M_{33})/2 - M_{23} \sin 4\Phi.$$
(17)

Now the experimental BSPM can be presented using the parameters, which do not depend on the coordinate system:

$$\mathbf{M} = \begin{pmatrix} A & B & 0 & H \\ B & E + F & 0 & 0 \\ 0 & 0 & -E + F & D \\ H & 0 & -D & C \end{pmatrix}. \tag{18}$$

Such a representation is very convenient for making a comparison among the matrices obtained in different experiments.

In addition to the properties inherent to any BSPM, the only supposition used for obtaining the matrix (18) was that about the presence of a subensemble of particles, for which the only plane  $P_0$ exists, relative to which it has the mirror symmetry. It was assumed that it is the vertical plane, and the wave vector of sounding radiation is directed toward zenith, because in this case the reference plane already exists, which contains the vectors of the gravity force and of another orienting factor. If the aforementioned conditions have been fulfilled, the experimental BSPM should be reducible to the form (18). The experimental BSPM published earlier in Ref. 1, and the matrix transformed by the rule (15) at the experimental value  $\Phi = 17.5^{\circ}$  are written below. The matrices are normalized to the element  $M_{11}$  so that  $m_{ij} = M_{ij}/M_{11}$ ;  $b = B/M_{11}$  and so on:

$$\mathbf{m}_{\rm exp} = \begin{pmatrix} 1 & -0.56 & 0.38 & -0.03 \\ -0.56 & 0.37 & -0.21 & 0.20 \\ -0.38 & 0.21 & -0.10 & -0.27 \\ -0.03 & 0.20 & 0.27 & 0.53 \end{pmatrix},$$
 
$$\mathbf{m}_{\rm r} = \begin{pmatrix} 1 & -0.66 & 0.01 & -0.03 \\ -0.66 & 0.45 & -0.06 & 0.01 \\ -0.01 & 0.06 & -0.01 & -0.34 \\ -0.03 & 0.01 & 0.34 & 0.53 \end{pmatrix}.$$

Elements of the anti-diagonal blocks of the reduced matrix deviate from zero values within the

limits of possible experimental errors, for which the estimate of standard deviation is  $\sigma = \pm 0.04$ . That means that the measured BSPM can be reduced to the form (18) with the normalized parameters  $a \equiv 1$ ; b = -0.66; c = 0.53; d = -0.34;  $h \approx 0$ ; e = 0.22; f = 0.23. Knowledge of the aforementioned parameters is important for interpretation of the BSMP. For example, in this case large absolute value of the parameter b together with also large f value allows us to suppose that the ensemble consists of strongly elongated and significantly oriented particles, and that there are little of the asymmetric randomly oriented particles if any, because the parameter h is close to zero, and so on. But, it is an issue for separate consideration, here let us consider only the parameter f, which is the characteristic of a disturbance of the axial symmetry of the ensemble of particles. According to expression (17) it is determined from the elements of the experimental BSPM as:

$$f = \cos 4\Phi \left( m_{22}^{\text{exp}} + m_{33}^{\text{exp}} \right) / 2 - m_{23}^{\text{exp}} \sin 4\Phi,$$
 (19)

and from the elements of the reduced matrix as:

$$f = (m_{22}^{\rm r} + m_{33}^{\rm r})/2 \tag{20}$$

[see definition (9)].

In Ref. 2 the parameter  $\chi$  was introduces, which is written in the notations accepted here as

$$\chi = (m_{22}^{\rm r} + m_{33}^{\rm r})/(1+c) = 2f/(1+c), \quad (21)$$

i.e., the parameter f is normalized to the invariant of the BSPM (a + c), where  $a \equiv 1$ .

The parameter  $\chi$  in the cited paper has the meaning of the ratio  $I_2(k)/I_0(k)=i_2(k)$ , where  $I_2(k)$  and  $I_0(k)$  are the modified Bessel functions of the first kind and of second and first order, respectively; k is the parameter of Mises distribution,  $i_2 \to 1$  at  $k \to \infty$ . In fact, already at k=10 the function  $i_2$  is close to 1, and that means that the axes of almost all axially symmetric particles are oriented along a preferred direction.

One can show, but we will not do this here, that the parameter  $\chi$  tends to 1 in increasing the degree of orientation of the axially symmetric particles, independently of the form of the angular distribution function of orientations. It is caused by the fact that if the axis of particle symmetry coincides with the orientation plane, then

$$A_{12} = A_{21} = 0.$$

It is not fulfilled for the asymmetric particles, so  $\chi < 1$ . In the absence of orientation, i.e., for the ensemble with axial symmetry relative to the wave vector direction, its BSPM has the form (4), and f, together with  $\chi$ , equals to zero. Hence, the domain of  $\chi$  variation is defined as  $0 \le \chi \le 1$ .

The parameter  $\chi$  for mixed ensembles can have no that direct relation to the degree of particles' orientation as in the case of a homogeneous ensemble considered in Ref. 2. The matter is that the parameter fis determined by the state of only sub-ensemble of oriented particles, but the denominator (1 + c) is related to the entire ensemble, and it is impossible to isolate the fraction of this invariant related to the oriented sub-ensemble. So the presence of small number of strongly oriented particles can be masked by the presence of a large number of the non-oriented ones. Hence, one can consider the parameter  $\chi$  rather than a qualitative characteristic showing the degree of the effect of orienting factor on the ensemble of particles as a whole. The discussion of the relation of the BSPM symmetry to the orientation of particles will be incomplete if we do not mention the problem on the ambiguity of Eq. (16). It is easy to show that two values of  $\Phi$  make the expression (16) hold:

$$\Phi_{1,2} = \arctan\left(-\frac{M_{21}}{M_{31}} \pm \sqrt{\left(\frac{M_{21}}{M_{31}}\right)^2 + 1}\right). (22)$$

The directions  $\Phi_1$  and  $\Phi_2$  are mutually orthogonal, and this reflects the fact that there is one more position of the reference plane different from the considered one  $P_0$ , at which the BSPM can be reduced to the form (18). This follows from the symmetry property of the amplitude scattering phase matrix. If turning the coordinate system by 90°, the particles which were mirror reflection of each other relative to the reference plane, become such relative to the bisector plane.<sup>4</sup> It has the same consequences for the BSPM as the mirror symmetry relative to the reference plane. But the signs of the elements  $M_{12}$  and  $M_{21}$  change, and the signs of the elements  $M_{34}$  and  $M_{43}$  invert. This is easy to show by substituting the angle  $\Phi$  of 0° and 90° to matrix (14).

To unambiguously select the angle  $\Phi$ , one needs to consider two cases (at coincidence of the reference plane xoz with the plane of symmetry of the ensemble of particles):

a) 
$$M_{12} = M_{21} = B > 0$$
; b)  $M_{12} = M_{21} = B < 0$ .

The following rules are fulfilled: a) B > 0.

a) D > 0.

b) B < 0.

If 
$$M_{21}$$
 and  $M_{31} > 0$ , then  $-\pi/2 < \Phi < -\pi/4$   
 $M_{21}$  and  $M_{31} < 0$ ,  $0 < \Phi < \pi/4$   
 $M_{21} > 0$ ,  $M_{31} < 0$ ,  $\pi/4 < \Phi < \pi/2$   
 $M_{21} < 0$ ,  $M_{31} > 0$ ,  $-\pi/4 < \Phi < 0$ .

Choosing between the alternatives "a" or "b" requires *a priori* information. This problem was discussed in Ref. 2, where it was shown that the

alternative "a" is realized at orientation of the elongated axially symmetric particles across the reference plane, and, on the contrary, the alternative "b" is realized at grouping of the particle axes near the reference plane.

Let us show that in some cases this problem can be solved experimentally. Let the ensemble contain a sub-ensemble with 2D chaotic orientation caused by gravitational orientation. Such a sub-ensemble has axial symmetry relative to the vertical direction, and its BSPM at sounding along the zenith direction has the form (4). Let us incline the optical axis of the lidar by the angle  $\theta$  in an arbitrary azimuth direction. Then the axial symmetry of the ensemble relative to the optical axis of the lidar is broken, and there appears a plane that contains the optical axis and the vertical. Superposing the plane xoz of the measurement basis of the lidar with this plane, we obtain a BSPM of the form (6) with possible non-zero elements  $M_{14}$  and  $M_{41}$ . This BSPM gives the answer to the question on the sign of  $B = M_{12}$ , because the position of the symmetry plane of the ensemble is known.

Let us summarize the above-stated material. Analysis has shown that the direction of action of the factor that orders the positions of particles relative to some vertical plane can be determined by means of lidar measurements of the BSPM independently of the shape of particles that undergo the orientation effect and of the presence of particles do not undergo such an impact. The parameter  $\chi$  is determined, which characterizes the strength of the orienting factor action on the ensemble of particles.

If there are no orienting factors apart from gravity, only the diagonal elements and, possibly, the elements  $M_{14}$  and  $M_{41}$  of the BSPM measured with a lidar directed toward zenith will differ from zero. Then  $M_{22} = -M_{33}$  equality should be fulfilled. If a subensemble with 2D random orientation has been formed under the effect of gravitational sedimentation, this state can be revealed by sounding along a slant direction. Such a state can be also revealed at sounding along zenith from the absolute values of the elements  $M_{22}$  and  $M_{33}$ . But, it requires knowledge of the microphysics of particles and its relation to the BSPM elements. These data can be obtained, for example, by means of calculating the BSPM elements for different models of aerosol ensembles. 6 However, these problems were not discussed here.

In this paper we considered some effects that result from the general properties of the BSPM symmetry and symmetry of aerosol ensembles at only minimal restrictions on the properties of the comprising particles.

It was supposed that particles are non-spherical and, at least, a fraction of them can take some preferred orientation under the effect of physical forces in the atmosphere. As to the assumptions concerning the symmetry of an ensemble of particles, the assumption of axial symmetry is quite an obvious one in the case of the gravitation-caused orientation only.

The assumption of the homogeneity of the field of forces forming the orientation of particles along a horizontal direction and, hence, of the presence of mirror symmetry of the ensemble sounded relative to vertical plane is not obvious, but it seems to be quite reasonable. The criterion of its fulfillment can be reducibility to the canonical form (18) of the BSPM obtained in sounding along the zenith.

The BSPM obtained at slant sounding can be irreducible to this form. If the effect of nongravitational orienting factor is significantly stronger than the gravitational one, so that one can ignore the latter, the formation is possible of an ensemble with axial symmetry about the direction of the effect of nongravitational factor. In this case, inclining the lidar at any angle, one can found the reference plane containing the direction of sounding and that of the action of the orienting factor. Then the reducibility of the matrix to the form (18) remains. If the joint effect of gravitational and another factor produces a set of parallel vertical planes at a certain azimuth, relative to which the ensemble of particles possesses mirror symmetry, the reducibility to the form (18) remains only at the optical axis of the lidar lying in the plane from this set and containing the location point of the lidar. Otherwise, the BSPM can contain 10 different parameters and becomes irreducible to a simpler form. But this variant is not interesting, because sounding along the zenith is sufficient for determining the direction of action of the orienting forces and the parameter  $\chi$ , and the necessity of inclination of the optical axis of the lidar can arise in the above-stated case of the axial symmetry of the ensemble of particles relative to the vertical direction. The inclination toward any azimuth direction makes up the plane of symmetry.

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