Theory and experimental results on laser sensing of oriented crystal particles in clouds

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Theoretical and methodological aspects of laser sensing of aerosol media with anisotropic distribution of non-spherical particles over angles of spatial orientation are discussed. Parameters characterizing the particle orientation are determined through elements of the backscattering phase matrix. The results of studying particle orientation in crystal clouds obtained from lidar measurements of the backscattering matrix are presented.

Introduction

Crystal clouds of the upper layer permanently cover up to one third of the Earth's surface and noticeably affect the Earth's climate. The arguments in favor of the study of even thinnest cirrus clouds can be found in Ref. 1, where insufficient level of knowledge of their radiative properties is noted as well. The anisotropy of cirrus clouds caused by such their peculiarity as spatial orientation of particles is mentioned among the problems that are still to be addressed. Particles have different size along different axes, and hence, are subject to orienting effect of aerodynamic forces stimulating them to occupy the positions, at which the aerodynamic resistance is maximum. The effect of this factor was observed in the experiment.[2] It begins to be observable as particles reach the size of about 40 μm and more, that, in general, is characteristic of crystal clouds. Due to orientation of particles, the coefficients of directed scattering are dependent on the direction of incidence and polarization state of radiation incident on the cloud layer.

The proofs of the presence of orientation of particles in crystal clouds were obtained for the first time in observations of such natural phenomena as parhelia, lower sun, arcs, sun pillars, crosses, etc., united by the general term halo. A lot of papers are devoted to explanation of different shapes of halo, the review of which is presented, for example, in Ref. 3. The majority of halos are explained by the phenomena of refraction, reflection, and diffraction of light on crystals of hexagonal shape, and some of them, for example, rings of unusual radii, different from the rings of big and small halos, – on crystals of more complicated shapes. It is essential that only ring aureoles can be explained by the effect of refraction from chaotically oriented crystals. All other shapes of halo are explained supposing that the significant fraction of particles is oriented to some extent.

Until recently, observations of halo have been the only source of information about orientation of particles in clouds. But the capabilities of this method are limited. First, observations are realizable only in the daylight time at certain elevation angles of the Sun and under certain meteorological conditions. Second, only particles of regular hexagonal shapes take part in the formation of a halo, and the residual variety of particles, which also take part in orientation process, form the diffuse, but not obligatory isotropic background. Anisotropy of scattering by oriented particles of irregular shapes can not be revealed from observations of halo.

Then, almost all of the halos can be explained supposing orientation at sedimentation due to gravity. There are only very few indications[4] of the orientation of particles by wind fluxes made on the basis of observations of the tilt of sun pillars, which is quite rare phenomenon. The problem on how essential is the role of the azimuth orientation, when the particles of a characteristic size can take some preferred position relative to the direction of the mean wind velocity, was not solved. And, finally, observations of halo do not provide for obtaining quantitative characteristics of orientation of the entire ensemble of cloud particles.

The significant step in the study of the problem of orientation of particles of crystal clouds was achieved thanks to the development of the optical-radar method for investigations of anisotropic aerosol media by studying the polarization of scattered light.[5] The method is based on remote measurement of the backscattering phase matrices (BSPM) by means of optical radar. In this paper, which is a review, the main theoretical principles of the method are stated, as well as the most important results obtained using it in studying crystal clouds.

Correlation of the BSPM symmetry properties with the orientation of particles

Let us define the terms of azimuth and polar orientations of particles in aerosol ensembles. The

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azimuth-oriented ensemble in the coordinate system of the lidar $\mathbf{e}_z = \mathbf{e}_x \times \mathbf{e}_y$ means such its state, at which the characteristic sizes of particles, for definiteness, their maximum diameters, are grouped predominantly near a selected plane containing the direction of sounding $\mathbf{e}_z$. The polar-oriented ensemble means the state, at which the characteristic size of particles occupies some predominant positions relative to the plane perpendicular to the direction of sounding. It is the horizontal plane when sounding along zenith or nadir.

In the absence of azimuth orientation the ensemble of particles has rotation symmetry in the meaning defined in Ref. 6. The form of BSPM does not change in the presence of this orientation, if the coordinate system in which it was defined, has been rotated around the direction $\mathbf{e}_z$. The form of BSPM is completely determined by three invariants of rotation, which are the elements of the matrix M_{11}, M_{44}, $M_{14} = M_{41}$. Then

$$M_{22} = (M_{11} - M_{44})/2, \; M_{33} = -(M_{11} - M_{44})/2, \quad (1)$$

and all other elements are equal to zero. It is shown[7] that any deviation from such form of the BSPM means the presence of a preferred azimuth direction. If one assumes that the particles are arranged mirror-symmetrical relative to it in presence of some preferred azimuth direction, then the BSPM with, in general case, all elements different from zero, can be reduced to the block diagonal form with possibly non-zero elements M_{14} and M_{41}. The latter is caused by the fact that the ensemble can contain sub-ensemble of particles which are not subject to orientation, for which the condition of mirror symmetry is not obligatory. The answer to the question on the correctness of the accepted admission lies in the fact, how "reducible" are real BSPMs to the block-diagonal form. To run ahead, let us note that the experiments have shown good validity of this assumption. The procedure of reducing is equivalent to the process of virtual superposition of the reference plane xOz of the polarization basis with the plane of symmetry of the distribution over azimuth directions. It is mathematically reduced to the search for the argument of the rotation operator $\mathbf{R}(\Phi)$ of the coordinate system around z-axis, which, as applied to the experimental matrix $\mathbf{M}_\mathrm{E}$ in the transform

$$\mathbf{R}(\Phi)\mathbf{M}_\mathrm{E}\,\mathbf{R}(\Phi) = \mathbf{M}, \quad (2)$$

reduces the matrix to the block diagonal form

$$\mathbf{M} = \begin{pmatrix} M_{11} & M_{12} & 0 & M_{14} \\ M_{21} & M_{22} & 0 & 0 \\ 0 & 0 & M_{33} & M_{34} \\ M_{41} & 0 & M_{43} & M_{44} \end{pmatrix}, \quad (3)$$

the following symmetry conditions characteristic of any BSPM are always fulfilled:

$$\begin{aligned} &M_{ij} = M_{ji}, \text{ if } i \text{ or } j \neq 3, \\ &M_{ij} = -M_{ji}, \text{ if } i \text{ or } j = 3, \\ &M_{11} - M_{22} + M_{33} - M_{44} = 0. \end{aligned} \quad (4)$$

Fulfillment of the condition (1) in general case is not obligatory.

Such matrices will further be called "reduced." Two values of the angle Φ determining two mutually orthogonal directions satisfy the procedure (2). The unambiguous choice is possible if one direction will conditionally be accepted as the direction of preferred orientation. In Ref. 5, the direction perpendicular to the maximum diameters of particles is selected as such a direction. The element M_{12} of the "reduced" BSPM in this case has minus sign. If the alternative direction has been chosen, M_{12} is positive with the same absolute value. Correspondence of the signs and direction is revealed from the results of mathematical simulation of the BSPM of ensembles of hexagonal plates and columns[8] and from the results of sounding along slant paths.

The parameter χ was introduced in Ref. 7 characterizing the degree of azimuth orientation or, in other words, the sharpness of orientation of the characteristic size near the mode of the distribution. It is represented by the elements $m_{ij} = M_{ij}/M_{11}$ of the normalized "reduced" BSPM by the following formula:

$$\chi = (m_{22} + m_{33})/(1 + m_{44}). \quad (5)$$

The parameter, similar to χ, was introduces earlier[9] for description of orientation of particles elongated along the axis of symmetry, in accordance with the distribution

$$f(\alpha, \alpha_\mathrm{m}, k) = \exp[k\cos 2(\alpha - \alpha_\mathrm{m})]/I_0(k), \quad (6)$$

where f is the probability density of orientation of the crystal axes along the azimuth angle α near the mode of distribution α_m. The parameter of the distribution k is related with the value χ by the following formula:

$$\chi = I_2(k)/I_0(k),$$

where I_2 and I_0 are the modified Bessel functions of the first kind of the second and zero orders, respectively.[9] The ratio I_2/I_0 is tabulated as a function of k. Then the parameter of the distribution (6) is determined from the value χ determined from the experiment.

It is shown[7] that the relationship (5) is general and is applicable to description of the azimuth orientation of particles of an arbitrary shape. The parameter χ is defined in the interval [0 1]. Equality to zero means the absence of the azimuth orientation. The upper boundary can be reached only at strong orientation of symmetric particles in one direction. The distribution (6) was introduced by authors[9] heuristically analogously to the Mises distribution.[10] But, it is shown in our investigations, which are planned to be published in the nearest future, that the distribution has such a form for monodisperse aerosol ensembles. The parameter k takes the specific physical meaning. The distribution of polydisperse ensemble of particles over azimuth orientation angle can be presented in the form of superposition of the distributions of the form (6).

Let us note that the presence of rotation symmetry does not exclude the possibility of polar orientation. The simple proof is presented in Ref. 7 of the fact that the parameter characterizing the polar orientation can be the element m_{44} of the normalized BSPM independent of the presence or absence of azimuth orientation. Quantitative relation between this element and the parameter of the distribution of the form (6) should be revealed from the results of mathematical simulation of BSPM of ensembles of hexagonal plates and columns. Preliminary estimates of this relation performed on the basis of the results from Ref. 8 are shown in Fig. 1.

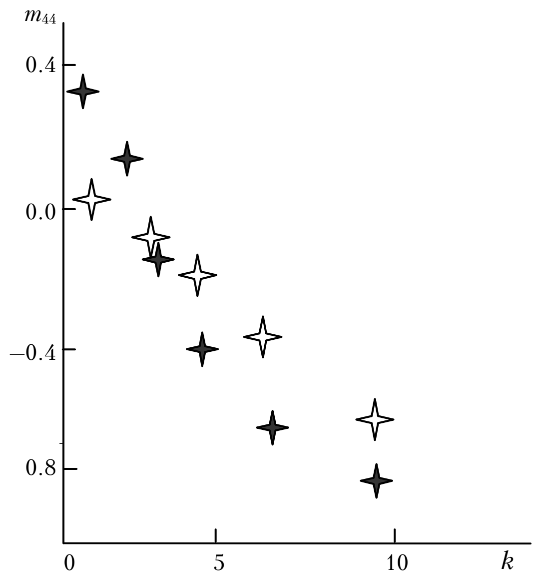

Fig. 1. Correlation between the parameter k of the distribution (6) over the angles of polar orientation and the element m_{44} of the normalized BSPM for hexagonal columns (dark signs) and hexagonal plates (light signs).

The general tendency is that m_{44} takes more and more negative values, tending to the asymptotic value of -1, as maximum diameters of particles tend to group near the plane perpendicular to the direction of sounding.

Aspects of a technique for determination of BSPM from lidar measurements of the Stokes parameters

The vector-matrix equation of laser sounding of anisotropic aerosol medium in the single scattering approximation has the form[11]

$$P(z)\mathbf{s}(z) = \frac{1}{2}cW_0Az^{-2}\,\mathbf{Y}(z, -\mathbf{e}_z)\,\mathbf{M}(z)\,\mathbf{Y}(z, \mathbf{e}_z)\mathbf{s}_0, \quad (7)$$

where $P(z)$ is the power of the scattered radiation incident on the receiving antenna; A is the area of the antenna, $\mathbf{s}(z) = (1, q(z), u(z), v(z))^{\mathrm{T}}$ and $\mathbf{s}_0 = (s_1, s_2, s_3, s_4)^{\mathrm{T}}$ are the dimensionless Stokes vectors, respectively, of scattered and incident light, c is the speed of light in the medium, W_0 is the pulse energy of sounding radiation, z is the distance to the scattering volume. The elements of the backscattering phase matrix $\mathbf{M}(z)$ have the dimension $[\mathrm{m}^{-1} \cdot \mathrm{sr}^{-1}]$. The matrix operators $\mathbf{Y}(z, \mathbf{e}_z)$ of transformation of the radiation at its propagation from the lidar to the scattering volume and back $\mathbf{Y}(z, -\mathbf{e}_z)$ have the following form:

$$\mathbf{Y}(z,z_0,\mathbf{e}_z) = \exp\{-\int_{z_0}^{z} \boldsymbol{\varepsilon}(z', \mathbf{e}_z)\mathrm{d}z'\}, \quad (8)$$

where $\boldsymbol{\varepsilon}(z', \mathbf{e}_z)$ is the transformation (extinction) matrix as a function of the position and direction of propagation of radiation $\mathbf{e}_z$. To calculate this matrix operator, the interval $[z, z_0]$ is divided into n elementary intervals Δz_i so that within the limits of one interval one can assume ε to be invariable and $|\varepsilon_{ij}|\Delta z \ll 1$. Then the operator $\mathbf{Y}(z, z_0, \mathbf{e}_z)$ can be presented in the form of the product

$$\mathbf{Y}(z,z_0,\mathbf{e}_z) = \prod_{i=1}^{n}(\mathbf{I} - \boldsymbol{\varepsilon}_i\Delta z_i),$$

where $\mathbf{I}$ is the unitary matrix.

One should consider Eq. (7) in the cases when one can not ignore the phenomena of linear of circular dichroism of the scattering medium. The possibility of ignoring these phenomena in the case of sounding of crystal clouds is justified in Ref. 12. It is shown that only for fine particles of the size $\cong 1$ μm oriented strongly in one direction the non-diagonal elements of the extinction matrix have the value of 0.02 of the values of the diagonal elements. Noticeable linear dichroism can be observed in this case. The non-diagonal elements for the particles of the size more than 10 μm are hundredths or even thousandths shares of percent of the diagonal ones. In this relation, the extinction matrices of crystal clouds can be considered diagonal with very good approximation. Then the effect of anisotropy of crystal clouds reduced in Eq. (7) to the dependence of the diagonal elements on the angles of radiation incidence on the cloud layer. An example of such a dependence in shown in Fig. 2.

The diagonal elements of the extinction matrix are represented by the same combination of the amplitudes of forward scattering and are equal to each other: $\varepsilon_{ii} = \varepsilon$. Then Eq. (7) is reduced to the form

$$P(z)\mathbf{s}(z) = \frac{1}{2}cW_0Az^{-2}\mathbf{M}(z)\mathbf{s}_0\exp\{-2\int_{0}^{z}\varepsilon(z', \gamma, \alpha)\mathrm{d}z'\}, \quad (9)$$

where $\varepsilon(z, \gamma, \alpha)$ is the extinction coefficient as a function of the distance along the sounding path, γ and α are the polar and azimuth angles of the path.

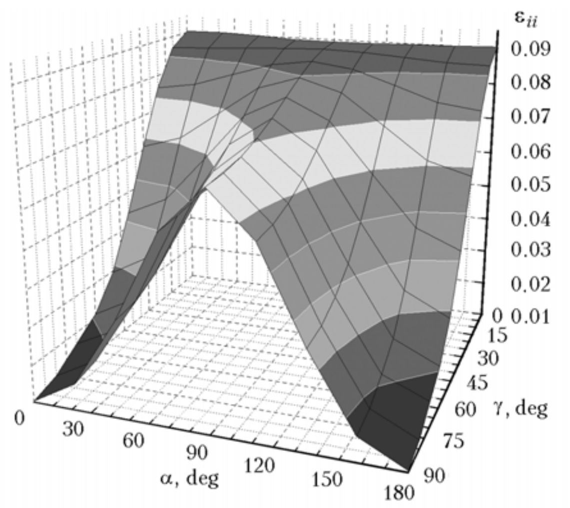

Fig. 2. Behavior of the elements of the extinction matrix ε_{ii} of the ensemble of oriented ice columns, the axes of which lie in the horizontal plane and oriented in the azimuth direction $\alpha = 0$ as a function of γ – the polar angle of incidence of radiation on the layer (read out from the vertical direction) and α – azimuth position of the plane of incidence. The latter is determined by the vertical direction and the direction of the wave vector of the incident radiation.

The equation in this form is used in interpretation of the experimental results on sounding crystal clouds. Let us note that $P(z)/A$ is equal to the intensity $I(z)$, and Eq. (9) is four scalar equations relative to the Stokes parameters of the scattered radiation $I(z)$, $Q(z)$, $U(z)$, $V(z)$. But experimentally it is possible to measure only the intensity at certain positions of the polarization devices installed in front of the detector of radiation. Operation of these devices can be described by means of the notion of the instrumental vector. It is the vector-row, which is the first row of the product of the Müller matrix operators of the polarization devices[13] installed in the receiving channel of the lidar.

Let us denote the instrumental vector as **G**. If applying this operator to both parts of Eq. (9), the left part of the equation will be the fraction of the total flux of scattered radiation, which is transmitted at the acting instrumental vector and the given polarization of the incident radiation flux. The scalar product **Gs** of the vectors determines this fraction. The matrix product $\mathbf{GM}(z)\mathbf{s}_0$ in the right-hand part will give some linear combination of the elements of the BSPM. To determine BSPM, it is necessary to perform a series of measurements at different states of the polarization of the sounding radiation $\mathbf{s}_0^i$ and different instrumental vectors $\mathbf{G}_j$, the number of which is sufficient to obtain the system of linear equations for determination of all elements of the matrix. Four states of polarization of the laser radiation and six instrumental vectors were used in experiments with the lidar "Stratosfera" described below.

The important aspect of the technique is the choice of such a measurement scheme, at which the effects of instability of the laser pulse energy and variability of the transmission on the path are excluded. According to these ideas, the receiving part of the lidar "Stratosfera" is designed so that the signals are simultaneously measured in pairs, which are determined by three pairs of mutually orthogonal instrumental vectors $\mathbf{G}_j, \mathbf{G}_j^*, j = 1, 2, 3$. Orthogonality of the vectors means that $\mathbf{G}_j^* \mathbf{G}_j^{\mathrm{T}} = 0$. The following pair of equations is realized at the ith state of polarization of the laser radiation and jth pair of the instrumental vectors:

$$I_k(z) = cW_0 z^{-2} \exp\left(-2\int_0^z \varepsilon(z', \gamma, \alpha)\mathrm{d}z'\right)\mathbf{G}_j \mathbf{M}(z)\mathbf{s}_0^i,$$
$$I_k^*(z) = cW_0 z^{-2} \exp\left(-2\int_0^z \varepsilon(z', \gamma, \alpha)\mathrm{d}z'\right)\mathbf{G}_k^* \mathbf{M}(z)\mathbf{s}_0^i, \qquad (10)$$

where

$$k = 3(i - 1) + j,\ i = 1, 2, 3, 4;\ j = 1, 2, 3.$$

In the full cycle of measurements necessary for determination of a BSPM, the index k passes the values from 1 to 12. Twelve equations of the following form are formed from Eq. (10):

$$C_k(z) = [\mathbf{G}_j \mathbf{M}(z) - \mathbf{G}_j^* \mathbf{M}(z)]\,\mathbf{s}_0^i / [\mathbf{G}_j \mathbf{M}(z) + \mathbf{G}_j^* \mathbf{M}(z)]\mathbf{s}_0^i, \qquad (11)$$

where

$$C_k(z) = [I_k(z) - I_k^*(z)]/[I_k(z) + I_k^*(z)]. \qquad (12)$$

Transition to equations of the form (11) has great significance, because the pulse energy and transparency of the path disappear in them. At exact observance of the projected parameters of the polarization block of the lidar, the values C_k are the Stokes parameters q, u, v, normalized to the intensity, when the index j has been equal to 1, 2, 3, respectively. When the parameters are deviated from the projected ones, C_k are the linear combinations of the parameters q, u, and v. In this paper we do not consider the problems of calibration of instrumentation and solution of the system of equations (11). They are described in details in Refs. 14 and 15. Let us only note that calibration of the lidar using the "molecular reference" and the algorithm for processing make it possible to separate out the molecular and aerosol **A** components of the BSPM.

After adding the system (11) by the conditions of symmetry of BSPM (4) we obtain the system of 12 linear equations for 8 elements a_{ij} of the normalized matrix of the aerosol component. The residual elements are determined from the conditions of symmetry. The normalized matrix is defined as follows: $a_{ij} = A_{ij}/A_{11}$. The surplus of the system of equations is used for minimization of the errors in determining the matrix elements with the method of least squares. Simultaneously it enables one to estimate the errors, what, in its turn, makes it possible to estimate the reliability of the obtained results and to exclude from consideration the BSPM with the errors exceeding a certain threshold.

Results of experimental investigations of the BSPM of crystal clouds

The results presented below are based on the statistical analysis of the sample of 453 experimental BSPM.

The first result is related to reducibility of the experimental matrices to the form (3). The algorithm for the search for the argument Φ of the transform (2) in the presence of experimental errors in determining the elements of BSPM is based on the use of the method of least squares and is described in Ref. 15. The criterion of reducibility is the one-moment vanishing of the elements a_{13}, a_{23}, a_{24} of the normalized BSPM at a certain value of the angle Φ. Exact observance of this condition, obviously, is impossible because of the errors in measurements. The sample-mean weighted error in determining the elements of BSPM is 0.04.

The histograms of the frequencies of occurrence of the elements a_{13}, a_{23}, a_{24} of the normalized reduced BSPM have well pronounced maximum in the range of zero values. Their joint distribution is well approximated by the normal distribution with zero mean value and the standard deviation $\sigma = 0.07$. Then it follows that 75% of events fall into the range of the doubled error ±0.08.

Based on this result, one can suppose that the hypothesis of the mirror symmetry of the ensemble of particles relative to a certain plane lying in the basis of the idea about reducibility of BSPM to the form (3) is fulfilled only partially. Nevertheless, it is useful, because in the cases when non-diagonal elements have been essentially greater than the measurement error, one can determine the angle Φ in the transform (2) with the error of a few degrees.

The second important result is that BSPM corresponding to the ensembles of particles with rotation and obeying the condition (1) form the minor part of the sample. The histogram of the frequencies of occurrence of the values of the element a_{12} is shown in Fig. 3.

In the range of values different from zero, only 33% of events fall into the corridor of double measurement error. The sample mean value is equal to –0.22. This means that some azimuth orientation of particles presents in a cloud in the majority of events. The shift of the mode of the parameter χ, Eq. (5), from zero to the value of 0.1 also is the evidence of this fact. Up to 80% of events fall into the range of the values χ from 0 to 0.2, that corresponds to the change of the parameter k in the distribution (6) from 0 to 1.5. Hence, strong manifestation of the azimuth orientation is quite a rare phenomenon. The values χ of about 0.6 are observed in some experiments, that correspond to $k \cong 5$. For clearness, let us note that at $k = 1$, in agreement with the distribution (6), the rms deviation from the direction of the preferred orientation is ±35°, and at $k = 5$ it is about ±15°. At the uniform distribution ($k \to 0$) this value is equal to ±52°.

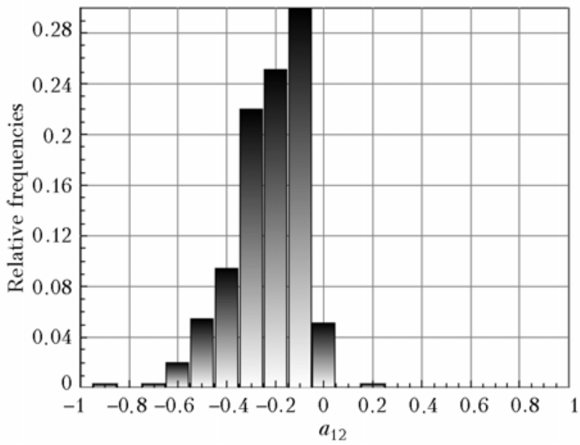

Fig. 3. Distribution of the frequency of occurrence of the values of the element a_{12} of the normalized BSPM. Determined from the sample of 453 BSPMs.

The correlation is observed between the directions of particles and predominant wind directions at the heights of existence of crystal clouds. It is seen in Fig. 4 that the most probable direction of orientation is the east-west line.

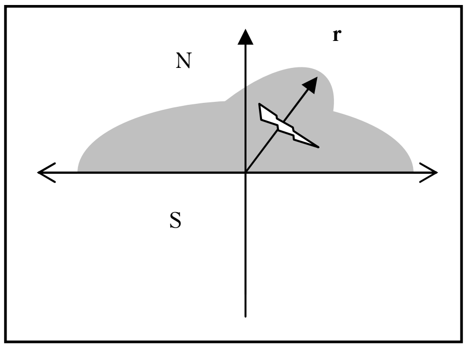

Fig. 4. Diagram of the directions of the predominant azimuth orientation of the maximum diameters of particles perpendicularly to the radius-vector **r**. The probability density is proportional to the length of the radius-vector.

According to the accepted rule of reducing BSPM to the form (3), one should assume that the maximum diameters of particles are oriented predominantly across the direction of orientation determined from the experimental matrix. Presumably, it is the direction across the wind velocity vector. As known, westerly winds are prevalent in the region of Tomsk in the troposphere. The possibility of orientations of particles by the pulsations of wind velocity is the working hypothesis, which we are testing now.

The histogram shown in Fig. 5 gives the idea of the orientation of the maximum diameters of particles in horizontal direction.

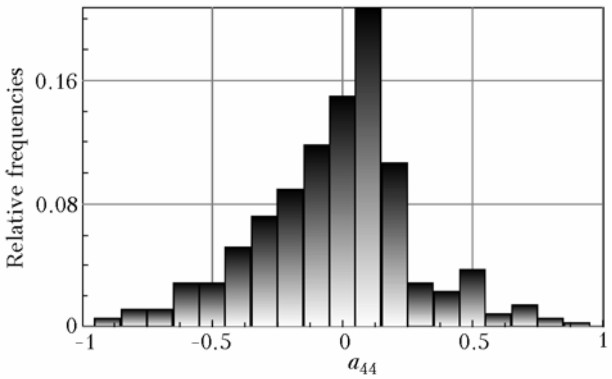

Fig. 5. Distribution of the frequency of occurrence of the values of the a_{44} element.

The sample-mean value of the a_{44} element is close to zero. From Fig. 1, one can see that this value corresponds to the parameter of the distribution $k \cong 1$, i.e., quite weak orientation. But the distribution has the abscess to the side of negative values. If taking the value $a_{44} = -0.1$ (that corresponds to $k \cong 3$) as a sign of essential orientation, one should refer about 50% of events with the essential and strong orientation. In other cases small values of $|a_{44}|$ can be the consequence of the presence in a cloud of big number of fine or large but isometric particles. Another possible reason of weak orientation can lie in strongly developed turbulence of air.[16]

Conclusion

The following problems were solved during development of the method for remote study of anisotropic aerosol media by means of optical-radar measurements:

– the equation of laser sounding of anisotropic medium is derived;

– applicability of this equation is justified for interpretation of the data of laser sounding of crystal clouds;

– the parameters are defined through the elements of BSPM, which quantitatively characterize the state of orientation of particles in cloud;

– new data are obtained on microphysical characteristics of cirrus clouds, which are important for more accurate calculations in the problems of radiation transfer in the atmosphere, but earlier could not be obtained because of the absence of the techniques for their instrumental determination.

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