# *LIGHT SCATTERING IN THE EARTH'S ATMOSPHERE*

(An essay on the 150th anniversary of the discovery by Arago of the polarization of light in the daytime sky, and on the 100th anniversary of the discovery by Govi of the polarization of light in scattering)

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## 1. INTRODUCTION

ATMOSPHERIC optics belongs to the number of ancient sciences whose origins are lost in prehistoric time. The various light phenomena in the atmosphere — the varying blue of the sky, the rosy dawn, rainbows and the extraordinary halo, fantastic mirages — have long capitvated poetic imagination, converting them into objects of religious cults. But even in ancient memorials of material culture and in the evidence of the historians, there are found traces of searching thought, attempting to discover the real nature of phenomena behind mystic coverings. In every age, very promising minds pursued this field. Thanks to them, atmospheric optics invariably occupied a conspicuous place in the process of understanding nature, never being relegated to scientific obscurity. It is true that its development was frequently marked by great mistakes and curious happenings, but it was no exception in this respect. It is far more important that it is connected with many great discoveries, which had a decisive effect on the history of science. For example, it suffices to recall it was precisely from attempts to explain the blue color of the sky (see below) that there arose one of the most important and widest fields of contemporary physics — the study of the scattering of radiation by matter. In the same way, measurements of transmission in the earth's atmosphere, as is well known, led at the beginning of our century to one of the most decisive proofs of the existence of molecules and the validity of the kinetic theory of gases. Also widely known is the role played in the development of spectroscopy by the discovery of selective absorption of light by the atmosphere.

It is not difficult to recall a number of similar examples. It is also not difficult to see that a basic factor in determining this role of investigations in the region of atmospheric optics is the scale of the earth's atmosphere, which makes it possible to carry to completion much finer observations that can be obtained (with the same measuring techniques) under laboratory conditions. It is just this circumstance that defines what to our point of view is the basic mark of atmospheric optics as a science. Setting aside the numerous and often very important problems of an applied character, we see that the development of atmospheric physics has always been directly connected with the leading advances of theoretical concepts and experimental technology, and sometimes has anticipated them, and that the fundamental difficulties of this science have always been the difficulties of physics in general. In particular, we recall that, in connection with the studies of atmospheric optics and the optics of planetary and stellar atmospheres, the theory of propagation (transfer) of radiation in a scattering medium sprang up and received its development, acquiring today great value in applied nuclear physics and already becoming a fundamental branch of mathematical physics. It is also impossible not to recall the direct connection of the development of the study of turbulence with investigations on the twinkling of stars and other distant light sources. Finally, we shall quickly see that the fundamental problems of the study of the optical properties of the atmosphere are not only tightly bound with the most important problems of colloidal optics, but are very close (methodologically, at any rate) to the fundamental problems of contemporary nuclear physics. In particular, this leads to the result that the solution (and even the correct formulation) of a whole series of, it would appear, especially classical problems of atmospheric physics would be impossible without bringing in the most advanced ideas of contemporary theoretical physics.

The practical value of atmospheric optics is also much broader than may appear at first glance. The optical properties of the atmosphere determine to a significant degree its optical and thermal states, and indeed the optical and thermal states of the earth's surface for sowing, engineering construction, etc. Knowledge of these states and skill in forecasting their change are highly necessary, both for the development of methods of weather forecasting and for the solution of all problems connected with conditions of visibility, illumination and exposure, including urgent problems of transport, construction, medicine and agrobiology. Moreover, optical methods can serve in the study of the atmosphere itself (including the stratosphere, which is scarcely accessible by other methods) and for the study of processes taking place in it. This has direct meteorological importance and acquires fundamental importance in connection with the development of stratospheric means of transportation and with the increasing necessity of the investigation of planetary atmospheres from without, in order to guarantee the possibility of penetration through them (to the surface of the planets) in space ships.

However, if we turn to the development of atmospheric optics in the last half century, a rather unusual picture is revealed. On the one hand, during this time, especially during the last two decades, our knowledge of the atmosphere and, in particular of its upper layers, experienced a remarkable progress, requiring a radical revision of most of the representations that had only recently become widely adopted. At the same time there was a vigorous intrusion of contemporary physico-mathematical means of investigation into the region of atmospheric physics, thus greatly changing the aspects of this science. On the other hand, the traditional aims and directions of investigation scarcely experienced the spirit of the time. The concepts of the modern investigations essentially preserve the same features as at the beginning of the century, differing chiefly in the scale and technology of the accomplishments, and not in the formulation of the problems.

Such a conflict between old-fashioned tendencies and the extreme modernization of methods of their accomplishment has quite a regular foundation, which will be clear from what follows. However, this does not do away with the necessity of surmounting it by means of a serious review both of the aims and methods of atmospheric-optical investigations, the more so that this conflict finds its concrete expression in a whole series of clearly painful facts.

In particular, contemporary atmospheric physics is characterized by a complete break between theory, which is rarely very refined, and experiment, occasionally very fine and highly perfected. As a rule, they developed very vigorously, but practically independently of one another, missing the decisive stage of mutual control. In addition, a situation often came about in which the cost of the experimental or theoretical investigations significantly exceeded the value of the results obtained — in some cases, it was the material expression of the lack of correspondence of the general direction of investigation, its methodology and the nature of the object. Below we shall establish that this lack of correspondence was due in the first place to a lack of those special characteristics with which the transition from passive, chiefly qualitative observations of natural phenomena was made to purposeful, quantitative analysis of its physical nature under conditions of controlled variability of the very object of investigation — the atmosphere.

Thus, in the process of its very intensive development, atmospheric optics came in very definite fashion to a boundary which required serious reconsideration both as to program and to methods of investigation. Therefore, it is appropriate to discuss briefly, but in historical aspect, the modern position of this science, which is the purpose of the present paper. The necessity of a detailed review is heightened by the fact that in recent years the leading role in atmospheric-optical investigation has been more and more clearly assumed by Soviet scientists, and to the fact that, notwithstanding this, for some years the problems of atmospheric optics have remained practically untreated in domestic literature. However, we shall limit ourselves to only one rather large group of problems, namely, problems touching on the properties of the atmosphere as a scattering medium and leading directly to the two notable discoveries mentioned in the subtitle.

## 2. THE PUZZLE OF THE DAYTIME SKY AND THE DISCOVERY OF LIGHT SCATTERING

Studies of the color and polarization of the daytime, cloudless sky in connection with the discovery and explanation of the phenomenon of light scattering form one of the most interesting pages in the history of science. One can find a detailed description, although somewhat unclear and in much already archaic, of the early stages of this investigation — approximately up to the first decade of the present century — in well-known texts of atmospheric optics, which have long become bibliographical rarities.[1,2]

Therefore, we shall dwell only on certain more important moments, without the knowledge of which it is not possible to make a correct estimate of the present position of atmospheric optics as a science.

It is difficult to show when the idea first arose that the brightness and color of the daytime sky were brought about by "reflection" of sunlight from the air or from particles contained in it — droplets and dust.

We find it already clearly formulated by one of the pioneers of experimental physics Al-Hazen[3] (XI century) and later by Kepler[4] and Leonardo da Vinci.[5] Newton[6] held these views and, following him, Mariotte, Bouguer, Euler,[7] Goethe,[8] Clausius[9] and others.

Such a point of view has been the prevailing one but by no means the only one, and even in our time there have been attempts to oppose it, if only in part by other representations. Thus, several authors, among them Brewster and others, and at the end of the XIX century Chapuis[10] and Spring,[11] have suggested that the dark blue of the sky is brought about completely by selective absorption of the light by the air or by solid or gaseous impurities contained in it (for example, ozone). Other authors — Lalleman,[12] Hartley,[13] et al., and more recently Cohn[14] and, even in 1953, a group of American authors,[15] attempted to find the reason for the blueness of the daytime sky in various phenomena of photo- and cathode-luminescence. Finally, for a long time, right down to the eighties of the last century, i.e., much later than the completion of the first spectrophotometric measurements of the cloudless sky, there existed hypotheses that the reason for the blueness lay in the peculiarities of the observer's perception, and had no physical character. In particular, in 1885, Pickering[16] published a description of researches especially set up for the purpose of proving this hypothesis.

However, one must not consider these hypotheses, which are clearly incompatible with the contemporary viewpoint, only as historical curiosity. Their popularity and endurance before the critical objectors testifies conclusively to the serious difficulties which arise in path of explanation of the illumination of the daytime sky by scattering of light. The object of most attention in this case was the color of the sky.

In passing, we note that still greater difficulties confronted the explanation of the blueness of the sea and other water bodies. From the time of Bunsen, who turned his attention to the sharply expressed selectivity of the absorption capability of water, which causes its greenish blue color in transmitted light, the viewpoint was established that the color of the sea is explained by precisely this circumstance. Aufsess[17] and Pietenpol[18] adhered persistently to this concept at the threshold of the XX century, emphasizing the possibility of complementary coloring of the water by all sorts of impurities. After the researches of Schwartzchild, Schuster et al. on the theory of propagation of light in scattering media, it was not difficult to see that such an explanation was insupportable, because, in the absence of scattering, the depth of the tank would not have reflected the light and it would have appeared black, independent of the absorption spectrum of water. However, the contrary point of view, according to which the color of the sea would be explained by scattering (by molecular impurities or impurities contained in the water — bubbles, plankton, etc) even though advanced by a number of authors, appeared to be even less viable. From these theoretical considerations, it followed that in the absence of absorption, any law of scattering leads to the independence of the reflection coefficient of the container on the wavelength of light, i.e., the ocean would be white like the clouds.

Actually, as is well known, the color of the sea and other bodies of water is determined by the combined action of both factors. This was first shown in 1921 by Raman[19] and Shuleĭkin,[20] independently, and a rigorous theoretical analysis was completed only in the forties by Ambartsumyan, Chandrasekhar, Sobolev, and others.[21] With this, the role of fluorescence in the coloring of bodies of water remains only partially clear at the present time.

We shall not dwell on the first attempt to penetrate into the secret of the explanation of the blue sky, advanced by Leonardo da Vinci. This attempt proceeded from an opinion, which was widespread in his day, that the color is determined by the proportion in which light and darkness are mixed, and is not of interest today. In the same way, we shall discuss the hypothesis according to which the color of the sky is determined by the actual coloring of the particles of the air or of impurities contained in it. This hypothesis, which was advanced especially by Euler, was refuted by the difference of coloring of the atmospheric air in transmitted and reflected light, and it played no significant role in the XIX century.

The viewpoint of Newton was more widespread in the first half of the XIX century; according to this the blue color of the sky arises as the result of the interference of light upon its reflection from tiny water droplets contained in the air (like the interference colors of thin film). In the middle of the XIX century, Clausius[9] advanced against the hypotheses of Newton objections which were very weighty for their time but, as we now understand, were not always justifiable. Fundamentally, they reduce to the following. If water is actually contained in the air in the form of droplets, thus causing the appearance of such intense interference coloring of the sky, then the diffraction phenomena on these droplets ought to lead to the formation of powerful coronas, i.e., to an appreciable washing out of the outlines of heavenly bodies, which is not observed in a clear sky. Moreover, it was shown by Clausius to be inexplicable how the droplets of water could float in the air and why, if these actually were droplets of water, a rainbow is not observed in clouds and fogs. This stimulated Clausius to make an attempt to save the hypothesis of Newton by replacing the water droplets in it by thin-walled bubbles with their characteristic interference coloring. Upon increase of humidity, the thickness of the walls of the bubbles would, according to Clausius, increase, bringing about a whitening of the light reflected by the bubbles. The discovery of light scattering soon compelled Clausius to give up his hypothesis, but for a long time it remained an object of discussion. In particular, as late as 1883, it was actively defended by Müller,[22] who made an attempt to improve the representation of the interference nature of the blue color of the sky by taking into account the effects of multiple reflection from the bubbles.

However, the imperfections of the idea that the atmosphere, with water droplets and bubbles constantly contained in it, was a turbid medium (colloid), and that the color of the sky was determined by conditions of "reflection" of light from particles producing atmospheric turbidity were not the only places to look for the reason for those difficulties which it encountered among the competing assumptions. A more important role in this was played on the one hand by the well known, and (at that time) sharp difference between the character of the phenomena observed in transmitted light through a turbid medium — clouds and fog — and the picture of the bright blue sky, and on the other hand, the total absence of laboratory experiments simulating in some degree the blueness of the sky. It must be noted that in the first quarter of the XIX century, not only were the most significant characteristics of rainbows and various halos well known and, fundamentally, correctly understood, but the diffraction nature of corona had been discovered by Jordan and considered in detail by Fraunhofer. We also note that not long before (in 1790), the first apparatus for quantitative measurement of the blue color of the sky was constructed by Saussure,[23] and the character of the change in color from the zenith to the horizon was made clear. In 1799, these measurements were extended by Humboldt, and were later frequently repeated in a number of variations by a whole series of authors right down to our own day. The differences discovered here between the optical phenomena in clouds and fogs and in a clear sky were so striking that there was almost no doubt remaining of the necessity of seeking explanations in the two cases.

The first attempt at a laboratory study of the optical properties of a turbid medium was undertaken by Goethe[8] and had as its purpose simulation of the blue color of the sky. However, the object of observation — the lower, non-luminous part of a spirit flame illuminated by the sun — was unsuccessfully chosen by him, and, in spite of the qualitative confirmation of the basic idea (on a white background a light blue was observed and on a dark background a dark blue color), the experiment of Goethe would not be considered as convincing, especially being "supported" by his mistaken ideas as to the nature of light.

Successful laboratory simulation of light blue color of the sky was first achieved by Brucke[24] in 1853. Irradiating a water emulsion of gum arabic and observing it against a background of a black screen. Brucke observed a bright blue color, while in transmitted light, the emulsion had a reddish-yellow color. By the same means, Brucke demonstrated experimentally that one could seek in the optical properties of the emulsion explanations of the coloring of the vault of the sky and the characteristic reddening of the sun upon its approach to the horizon.

In 1860, Govi[25] carried out investigations with smoke from alcohol and tobacco, in which it was shown that color effects are produced in the scattering of light in smoke similar to those observed in the scattering of light by clouds and fogs. Finally, in 1869, that is, sixteen years after the experiments of Brucke, Tyndall[26] completed his brilliant experiment (which rapidly achieved wide fame) with the so-called "actinic clouds." The substance of these experiments was that as a result of chemical decomposition of the vapors of certain compounds in an illuminated vessel, an aerosol cloud was obtained, the particles of which gradually increased in size. In particular, the growth of the particles was manifest in changes of the coloring of the light scattered by them — a gradual transition was observed from azure blue, which reproduced completely the coloring of the southern sky, to white, characteristic of clouds, smokes, and fogs.

Thus the experiment of Brucke, as Clausius[27] correctly noted in 1853, demonstrated that the reflection of light from small particles takes place according to other laws than that from massive bodies, i.e., that in the case of small particles there is no longer a reflection of light, but a certain phenomenon which has obtained the name of scattering. Furthermore, the experiments of Brucke, Govi, and Tyndall have shown that the character of the scattering depends significantly on the character of the scattering particles, especially on their dimensions. Thus strong arguments appeared against the theory of Newton and Clausius, which the latter had pointed out.[27] However, the experiment of Brucke did not yield decisive arguments in support of the explanation of the brightness and color of the daytime sky by scattering. The blue of the light scattered by a colloid merely gave evidence of the admissibility of a similar explanation, but nowhere did it prove its validity. One had to seek the proof in other directions, and one did not have long to wait.

Even in 1809, François Arago,[28] viewing the daytime sky through a Nicol prism, discovered that the light coming from the sky was strongly polarized. Not only was the light of the sky polarized in this case, but also the light of a haze separating an observer from distant objects such as mountains. Therefore, there was no doubt that the very illumination of the air (or of impurities contained in it) was polarized. During the next 150 years, this phenomenon has served constantly as the object of persistent and numerous investigations, the results of which will be given below. But all attempts at its explanation by starting out from representations of the reflection of the sun's rays by particles of air or of impurities contained in it, put forth by Arago, Babinet, Brewster, Clausius and others, proved to be unsuccessful. In particular, this was in its time a strong argument against the Newton-Clausius picture. The situation was no better relative to competing hypotheses, of which we mention only the hypothesis of Brewster,[29] actively supported by Rubenson.[68] Starting out from the fact that the maximum of the polarization of light of the daytime sky, discovered by Arago and investigated in detail by Brewster and Rubenson, made an angle $\varphi$ with the vertical to the sun of about 90° (this corresponds approximately to twice Brewster's angle at the boundary of two media with similar indices of refraction), These authors expressed the conviction that the reflecting substances were not foreign particles but the air molecules themselves.

The solution of the intriguing puzzle of the polarization of the illumination of the daytime sky required a full half-century and was first obtained in the researches of the Turin astronomer Professor Govi, to which reference has already been made.[25] The investigation, the results of which were given in the form of two short letters to the Proceedings of the French Academy of Sciences, was undertaken with the aim of showing that the illumination of the daytime sky was brought about by the scattering of light, and that the illumination of comet tails, the light of which is also polarized, could be explained by the same phenomenon. Govi filled a closed and carefully shaded room with different smokes and directed a beam of sunlight into it through a slit in the shutter. Observing the luminous shaft through a polariscope, Govi discovered that the scattered light was strongly polarized, and that the maximum of the polarization did not occur at a 90° scattering angle. By subsequent experiments, Govi established the fact that if the scattering angle is changed over a wide range, then directions are observed in which the scattered light is completely unpolarized (neutral points according to the terminology of Arago) and intervals of angles are observed where the plane of polarization is rotated by 90° relative to the plane of scattering (negative polarization according to the terminology of Arago). In Sec. 5 we shall see that both these circumstances are characteristic of the illumination of the daytime sky (as demonstrated by Govi) and were first observed by Arago. On the basis of his experiments, Govi definitely concluded that the scattering of light by small particles had nothing in common with the reflection of light and represented an independent phenomenon.

Ten years later, there appeared the famous work of Tyndall[26] under the title "On the Blue Color of the Sky, The Polarization of Sky Light, and On the Polarization of Light by Cloudy Matter in General." Together with the evidence already mentioned on the effect of the dimensions of scattering particles on the coloring of light scattered by them, Tyndall reported the results of his very careful polarization measurements. It was established by him that the character of polarization changes sharply with change in the dimensions of the particles. If the particles are very small (dark blue color of the scattered light), then the degree of polarization is large and its maximum is observed at a scattering angle of 90°. With increase in the dimensions of the particles (whitening of the scattered light), the degree of polarization decreases and the maximum is displaced to one side while, in accordance with Govi, "neutral points" are observed and also regions of "negative polarization." Now the light scattered by colloids under laboratory conditions and the light emitted from the sky displayed at least two common features — the character of color and the character of polarization. This gave Tyndall a sufficiently strong foundation for assuming the identity of these phenomena, which immediately attracted great attention to him.

Along with this, a discovery was made which was most important in its consequences — the discovery of the change of polarization (i.e., spin) of radiation in the act of its scattering by matter. As is seen from the above, this discovery was made in 1809 by Arago under field conditions, but it was understood a half-century later and only after the independent laboratory experiments of Govi. We add that history has been unfair to both scholars. Knowledge of the discovery of Arago, which forms essentially one of the cornerstones of the organized structure of modern molecular optics, escaped the attention of later generations. The brilliant experiments of Tyndall not only underscored the remarkable discoveries of Brucke and Govi, but even eclipsed them — the name "Tyndall effect" was undeservedly but firmly attached to the phenomenon of light scattering by colloids.

In addition, the experiments of Tyndall again awakened an active interest in the problem of the daytime sky. Scarcely two years had passed before Rayleigh (then still Strutt) began the publication of a series of papers devoted to a theoretical discussion of this problem. These papers formed the basis of the future science of the scattering of radiation by matter.[30] Here Rayleigh started out from the concept that the scattering of light took place (similarly to the experiment of Tyndall) on small particles suspended in the air (the particles were supposedly of spherical shape), and considered light waves as waves in an elastic ether. Later, in 1899, he revised his theory on the basis of the electromagnetic theory of light,[31] which, however, did not change the fundamental conclusions: 1) outside of absorption bands of scattering particles, the intensity of light scattered by particles whose dimensions are much smaller than the wavelength of the light $\lambda$ is proportional to $\lambda^{-4}$, and 2) the degree of polarization p of the scattered light depends upon the angle of scattering $\varphi$, and is equal to

$$p = \frac{\sin^2\varphi}{1+\cos^2\varphi}\,. \qquad (1)$$

Naturally, an attempt was immediately made to compare these conclusions with the observation of the daytime sky. In part, so far as the coloring of the daytime sky is concerned, such measurements were first completed by Rayleigh himself, and, after him, by Vogel, Krov, Tsetvukha, Bock and, especially careful measurements in 1886 by Abney and Festing. The measurements showed that in very clear weather the spectral distribution of the brightness of the daytime sky corresponded approximately to $\sim \lambda^{-4}$, but that, even under the most favorable conditions, the departures from this relation were considerable. As far as polarization is concerned, the polarization picture of the sky, which was rather well studied at this time, as a whole corresponded approximately to Eq. (1), departing, however, from theoretical expectations in a number of important details. Finally, spectral measurements of the transmission in the atmosphere which were completed, in particular by Abney and Festing, and also by Becquerel in the eighties of the last century, again demonstrated that, for very high transmission in the atmosphere, the atmospheric attenuation of light changes with the wavelength also in approximate correspondence with the Rayleigh law $\sim \lambda^{-4}$ (in contrast to the proportionality $\lambda^{-2}$, following from the theory of Clausius), but with very appreciable and variable departures from it. Below we shall return to a discussion of the nature of all of these deviations; here we shall only note that such excellent, semi-quantitative agreement of the spectral and polarization characteristics of the light emitted from the sky with theoretical predictions could be regarded as convincing evidence in support of the validity of the explanation of the illumination of the sky as the scattering of sunlight from small particles suspended in the air.

However, Rayleigh in 1899 gave up these assumptions and went over to the hypothesis that the scattering particles were the air molecules themselves.[31] This step, which returned to the ideas of Brewster, was all the more daring since Tyndall directly confirmed that, according to his measurements, air, dust particles and water droplets do not scatter light at all.[26] As is well known, an important discussion between L. I. Mandel'shtam and M. Planck[32] quickly arose over this hypothesis, the result of which was the creation in 1908 — 1910 of the fluctuation theory of light scattering.[33] In particular, Planck showed that, in spite of the error discovered by Mandel'shtam in the initial assumption of Rayleigh on the incoherence of waves scattered by the separate molecules, the connection established by Rayleigh between the scattering power of the medium and its index of refraction was valid. He also obtained a reliable foundation of the theory of light scattering in finely dispersed colloids put forward in 1904 by Maxwell-Garnett,[34] which qualitatively explained the color of colloidal solutions. The starting point in this theory was the assumption that, if the colloidal particles and the distances between them are small in comparison with the wavelength, then one can attribute an index of refraction to the medium which will be connected with the polarizability of the individual particles by the Lorentz-Lorenz formula. Thus the content of the theory of Maxwell-Garnett was essentially reduced to consideration of cooperative (namely, dispersive) effects in scattering on a set of colloidal particles and the region of its applicability was limited only to extremely finely dispersed and very concentrated colloids, as a consequence of which this theory played no role at all in atmospheric optics.

The fact that the Rayleigh theory gave a correct qualitative description of the fundamental features of the illumination of the bright daytime sky — its color and polarization, and also the spectral dependence of transmission in the atmosphere — by means of very simple and graphic formulas, immediately brought forth universal acknowledgment of this theory. However, its triumph, which not only confirmed the hypothesis of Rayleigh that the atmospheric scattering of light bore an essentially molecular character, but which also demonstrated its decisive effect on the process of establishment of the molecular-kinetic point of view generally, was the determination of the Loschmidt number from measurements of the transmission of light (see Sec. 4).

Nevertheless, there did not exist any basis for abandoning the idea that aerosol particles suspended in air have a significant effect on the optical properties of the atmosphere, the more so since not all facts were explained by the Rayleigh theory. In particular, the brightness picture of the daytime sky and also the existence in it of neutral points were by no means compatible with it.

In the same year (1899) when the Rayleigh theory apparently gave the keys for the solution of the puzzle of the daytime sky, the theory of light scattering on foreign particles was first set forth. Love[35] rigorously investigated the problem of the diffraction of electromagnetic waves on a sphere. Within nine years, Mie again solved the same problem. But, in contrast with Love, he did not limit himself to the mathematical side of the problem, but compared in detail the conclusions of the theory with the optical properties of the colloidal suspensions of metals, which immediately gave publicity to this theory among experimentalists.

Subsequently, the experimental and theoretical investigations of the phenomena of radiation scattering by matter were developed very intensively and rapidly became the object of one of the fundamental branches of contemporary science. But the connection with the problems of atmospheric optics was not lost, and the effect of atmospheric-optical problems on the development of the theory of scattering continued to be decisive in many connections. Discussion of this connection will be to a significant extent the theme of what follows. However, it is first necessary to consider what parameters are needed to characterize the scattering medium and the scattering act itself.

## 3. DESCRIPTION OF LIGHT AND THE PROPERTIES OF THE MEDIUM IN THE SCATTERING ACT

If we turn to the phenomena of light scattering, we must characterize the atmospheric air as well as each colloid by their ability to absorb and scatter light of different wavelengths. However, upon close examination, it is clear that this ability, generally speaking, depends not only on the properties of the air itself but also on the character of polarization of the light.[37-40] Therefore, it is first necessary to seek out such optical characteristics of the scattering medium (in particular, atmospheric air) which would reflect the properties of the latter independently of the character of the light field. It is shown that this is possible only in the case when a transition is made in the description of the absorbed and scattered light beams from the ordinary intensities of the electric and magnetic fields of the light waves to other, specialized parameters.

A basic feature which we encounter in considering the propagation of light in a scattering medium is the multiplicity of scattering acts which are mutually incoherent, as a result of which a constant mixing of light beams with very different previous histories takes place. Inasmuch as not only an angular redistribution of the intensity of the light waves takes place in each scattering act, but also a change in its polarization, the scattered light is an incoherent, statistical mixture of beams of very different intensities and with various states of polarization.

However, the character and the result of this scattering act depend strongly on the polarization of the scattered light. Therefore, in problems of the scattering of radiation, one must characterize the latter by parameters which are additive for incoherent light beams and which cover descriptions such as their energy and the state of their polarization. Such parameters were first suggested by Stokes[41] in 1852, i.e., even before the discovery of light scattering, and for a long time were disregarded "owing to their uselessness." Subsequently they appeared in different variants in the works of Rayleigh, Poincaré, Becquerel, Wiener, Soleil, and others in connection with certain "exotic" problems, but remained completely unknown to a wide circle of physicists. Interest in them was reborn only in the 1940's precisely in connection with problems of scattering and propagation of light in turbid media, and, most important of all, in the atmosphere (references 42 — 47 and others; for more detail see reference 38). It was quickly shown that the Stokes parameters are connected in most direct fashion with the quantum mechanical radiation density matrices and historically form their first formulation. In this connection, it is impossible not to note that even the correct statement of such a characteristically

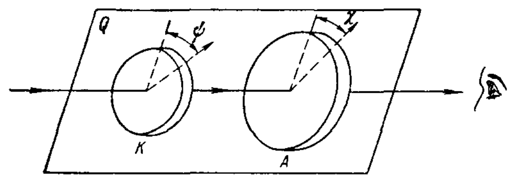



FIG. 1

classical problem as the multiple scattering of light in the atmosphere was completely impossible without taking on the most typical apparatus of quantum mechanics — the density matrix.

Bypassing the later generalization of the Stokes parameters (see, for example, reference 38), we can introduce them in their simplest form in the following way,[44,47] connecting them directly with one of the possible and most useful procedures of measurement. We shall assume that a compensator K, having a path length difference of a quarter-wavelength, and an analyzer A are placed successively in the path of the light beam (Fig. 1). We choose an arbitrary plane of reference Q, containing the direction of the ray, while the angles of rotation of the compensator $\psi$ and of the analyzer $\chi$ about the direction of the light beam will be measured from the plane Q counterclockwise looking toward the ray. Then, the Stokes parameters of the light beam are, by definition,

$$\left.\begin{aligned} S_1 &= I(\psi=0,\ \chi=0)+I(\psi=90^\circ,\ \chi=90^\circ),\\ S_2 &= I(\psi=0,\ \chi=0)-I(\psi=90^\circ,\ \chi=90^\circ),\\ S_3 &= 2I(\psi=45^\circ,\ \chi=45^\circ)-S_1,\\ S_4 &= S_1-2I(\psi=0^\circ,\ \chi=45^\circ), \end{aligned}\right\} \quad (2)$$

where $I(\psi, \chi)$ is the light intensity passing through the compensator and the analyzer for given values of the angles $\psi$ and $\chi$. It is easy to show[38] that

$$S_1=I, \quad S_2=Ip\cos 2\psi_0, \quad S_3=Ip\sin 2\psi_0, \quad S_4=Iq, \quad (3)$$

where I is the total intensity of the light beam, p is its degree of polarization, q is the so-called degree of ellipticity of the polarization, and $\psi_0$ is the angle of rotation of the direction of maximum polarization relative to the reference plane Q (Figs. 1 and 2).

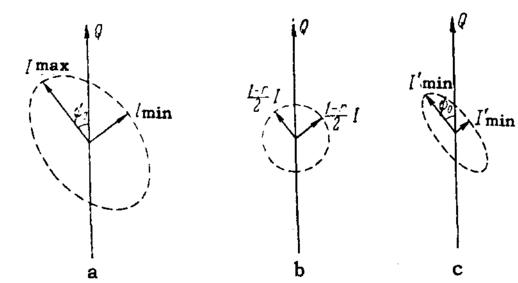



FIG. 2. a) Partially polarized beam
$I=I_{max}+I_{min}=I'+I''$, $p=\frac{I_{max}-I_{min}}{I}=rp'$, $q=rq'$.

b) Depolarized components
$I''=(1-r)I$.

c) Completely polarized component
$I'=I'_{max}+I'_{min}=rI$, $p'=\frac{I'_{max}-I'_{min}}{I'}$, $q'=\pm\frac{2\sqrt{I'_{max}-I'_{min}}}{I'}$

Generally speaking, an arbitrary partially polarized light beam of intensity I can be represented as the sum of two incoherent beams — a completely (generally speaking, elliptically) polarized beam of intensity rI and a completely depolarized beam of intensity $(1-r)I$ (Fig. 2). The quantity $r \equiv \sqrt{p^2+q^2}$ is the value of the polarization[48] or degree of homogeneity[44] of the light beam.

The four Stokes parameters $S_i$ (i = 1, 2, 3, 4) can be regarded as the components of a single Stokes vector-parameter $\vec{S}$ in four-dimensional functional space,[44,38] which materially simplifies the writing down of the formulas. Therefore, the different letter symbols for the Stokes parameters usually employed in foreign literature (for example, in reference 40) appear to us to be irrational, the more so since a universally adopted system of notation does not exist.

If the plane of reference is turned by an angle $\psi'$ in a counterclockwise direction (looking towards the ray) then the components of the Stokes parameter $\vec{S}$ change their values:

$$S_i' = \sum_j K_{ij}(\psi') S_j, \tag{4}$$

where the transformation matrix $K_{ij}$ has the form

$$K = \begin{pmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos 2\psi' & \sin 2\psi' & 0 \\ 0 & -\sin 2\psi' & \cos 2\psi' & 0 \\ 0 & 0 & 0 & 1 \end{pmatrix}, \tag{5}$$

and the quantities $S_1 = I$, p, q and r will be invariant relative to the transformation (5). For details on the properties of the Stokes vector-parameter, see reference 38.

The light ray undergoes attenuation in passage through the absorbing and scattering medium, and in the case of anisotropy of the medium for the scattering particles filling it, a change in the character of the polarization results. Both this and the other are recorded as the change of the vector-parameter of the light beam in its passage through an element of f path:[38]

$$dS_i = -\sum_j \varkappa_{ij} S_j \, dl, \tag{6}$$

where $\kappa_{ij}$ (i, j = 1, 2, 3, 4) is the so-called extinction matrix. In the case of an isotropic medium, it degenerates into a scalar — the attenuation (extinction) coefficient k:

$$\varkappa_{ij} = k\delta_{ij}, \tag{7}$$

where $\delta_{ij}$ is the Kronecker symbol (for details see reference 39).

We now turn to the description of the event of light scattering. We shall assume that the element of volume dV is irradiated in the direction $\mathbf{l}^0$ by a light beam with Stokes vector-parameter $\vec{S}_0$, and we shall consider the light beam scattered by the element of volume dV in a certain direction $\mathbf{l}$ (Fig. 3). As a

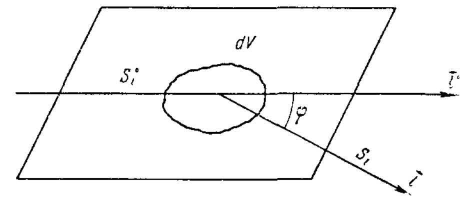



FIG. 3

reference plane Q both for the scattered and for the scattering beams, we shall take the plane of scattering which includes both the directions $\mathbf{l}^0$ and $\mathbf{l}$. Then, from the linearity of the equations of electrodynamics, and from the additivity of the Stokes vector-parameters for incoherent light beams, it follows[37,38,42-44] that the components of the Stokes vector-parameter of the scattering ($\vec{S}$) and radiating ($\vec{S^0}$) beams are connected by the relation

$$dS_i(\mathbf{l}) = \frac{1}{r^2} \sum_j D_{ij}(\mathbf{l}, \mathbf{l}^0) S_j^0(\mathbf{l}^0)\, dV, \tag{8}$$

where r is the distance of the point of observation from the scattering element of volume, and $D_{ij}$ are the components of a matrix of fourth rank which characterizes the scattering properties of the medium, independent of the state of polarization of the scattered light and referred to unit volume. We shall call it the light scattering matrix of the medium: its component $D_{11}$ is called coefficient of directed light scattering or the differential transverse scattering cross section. If the scattering medium is isotropic (for example, as a result of a random distribution of orientation of anisotropic particles), then the components of the scattering matrix $D_{ij}$ depend not on the directions $\mathbf{l}^0$ and $\mathbf{l}$, but only on the scattering angle $\varphi$ between them (Fig. 3). In this (and only in this!) case is it possible[37-40] to introduce the concept of a scattering coefficient of the medium $\sigma$ (or its total scattering cross section), independent of the state of polarization of the scattered light, by defining it as the fraction of the intensity of the light wave (referred to unit volume) incident on the scattering volume dV, which is scattered by the latter in all directions:

$$\sigma = \oint D_{11}\, d\omega, \tag{9}$$

where $d\omega$ is the element of solid angle of the scattered light beam. Then, in place of the matrix $D_{ij}(\varphi)$, we can introduce the normalized scattering matrix $f_{ij}(\varphi)$:

$$D_{ij}(\varphi) = \frac{\sigma}{4\pi} f_{ij}(\varphi), \tag{10}$$

where the component $f_{11}(\varphi)$, which satisfies the normalization condition

$$\frac{1}{4\pi} \oint f_{11}(\varphi)\, d\omega = 1, \tag{11}$$

in agreement with (9), is usually called the scattering function or indicatrix.

The energy carried away by the particles (or by the medium) from the wave irradiating it is not only scattered but is also partially absorbed, being transformed into other forms. It is evident that the fraction of the incident light intensity (referred to unit volume) absorbed by the medium, i.e., the coefficient (or cross section) of absorption $\alpha$ of the medium is determined as the difference of the attenuation and scattering coefficients

$$k=\alpha+\sigma, \tag{12}$$

where $\alpha$ will be a scalar, generally speaking, only in an isotropic medium. We note that all three quantities in (12) have the dimensions of $cm^{-1}$ or more conveniently, $km^{-1}$ (the cross section referred to unit volume).

It is very important that the quantities (matrices) $\alpha$, $\sigma$, and k be expressed in terms of the components of the scattering matrix $D_{ij}(\varphi)$ and the extinction matrix $\kappa_{ij}$, as a consequence of which the spectral and angular dependences of all 16 + 16 = 32 components of these matrices exhaust all the information which can be obtained about the properties of the medium by studying the phenomena of scattering, absorption of light, or other radiation. Nevertheless, preference is usually given to direct measurements of the extinction coefficient k and, in certain cases, of the specific absorption coefficient $\beta = \alpha/\sigma$ (for further details, see reference 39).

Often it is convenient to consider the reduced scattering matrix in place of the matrices $D_{ij}$ or $f_{ij}$; the components of this matrix, $\tilde{f}_{ij}$, are given by the relation

$$\tilde{f}_{ij}(\varphi)=\frac{D_{ij}(\varphi)}{D_{11}(\varphi)}=\frac{f_{ij}(\varphi)}{f_{11}(\varphi)}. \tag{13}$$

The form of the scattering matrix $D_{ij}(\varphi)$ depends essentially on the properties of the scattering medium, in particular on the composition, dimensions, form and orientation of the particles suspended in it. The character of the anisotropy of the medium or the symmetry of the scattering particles is directly reflected in the number of independent and non-vanishing components of the scattering matrix.[40,43,44,49] However, the concrete form of the scattering matrix is known only for molecular scattering. For non-absorbing gases, with corrections for the anisotropy of the molecules in correspondence with the theory of Rayleigh-Cabanne (see Sec. 5), it has the form:[44,38]

$$f(\varphi)=\frac{3}{4+3d}\begin{pmatrix}1+\cos^2\varphi+d & -\sin^2\varphi & 0 & 0\\ -\sin^2\varphi & 1+\cos^2\varphi & 0 & 0\\ 0 & 0 & 2\cos\varphi & 0\\ 0 & 0 & 0 & 2\cos\varphi\end{pmatrix}, \tag{14}$$

where the scattering coefficient (as the result of the absence of absorption, $k = \sigma$) is equal to*

$$\sigma=\frac{8\pi^3(n^2-1)^2}{\lambda^4 N}\,\frac{4+3d}{12-d}. \tag{15}$$

*In reference 38, the factor 1/2 is incorrectly omitted in Eq. (57).

Here $\varphi$ is the scattering angle, N is the number of molecules per unit volume, n is the index of refraction of the medium, $\lambda$ is the wavelength, $d = 4\Delta/(1-\Delta)$, and $\Delta$ is the depolarization of the scattered light at $\varphi = 90°$ and exposure of the scattering volume to linearly polarized light with an electric vector perpendicular to the plane of scattering.* It is essential that the plane of scattering is chosen as the plane of reference both for the incident and for the scattered beams.

In the case of scattering by spherical particles (Mie scattering), retaining the plane of scattering as the plane of reference Q for the incident and scattered beams, we have

$$f(\varphi)=\begin{pmatrix}f_1(\varphi) & f_2(\varphi) & 0 & 0\\ f_2(\varphi) & f_1(\varphi) & 0 & 0\\ 0 & 0 & f_3(\varphi) & f_4(\varphi)\\ 0 & 0 & -f_4(\varphi) & f_3(\varphi)\end{pmatrix}, \tag{16}$$

i.e., the scattering matrix contains only four independent components. Both the scattering coefficient $\sigma$ and the components of the scattering matrix are shown to be very sensitive to the wavelength, and the functions $f_i(\varphi)$ depend in complicated fashion on the scattering angle. The Mie theory makes it possible in principle to compute $\sigma(\lambda)$ and $f_i(\varphi, \lambda)$ for particles of a given dimension and with a given index of refraction. However, these calculations are actually so cumbersome that, in spite of the use of mathematical computers, they have been carried out only for a few cases, mostly in the absence of absorption and exclusively for $\sigma$ (or k) and $f_1(\varphi)$ [in a few cases also for $f_2(\varphi)$]. As far as the functions $f_3(\varphi)$ and $f_4(\varphi)$ are concerned, they have never been computed up to the present time. It must also be kept in mind that qualitative discussions in this region, and also interpolation of computed data, must be used with extreme care in view of the very capricious character of the change of most of the computed quantities.

In addition to scattering by spherical particles, the problem of scattering by elliptical particles has been considered in principle. Other forms of particles have not yet been amenable to the theory. Further details on the character of the scattering of light on solid particles can be found in references 40, 50.

## 4. ATMOSPHERIC TRANSMISSION AND AEROSOLS

The transmission in the atmosphere in various parts of the spectrum is among its important optical characteristics. It determines the conditions of visibility of distant objects and, generally, the lighting conditions of our existence. On it also depend the radiative and thermal conditions both on the surface of the earth and in the atmosphere. It is not surpris-

*Frequently the quantity $\rho = \frac{2\Delta}{1+\Delta}$ is introduced in place of $\Delta$ in the equations, i.e., depolarization at $\phi = 90°$ and irradiation by natural light, which is sometimes also denoted by $\Delta$.

ing that, from the time of Bourguer and Lambert, who first definitely formulated the problem, and Saussure, who first attempted observations, a great deal of effort has been spent on measurements of atmospheric transmission. Setting aside the extensive and practically important group of operational and climatological problems, which go beyond the limits of this article, we consider the material accumulated during the course of two centuries from a single point of view only, namely, knowledge of the properties of the atmosphere itself.

The widest category of investigations is made up of the numerous observations and measurements carried out by meteorologists in the hope of getting some sort of connection between transmission and weather, and in this fashion to make it possible to forecast the latter. Lacking any sort of theoretical foundations and, as a rule, limited only to surface correlation estimates on the basis of comparatively short time observations, carried out only under favorable meteorological conditions, these measurements bore no fruit which would be of interest from our point of view. In addition to a certain quantity of curious observations, lost in a wide and varied collection of various cases not amenable to scientific analysis, one can extract from them only the conviction of the extremely and universally uncontrolled variability of the optical state of the atmosphere, even on completely clear days.

Such a paucity of results also exists for the repeated measurements of transmission, carried out by astronomers with the purpose of studying interferences which take place in their investigations of the earth's atmosphere. In spite of well-known facts, the variability of the atmosphere has generally been ignored in these measurements, and efforts have been directed toward the determination of some average (at best, seasonal transmission on clear days) somehow characteristic for a given observatory, i.e., quantities having no real importance, as we shall see below.

Against this background, which is most heterogeneous in purposes and methods of investigation, there stand out a comparatively small number of researches which are clearly directed toward the investigation of the optical properties of the atmosphere itself, and which are distinguished by their clearly thought out methodology and the perfection of the apparatus employed. The framework of this paper does not permit us to give a full account of each of these researches which left a notable mark in science. Below we shall only sketch rapidly contemporary information relating to transmission in the atmosphere and note some conclusions following from it. Here we shall put aside the broad and self-contained problem of the selective absorption of the gaseous phase of the atmosphere, including also its impurities, such as water vapor, ozone, and carbon dioxide.

We shall also not concern ourselves with the rather difficult and still far from satisfactory solution of the problems of methodology nnd technology of carrying out transmission measurements (see, for example, reference 51 and the literature cited there). We shall only remark that one usually measures either the attenuation coefficient k, averaged over a long or short distance (usually horizontal) or the vertical optical thickness of the atmosphere above the head of the observer (who is located at an altitude h)

$$\tau(h) = \int_h^\infty k(h)\, dh, \tag{17}$$

which is connected with the vertical transmission of the atmosphere at the same level by the relation

$$P(h) = e^{-\tau(h)} \tag{18}$$

and with the oblique transmission at an angle $\zeta$ to the zenith by the relation

$$T(\zeta, h) = P(h)^{m(\zeta, h)}, \tag{19}$$

where $m(\zeta, h)$ is the so-called air mass, which is accurately described (for not too large $\zeta$) by the formula

$$m = \sec \zeta. \tag{20}$$

In place of $\tau$, one frequently uses a quantity proportional to it, the vertical optical density of the atmosphere: $D = 0.43\tau$. We note that Eqs. (17) — (19) are valid only for monochromatic light or under conditions of the invariance of k within the measured wavelength interval.

The discovery of molecular light scattering raised the problem of determining the optical thickness of the earth's atmosphere, which is evaluated by means of this phenomenon. In fact, in accord with the Lorentz-Lorenz formula (see, for example, reference 52, page 42) for gases,

$$(n^2 - 1) = 4\pi N\left(\alpha + \frac{P_0^2}{3kT}\right), \tag{21}$$

where $\alpha$ is the polarizability of the molecules and $P_0$ is their constant dipole moment. Denoting by $N_0$ Loschmidt's number and by $\rho_0$ and $n_0$ the density and the index of refraction of air under normal conditions, and assuming that the composition of the air remains constant at least up through those altitudes where the attenuation of light (as a result of scattering) remains appreciable, we find that at any altitude

$$N = N_0 \frac{\varrho}{\varrho_0}, \quad (n^2 - 1) = (n_0^2 - 1)\frac{\varrho}{\varrho_0}. \tag{22}$$

Making use of Eq. (15), and taking it into account that the pressure at the point of observation is given by $p = g\int_h^\infty \rho\, dh$, we find, in accord with Eq. (17),

$$\tau(h) = \frac{8\pi^3 (n_0^2 - 1)^2 p}{\lambda^4 N_0 \varrho_0} \frac{4 + 3d}{12 - d}. \tag{23}$$

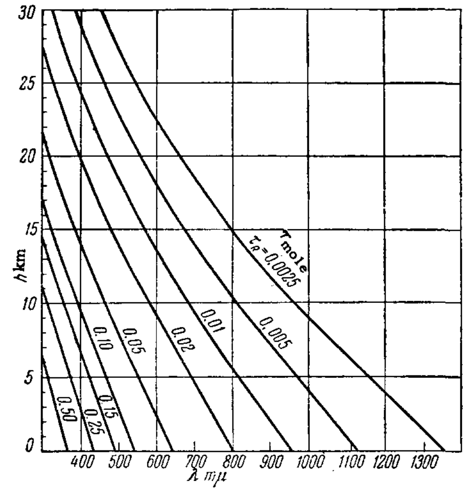
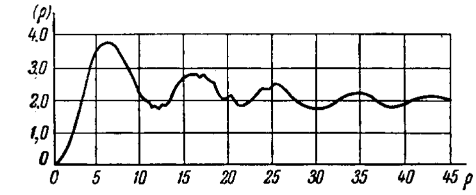


FIG. 4. Lines of equal optical thickness for an ideally pure atmosphere in altitude vs. wavelength coordinates.

Corresponding values of $\tau$ as a function of the wavelength and altitude of the point of observation, according to the calculations of Penndorf,[53] are shown in Fig. 4, which takes into account contemporary data on the mean altitude variation of the atmospheric pressure. From (23) and from the absence of correlation between the value of the transmission and pressure at a fixed altitude of the point of observation, the conclusion immediately follows that aerosols, which are always present in some measure in the air (as we now know, at all altitudes at least up to 80 km), are almost completely responsible for the optical variation of the atmosphere (see below).

However, if we choose days with especially high transmission, then we can expect fulfilment of Eq. (23). In the early years of this century, the least accurate quantity in this formula was Loschmidt's number and, therefore, attempts were undertaken to determine it from this equation (without account of corrections then unknown for the anisotropy of molecules — see Sec. 5). Early calculations, both of Rayleigh and of Kelvin (1902), showed agreement that was completely satisfactory for that time (according to Kelvin, $N_0 = 2.47 \times 10^{19}$ instead of the actual value $2.67 \times 10^{19}$), which served not only as a first proof of the validity of the Rayleigh hypothesis on molecular light scattering, but was also a strong argument in favor of the molecular kinetic picture generally (see reference 54). However, a more careful analysis of the data (in particular, the excellent observations of the Smithsonian Institution over a period of many years) showed that atmospheric air was never free from aerosols, and that it was further necessary to introduce corrections for the selective absorption of the gas phase. Introduction of these corrections made it possible to improve the agreement appreciably — the error was lowered by Cabanne to ~ 8 per cent, by Kew to ~ 2 per cent, by Vassy to 1 per cent and recently by Toropova to ~ 0.5 per cent. At the same time it was shown that by com-

FIG. 5. The function $K(\rho)$ for drops of water (index of refraction 1.33).



puting the part caused by molecular scattering and selective absorption of the gas phase from the total optical thickness (or the attenuation coefficient k) one could correctly determine the absorption due to the aerosol.

Such a separation of the aerosol components of k or $\tau$, carried out by a number of authors, immediately brought to the forefront its variability both in absolute value and in spectral dependence. According to the Mie theory for spherical particles* $k = N\pi a^2 K(\rho)$, where N is the particle concentration and a is thin radius, $\rho = (2\pi a/\lambda) m$, and m is the relative index of refraction of the matter composing them. In particular, for water droplets (without consideration of dispersion) the function $K(\rho)$ has the form[55] shown in Fig. 5. It is at once evident that the spectral variability of atmospheric transmission is connected with the change in the character of the particles suspended in the air, particularly, the change in their dimensions. As the measurements of Vassy,[56] Rodionov[57] and others (for details, see reference 51) have shown, the aerosol attenuation of light often has a sharply expressed selective character. In the case of a semidispersed aerosol, which we usually encounter under ordinary conditions, the selectivity of the attenuation should be more or less smoothed out. If we average the data of measurements of different authors or of a single author for a series of days, then the aerosol attenuation of light is found to be independent of the wavelength. Since the aerosol is added to the air, which scatters in proportion to $\lambda^{-4}$, its effect is seen to be much greater in the long-wave than in the short-wave region of the spectrum. Therefore, under ordinary conditions, the spectral dependence of k or $\tau$ is weaker than follows from the Rayleigh law (the sky has a whitish light), and can sometimes be approximately expressed in the form $k \sim \lambda^{-n}$, where n varies from zero to 2 or 3.

On cloudless days, the attenuation coefficient of atmospheric air generally increases with increase in altitude. At low altitudes (up to 0.5 — 1 km), this falling off takes place rapidly, chiefly as a result of the decrease in concentration of dust or water droplets, raised up by local air streams flowing over the earth. This decrease is then rapidly slowed and sometimes even changes to a weak increase up to an altitude of 3 — 5 km, i.e., approximately to the upper boundary

*In the visible portion of the spectrum, cooperative effects (see reference 39) which can alter the formula given for k are not important for aerosols, and for molecular scattering play a role only at altitudes of around 100 km.

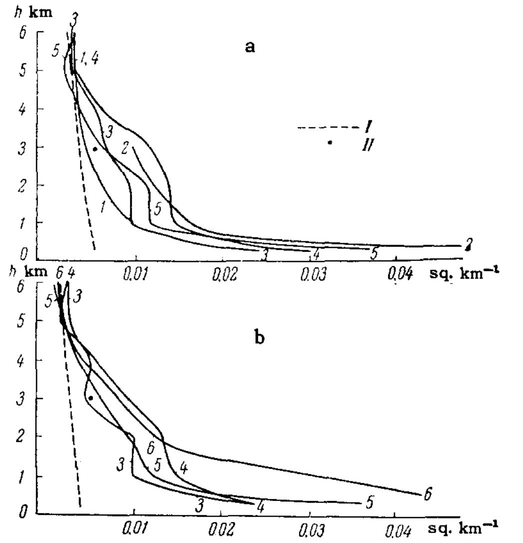


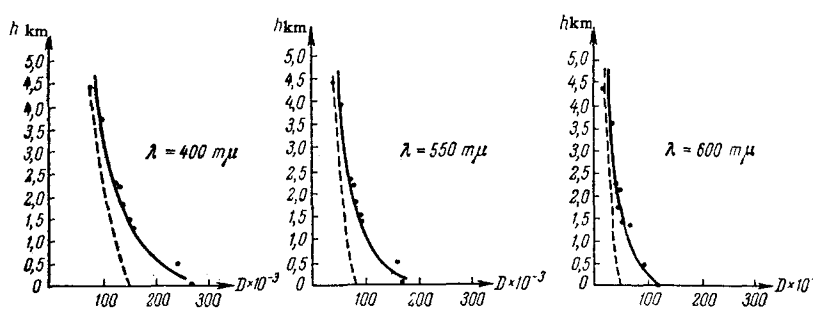

FIG. 6. Mean values of the attenuation coefficient k at different altitudes. 1 – Moscow oblast' (1956-57), 2 – Northern Kazakhstan (1956), 3 – Khar'kov oblast' (1957), 4 – Northern Caucasus (1957), 5 – Moscow oblast' (1958), 6 – West Germany (1944), I – Absolutely pure atmosphere, II – on the slopes of Elbrus (1957).

of the convective layer. Beyond that point, k decreases with altitude on the average, approximately according to the barometric formula, with frequent and very changeable disruptions as a result of aerosol (clouds) layers. As an example, we give the data obtained by Faraponova for measurements with an airplane[58] (Fig. 6). For measurements on mountain slopes, the altitude dependence is shown on the average to be more gradual than in the free atmosphere, without protruberances in the 3 — 5 region (Fig. 7), which makes it possible to explain the effect of the spreading out of the surface and mountain-valley circulations.[51] It remains to add that in the absence of clearly expressed cloud layers, the relative concentration of aerosols above 3 — 5 km varies but little with altitude and that on especially clear days the share of the total light attenuation of the atmosphere due to aerosols amounts to 30 — 50 per cent, which also explains the success of the first determinations of Loschmidt's number.

Aerosols, which include the liquid-drop phase of water, belong to the most variable component of the atmosphere both in a quantitative and in a qualitative sense. Numerous measurements, both of the transmission and the brightness and polarization of the daytime sky (see Secs. 5 and 6) testify to the fact that the aerosol is distributed in the atmosphere not in homogeneous layers but in the form of separate flocculent accumulations, which are carried along by the wind and which undergo constant qualitative changes, including the results of condensation processes. Therefore, the picture of a horizontally homogeneous atmosphere does not correspond to reality even on very clear and calm days. This picture finds its clear expression, in particular, in the variability already noted in the transmission with respect to time, in the dependence on the direction (azimuth) of observation, and in the displacement along the earth's surface.[51] The absence of correlation between the variability of the transmission at different points in the spectrum is quite characteristic, and supports not only the quantitative but also the qualitative variability of the aerosol. These effects are exhibited in Fig. 8.[51] No less characteristic is the absence (in spite of the prevailing point of view) of correlation between horizontal and vertical transmissions of the atmosphere.[51]

If we now return to cases of strong turbidity of the air — clouds, fogs, precipitation, etc, then the given observations present a still more varied picture, which finds its natural explanation in the extreme variation of the microstructure of these formations. In general, there is now no doubt that all the effects observed here lie within the framework of the theory advanced by Mie (see, for example, references 40, 50, 59-61). However, as a result of the characteristic semi-dispersed character of the water aerosol and the varied nature of its size distribution, the observed phenomena lend themselves to quantitative analysis only with difficulty. In this case the chief role is played by barriers of two kinds. Firstly, the calculations of attenuation coefficients by the Mie theory far from cover all cases which one encounters in practice in water droplets. In particular, data referring to water absorption bands are almost completely absent, while very appreciable and peculiar anomalies are known to exist.[62] In the second place, we have no reliable data on the sub-microscopic fraction of water droplets, which is most active in optical behavior. Moreover, measurements under conditions of very high turbidity are complicated by effects

FIG. 7. Dependence of the mean vertical optical density of the atmosphere on altitude above sea level according to the measurements of various authors on the slopes of mountains. Dashed line – dust-free atmosphere.



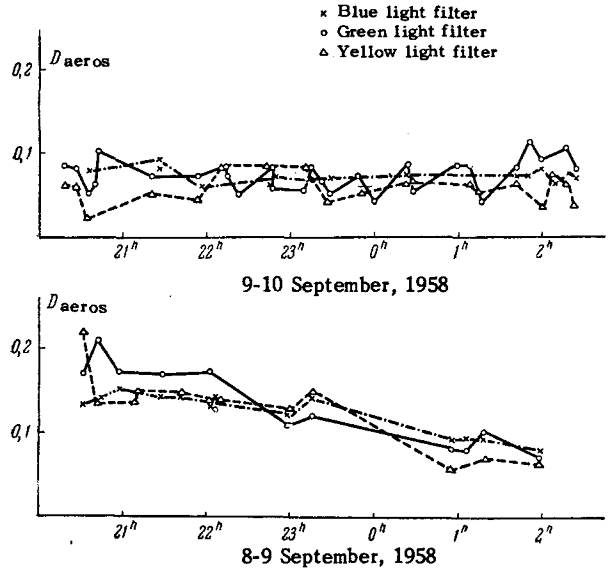



FIG. 8. Examples of variation of aerosol component of vertical optical density of the atmosphere in different parts of the spectrum.

of multiple light scattering, which are difficult to separate. Therefore, numerous efforts directed toward the use of data on transmission for the determination of the microstructure of clouds and fog or of atmospheric aerosols in general have not yet led to expected results, although they have created confidence in the significance of this problem.

Such, in general terms, are the results of 200 years of investigation of the transmission qualities of the atmosphere in the visible portion of the spectrum (for details, see references 40, 50, 51). We shall now consider what can be extracted from the data on the illumination of daytime sky.

## 5. THE POLARIZATION PICTURE OF THE SKY; THE ANISOTROPY OF MOLECULES AND MULTIPLE SCATTERING

The observation of Arago not only made possible the discovery of light scattering, but also had a permanent value in itself. At the same time, it raised questions whose solution required all the power of the physics of the twentieth century and directly stimulated the development of two of its important sections — molecular optics and the theory of transfer of radiation in turbid media.

Having discovered the polarization of the light received from the firmament, Arago went on to plot the polarization map of the sky and developed almost all its characteristic peculiarities. He established the fact that the maximum polarization is observed at an angle of about 90° to the direction of the sun's rays, and that (expressing it in our modern language) the plane of polarization coincides with the plane of scattering ("positive" polarization). He also discovered (in the vicinity of an anti-solar point) a region where

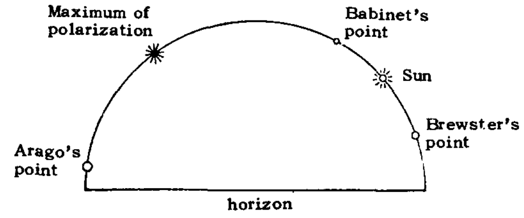



FIG. 9. Position of the neutral points and maximum polarization in the vertical of the sun.

the plane of polarization is perpendicular to the plane of scattering ("negative" polarization), while at the boundary of these two regions a "neutral point" is located in the vertical of the sun, from which completely depolarized light emerges. A similar picture is observed at night in moonlight, as was established by Arago, and confirmed 80 years later by Cornu and Pilchikov. The magnitude of the maximum polarization and the position of the neutral point, according to the observations of Arago, depend strongly on the state of the atmosphere, while sometimes the neutral point is shifted in the direction of the normal of the sun. In 1840, Babinet added still another neutral point to this picture, located not far from the sun, and later Brewster discovered a third neutral point located under the sun (Fig. 9).

Subsequent observations, pursued diligently up to our own day, have merely confirmed and made somewhat more accurate this qualitatively correct picture, and have added to it almost nothing new, although among the investigators we meet (in addition to the above) such names as Becquerel, Weber, Wild, Jensen, Forno, Süring, Pernter, Tikhanovskiĭ, and many others. Perhaps the only discoveries which contain points of importance were: 1) the observation by Pilchikov[63] (1892)(which is completely unnatural from the point of view of the Rayleigh theory) of the dependence

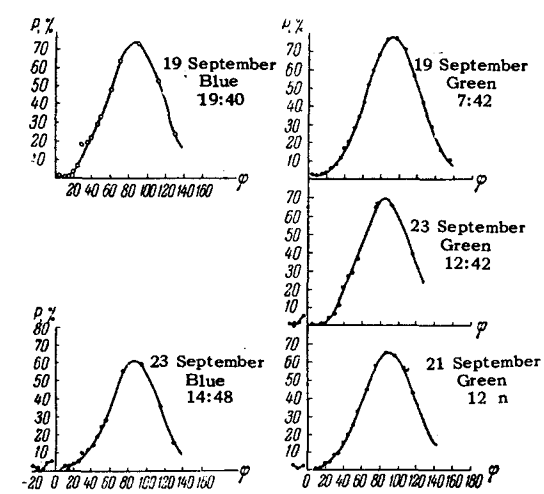



FIG. 10. Examples of the dependence of the degree of polarization of the illumination of the daytime sky on the angle of scattering on different days.

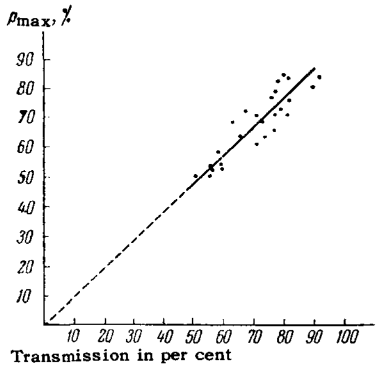



FIG. 11. Correlation between the maximum polarization of light of a daytime sky and the vertical transmission of the atmosphere.

of the degree of polarization on the wavelength ("dispersed polarization" in the terminology of Schirmann), which was subsequently confirmed by the observations of Tikhanovskiĭ and Pernter, and also 2) the discovery by Soret, Dorno, MacConnell and others of the effect of the albedo of the earth's surface on the polarization. One must also note the observation of Rubenson[68] (1864), confirmed later by Wild and Jensen, that the position of the maximum polarization, as a function of the weather, can be displaced somewhat along the vertical to the sun both in the direction of lower and in the direction of higher angles. As an illustration, a typical dependence of the degree of polarization on the scattering angle along the vertical to the sun, according to the data of Rozenberg and Turikov, is given in Fig. 10, while Fig. 11 gives the dependence obtained by the same authors of the degree of polarization at the maximum on the vertical transmission of the atmosphere.[51] The latter drawing shows that, in spite of the appreciable individual variations, there is a clearly expressed correlation, according to which the degree of polarization at the maximum is equal to the vertical transmission of the atmosphere, independently of the altitude of the sun, which is in good agreement with the observations both of Arago himself and also of all subsequent investigators of the sky. It is not possible for us to linger on subsequent details, and we must refer the readers to the excellent reviews of Jensen[64] and Dorno,[65,66] particularly because little has changed in this field since their time. Reviewing the observational material, it remains to be said that in the main it was re-obtained in searches for empirical connections with the weather and, in spite of its immensity, it presents no interest for subsequent analysis. As a conspicuous example, we must introduce the researches of Roggenkamp,[67] who during his life carried out more than 15,000 sets of the position of the neutral points in the twilight hours (i.e., 15,000 evenings!) and carefully obtained from them . . . the arithmetic mean.

We saw above that the experiments of Govi and Tyndall on the scattering of light in colloids discovered practically all of the peculiarities noted by Arago in the polarization of the light of the daytime sky, and that precisely these peculiarities strengthened the conviction in that day of the identity of the two phenomena. On this point Rubenson in 1864 directly pointed out[68] that one must see the reason for all the variations of the polarization of the illumination of the daytime sky in the contamination of the air by dust and water droplets. However, with the appearance of the theory of Rayleigh, the general character of the picture was so well explained, and all other points of view being decisively relegated to second place, these peculiarities were regarded as surprising anomalies, requiring for their explanation the point of view of molecular scattering.

The pioneer in these searches was one of the most active students of the polarization of the illumination of the daytime sky, Soret,[69] who advanced in 1888 an idea that was completely new for his time on the possibility of multiple scattering of light (by analogy with the idea advanced by Muller[22] of its multiple reflection), and who attempted to apply it to an explanation of the existence of the neutral points. In essence this was first worked out in the region of the transfer of radiation into the scattering medium, showing, in spite of its extreme roughness of calculation, the serious effect on the subsequent form of all problems on the whole. The first equations of radiation transfer were quickly formulated by Khvol'son), and also by Schwartzchild and Schuster. They first started development of this branch of knowledge within the framework of

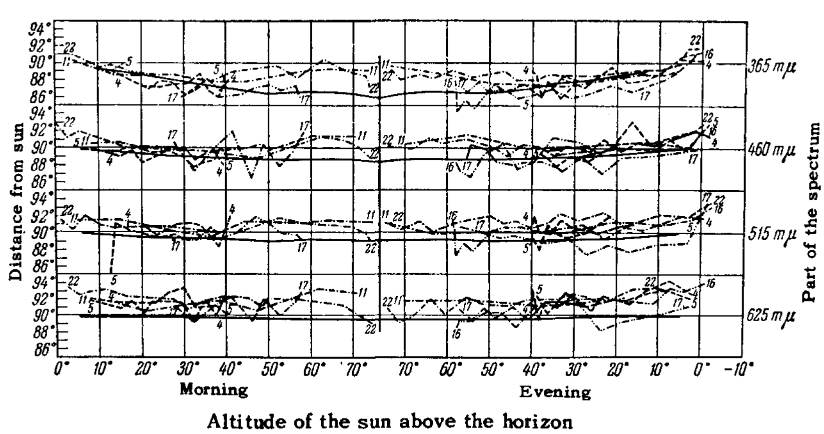



FIG. 12. Real and observed displacements of the position of the maximum polarization in different parts of the spectrum according to observations at different points and on different days. The solid line – theoretical expectation for the Rayleigh scattering and albedo of the earth's atmosphere 0.25.

mathematical physics. However, in the initial formulation, there were only the equations of energy transfer, not taking into account polarization effects and of Soret's problem unsuitable for solution. Therefore it was again necessary for its solution to fall back on homemade methods (although more reliable than those used by Soret). This was done in 1914 by Ahlgrimm[70] and in 1927 by Tikhanovskiĭ,[71] who by means of long and complicated calculations showed that the idea of Soret was correct, and that actually secondary scattering of light in the atmosphere leads to the appearance in the sky of neutral points and to a certain $\tau$-dependent decrease in the degree of polarization at the maximum. Here, the dispersion polarization discovered by Pilchikov found its explanation.

At that time an event took place which directed the thoughts of investigators along another path. In 1917-1918, Max Born advanced the idea of the anisotropy of molecules.[72] Simultaneously, Strutt (Rayleigh) the younger discovered[73] that in light scattering in vapors and gases the polarization is never complete in observations at a scattering angle $\varphi = 90°$. The latter phenomenon especially attracted attention in connection with the picture of the polarization of the sky, and in 1921 Cabanne[74] explained it by starting out from the idea of Born, and improved the Rayleigh theory by introducing in it an account of the anisotropy — see Eqs. (14) and (15) (see also references 52, 29, and 75). However, the correction of Cabanne did not solve the problem. For pure air it led to a value of the degree of polarization at $\varphi = 90°$ equal to 92 per cent, in place of the 84.5 per cent known from the data of Tikhanovskiĭ as the maximum observed in the atmosphere (see Fig. 14). Even account of secondary scattering with the Cabanne correction, carried out in 1927 by Tikhanovskiĭ, did not correct the situation. Even more incomprehensible was the effect which the weather had not only on the value of the polarization but also on the position of the neutral points. This stimulated Schirmann and Milch[76] to return to the hypothesis of Rubenson,[68] that one must seek the reason for polarization effects in the scattering of light by large particles, in correspondence with the theory of Mie, which was already well developed at this time. However, this attempt remained within the realm of abstract discussion, because concrete calculations of the polarization of light scattered by such particles are very few even today, and 20 years ago there were almost none at all.

The next step could be taken only when in 1946, the equation of radiation transfer, taking into account the polarization of the latter was formulated independently by Chandrasekhar[45] and Rozenberg[44] (see references 21 and 38). This matrix equation, now so widely used in atomic physics, was formulated by these authors, and also by V. V. Sobolev,[46] in connection with problems of polarization of light emitted from the sky. This formulation was shown to be possible only by use of the Stokes vector-parameter and the scattering matrix, and immediately made it possible to increase appreciably the number of problems that could be solved. In particular, it was clearly established that the polarization effects have an important influence not only on the polarization but on the intensity of multiply scattered light, even in the depth of the scattering medium.[49] At the same time, it was made clear that the calculations of Soret, Ahlgrimm, and Tikhanovskiĭ contained methodological errors. However, qualitative calculations on the rough scheme of Soret, carried out by Rozenberg,[77] showed that these errors did not change the general picture, and that the neutral points and their position in the sky are explained by secondary scattering. Finally, in 1954, Chandrasekhar and Elbert[78] published detailed tables of the brightness and polarization of the daytime sky at different altitudes of the sun under the assumption of purely molecular scattering. These tables, which were the result of careful solution of the rigorous equation of transfer for different assumptions on the albedo of the earth's surface essentially completed the solution of the problem of the dust-free atmosphere.

Sekera[79] quickly carried out careful investigations of the polarization of daylight sky by means of an automatic polarimeter, and established the fact that on average the picture drawn by Chandrasekhar and Elbert did not correspond at all badly with reality, but that the agreement again bore only a semi-quantitative character. In addition, systematic departures from this picture and also random variations, including short period ones are definitely observed. As an example of this we have the wandering of the maximum of the polarization for different parts of the spectrum, shown in Fig. 12. Recently Lipskiĭ,[80] measuring the polarization of the light of the daytime sky with very high angular and spectral resolution, also discovered fast and deep-seated vibrations of the degree of polarization, which were not correlated for different wavelengths. This leaves no doubt of the validity of the point of view already expressed by Rubenson, and also by Schirmann, that atmospheric aerosols are responsible for all variations of the polarization of the daytime sky light, that these aerosols are extremely variable, both quantitatively and qualitatively, and that their role in the scattering of the light by the atmosphere is never small. Inasmuch as the effects of secondary scattering are also not small, the division of the effects of molecular and aerosol scattering in the daytime sky is not possible, as Sekera[77] emphasized, because of the presence of nonlinear effects which cannot be neglected even at very high transmission. For the same reason, it is impossible to draw any reliable information from the data on the illumination of the daytime sky on the scattering properties of air in every case, inasmuch as one is dealing with such a fine-grained effect as polarization, i.e., with the components $f_{21}(\varphi)$ and $f_{31}(\varphi)$ of the scattering matrix — see Sec. 3. Such is the some-

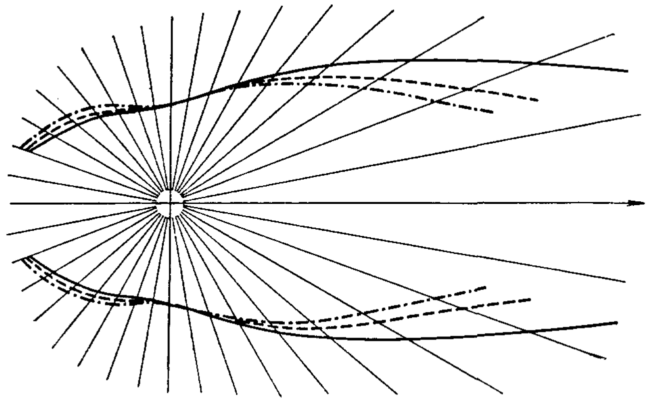


FIG. 13. Examples of the atmospheric indicatrices of scattering found from measurements on the brightness of the daytime sky.

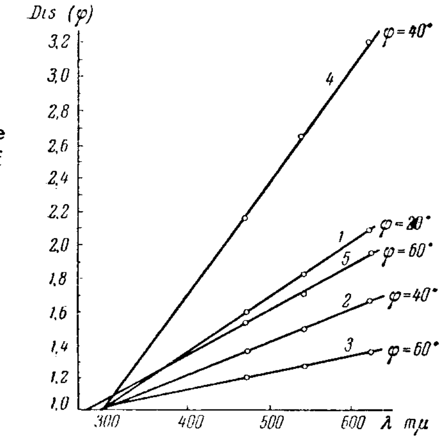



FIG. 14. Increase of the dissymmetry of the indicatrices of scattering with increase in the wavelength: 1, 2 and 3 – for atmospheric air; 4 and 5 – for aerosol component.

what unexpected result of the very intensive investigations over a century and a half, which as we have seen exerted an important effect on the development of optics and made possible a whole series of important discoveries.

## 6. THE BRIGHTNESS MAP OF THE SKY AND THE SCATTERING FUNCTION

Systematic measurements of the brightness of the daytime cloudless sky were begun in 1898 by Jensen[64] and continued to our own day by many authors. Especially broad and long-term observations were completed by Dorno[65] and in recent years by Pyaskovskaya-Fesenkova.[81] As a result of this, a very rich collection of brightness pictures of the sky has been accumulated under different meteorological and geographic conditions. However, the first comparisons of these pictures with the Rayleigh theory led to a remarkable result. If the polarization and the coloring of the sky corresponded approximately to theoretical expectations, then the angular distribution of the brightness had nothing in common with them. The idea then arose of using these data for experimental determination of the scattering function $f_{11}(\varphi)$ of atmospheric air, the more so since other ways for this measurement were not seen at that time. In broad outline (for details, see reference 81), the idea was the following. If one measures the brightness of the sky at different azimuths, but always along the almucantar of the sun (that is at points located below the horizon at the same angle as the sun is above it), then the attenuation of the sun's rays on their path to the scattering region and beyond it to the eye of the observer will be the same, and the difference in brightness will not be brought about by the angular dependence of the component $f_{11}(\varphi)$ of the scattering matrix. In this case, naturally, it is assumed that the secondary scattering does not greatly distort the picture, and that the atmosphere is homogeneous in the horizontal direction.

Furthermore, since the properties of the atmosphere change with altitude, the data obtained on the form of the scattering function $f_{11}(\varphi)$ will be certain weighted averages over the altitude.[82] A detailed analysis of data obtained in this fashion is contained in reference 81 (see also reference 51), and we shall limit ourselves to a certain amount of fundamental information.

First of all, it has been pointed out that the scattering indicatrices (functions) are extremely strongly elongated in the forward direction and terminate in a diffraction "nose," which is characteristic of the Mie theory, and which is responsible for the aureole around the sun. Examples of observed indicatrices[81] are shown in Fig. 13. It has been further shown that the form of the indicatrix is very sensitive to change of atmospheric conditions, not only in the region of small and large angles of scattering, especially in the region of the aureole, since this sensitivity is much less in the region of medium angles of scattering. Finally, it was discovered[81] that the prolateness of the indicatrix increases with increase in the wavelength, which is illustrated in Fig. 14,[81] where the so-called dissymmetry of the scattering function

$$Dis(\varphi) = \frac{f_{11}(\varphi)}{f_{11}(\pi - \varphi)}$$

is plotted along the ordinate. As Kastrov[83] has shown, this is the result of the decrease of the relative role of molecular scattering (or scattering by the submicroscopic fraction) as the wavelength increases and the scattering angle decreases.

Thus the brightness map of the sky testifies unambiguously to the fact that the scattering in the atmosphere bears an essentially aerosol character. It was very unexpected to learn that this character of the scattering is preserved up to very high frequencies, as follows from Fig. 15, which was obtained by measurement of the brightness of the sky with the aid of an "autostratostat."[84]

The methodology of the theoretical calculation of the brightness of the sky under different conditions on the basis of a solution of the equation of radiation transfer in a scattering medium was first developed by Kuznetsov,[85] and then by Chandrasekhar and Sobo-

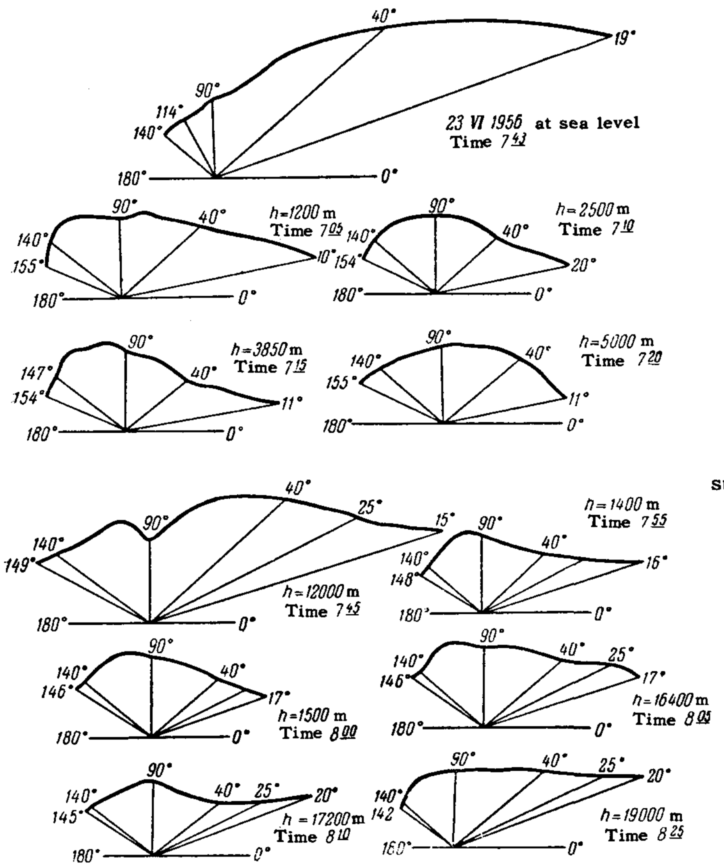



FIG. 15. Indicatrices of scattering of atmospheric air, measured by the brightness of the daytime sky at different altitudes.

lev,[21] unfortunately, without account of polarization effects. Making use of this method, a number of authors have carried out numerical calculations of the brightness map of the sky for different assumptions of the form of the indicatrix, and also the altitude dependence of the scattering coefficient, as a result of which we now have detailed tables covering a wide range of possible variations of these quantities.[86]

Analysis of these tables makes it possible, in particular, to estimate how much the effects of secondary scattering, and also reflections from the earth's surface, can distort the form of the scattering function drawn from data on the brightness of the daytime sky. It is shown[82] that these distortions are very significant as soon as the transmission of the entire thickness of the atmosphere becomes lower than approximately 85 percent, i.e., practically always, and do not decrease with increasing height of the observer. Therefore, it is not possible to draw anything except qualitative and highly tentative conclusions relative to $f_{11}(\varphi)$ from analysis of the light of the daytime sky. An exception is the case of extremely high transmission, which is less characteristic for the atmosphere. Besides, knowledge of the brightness and polarization maps of the sky and their variation has large importance by itself, both conceptually and in an applied sense, independent of the problem of the determination of the form of the scattering matrix of atmospheric air. For example, we recall that the polarization of the light of the daytime sky can be used for the purpose of orientation in space (bees, as is well known, orient themselves in just this fashion) and the illumination of our houses is determined by this brightness map.

Thus, we have shown that all the integral methods, based on the study of the illumination of the daytime sky, do not warrant placing any confidence in them and cannot supply us with trustworthy information on the form of the scattering matrix (we also add, and because this matrix is subjected to material and completely random changes with altitude and time). Therefore the naturally reliable method of study of the character of the scattering matrix and its variations is the method of local investigation of the scattering act by means of a directed light beam. However, the possibility of carrying out detailed measurements, which require the use of rather ideal lighting and observing means, has existed only in recent times, and measurements to date have been limited exclusively to the angular dependence of the component $f_{11}(\varphi)$, with no studies of either its spectral behavior or of the other components of the scattering matrix.

As was to be expected, there exists only a qualitative and very uncertain agreement among the (as yet) small amount of data of different authors. At sea level (and at the present time there is no basis for expecting [see Sec. 7] that the situation will be different elsewhere, at least up to altitudes of the order of 80 – 90 km) the indicatrices of scattering have a typical aero-

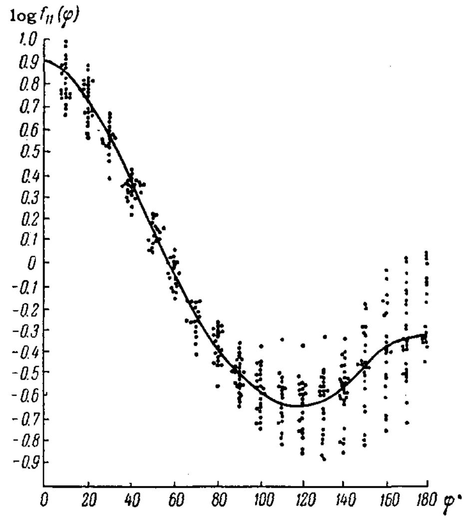



FIG. 16. Scattering function of atmospheric air at sea level (in logarithmic scale) from measurements over a series of nights.

sol character, with a sharp elongation forward and a more or less clearly marked aureole part. As an example, indicatrices measured at sea level by Chesterman and Stiles[87] are shown in Fig. 16. The protracted measurements of Barten'eva[88] have shown (in agreement with other authors) that the degree of elongation of the indicatrix corresponds well with the value of the scattering coefficient, increasing simultaneously with it, and the absence of any geographical dependence is clearly noted, except the frequency of repetition of this or that condition of transmission. Furthermore, it is observed that all the indicatrices intersect in a comparatively narrow range of scattering angles around $\varphi = 45°$, so that the coefficient of directed light scattering $D_{11}(45°)$ can be regarded with accuracy as a measure of the transmission of air. (According to the data of Pyaskovskaya-Fesenkova,[81] the intersection of all indicatrices takes place at $\varphi = 60°$, which can be explained, in particular, by their distortion as a result of secondary scattering).

Any further judgments on the form of the scattering function for atmospheric air are impossible at the present time, because all measurements have been carried out without an accompanying control on the nature and microstructure of the aerosol component.

The other components of the scattering matrix have in general not been measured to date. An exception are the still preliminary experiments recently completed by Rozenberg and Rudometkina.[51] One can obtain a general picture of the character of polarization effects taking place in light scattering in the atmosphere from Fig. 17, in which a collection of photographs of a given part of a linearly polarized projected beam are mounted; these were carried out on a single night by means of a photocamera equipped with polaroids and filters ($\lambda_{eff} = 420 \pm 20\ m\mu$) directed at different angles $\varphi$ to the beam (the visual angle of the camera is $\pm 15°$, and the ends of the picture of the ray are distorted by the vignetting effect). The arrows to the right show the direction (relative to the vertical) of the electric field vector of the wave in the projected beam (P) and passed by the analyzer on to the camera (A).

Furthermore, it was established experimentally by Rozenberg and Mikhaĭlin[89] that for known conditions (corresponding to the conclusions from the theory of Mie) the light scattered by the atmosphere (i.e., with particles of the aerosols impregnated in it) is elliptically polarized, while the degree of ellipticity q

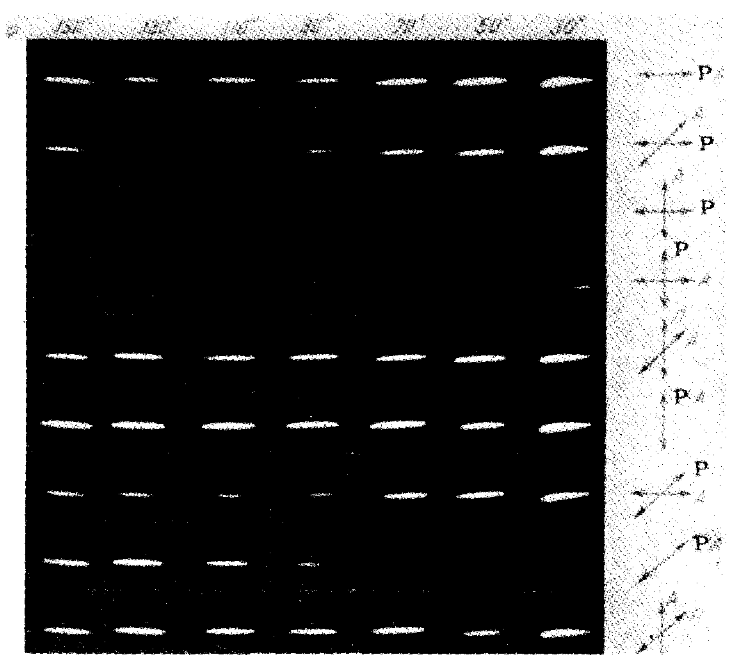



FIG. 17. Mosaic representation of the angular dependence of the brightness of a horizontal projector beam, on one of the nights at different states of beam polarization (P), observed through differently oriented analyzers (A). Each picture subtends an angle interval of ±15° relative to the value of φ indicated on top, and distorted at the edges by vignetting.

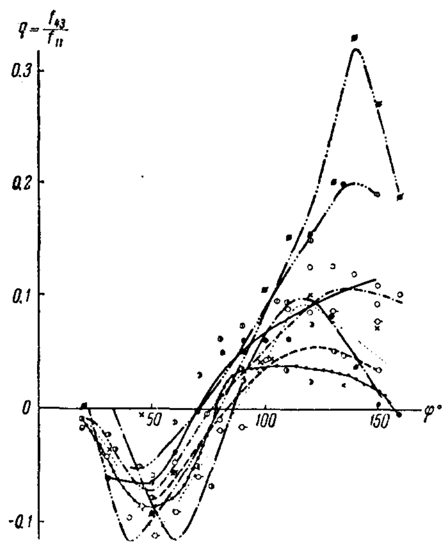



FIG. 18. Angular dependence of ellipticity of light scattered in the surface layer of the atmosphere, from measurements made in a series of successive nights.

$= f_{43}/f_{11}$ — see Eq. (16) — is never large, especially in the region of the rainbow (Fig. 18). It was shown here that the components of the matrix $f_{41}$ and $f_{42}$, if they are not equal to zero, are in any event very small. But a more detailed investigation of the scattering matrix and its angular and spectral dependences is a thing of the future.

The problem of the investigation of the aureole is a special one. It follows from the Mie theory that the aureole appears only in the presence of sufficiently large particles and has a diffraction character, which allows one to describe it with a comparatively simple and general formula. In the case of a semi-dispersed aerosol, the angular structure of the aureole is obtained as a result of simple mutual superposition of the aureoles from all particles. As shown by Sliepcevich and later by Shifrin,[90] this makes it possible by comparatively simple means to calculate the distribution of the dimensionally large (more than several microns) droplet fraction of the aerosol which account for the formation of the auerole; this can be done from a knowledge of the angular structure of the auerole. There can be no doubt of the future of this method which attracts ever greater attention.

Finally, the problem considered by us of the form of the scattering matrix is directly related to a wide circle of various phenomena connected in their origin with the reflection, refraction and diffraction of light on water droplets and ice crystals — all sorts of rainbows, halos, coronas, and glories. For a long time they were the chief object of attention in atmospheric optics and many volumes have been devoted to their detailed description (see, for example, references 1, 2, and 91). In spite of the fact that over a period of 1000 years an uncountable number of investigators, frequently very experienced and notable, have taken part in their investigation, for the majority of them there exists, besides the general description of the characteristic features and a detailed classification, only a qualitative explanation of the reasons on which they depend. Rigorous theory has been limited only to those phenomena which follow from the Mie theory, and principally (with the exception of rainbows) it touches only the general features of the phenomenon, omitting its details. So far as phenomena produced by ice crystals are concerned, there is no theory at all. Experimental data in this region are also very sketchy and there is no systematization, chiefly because of the absence of data on real conditions of the scattering medium to which they refer. But then this situation is met with in all regions of colloidal optics, and it provides the justification for the fact that the Mie theory remains today practically untested from a quantitative viewpoint after half a century.

## 7. OPTICAL PROBING OF THE ATMOSPHERE AND THE PROBLEM OF INTERPRETATION OF DATA

The idea of optical testing of the atmosphere is very simple. If we know how the laws of scattering are related to the properties of the scattering medium, then we can draw conclusions on the state of the latter (in the given case, the atmosphere) from the character of the light scattered by it. We have seen above that, as a consequence of the optical inhomogeneity of the atmosphere and of the obstacles presented by multiple scattering, use of integral effects (such as the illumination of the daytime sky or the transmission of the entire thickness of the atmosphere) does not provide reliable quantitative information on the optical state of the atmosphere at different altitudes at a given instant of time. We add that the decisive obstacle in this case is the complication of the theory of light propagation in scattering media which does not allow us to put into general form the solution of the inverse problem, i.e., the problem of the determination of the properties of a medium from its light field, including the angular dependence of all components of the scattering matrix.[39] Therefore, investigation of the optical properties of the atmosphere must necessarily have a doubly local character, in which the effects reliably measured are only those of single scattering in a distinctly limited and comparatively small volume. Evidently, this can be done if the atmosphere is irradiated by a sharply bounded, directed light beam, the tracing and displacement of which makes it possible to test the different regions of the atmosphere.

This idea was first stated by Al-Hazen.[3] He noted that during twilight the shadow of the earth rose higher and higher and that night began when the entire atmosphere was in the shade. This permitted Al-Hazen, by measuring the duration of the twilight, to estimate the altitude of the atmosphere at 52,000 paces (later this calculation was improved by Kepler[4]), which was very high accuracy for the eleventh century.

In the period following, the problem of twilight, which embraces about 10 percent of the year at the equator and not less than 30 percent at the poles attracted the attention of J. and N. Bernoulli, Maupertuis, D'Alembert, Clausius and many others, remaining constantly in the field of view both of observers and of theoreticians. The modern explanation of twilight phenomena as caused by scattering and attenuation of the light of the sun by the earth's atmosphere was first advanced by Berzold[92] in 1863, i.e., immediately after the discovery of Govi, where the necessity of explaining the character of the polarization of the light of the twilight sky played no small role.

Subsequent development of the theory of twilight led along the line of explanation of the redness of daybreak. Only in 1923, did Fesenkov, for the first time after Al-Hazen and Kepler, again set up the inverse problem of using the twilight phenomena for optical testing of the high altitude layers of the atmosphere.[93] Careful analysis of the effect of the various factors on the brightness of the twilight sky permitted Fesenkov in 1930 to advance the very fruitful idea of the "twilight ray," later developed in detail by Staude and in a somewhat different form by Link. The substance of this idea briefly is that at each given moment the twilight light proceeds from the comparatively thin effective layer of the atmosphere, bounded above as a consequence of the rapid decrease in the air density with altitude and below because of the rapid increase (upon approach to earth) of the attenuation of the emitted rays of the sun in the lower levels of the atmosphere. During the twilight period this effective layer gradually rises (or falls), which makes it possible to examine systematically the scattering ability of the different layers of the atmosphere, beginning approximately at 20 km above sea level and higher, so long as we can generally distinguish against the background of the sky the light scattered at the high layers of the atmosphere.

During the twenties and forties, no doubts were raised by anyone that, beyond the limits of the troposphere, the atmospheric air is perfectly pure, and the problem of testing was regarded as the determination of the altitude dependence of the density and the temperature of the air at altitudes up to 100 or even 300 — 400 km, not attainable at that time by other means. The treatment of data of twilight observations, completed during this period by a number of authors, led to values that were completely reasonable in order of magnitude, in agreement with other indirect estimates, values which in their time played an important role as the stimulus toward reexamination of the traditional picture of an isothermal stratosphere.[95] The tendency to raise the accuracy of the estimate led in the middle of this period to the necessity of analyzing the role which secondary light scattering plays in the formation of the twilight. The difficulty of the problem is such that a rigorous solution of it has not been obtained to date. Approximate solutions, valid for different initial assumptions, have been shown to be extremely contradictory, but in general prejudicial for the method as a whole, at least in its application to altitudes above 100 km. Such an uncertainty in the competence of the method on the one hand and the simultaneous development of rocket methods of investigation of the stratosphere on the other, has for a long time moved the twilight method to a back seat. However, the abundant observational material accumulated since that time makes it possible to draw the conclusion that for a suitable arrangement of observations and suitable treatment of observational data, one can, by studying the twilight, obtain rather trustworthy information on the scattering properties of the atmosphere in the range of altitudes approximately from 20 to 100 km.[96] Here attention[96] is turned to the extreme variability of the brightness, color,[97] and polarization[98] of the light of the twilight sky. The variability of this is so large and so characteristic that one can attribute it only to a single reason[44,96] — that, at all altitudes in the stratosphere, at least up to 80 — 90 km, i.e., up to the level of existence of the so-called silver clouds,[99] the basic substance for light scattering is not the air but aerosols with an inherent time and space variability both in quantity and in dimensions and nature of particles. In particular, if these aerosols have meteoric origin, then their appearance must be connected with the increase of ionic concentration, which makes understandable the connection noted by Khvostikov between the twilight polarization anomalies at the altitude of twilight light of around 80 km and the critical frequency of reflection of radio waves.[98] We add that an estimate of the concentration of aerosol particles which is necessary in order that scattering at altitudes of 50 — 80 km have an aerosol character[44] leads to quite reasonable numbers — of the order of 1 to $10^{-2}$ particles per cubic centimeter with dimensions of 0.1 to $0.3\,\mu$ (see reference 99). Thus the problem of twilight testing of the stratosphere has changed sharply, and concerns now the observation of the transfer, sedimentation, and transformation of the aerosol in the mesosphere, i.e., investigations of the meteorology of extremely high altitudes, which have assumed a practical importance in recent years. The significance of the revival of the twilight method can be estimated by recalling the high expense of rocket investigations, which limits their widespread use, and the difficulty of using rocket technology for the investigation of aerosols.

In 1930, Synge (see reference 51) pointed out the possibility of testing the atmosphere by a ray from a projector. The progress of projector and measurement technology has made it possible to realize this possibility very quickly, while the "ceiling" of testing has steadily increased, although not reaching imposing figures in the 1950's — about 70 km. Here every American investigator, without exception, aimed at extracting from the probing data information on the density and temperature of the air at these altitudes, while in the Soviet Union, I. A. Khvostikov had already clearly

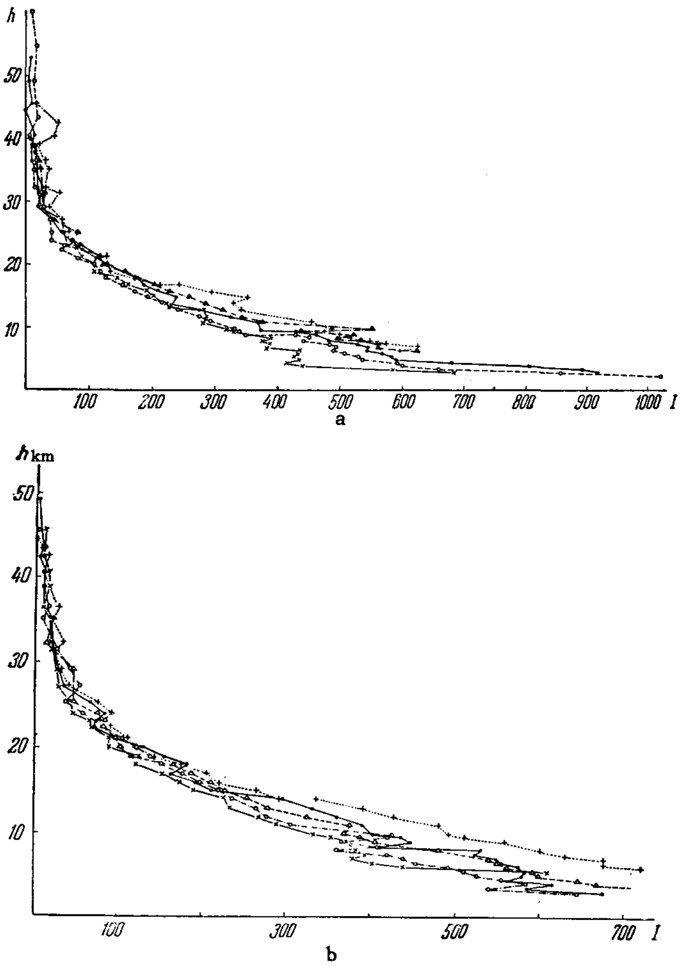



FIG. 19. Dependence of the brightness of the scattered light of the projector (in arbitrary units) on the altitude of the scattering volume according to measurements of a number of consecutive nights in August (a) and September (b), 1954.

aimed the investigation in 1944 in the direction of a study of the atmospheric aerosol, for which measurements of the brightness of the scattered light of the projector were from the outset accompanied by measurements of its polarization (for details, see reference 51).

In the period 1944-1958 many hundreds of optical profiles of the atmosphere were obtained in the laboratory of atmospheric optics of the Institute of Atmospheric Physics, Academy of Sciences, U.S.S.R., by means of projection probing by different methods and under different conditions; these made it possible to draw a number of completely unambiguous conclusions.[51]

It has been shown that the scattering ability of the atmosphere, even on very clear days and at all altitudes acceptable to projection probing, was subject to comparatively rapid and random changes, by a factor of at least 2 or 3. This is clearly seen in Fig. 19, where the intensities of the scattered light of a projector are plotted (in arbitrary units) as a function of the altitude of the scattering volume for two series of successive nights according to observations in Bakuryani (1954). Almost always, at one altitude or another, more or less clearly pronounced layers of aerosol were discovered, which frequently had an explicit cloud structure and which were made evident both by increase in the brightness of the scattered light of the projector, and also by the change in its polarization. An example of such a vertical optical profile of the earth's atmosphere can be seen in Fig. 20, in which data of measurements of the intensities of two linear components of the scattered light of the projector are shown for a single night ($I_1$ is the polarization in the plane of the scattering, $I_2$ is perpendicular to it). The measurements were carried out for the blue region of the spectrum and as a function of the altitude of the scattering volume. The degree of polarization of the scattered light almost never corresponds to the Rayleigh law, but falls off more slowly or more rapidly, while the polarization in the aerosol layers is frequently negative, as for example is the case in Fig. 21 at an altitude of around 22 – 23 km (the altitudes are shown on the right along the ordinate, while the corresponding scattering angle is shown on the left of the ordinate). It is noted that the aerosol layers are most frequently observed in the region of the tropopause (10 – 12 km in our latitudes) and at the altitudes 22 – 25 km, which corresponds to the usual altitude of the phenomenon of the so-called mother-of-pearl clouds. In the latter case, the data of projection testing made it possible for Driving to establish the fact that the clouds are formed by the supercooling of water droplets; he estimated their

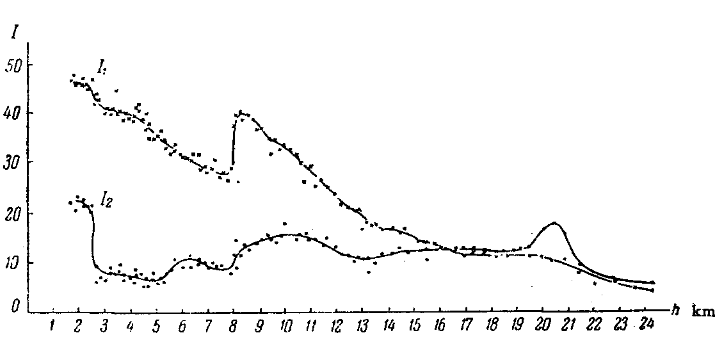



FIG. 20. Dependence of two mutually perpendicular polarized components of the brightness of the scattered light of a projector on the altitude of the scattering volume for a single night.

dimensions and concentration.[51] Clearly expressed aerosol layers were frequently observed even higher, in every case up to 40 km. Sharp variations of the ceiling of projection testing are characteristic; they can be attributed only to strong variations of the scattering ability of the atmosphere at the corresponding altitudes (40 — 70 km), i.e., the variability of the aerosol located there,[51] which is in excellent agreement with the conclusions based on twilight observations.

Thus, the data of projection and twilight testing of the atmosphere, together with the data of measurements of the brightness of the sky with "autostratostats" (Sec. 6) leave no doubt that at all altitudes up to 80 — 90 km aerosols are chiefly responsible for the scattering of light in the atmosphere. Attempts at the determination of densities and temperatures of the air by optical means have led, as is well known, to a significant scatter of values, and to agreement with the data by other methods only in order of magnitude. Now this can be explained by the fact that, on an average, the relative concentration of the aerosol varies slightly with altitude, and that in the visible portion of the spectrum the scattering ability of the atmospheric aerosol is, in the mean, close in order of magnitude to the scattering ability of pure air. Thus the only real result of optical studies of the atmosphere at all altitudes can be exclusively (if one puts aside the investigation of the selective absorption of the gas phase) the study of optical properties of the atmospheric aerosol and the changes experienced by it — transfer, sedimentation, crystallization, condensation, evaporation, etc. This means that the use of optical methods for the investigation of such important meteorological processes as those mentioned becomes the fundamental problem of atmospheric optics, the more so since to date no effective means of investigation of these processes exist (especially if one is discussing processes in which submicroscopic particles take part).

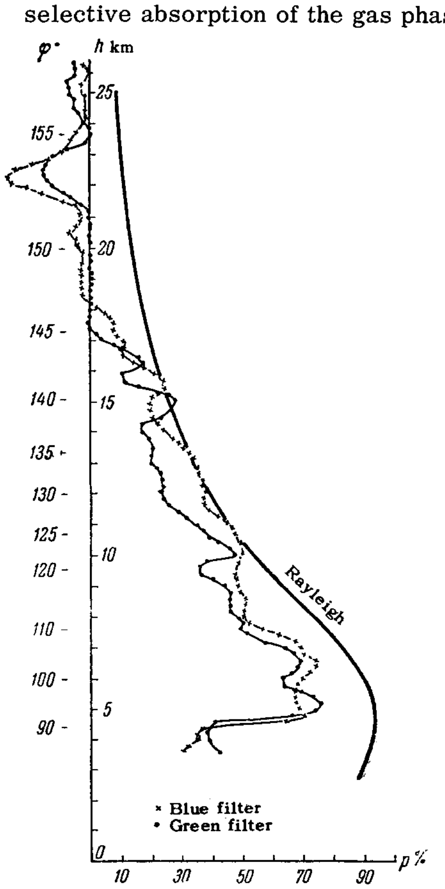



FIG. 21. Dependence of the degree of polarization of the scattered light of a projector on the altitude of the scattering volume in two parts of the spectrum for a single night.

However, we immediately come across the problem of the interpretation of sounding data. Essentially, the problems facing us are identical with those of nuclear physics: it is necessary, by means of the scattering matrix, by its angular and spectral dependences, to make clear with the maximum completeness obtainable the properties of the scattering medium (the nature of the scattering particles, their size distribution etc.), i.e., one must solve the inverse problem of the theory of light scattering by the set of heterogeneous particles of the aerosol. After what has been pointed out in the previous paragraph, it is scarcely necessary to add that at the present time the state of the theory makes it possible to carry out such an analysis only in very few specially favorable cases, and on a very limited scale, as a consequence of which the effectiveness of the testing is negligible at the present time. Mostly, the observational data clearly reveal certain processes, but interpretations are not supplied, and the result is only an increase in the collection of barren puzzles. Therefore, the central problem of modern atmospheric optics is undoubtedly the detailed explanation of the laws of light scattering by particles of the aerosol, primarily by water droplets and ice crystals, but also ensembles of these with different distribution laws.

This is the very general problem of colloidal optics, and it must be solved by employing the most effective means of contemporary experiment through a direct combination of field and laboratory investigations with simultaneous treatment of theoretical representations. The greatest difficulty lies in the fact that the submicroscopic fraction of the aerosol, which is very active in its optical behavior, does not yield to detailed study by other means at the present time. This leads to the impossibility of direct comparison of the optical characteristics of the medium with its micro-physical parameters, which can be compensated only by an abundance of optical information, i.e., by the simultaneous operation of different optical means of investigation in a joint study of the different aspects of a single phenomenon. Only such a complex optical experiment will be in a position to guarantee the representation and the possibility of analysis of the results, and at the same time the creation of bases for the interpretation of the data of the optical sounding of the atmosphere.

## 8. PROPAGATION OF LIGHT IN CLOUDS AND FOGS AND SIMILAR PROBLEMS

In addition to the investigation of the dependence of the scattering matrix on the microphysical characteristics of the aerosol, the 20th century has put forth still another round of problems, which are very important for atmospheric optics both from an applied and from a theoretical point of view. The formulation of the equation of radiation transfer and the development of methods of its solution, in particular with the use of computer technology, has made possible the investigation of processes of propagation of radiation and radiation conditions in extremely turbid media — clouds, fogs, the sea, etc. The problems arising from the demands of atmospheric optics, among them the requirement of forecasting visibility, have long since pushed past its framework and achieved first-rate importance in astrophysics and nuclear physics, which makes it impossible to touch on them in any detail within the framework of this paper (see reference 21). Here we can only call to mind certain problems whose solution is most important from the point of view of an understanding of atmospheric-optical phenomena.

We immediately note that the biggest obstacle to the development of a modern theory of light propagation in strongly scattering media is again the absence of information on the form of the scattering matrix. As a result, the problem of calculating or estimating polarization effects, which are nowhere small, can neither be completely furnished nor solved with any certainty, with the exception of the trivial case of Rayleigh scattering, which has no relation to reality. This at once deprives the theory, no matter how inventive it might be, of practical significance, and sharply limits the possibility of its comparison with experimental data.

Even the scattering functions $f_{11}(\varphi)$ for real polydispersed systems remain today but little studied, either theoretically or experimentally. As a consequence, all the computations must be carried out relative to highly idealized systems, and their comparison with reality inevitably involves a highly qualitative, descriptive character, the more so since the experimental data on the light conditions in the scattering medium are very poor. Such a situation is emphasized by the absence of effective approximate methods of solution of the transfer equation, which would permit one to explain even qualitatively the general regularities, which forces theoreticians to direct their efforts to the collection of supposedly "rigorous" solutions of more or less typical, but, as a rule, barbarously stylized examples. Therefore, in spite of the serious progress in this region, with which the last 150 years have been marked, the situation remains a sad one, chiefly because of the complete separation between theory and experiment.

Turning to problems which await their solutions we must first of all point to the investigation of the laws of reflection of light from formations that strongly scatter it — clouds, snow, the sea, sand, soil, plant cover, etc., as a function of their microstructure, their absorbing ability and the angles of incidence and observation of the light beam. Here a large amount of observational material has been accumulated which finds to date no exhaustive theoretical explanation and generalization (see references 21, 39), consequences that are the more necessary in that they would permit a large step forward in a number of practically important problems.

Furthermore, detailed experimental and theoretical investigations of the light regime inside the scattering medium are necessary — from its boundaries to its depths, where the character of the light conditions is determined not from the properties of the illumination but from the properties of the medium itself.[21,39]

The whole region of three-dimensional problems of heat transfer remains today completely unstudied. To this is related the theory of twilight phenomena, and the theory of the so-called "ice" and "water" sky and, of first importance, the wide and practically important problem of the penetration of a projected ray through fog or other strongly scattering medium.[51] The last problem has a great methodological significance, because it is not clear at the present time to what measure the effects of multiple scattering are capable of preventing the interpretation of experiments on the determination of the coefficients and the scattering matrix in clouds and fogs, while the ways for the choice of the most rational measuring systems are very unclear.

Let us mention one of the most important (from our point of view) problems of modern optics — the creation of the foundations of the spectroscopy of dispersive media by the search for means of utilization of the effects of multiple scattering for experimental separation of the coefficients of absorption and scattering of dispersed phases.[39] In particular, in this way one should expect to obtain data currently absent on the absorptive ability of clouds and fogs in different portions of the spectrum.

Realization of the investigations mentioned requires the joint and directed efforts of theoreticians and experimentalists, while, as in other problems of light scattering, it is reasonable to join laboratory experiments with natural measurements on clouds, fogs, or oceanic water, subordinating these measurements to carefully thought-out general purposes.

## 9. RADIATION CLIMATOLOGY AND THE OPTICS OF THE AEROSOL

For centuries, and up to very recent times, the purposes of atmospheric optics as a science have been connected with hopes of using optical "objects" for weather forecasting. For example, evidence of this is clearly expressed by the subtitle of a book on atmospheric optics published in the Russian language as late as in 1924 by Brounov: "Optical Phenomena of the Sky in Connection with Weather Prediction." However, successes in this region were minimal and we now understand very well the reason why. The connection of the optical situation with the weather bears a doubly uncertain character. Meteorological processes in some fashion not yet clear to us in all the necessary detail bear on the fate and characteristics of the atmospheric aerosol. The latter in turn has a strong effect on the radiation and thermal conditions of the atmosphere, and at the same time on the behavior of meteorological processes. The optical state of the atmosphere reflects, again in a form not completely understandable to us, the instantaneous state of the atmospheric aerosol. Therefore, the way to the utilization of optical "objects" for weather forecasting lies, in the first place, in the discovery of relations between the nature of the aerosol and its optical properties and, in the second place, in clarification of the connection between the weather-producing processes and processes of transport and transformation of the aerosol. The lack of a future for these empirical searches of various "objects" is amply illustrated by their history.

Nevertheless, numerous and unremitting investigations in these directions have not been fruitless. They have led to the discovery and understanding of a great collection of atmospheric-optical phenomena, and if this period is regarded in retrospect, it is not difficult to establish the fact that precisely this qualitative description and explanation of all these phenomena constitutes the fundamental content of atmospheric optics. It is also not difficult to show that this stage of initial accumulation of facts and the creation of general qualitative representations on the nature of atmospheric-optical phenomena has already been exhausted. We shall set forth the general information on the basic optical characteristics of the atmosphere and on their variability in the measure in which they are attained by means of comparatively simple apparatus and uncomplicated theory. This information forms a reliable basis for general orientation on the behavior of the phenomena. An unusual epitaph of this purely observational stage is given by the book of M. Minnaert which has recently come into our hands: "Light and Color in Nature."[91] However, it is impossible to close one's eyes to the fact that this trend becomes archaic and by no means contains the fundamental tendencies of modern science. And these tendencies are connected with the transition from passive observation of natural phenomena to single-minded quantitative analysis of their physical nature; the analysis operates steadily on a broad invasion in this region of contemporary physico-mathematical means of investigation, both experimental and theoretical. This is most evident in the sharp change of problems and methods of investigation.

The observational aspect of atmospheric optics apparently has not lost its value, but it is now regarded in a completely different fashion. This is the development of radiation climatology on the basis of a statistical analysis of data, for example, on the transmission in the atmosphere in different parts of the spectrum, obtained on a far-flung grid of observation stations with the aid of mass-produced and inexpensive measuring apparatus. The aim of such a type of regular service must be to provide the national economy with operational and climatological information, which permits rational solutions of problems of transport, construction, illumination, agrotechnology, health science, etc. In addition there stands out sharply a completely independent group of problems, which requires a different approach in principle, which can conditionally be called the optics of the aerosol and which, in particular, should uncover the way for the investigation of meteorological problems by optical means.

The outstanding problems here have already been mentioned above. These are the study of the angular and spectral dependences of the scattering matrix as a way to a clarification of the microstructure of the aerosol, and the study of the laws of radiation propagation in strongly scattering media. However, in a real atmosphere we meet up with two circumstances which hinder the path of investigation. In the first place, there is the constant and uncontrolled variability of the atmosphere as an object of investigation, which does not permit one to reproduce the conditions of measurements. In the second place, there is the presence of the submicroscopic fraction, which is optically active, but which does not submit to identification by other methods. Therefore, the contemporary atmospheric-optical experiment takes on very specific marks. It must be a complex, many-sided optical investigation of isolated special cases, in which the volume of optical and other auxiliary information obtained is so large that it admits of an unambiguous theoretical analysis both in the sense of identification of the parameters of the scattering medium and in relation to comparing it with the different optical properties. Of course, such a rich, complex study can be carried out only by use of the most advanced means of contemporary measurement technology, and requires serious concentration of forces of highly skilled staffs of scientists on isolated, comparatively narrow, and clearly defined aspects of the problem. The complexity and purposefulness of the investigations are the most characteristic marks of the modern problems of optics and of the aerosol. At the same time, it is evident that isolated experiments in this direction, no matter how fine the apparatus with which they are carried out, lead only to additions to the collection of observed cases, but do not help in advancing the understanding of the physical laws.

It is perfectly natural that laboratory investigations on colloidal optics serve as a necessary complement to natural measurements, but, as at an earlier time, they cannot replace it, because the scale of the atmosphere permits observations, for modern measurement technology, of certain phenomena not yet accessible to laboratory conditions. Therefore, atmospheric optics in the correct understanding of its problems, as before, maintains its position as one of the leading advance posts in the study of the scattering of light and of its propagation in scattering media, and this circle of problems is undoubtedly related to a number of fundamental problems of modern optics generally.

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