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On the size, shape, and orientation of noctilucent cloud particles

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ABSTRACT

Conclusions about the size of noctilucent cloud particles are often made under the assumption that they are spherical. The small degree of circular polarization observed in the light from noctilucent clouds suggests, however, that the particles may be aligned, and hence nonspherical: unpolarized light is elliptically polarized upon single scattering by aligned particles. Yet no suitable mechanism—aerodynamic, electrical, magnetic—has been proposed which will align small (< 10 μm) mesospheric particles. A more likely explanation for the observed circular polarization is that the light illuminating noctilucent clouds has acquired ellipticity because of multiple scattering in the long atmospheric paths it traverses. Even if noctilucent cloud particles are nonspherical, however, presently accepted upper limits on their size (~ 0.1 μm) and volumetric scattering need not be revised: scattering by small ice spheres as well as ice ellipsoids of any shape is consistent with the observed angular dependence of the degree of linear polarization. Moreover, particles much larger than 0.1 μm, regardless of shape, will lower the maximum degree of linear polarization below that which has been observed.

1. Introduction

The size of noctilucent cloud particles has been the subject of recent controversy. Linear polarization measurements by Witt (1969), Tozer and Beeson (1974), and Witt *et al.* (1976) suggested to these authors that the upper limit of particle size is less than about 0.13 μm. Hummel and Olivero (1976) analysed the satellite radiance measurements of Donahue *et al.* (1972) and arrived at the same upper limit. Gadsden (1978), however, has argued that the evidence for this upper limit is not compelling. His reasons are twofold:

(1) Scattering per unit volume (or mass) of particles strongly depends on their size. For example, volumetric scattering by particles small compared with the wavelength is proportional to the cube of their (linear) size; at the other extreme, for particles much larger than the wavelength, volumetric scattering is inversely proportional to size. A greater mass of small particles (say smaller than 0.13 μm) than somewhat larger particles (say between 0.2 and 0.4 μm) is necessary to give the observed brightness of noctilucent clouds. This in turn implies that the smaller the particles the greater the amount of mesospheric water vapor necessary for a given brightness.

(2) Small degrees of circular polarization (i.e., ellipticity) have been observed in light from noctilucent clouds (Gadsden, 1975; 1977; Gadsden *et al.*, 1979), which suggests partially oriented, hence nonspherical, particles. Previous estimates of particle size have been made from analyses based on the assumption of spherical particles. If the particles are not spherical, however, such analyses are suspect.

We shall not delve into the matter of whether or not there is sufficient water vapor in the mesosphere to provide enough mass for noctilucent clouds composed of ice particles no larger than 0.13 μm. In this paper we shall address the consequences of nonsphericity for conclusions about noctilucent cloud particles. We begin by considering some of the constraints imposed on particles if they are to elliptically polarize light upon scattering.

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2. Polarization by scattering: the scattering matrix

The state of polarization of a beam of light is specified by the four Stokes parameters, which are denoted by I, Q, U, V; I is the intensity (or radiance or irradiance) of the beam and the other three parameters are related to the vibration ellipse (see, for example, Shurcliff, 1962). The Stokes parameters satisfy the inequality $I^2 \geqslant Q^2 + U^2 + V^2$. Equality holds if the beam is completely polarized; unpolarized light is characterized by $Q = U = V = 0$. Between these two extremes lies light of various degrees of partial polarization. Of particular interest here is elliptically polarized light, which is characterized by nonzero V (to avoid cumbersome phrases, elliptically polarized light will be shorthand for partially polarized light with nonzero V). V/I is defined as the degree of circular polarization.

Upon scattering by a suspension of particles the polarization state of light may be changed. The Stokes parameters of incident and scattered light are related by the scattering matrix (or Mueller matrix)

$$\begin{bmatrix} I_s \\ Q_s \\ U_s \\ V_s \end{bmatrix} = \begin{bmatrix} S_{11} & S_{12} & S_{13} & S_{14} \\ S_{21} & S_{22} & S_{23} & S_{24} \\ S_{31} & S_{32} & S_{33} & S_{34} \\ S_{41} & S_{42} & S_{43} & S_{44} \end{bmatrix} \begin{bmatrix} I_i \\ Q_i \\ U_i \\ V_i \end{bmatrix}. \tag{1}$$

If the particles are randomly separated by sufficiently large distances then the scattering matrix of the suspension is the sum of scattering matrices of the individual particles. For the moment, single scattering is assumed. The matrix elements S_{ij} depend on particle characteristics—size, shape, and composition—as well as on the directions of incident and scattered light.

If incident unpolarized light is transformed by scattering into elliptically polarized light ($V_s \neq 0$) then it follows necessarily from (1) that S_{41} must not be zero.

The 16-element matrix in (1) is the most general scattering matrix. Symmetry, however, reduces the number of nonzero, independent elements. For example, the scattering matrix for a suspension that is invariant under all rotations and reflections has the form (Perrin, 1942)

$$\begin{bmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{22} & 0 & 0 \\ 0 & 0 & S_{33} & S_{34} \\ 0 & 0 & -S_{34} & S_{44} \end{bmatrix}. \tag{2}$$

This includes a suspension of randomly oriented particles each of which has a plane of symmetry. If each particle is spherically symmetric then $S_{33} = S_{44}$ and $S_{11} = S_{22}$. It is clear that a suspension described by the scattering matrix (2) cannot elliptically polarize unpolarized light. Nor can it elliptically polarize horizontally ($Q > 0$, $U = V = 0$) or vertically ($Q < 0$, $U = V = 0$) polarized incident light. Horizontal and vertical in this instance mean parallel and perpendicular to the scattering plane, which is determined by the directions of incident and scattered light. Obliquely polarized incident light ($U \neq 0$) can, however, acquire a degree of circular polarization upon scattering provided that S_{34} is not zero.

Multiple scattering by particles described by the scattering matrix (2) can yield elliptically polarized light. After single scattering, initially unpolarized light will be linearly polarized either parallel or perpendicular to the scattering plane depending on the sign of S_{12}. This scattered light is incident on other particles; moreover, it is, in general, polarized obliquely to scattering planes determined by the directions of incident (scattered once) and twice-scattered light. So elliptical polarization is possible.

If aligned particles are to elliptically polarize unpolarized light upon *single* scattering, then S_{41} must be nonzero. If randomly oriented particles are to elliptically polarize unpolarized light upon *multiple* scattering, then S_{34} must be nonzero. A specific example is given in the following section, but first, some additional results are needed.

The relation between the components of the incident field and those of the field scattered by a single particle may be written in matrix form

$$\begin{bmatrix} E_{\parallel s} \\ E_{\perp s} \end{bmatrix} = \begin{bmatrix} S_2 & S_3 \\ S_4 & S_1 \end{bmatrix} \begin{bmatrix} E_{\parallel i} \\ E_{\perp i} \end{bmatrix}, \tag{3}$$

where $\parallel$ and $\perp$ denote components parallel and perpendicular to the scattering plane. For clarity, multiplicative factors which are irrelevant to the discussion here are omitted from the amplitude scattering matrix in (3) as well as from (4×4)

scattering matrices. The scattering matrix elements S_{ij} are functions of the amplitude scattering matrix elements S_j. Only a few of these functions are needed here:

$$
\begin{aligned}
S_{34} &= \mathrm{Im}\{S_2 S_1^* + S_4 S_3^*\},\\
S_{41} &= \mathrm{Im}\{S_2^* S_4 + S_3^* S_1\},\\
S_{42} &= \mathrm{Im}\{S_2^* S_4 - S_3^* S_1\},\\
S_{43} &= \mathrm{Im}\{S_1 S_2^* - S_3 S_4^*\}.
\end{aligned} \tag{4}
$$

These equations can be extracted from van de Hulst (1957, Ch. 5), although here, as elsewhere in this paper, the conventions and notation are different from those of van de Hulst.

3. Scattering in the Rayleigh limit

Scattering and absorption by a particle small compared with the wavelength are completely determined by the dipole moment **p** induced by the electric field of the incident light. Although **p** is not necessarily parallel to the electric field, it is linearly related to the field through the polarizability tensor, the components of which are denoted by α_{ij}. The amplitude scattering matrix elements for an anisotropic dipole are

$$
\begin{aligned}
S_1 &= \frac{-ik^3}{4\pi}\\
&\quad (\alpha_{11}\sin^2\phi - 2\alpha_{12}\sin\phi\cos\phi + \alpha_{22}\cos^2\phi),\\
S_2 &= \frac{-ik^3}{4\pi}\\
&\quad [\cos\theta(\alpha_{11}\cos^2\phi + 2\alpha_{12}\sin\phi\cos\phi + \alpha_{22}\sin^2\phi)\\
&\qquad - \sin\theta(\alpha_{13}\cos\phi + \alpha_{23}\sin\phi)],\\
S_3 &= \frac{-ik^3}{4\pi}\{\cos\theta[\alpha_{11}\sin\phi\cos\phi + \alpha_{12}(\sin^2\phi - \cos^2\phi)\\
&\quad - \alpha_{22}\sin\phi\cos\phi] - \sin\theta(\alpha_{13}\sin\phi - \alpha_{23}\cos\phi)\},\\
S_4 &= \frac{-ik^3}{4\pi}[\alpha_{11}\sin\phi\cos\phi + \alpha_{12}(\sin^2\phi - \cos^2\phi)\\
&\quad - \alpha_{22}\sin\phi\cos\phi],
\end{aligned} \tag{5}
$$

where $k = 2\pi/\lambda$ is the wavenumber; θ, the scattering angle, is the angle between the directions of incident and scattered light; and ϕ is the azimuth of the scattered light. The components of the polarizability tensor depend on the orientation of the dipole relative to the incident beam.

Unless a particle is absorbing, α_{ij} is real. It therefore follows from (4) and (5) that in the Rayleigh limit the matrix elements S_{41}, S_{42}, S_{43}, S_{34} for a *nonabsorbing* particle vanish regardless of its orientation. Thus, a suspension of such particles cannot elliptically polarize unpolarized light by either single or multiple scattering. Only if the incident light is elliptically polarized will the scattered light be elliptically polarized.

Noctilucent cloud particles are generally believed to be composed of ice, although there is no direct evidence to support this belief. Ice is very weakly absorbing at visible wavelengths (Grenfell and Perovich, 1981), so much so that for our purposes it may be considered strictly nonabsorbing without error. The possibility remains, however, that the ice particles nucleate on absorbing particles (see, for example, Hunten *et al.*, 1980). But the trend of opinion is that solid nuclei are not essential to the formation of noctilucent cloud particles and that hydrated ions are sufficient for nucleation (Witt, 1969).

If noctilucent clouds are composed of small ice particles nucleated on ions, then the only way they can yield elliptically polarized light upon scattering is if the light illuminating them is elliptically polarized. Such elliptically polarized light may be produced by multiple scattering in the long paths traversed by the illuminating light, provided that the scatterers are larger, or more absorbing, than those in the clouds.

The validity of the conclusions above about noctilucent cloud particles requires that Rayleigh theory describes their scattering properties to good approximation. In the following section, therefore, the evidence for this is discussed. It is also well to note that, at least for particles smaller than the wavelength, there is no sharp cutoff beyond which Rayleigh theory is completely invalid: the transition is gradual. So the Rayleigh theory may be extended slightly beyond its strict bounds, especially when we consider that 0.13 μm is assumed to be the *upper* limit of particle size.

4. The evidence for small particles: linear polarization

The strongest evidence supporting small particles (<0.13 μm) is provided by measurements of the degree of linear polarization of light from noctilucent clouds. Linear polarization measured from the ground by Witt (1960) increased monotonically with scattering angle (between 20 and 60°) to maximum values of about 0.4 for blue light ($\lambda = 490$ nm) and about 0.5 for red light ($\lambda = 610$ nm). Witt (1969) observed high polarization at scattering angles between 83 and 85 degrees with rocket-borne photoelectric photometers ($\lambda = 534$ nm and $\lambda = 366$ nm) from which he concluded that the upper limit of particle radius was between 0.1 and 0.2 μm depending on the assumed refractive index. Tozer and Beeson (1974) also extended the range of scattering angles to 84° by using rockets. They measured quite large linear polarizations, as high as 0.94–0.96 at 80–84°. From this they inferred that the particles were small (less then 0.13 μm). Further rocket-borne measurements, both at visible (536 nm) and ultraviolet (236 nm) wavelengths, by Witt *et al.* (1976) suggested to them an upper size limit of about 0.1 μm or smaller. But inferences about particle size have been criticized by Gadsden (1978) on the grounds that, although the particles may not be spherical, the data were analysed with theory for spheres (Mie theory). So we must ask: to what extent are conclusions about upper limits of particle size modified if the particles are not spherical? This question can be answered within the framework of Rayleigh theory, as well as beyond its purview by appealing to recent calculations.

The scattering matrix for a suspension of randomly oriented, homogeneous Rayleigh ellipsoids is (van de Hulst, 1957, Ch. 6)

$$\begin{bmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{22} & 0 & 0 \\ 0 & 0 & S_{33} & 0 \\ 0 & 0 & 0 & S_{44} \end{bmatrix}$$

$$S_{11} = \frac{3k^2\langle C_{\mathrm{sca}}\rangle}{8\pi}\frac{1}{2}\left(\frac{6-M}{5} + \frac{2+3M}{5}\cos^2\theta\right),$$

$$S_{12} = \frac{3k^2\langle C_{\mathrm{sca}}\rangle}{8\pi}\frac{1}{2}(\cos^2\theta - 1)\frac{2+3M}{5},$$

$$S_{22} = \frac{3k^2\langle C_{\mathrm{sca}}\rangle}{8\pi}\frac{1}{2}(\cos^2\theta + 1)\frac{2+3M}{5},$$

$$S_{33} = \frac{3k^2\langle C_{\mathrm{sca}}\rangle}{8\pi}\frac{2+3M}{5}\cos\theta,$$

$$S_{44} = \frac{3k^2\langle C_{\mathrm{sca}}\rangle}{8\pi}M\cos\theta, \tag{6}$$

where the scattering cross section averaged over all orientations is

$$\langle C_{\mathrm{sca}}\rangle = \frac{k^4}{6\pi}(\tfrac{1}{3}|\alpha_1|^2 + \tfrac{1}{3}|\alpha_2|^2 + \tfrac{1}{3}|\alpha_3|^2)$$

and

$$M = \frac{\mathrm{Re}\{\alpha_1^*\alpha_2 + \alpha_1^*\alpha_3 + \alpha_2^*\alpha_3\}}{|\alpha_1|^2 + |\alpha_2|^2 + |\alpha_3|^2}.$$

The principal components of the polarizability tensor are

$$\alpha_j = v\frac{\varepsilon - 1}{1 + L_j(\varepsilon - 1)}, \qquad (j = 1, 2, 3)$$

where v is the particle volume and $\varepsilon = \varepsilon' + i\varepsilon''$ is its dielectric function relative to that of the surrounding medium. The geometrical factors L_j are elliptical integrals of the form

$$L = \frac{abc}{2}\int_0^\infty \frac{\mathrm{d}q}{(d^2+q)\sqrt{(a^2+q)(b^2+q)(c^2+q)}},$$

where d is one of the three lengths $a \geqslant b \geqslant c$ of the semiaxes. In general, there are three distinct geometrical factors $L_1 \leqslant L_2 \leqslant L_3$; they satisfy the conditions $0 \leqslant L_j \leqslant 1$ and $L_1 + L_2 + L_3 = 1$. All the geometrical factors for a sphere are $\frac{1}{3}$; for a spheroid two of them are equal.

The degree of linear polarization P of scattered light, given incident unpolarized light, is defined as

$$P = -\frac{S_{12}}{S_{11}}$$

for any system of scatterers. If P is positive the scattered light is partially polarized perpendicular to the scattering plane; if negative it is polarized parallel to the scattering plane.

The degree of polarization of light scattered by randomly oriented Rayleigh ellipsoids is

$$P(\theta) = \frac{1 - \cos^2\theta}{1/P(90°) + \cos^2\theta},$$

where the polarization at 90° is

$$P(90°) = \frac{2+3M}{6-M}$$

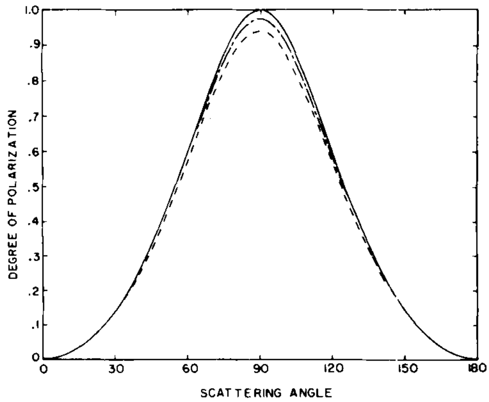

Fig. 1. Degree of linear polarization acquired by initially unpolarized visible light upon scattering by small (Rayleigh limit) ice particles of various shapes: spheres (——), randomly oriented prolate spheroids (— — —), and randomly oriented oblate spheroids (---).

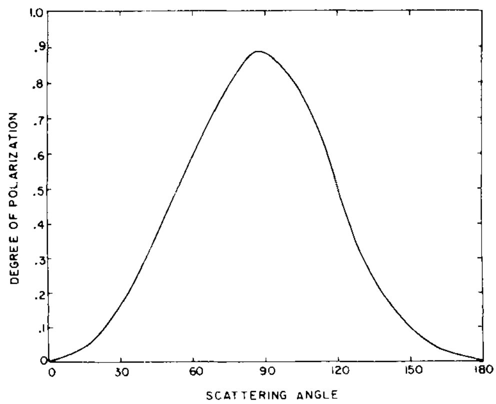

Fig. 2. Degree of linear polarization acquired by initially unpolarized light upon scattering by randomly oriented oblate spheroids with axial ratio $a/c = 5$; the refractive index is 1.33 and $2\pi a/\lambda = 5$ (from Asano and Sato, 1980. Reproduced with permission of the Optical Society of America).

and lies between $\frac{1}{13}$ and 1 depending on the value of M, which satisfies the inequality $-\frac{1}{2} \leqslant M \leqslant 1$.

The angular dependence of P for ice spheres, plates ($L_1 = L_2 = 0$), and needles ($L_2 = L_3 = \frac{1}{2}$) is shown in Fig. 1, where the refractive index at visible wavelengths is taken to be 1.305; these three shapes bracket the range of possibilities for randomly oriented Rayleigh ellipsoids. Note that plates give the least polarization at 90°, which is still over 0.94; polarization by needles is closer to that by spheres.

The optical properties of some small particles at certain wavelengths (e.g., in the far infrared) can be quite shape dependent (Bohren and Huffman, 1981), but not small ice particles at visible wavelengths. In addition to the degree of polarization, the scattering cross sections are nearly independent of shape:

$$\frac{\langle C_{\text{sca}} \rangle_{\text{needle}}}{\langle C_{\text{sca}} \rangle_{\text{sphere}}} = 1.06 \quad \frac{\langle C_{\text{sca}} \rangle_{\text{plate}}}{\langle C_{\text{sca}} \rangle_{\text{sphere}}} = 1.19.$$

Within the framework of Rayleigh theory, therefore, shape considerations do not enter into estimates of the amount of scattering per unit volume of small ice particles.

Some insight into how departures from Rayleigh theory affect linear polarization can be obtained from recent calculations using the exact theory of scattering by arbitrary spheroids (Asano and Sato, 1980). The curve in Fig. 2 shows results for randomly oriented oblate spheroids with refractive index 1.33, which is near enough to that of ice: $a/c = 5$ and $2\pi a/\lambda = 5$. For $\lambda = 0.5$ μm this corresponds to a particle with the same volume as that of a sphere with radius 0.23 μm. Note that the maximum degree of polarization for these spheroids is about 0.89, which is less than the greatest values measured by Tozer and Beeson. If the equivalent radius is doubled then the calculations of Asano and Sato show greatly reduced linear polarization.

Gadsden (1978) quite rightly criticizes the common—and generally unwarranted—practice of analyzing experimental data for non-spherical particles with Mie theory. That randomly oriented nonspherical particles do not scatter light like equivalent spheres has been convincingly demonstrated both by experiment (e.g. Holland and Gagne, 1970; Perry *et al.*, 1978) and by calculation (Asano and Sato, 1980). Indeed, P for nonspherical and for spherical particles are often opposite in sign. But the size at which the scattering properties of spherical and nonspherical particles (composed of material with optical constants similar to those of ice at visible wavelengths) drastically diverge—perhaps 0.4–0.5 μm—is somewhat greater than the greatest size suggested for noctilucent cloud particles.

Before proceeding, there is one more question to be answered: is the degree of circular polarization observed in light from noctilucent clouds consistent with the hypothesis that they are composed of ice particles, randomly oriented and nearly Rayleigh-like, and illuminated by light that has acquired ellipticity by multiple scattering in the long atmospheric paths it traverses on its way to the clouds? More specifically, does scattering by such particles reduce the degree of circular polarization of the light illuminating them? Fortunately, everything needed to answer this question is at hand.

Let elliptically polarized light be incident on particles described by (6); if its degree of circular polarization is V_i/I_i then that of the scattered light is

$$\frac{V_s}{I_s} = \frac{S_{44}}{S_{11}} \frac{V_i}{I_i},$$

where we have set $Q_i = 0$ for convenience. M is nearly unity for ice particles in the Rayleigh ellipsoid approximation (see Fig. 1), so to good approximation

$$\frac{V_s}{I_s} \simeq \frac{2\cos\theta}{1+\cos^2\theta} \frac{V_i}{I_i}.$$

Except for scattering angles within 15° of either side of 90°, the degree of circular polarization of the scattered light does not fall to less than half that of the incident light.

The preceding analysis, upon which the major conclusions of this paper are based, contains the underlying assumption that the particles are randomly oriented. Before discussing why this is likely, there is one more matter to be disposed of: the implications of Rayleigh-Gans theory for the problem at hand.

5. Scattering in the Rayleigh-Gans approximation

The Rayleigh-Gans (RG) theory applies to "soft" or "tenuous" particles, those that are optically similar to their surroundings, although they may be larger than allowed by Rayleigh theory. The two conditions for the validity of the RG theory are (van de Hulst, 1957, Ch. 7)

$$|m-1| \ll 1, \; kd|m-1| \ll 1,$$

where d is a characteristic linear dimension of the particle and m is its refractive index relative to that of the surrounding medium; $k = 2\pi/\lambda$. The RG theory is of uncertain applicability to ice particles in the atmosphere because the value of $|m-1|$ is near that (for spheres, at least) where this theory begins to give large errors (see Kerker, 1969, p. 428). Nevertheless, the RG theory has been raised in the context of noctilucent clouds (Gadsden, 1977) so it is worth brief consideration to see if it leads to any new conclusions.

Within the framework of the RG theory the scattering matrix for *any* particle, regardless of its shape or orientation, is

$$\begin{bmatrix} S_{11} & S_{12} & 0 & 0 \\ S_{12} & S_{11} & 0 & 0 \\ 0 & 0 & S_{33} & 0 \\ 0 & 0 & 0 & S_{33} \end{bmatrix}. \tag{7}$$

Particles with the scattering matrix (7) do not, therefore, elliptically polarize unpolarized or linearly polarized light. The only circumstance under which elliptically polarized light is produced is if the incident light is elliptically polarized.

Particles described by the RG theory are highly polarizing: the degree of linear polarization at 90° is 1.0. Such particles are therefore candidates for noctilucent cloud particles. If the RG theory is valid in this instance then ice particles cannot be much larger than about 0.1 μm.

6. Alignment of particles

One possible explanation for the elliptical polarization observed in light from noctilucent clouds is that the particles are aligned (as well as either larger than Rayleigh theory allows or highly absorbing). This is the view favored by Gadsden (1975), who states that "if the needles are falling through the atmosphere they will tend to line up with their long axes horizontal". But aerodynamic forces greatly weaken at altitudes well below that of noctilucent clouds. Indeed, applying continuum aerodynamics to small particles at the mesopause, where the mean free path of molecules is about 0.5 cm, is not strictly valid. Nevertheless, if we appeal to analysis intended for tropospheric particles we can obtain some idea about the size particles must be if they are to be aligned by aerodynamic forces and how this size depends on temperature and pressure.

The stable orientation of an elongated particle falling in a viscous fluid is with its axis perpendicular to the direction of fall. Counteracting this tendency toward alignment, however, is thermal buffeting (i.e., Brownian rotation). Fraser (1979) calculated the cumulative probability distribution for particle orientation as a function of size for columns (needles) and plates. There is a very sharp transition between completely aligned and randomly oriented particles; for needles near sea level, the critical size below which the needles are randomly oriented is about 12 μm. The value of the critical size depends on the parameter $Q/k_B T$, where k_B is the Boltzmann constant, T is the absolute temperature, and Q is related to the aerodynamic couple acting on the particle. This parameter is proportional to pc^7/T^3, where c is the particle size and p is the pressure. Therefore, the critical size is not very sensitive to pressure and temperature, although it increases slightly with increasing altitude.

Extrapolation from the troposphere to the mesosphere is, in general, invalid. But in this instance the critical size increases even faster than predicted by extrapolating Fraser's results. Indeed, Reid (1975) has calculated the force on an arbitrarily shaped particle, small compared with the molecular mean free path (molecular flow regime), from which it follows that the net couple acting on such a particle is zero.

If aerodynamic forces do not align mesospheric particles then what other mechanisms are there? If we accept that the particles are composed of ice, then magnetic alignment is not possible. The only remaining possibility is electrical alignment, which requires that the particles have a dipole moment, either induced or permanent; merely being charged is not enough.

The maximum dipole moment p induced in a particle of radius a by a field E is $p = \varepsilon_0 \alpha E$, where ε_0 is the permittivity of free space and the (static) polarizability α is $4\pi a^3$. The couple required to rotate the dipole is $pE = 4\pi\varepsilon_0 a^3 E^2$, which is proportional to the energy required to rotate the dipole from its stable orientation through 90°; this must be greater than $k_B T$, the molecular kinetic energy, if the particle is to be aligned by the field. If a is taken to be 0.5 μm and T is 150 K, then the electric field for which pE equals $k_B T$ is about 12,000 V m^{-1}. The electric field (fair weather) is about 100 V m^{-1} at sea level and decreases rapidly with altitude. Electrical alignment by this mechanism may therefore be ruled out.

Suppose that a particle has a permanent dipole moment qd by virtue of having acquired an equal number of positive and negative charges, both with total charge q; d is the charge separation. If E is taken to be 1 V m^{-1} and $d = 0.5$ μm, then q must be more than $10^4 e$, where e is the electronic charge. This is highly unlikely: the electron density in the mesosphere is perhaps 10^3 cm^{-3}; if the number density of particles in noctilucent clouds is 1 cm^{-3}, then each particle must acquire more than ten times the maximum number of charges available to it!

7. Conclusions

The high degree of linear polarization observed in light from noctilucent clouds at scattering angles near 90°, particularly at ultraviolet wavelengths, together with the monotonic increase of polarization with scattering angle, strongly suggests that the particles cannot be much larger than about 0.1 μm. This is beyond the limit where Rayleigh theory is strictly valid but not greatly so. Even if some hitherto unsuspected mechanism for aligning noctilucent cloud particles were to come to light, thereby providing a possible explanation for the small amount of observed circular polarization, this would still not vitiate the constraints imposed on the maximum size of particles by the linear polarization measurements.

Whatever the shape of noctilucent cloud particles, they are randomly oriented in the absence of an alignment mechanism. A suitable mechanism for aligning small (~0.5 μm) particles in the mesosphere has yet to be proposed. But only a suspension of aligned particles (assuming that it is superposable on its mirror image, which rules out particles with helicity, either microscopic or macroscopic) can elliptically polarize unpolarized light upon single scattering (noctilucent clouds are sufficiently thin that multiple scattering within them is negligible).

A possible explanation for the circular polarization observed in light from noctilucent clouds is that the light illuminating them has acquired a degree of circular polarization because of multiple scattering in the long atmospheric paths it traverses; this is not inconsistent with the supposition that the cloud particles are composed of ice, randomly oriented, and sufficiently small that Rayleigh theory gives a reasonable approximation to their scattering properties. In this context, a statement by Witt *et al.* (1976) is apposite: "... although multiple scattering effects within the mesosphere itself need not be considered, the additional source of light from the lower atmosphere must be fully taken into account to allow the proper interpretation of upper-atmospheric polarization measurements."

The common practice of analyzing scattering data for nonspherical particles with Mie theory is not valid in general and can lead to both qualitatively and quantitatively incorrect conclusions. But Mie theory is a reasonably good approximation if the particles are not too large— but larger than allowed by Rayleigh theory—and if their optical constants are similar to those of ice at visible wavelengths.

Noctilucent cloud particles may be needle shaped. Even if they are, however, presently accepted estimates of their maximum size and volumetric scattering need not be revised.

8. Acknowledgements

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