Intensity pulsations at the aerosol particle center and in the main maximum of the internal optical field

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The paper presents the results of calculation of the optical field intensity distribution in particles of inorganic and organic materials with the refractive index of 1.4. It is shown that the optical field intensity at a drop center varies quasi-periodically with changing the diffraction parameter. The modulation period of the optical field dependence on the droplet diffraction is about unity. This makes it possible to find the intensity at the droplet center from the standard dependence calculated for a transparent particle. The calculation of optical field intensity in the main droplet maximum provides a more complicated dependence on the droplet diffraction parameter. It is shown that for the high-transparent particles with increasing size the quasi-periodic dependence of intensity in the main maximum on the diffraction parameter is replaced by a more complex pattern of interference pulsations. For high-absorption materials, two pulsation dependences, namely, the modulation dependence (the period is about 20) and the high-frequency one (the period is about unity) can be recognized. The results obtained can be used for direct calculation of evaporation of the particle material with the refractive index about 1.4 and different values of the imaginary part of the refractive index.

Introduction

A knowledge of the optical field distribution in the volume of aerosol particles is of critical importance for investigation of the behavior of aerosol particles in a laser beam (photophoresis of particles, explosion of particles in laser beams of a single-particle laser mass-spectrometer,[1] and so on).

To date the data on optical fields in aerosol particles are very scanty. As a result, many investigations of different processes, important for aerosol science, are based on simplified and often wrong concepts of the optical field distribution in aerosol particles and mechanisms of interaction of laser radiation with the particulate matter.

For example, in some works (Refs. 2 and 3), where the processes are simulated in a single-particle laser mass spectrometer, the Bouguer law is used to describe the optical field distribution in particles. In accordance with this assumption, maximal intensity of laser radiation affecting the particulate matter is on the illuminated surface of the particle. Moreover, it is assumed that the radiation intensity in the main maximum does not exceed the intensity of the incident radiation. At the same time, calculations of the optical fields in aerosol particles with high refractive index show the availability of the optical field maximum, which, as a rule, lies in the shadow hemisphere of the particle and greatly exceeds the intensity of unperturbed radiation.[4]

Analogous results were obtained for a wide spectrum of weakly absorbing particles.[4] However, for particles of strongly absorbing materials there may exist conditions when the main maximum of the optical field is located in the shadow hemisphere. In this case the radiation intensity in the main maximum of such a particle can significantly exceed the radiation intensity outside the particle.[5] It is self-evident that the conclusions of Refs. 2 and 3 based on such arbitrary assumptions on the optical field distribution in aerosol particles cannot be considered as justified.

In this paper we investigate the optical fields in aerosol particles with the refractive index of 1.4. This value of the refractive index is often observed in many organic liquids, solutions of sulfuric acid, different minerals, typical of atmospheric dusts.

1. Calculations of optical fields in particles

To calculate internal optical fields in a particle, we used the Mie series summation by the algorithms described in Ref. 6. Calculations for the center of a particle of radius a were performed by the relations from Ref. 7. The main maximum was found by comparing the intensities at the nodes of the grid spaced by $\Delta r = \lambda/(40\pi|m|^2)$ on the radius, where λ is the wavelength of the incident radiation, $\pi = 3.14...$, $m = n - i\kappa$ is the complex refractive index of the particulate matter. Angular distance between the grid nodes was $\Delta\theta = \lambda/(40a|m|^2)$.

Below the results of comparison of dependences of the optical field intensities on the diffraction parameter at the aerosol particle center and at the point of location of the main maximum of the internal optical field are given. In Ref. 4 it is shown that in the shadow hemisphere of weakly absorbing spherical particles, such as, for example, the drops of nitromethane, a giant

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maximum of the optical field is formed. In Ref. 8 it is shown that the results of investigation of optical fields, obtained for one wavelength, can be used for other wavelengths if the following similarity criteria are met:

$$n(\lambda_1) \approx n(\lambda_2); \tag{1}$$

$$\rho_1 \approx \rho_2; \tag{2}$$

$$\kappa_1 a_1 \approx \kappa_2 a_2, \tag{3}$$

where

$$\rho_i = 2\pi a_i/\lambda_i.$$

In Ref. 9 an important step was made to expand the similarity conditions of optical fields of aerosol particles. It is shown that the optical fields in particles coincide, if, in addition to (1) and (2), the following condition is fulfilled:

$$\kappa\rho \ll 0.005. \tag{4}$$

In this paper we describe a possibility of further widening of the similarity conditions for optical fields in the particles with different absorption coefficients of their matters.

The similarity criterion (1) requires a proximity of refractive indices of the compared matters. Therefore, we have selected a series of different sets of the matters and wavelengths (Table 1), for which the proximity of values of real parts of the refractive index to 1.4 is typical.[10–12] It should be noted that, in the wavelength range of 10.6 μm important for atmospheric optics, the real part of the complex refractive index of water is 1.264 (see Ref. 13). This magnitude increases as λ approaches to the visible range of the spectrum. As a rule, the data in Table 1 are given in order of increasing value of the imaginary part of the refractive index.

Table 1

No.	Particulate matter	Wave-length, μm	Refractive index		Ref.
			n	κ	
1	H_2O	2.5	1.395	9.307e−9	11
2	CaF_2	4.8	1.401	3.5e−8	11
3	CaF_2	5.5	1.392	3.5e−8	11
4	MgO	8.929	1.404	5.76e−4	11
5	KCl	20.0	1.395	7.64e−5	11
6	KBr	32.0	1.406	8.5e−4	11
7	ThF_4	7.0	1.41	1.3e−3	11
8	ThF_4	7.5	1.39	1.3e−3	11
9	$MgAl_2O_4$	7.6923	1.398	4.4e−3	11
10	Spinel (aluminum oxynitrite)	7.6923	1.39	7.5e−3	11
11	H_2O	4.4722	1.396	0.01064	11
12	H_2O	3.4483	1.401	0.01243	11
13	SiO_2	17.50	1.4	0.01361	10
14	$SrTiO_3$	10.2	1.402	0.019	11
15	CsI	83.3	1.4	0.028	11
16	Y_2O_3	12.195	1.41	0.035	11
17	$BaTiO_3$	11.36	1.4	0.0434	10
18	Hexafluorine-benzene	10.05	1.4	1.22	12
19	Monobromine-trichlormethane	10.6	1.4	3.2300e−3	12

Figure 1 shows the dependences B_c of the optical field at the particle center with different absorption indices on the diffraction parameter. It should be noted that the curve *1* reflects the results of calculations made for all the matters from Table 1 with numbers less than 13. Thus, this curve describes the dependence of the optical field relative intensity on the radius at different wavelengths for particles of different matters having the imaginary parts of complex refractive indices differed by many orders of magnitude. In practice, curve *1* can characterize the optical field at the center of any weakly absorbing sphere. This curve shows the precise value of the internal optical field for a wide diversity of sets of optical parameters ρ and m characterizing weakly absorbing particles.

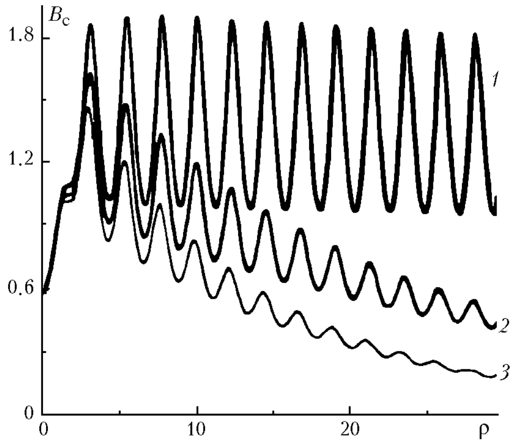

Fig. 1. Dependence of the optical field intensity at the center of a microsphere on the diffraction parameter for the microsphere's materials having the real part of the refractive index close to 1.4. Curve *1* is plotted for water droplets irradiated at λ = 3.4483 μm, $m = 1.401 - i0.01243$. Curve *2* – for microspheres of cesium iodine at λ = 83.3 μm, $m = 1.4 - i0.028$. Curve *3* – for $BaTiO_3$ at λ = 11.36 μm, $m = 1.4 - i0.0434$.

The calculating results show that for identical diffraction parameters and for matters with the same values of real parts of the refractive indices the optical field intensities at the center of different particles coincide until the difference between imaginary parts is so strong that the condition

$$|\kappa_1\rho_1 - \kappa_2\rho_2| \ll 1 \tag{5}$$

becomes broken.

As the imaginary part of the refractive index of the particulate matter increases, the dependence of the optical field intensity at the particle center more and more deviates from the dependence found for transparent particles.

The calculation of the optical field intensity at the droplet centers of all other materials given in Table presents the values intermediate between those shown in Fig. 1.

Figure 1 shows that the effect of the absorption index of the particulate matter on the field intensity at the droplet center increases with the growth of ρ, i.e., with increasing particle radius or with decreasing wavelength of incident radiation.

Now we consider two weakly absorbing particles. As it was mentioned above, the intensities of optical fields at the particle centers coincide if conditions (1), (2), and (5) are fulfilled. Let us use the condition of smallness of radiation absorption by particulate matter. Mathematically, the condition of smallness has the form:

$$\begin{cases} 4\pi \dfrac{\kappa_1\, a_1}{\lambda_1} \ll 1, \\ 4\pi \dfrac{\kappa_2\, a_2}{\lambda_2} \ll 1. \end{cases} \qquad (6)$$

As follows from Eq. (6), the condition (5) holds automatically for all weakly absorbing particles.

It follows herefrom that all weakly absorbing particles with the same values of the diffraction parameter and real part of the refractive index have close values of intensity of the optical field in the centers until the value of the product of the absorption index of the particulate matter by the particle radius remains small as compared to 1. This conclusion agrees well with the results of a wide series of calculations by the Mie theory (see Fig. 1).

This enables us to construct standard dependences of intensity at the particle center on parameters of the problem of aerosol optics of weakly absorbing particles.

If one of the matters is transparent, then the second condition from (6) is identically equal to zero. In this case the dependence of the optical field intensity at the particle center, found for the transparent matter, can be extended to any particles provided the conditions (1) and (2), as well as the condition of smallness

$$4\pi \frac{\kappa a}{\lambda} \ll 1 \qquad (7)$$

are fulfilled.

It should be noted that the condition (7) holds not only in the case of weakly absorbing particles, but with high degree of accuracy for any type of the particulate matter provided the particle radius is sufficiently small.

Thus, the dependence of the optical field intensity at the transparent particle center on the diffraction parameter can be treated as standard, i.e., the optical field intensity of any other particle with the real part of the complex refractive index close to that, for which the standard dependence is found, coincides with this dependence providing that condition (7) is fulfilled.

The obtained results simplify considerably the problem of finding a typical dependence of the optical field intensity at the droplet center on optical parameters of the problem. They give a possibility to find many characteristics of internal optical field from simple relations of similarity and standard curves.

The calculations show that any standard dependence of the optical field intensity at the droplet center on the diffraction parameter, found for a certain value of the real part of the refractive index, describes a certain (nonzero) range of radii of particles having the same value of the real parts of the complex refractive index and different values of its imaginary parts.

The lower is the value of the diffraction parameter, the better the condition (5) is fulfilled and the smaller is the error of description of the optical characteristic of this matter using the standard dependence.

2. Optical fields in the main maximum of the particle

Figure 2 illustrates a change of the intensity dependence in the main maximum of the internal optical field on the diffraction parameter for particles with close values of the real part of the refractive index ($n \approx 1.4$) as the absorption index κ of the particulate matter increases. Each curve was calculated for its own set of optical parameters of the matters (wavelength, real and imaginary parts of the complex refractive index) from the corresponding line of Table 1.

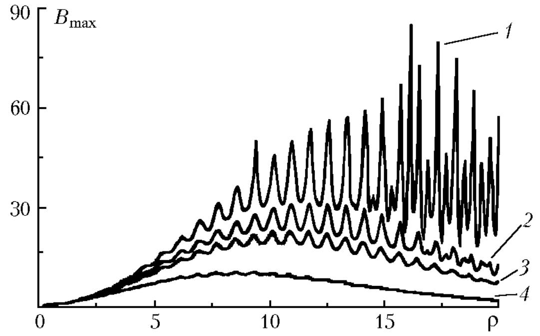

Fig. 2. Dependences of intensities in the main maximum on the diffraction parameter ρ in the particles with the refractive index $n \approx 1.4$. With increasing curve number the absorption coefficient κ of the particulate matter grows. $\kappa = 9.307 \cdot 10^{-9}$ for water droplets at $\lambda = 2.5$ μm (*1*), $\kappa = 0.01243$ for water at $\lambda = 3.4483$ μm (*2*), $\kappa = 0.019$ for $SrTiO_3$ at $\lambda = 10.2$ μm (*3*), $\kappa = 0.0434$ for $BaTiO_3$ at $\lambda = 11.36$ μm (*4*).

The calculations show that the dependences of the radiation intensity in the main maximum undergo modulations of at least two types. The period of the low-frequency modulation is about 24 ρ for transparent particles and decreases to 18 ρ with increasing imaginary part of the complex refractive index. The high-frequency modulation has the oscillation period about 1 ρ, which decreases to 0.8 ρ with increasing imaginary part of the complex refractive index (Fig. 2).

Thus, the variation of the imaginary part of the refractive index by more than six orders of magnitude leads to variation of the oscillation period by no more than 20 percent.

The intensity of the main maximum stronger depends on the imaginary part of the refractive index of the particulate matter. It is interesting that there is a certain range of values of the diffraction parameter where the radiation intensity in the main maximum rises with increasing the radiation absorption by the particulate matter. As a rule, this is due to the optical field resonances.

With increasing diffraction parameter the intensity of the main maximum, as a rule, rises. For all the matters, given in Table, such a situation is observed, at least, up to values of the diffraction parameter close to 6.8 (see Fig. 2). For large values of ρ the situation is complicated by a new level of interference effects.

Interesting results were obtained from calculations conducted for highly transparent matters. For example, curve *1* shows the intensity variation in the main maximum in water droplets at $\lambda = 2.5$ μm ($m = 1.395-i9.307\cdot10^{-9}$). In this range of wavelengths the water droplets are highly transparent. The curve of dependence of the main maximum on the diffraction parameter becomes more complicated with increase of the latter. For high values of $\kappa\rho$ the radiation intensity in the main maximum of a particle decreases with increasing imaginary part of the complex refractive index.

Numerical investigation has shown that the influence of variation of the absorption coefficient of high-transparent matters ($\kappa\rho \le 0.01$) on the intensity in the main maximum is nonmonotone. As the imaginary part of the refractive index increases, the radiation intensity in the main maximum can decrease or increase with equal probability. This is due to the fact that the main maximum is the result of complex interference of internal reflected waves. And small detuning in the form of increasing imaginary part of the refractive index can result in appearance of the resonance with corresponding rise of intensity in the main maximum.

The best example is the resonance close to the diffraction parameter of 16.2. In this case the increase of the imaginary part of the refractive index from $9.307\cdot10^{-9}$ to $3.5\cdot10^{-8}$ results in occurrence of a giant maximum of the optical field with the intensity 6300 times higher than that of the incident radiation. It should be noted that this intensity hundred times exceeds the intensity of any maxima detected by now in this range of optical characteristics.

The calculation results indicate that the dependence of the optical field intensity in the main maximum on the diffraction parameter and the imaginary part of the refractive index of a particle is more complicated than the same dependence for the particle center. We failed to construct, based on the standard curves, the criteria of similarity for a search for the dependence of the optical field intensity in the main maximum on the incident optical field intensity as it has been gained for the particle center. Systematization of data on the intensities in the main maximum is very complicated and requires looking over all dependences for different real and imaginary parts of complex refractive indices and diffraction parameters.

Conclusions

We have shown that the dependence of the optical field intensity at the droplet center on the diffraction parameter for a spherical particle with nonzero value of the imaginary part of the refractive index can be found from the standard curve constructed for spherical particles with the same values of real part of the complex refractive index.

It is shown that dependence of intensity in the main maximum on diffraction parameter ρ has the low-frequency modulation with a period about 20 ρ and high-frequency quasi-periodic oscillations with the frequency about 1 ρ. It is interesting to note that both modulation periods decrease slightly (no more than by 20 percent) when passing from high-transparent matters to high-absorbing ones.

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