SOME PECULIARITIES IN THE RESONANCE STRUCTURE OF THE SCATTERING PARAMETER FOR OPTICALLY ACTIVE SPHERICAL PARTICLES

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The influence of the optical activity particle material on their light scattering characteristics is investigated. Results of a numerical analysis of the resonance structure of the functional dependence of the scattering and absorption coefficients on the diffraction size of the particles are presented. A number of peculiarities are noted for particles composed of optically active materials, along with the tendencies inherent in the behavior of these quantities for optically inactive particles.

The continued improvement of the optical methods used for investigating the properties of aerosol necessitates the analysis of finer scattering and absorption effects, in particular, resonance effects.

In this paper we present the results of computations of the extinction efficiency factor $K_E(m, p)$ and the absorption efficiency factor $K_A(m, p)$ having resonance structures due to resonance conditions of the electric and magnetic partial waves inside dielectric, weakly absorbing spheres as functions of their optical activity.

The extinction efficiency factor $K_E(m, p)$ and the scattering efficiency factor $K_S(m, p)$ turn out to be different in the case of optically active particles and different directions of circular polarization of the incident light[1,2]:

$$K_{SR}(m,p) = \frac{2}{\rho^2}\sum_{n=1}^{\infty}(2n+1)\,[\,|a_n|^2 + |b_n|^2 + 2|c_n|^2 + 2\,\mathrm{Im}\,\{(a_n + b_n)\,c_n^*\}];$$

$$K_{SL}(m,p) = \frac{2}{\rho^2}\sum_{n=1}^{\infty}(2n+1)\,[\,|a_n|^2 + |b_n|^2 + 2|c_n|^2 - 2\,\mathrm{Im}\,\{(a_n + b_n)\,c_n^*\}];$$

$$K_{ER}(m,p) = \frac{4}{\rho^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}\,\{a_n + b_n + 2ic_n\};$$

$$K_{EL}(m,p) = \frac{4}{\rho^2}\sum_{n=1}^{\infty}(2n+1)\,\mathrm{Re}\,\{a_n + b_n - 2ic_n\}.$$

where $K_{SL}(m, p)$ and $K_{SR}(m, p)$ are the scattering efficiency factors for clockwise and counterclockwise circular polarizations of the incident radiation, respectively, and $K_{ER}(m, p)$ and $K_{EL}(m, p)$ are the extinction efficiency factors for the same polarizations, respectively.

The calculational program for the evaluation of the light scattering parameters of optically active spherical particles with complex index of refraction $m = n - i\kappa = 1.333 - i\times5\times10^5$ (where n is the index of refraction, κ is the index of absorption) was realized following a scheme analogous to that in the case of optically inactive particles[3]. A numerical analysis of the interference and fine resonance structure was carried out for $K_A(m, p)$ and $K_E(m, p)$, for diffraction parameter $\rho = 2\pi R/\lambda$ (where R is the particle radius and λ is the wavelength of the incident radiation, equal to 0.53 μm) in the range 20 to 30. The results of the first estimates showed that with increase of the optical activity of the particulate material ($\Delta m/m$, birefringence effects $\Delta n/n$, and .the effects of dichroism $\Delta\kappa l/\kappa$) some specific peculiarities are observed, namely transformations of resonance structure of the extinction and absorption efficiency factors, shifts of the resonance peaks toward smaller values of ρ, and others, along with tendencies already known for inactive materials[4–6].

The analysis of the problem of the influence of the optical activity of the particle material on the resonance structure of the field inside a particle and on the light scattering coefficients can be conditionally divided into three stages:

1. The change of the optical activity due to the change of the particles' total complex index of refraction m.

2. The influence of the birefringence (changes of the real part of the refractive index for one of the directions of circular polarization or for both directions but with different signs).

3. The influence of the phenomenon of dichroism (deviation of the imaginary part of the refraction index κ_R or κL from its mean value κ_m equal to κ of an inactive particle).

The ratio of the difference $m_{L(R)} - m_{R(L)} = \Delta m$ to the value of m for an inactive particle $\Delta m/m$ was chosen as the parameter characterizing the degree of the particle's optical activity. By analogy, the values $\Delta n/n$ and $\Delta\kappa/\kappa$ were chosen to characterize the effects of birefringence and dichroism, respectively. Let us consider in detail each of the above cases.

The range of variation of $\Delta m/m$ in the first series of computations was 0.03; 0.06; 0.12. Figure 1a illustrates the results of a comparison of the scattering coefficients for optically active particles with the corresponding values for optically inactive particles, provided that $m_L = m_R = m$. Marked changes of not only the interference structure of the extinction coefficient $K_E(m, p)$, but also the resonant structure are observed with increase of the optical activity. Against a background of a decrease of the interference component of $K_E(m, p)$ there is observed a tendency toward a degeneration of the number of elements in the peak structure, simultaneous with their shift to the region of smaller values of the diffraction parameter. For all this, an ambiguity of the contribution from the interference component of the waves with clockwise and counterclockwise circular polarizations was noted.

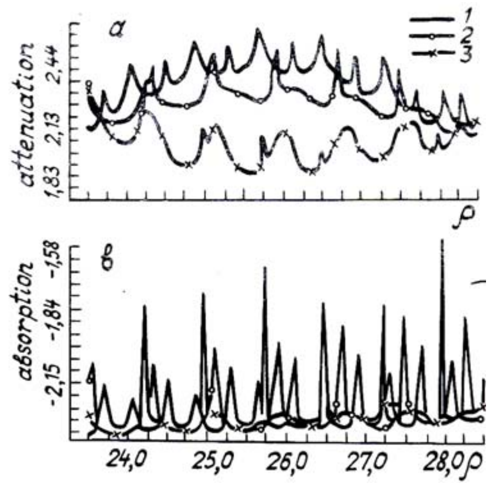

FIG. 1. Functional dependence of the efficiency: (a) represents weakening $K_E(m, p)$; (b) represents absorption $K_A(m, p)$. Curve 1 is for inactive spherical particles with $m = 1.333 - i{\times}5{\times}10^{-5}$; curve 2 is for optically active particles with $\Delta m/m = 0.03$; curve 3 is for $\Delta m/m = 0.12$.

A characteristic feature of the behavior of the absorption coefficient of the optically active particles $K_A(m, p)$, shown in Fig. 1b, for the same rates of increase of the optical activity, is the gradual decrease of the number of maxima (from 13 for the inactive particles to 7–8 at 0.03 and 0.06 and 6 at 0.12), owing to the suppression of the resonance effects of the electric partial wave with simultaneous growth by a factor of 3 to 5 of the absorption coefficients due to the magnetic component for $\Delta m/m$ relative to that for inactive particles. Quasiperiodic behavior of K_A in the considered range of the diffraction parameter ρ for active particles is preserved and is characterized by the amplitude $\Delta\rho \sim 0.55$. It should be noted that in this case $m_R < m_L$. The calculations showed that for $m_R > m_L$ the wave with counterclockwise circular polarization exhibits a stronger resonance behavior. As to the increase of the amplitudes of the resonance peaks $K_A(m, p)$ of the electric and magnetic partial waves with increase of the order of the wave for the inactive particles, for $\Delta m/m = 0.03{\div}0.12$ it is preserved for peaks of the type $b_n^{(1)}$, as was shown by calculations with greater resolution (~ 0.001) of the diffraction parameter ρ. In Fig. 1b the dips in the envelope of the maxima of $K_A(m, p)$ for $\Delta m/m = 0.12$ are caused by insufficient resolution in ρ (~0.01).

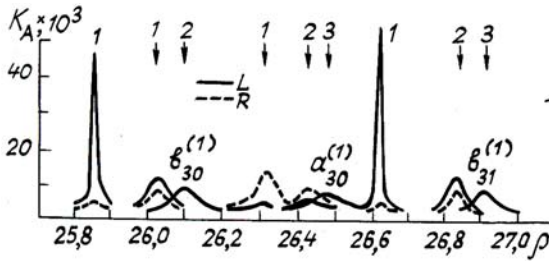

FIG. 2. Dynamics of the growth of the peak values of $K_{AL}(m, p)$ and $K_{AR}(m, p)$ with increase of the degree of the birefringence phenomenon: (1) $\Delta n/n = 0.05$; (2) $\Delta n/n = 0.005$; (3) $n_L = n_R = n$ – are optically inactive particles.

The importance of the birefringence phenomenon considered as a factor weakening the optical activity of the medium can be judged on the basis of the results of calculations of the absorption coefficients $K_A(m, p)$ and K_E(m, p) presented in Figs. 2 and 3 for $\Delta n/n$ = 0.0005–0.05 and in Table 1 for $\Delta n/n$ = 0.55–0.105. The calculations carried out for the extinction coefficients $K_{ER}(m, p)$ and $K_{EL}(m, p)$ demonstrated their marginal variation (by about ±5%) for $\Delta n/n < 0.05$. For $\Delta n/n \geq 0.05$ some correlation in the behavior of the extinction and absorption coefficients is observed.

The main features of the birefringence phenomenon are most explicitly seen in an analysis of the behavior of the absorption coefficient. Figure 2 enables one to trace out the dynamics of the growth of the peak values of $K_{AL}(m, p)$ and $K_{AR}(m, p)$ with the increase of the order of the partial wave, where $K_{AL}(m, p)$ is larger than $K_{AR}(m, p)$ for the given range of ρ, being five to six times greater than the amplitude of the corresponding partial wave for particles of optically inactive material, while n increases by only 5%. Simultaneously there is observed a shift of the maxima towards smaller values of the parameter ρ.

In addition to the peculiarities noted in Fig. 2, Fig. 3 allows one to see the conservation of quasiperiodicity of the maxima of the absorption coefficients. Thus, for $b_n^{(1)}$ $\Delta\rho$ = 0.75–0.76, while for $a_n^{(1)}$ $\Delta\rho$ varies from 0.802 to 0.806 with nearly linear growth of the peaks of the magnetic and electric partial waves $K_A(m, p)$ for ρ varying from 25.8 to 28.2.

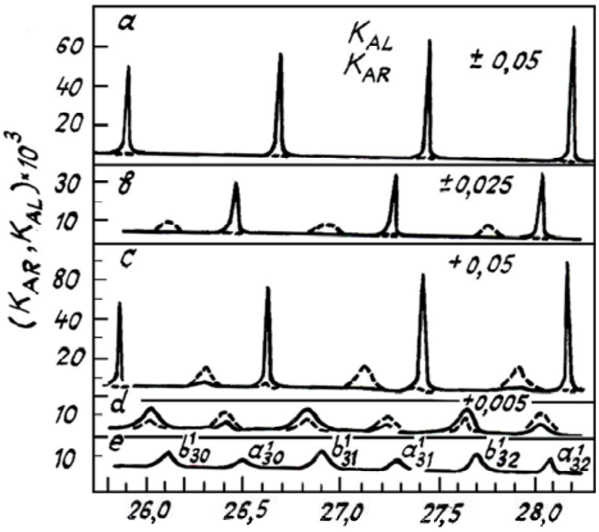

FIG. 3. Dependence of the absorption coefficients on the diffraction parameter for optically active particles (dashed curves correspond to $K_{AR}(m, \rho)$; solid curves represent $K_{AL}(m, \rho)$ and $K_A(m, \pi)$ for inactive particles):
a) $n_L = n \times (1 - 0.05)$, $n_R = n \times (1 + 0.05)$;
b) $n_L = n \times (1 - 0.025)$, $n_R = n \times (1 + 0.025)$;
c) $n_L = n$, $n_R = n \times (1 + 0.05)$; d) $n_L = n$, $n_R = n \times (1 + 0.005)$; e) $n_L = n_R = n$ — are inactive particles.

It is important to note that for weak optical activity ($\Delta n/n \sim 0.005$) resonance peaks are observed for radiation both with the clockwise and counter clockwise circular polarization with amplitudes only marginally different from those for the case of the inactive particles. When ЛIn/n increases up to a value of 0.05, there takes place a substantial (five- to sixfold) growth of the resonance peaks, caused by the magnetic component of the partial wave, changes of the peaks of the electric component of the partial wave being insignificant.

Interesting peculiarities in the behavior of the absorption coefficient of the optically active particles can be observed upon simultaneous variation of the index of refraction for radiation with either sense of circular polarization while keeping its mean value constant (Fig. 3a and 3b). Whereas a resonance structure can be seen in Fig. 3b for $n_R = n(1+0.025)$ and $n_L = n(1-0.025)$ similar to that shown in Fig. 3c, only with wider-based peaks, for $n_R = n(1+0.05)$ and $n_L = n(1-0.05)$ (Fig. 3a) the resonances of $K_{AR}(m, \rho)$ (or of $K_{AL}(m, \rho)$ with n_L and n_R reserved) caused by the electric partial wave degenerate. Calculations carried out for $n_R = n$ and $n_L = n(1-0.05)$ also showed that the peaks caused by resonances of the magnetic partial waves conserve their amplitudes, equal to the amplitudes of the resonances in the case of the optically inactive particles, but in this case the electric peaks degenerate.

As the birefrigence effect becomes more pronounced, as can be seen from Table 1 (here ρ_i are the positions of the centers resonance values of K_{AR} and K_{AL}; $\Delta n/n = 0.055, 0.06; 0.075; 0.105$), the main features noted for $\Delta n \le 0.05$ are preserved, and only for $\Delta n/n = 0.105$ and $\rho > 24.937$ do the amplitudes of the resonance peaks of the magnetic component of the partial wave begin to decrease, at the same time that $a_n^{(1)}$ (K_{AR} for $n_R > n_L$) continues to grow. Thus, there takes place a shift of the interference structure of $K_A(m, \rho)$ into the region of smaller ρ. Here it is possible to see an analogy with the results noted in Ref. 6, where the influence of the size of the water droplets on the structure of the internal particle field under resonance conditions was studied, in the fact that there exist regions of the diffraction parameter where the peak values a_n continue to grow while the peaks due to the magnetic component b_n begin to decrease since the envelopes of the electric and magnetic partial waves are shifted by one order of magnitude in ρ.

The importance of circular dichroism in the formation of the resonance structure of the internal field of optically active spherical particles and of the light scattering parameters of the medium can be judged from the results presented in Table 2. The increments of the absorption coefficients ΔK_{AR}, ΔK_{AL}, expressed as percents of the corresponding values of the coefficient K_A of the inactive particles at. the centers of the resonance peaks $\Delta A_{(R,L)} = \Delta K_{A(R,L)}/K_A$ can be used as parameters: characterizing the effect of dichroism. In contrast with the cases of variation of $\Delta m/m$ and $\Delta n/n$, over the entire range of variation off $\Delta\kappa/\kappa$ (0.5 to 100%) the positions of the resonance; peaks and their number in the examined interval $\rho = 23.5-28.5$ remain unchanged. With increase of the degree of dichroism only for one sense off circular polarization, e.g., $\kappa_{R(L)} = \kappa\left(1 + \frac{\Delta\kappa}{\kappa}\right)$, $\kappa_{L(R)} = \kappa$, a growth of the absorption coefficients, of both $K_{AR}(m, \rho)$ and $K_{AL}(m, \rho)$ by an amount (using' their sum as a basis of comparison) comparable with the change in the degree of dichroism,, i.e.,. $(\Delta_{AR} + \Delta_{AL}) \approx \Delta\kappa/\kappa$, is observed. The main contribution to $K_A(m, \rho) \sim 3/4$ comes from the radiation with unchanged κ, and the extinction coefficients and K_{ER} and K_{EL} decrease: insignificantly.

In the case of an increase: of the degree of dichroism of both senses of polarization $\kappa_{R(L)} = \kappa\left(1 + \frac{\Delta\kappa}{\kappa}\right)$, and $\kappa_{L(R)} = \kappa\left(1 + \frac{\Delta\kappa}{\kappa}\right)$ the absorption coefficient grows by an amount $\sim 1/4\Delta\kappa/\kappa$ for the radiation with the decreased values of κ and decreases by the same amount for the radiation with the increased values of κ. For the dichroism increasing in this way, the trends of the variation of the light scattering coefficients are weaker and the sum of absolute values $(\Delta_{AR} + \Delta_{AL}) \approx (1/2)\Delta\kappa/\kappa$ and $K_A = (K_{AL} + K_{AR})/2$

remains, practically unchanged due to the compensation of K_{AR} and K_{AL}. The extinction coefficient vary insignificantly (by ~ 0.005 to 2%) over the entire range of the degree of dichroism from 5% to 100%.

Table 1. Influence of birefringence on the absorption coefficient

$\Delta n/n$											
0.055			0.06			0.075			0.105		
ρ_i	K_{AR}	K_{AL}	ρ_i	K_{AB}	K_{AL}	ρ_i	K_{AR}	K_{AL}	ρ_i	K_{AR}	K_{AL}
23.899	10.42	4.82	23.899	10.42	4.82	23.887	14.74	61.20	23.867	10.93	4.80
24.262	5.76	43.35	24.186	5.81	44.85	24.634	8.13	70.46	24.026	6.34	105.50
24.703	11.64	4.96	24.702	11.70	4.94	24.692	11.90	6.11	24.673	12.55	5.02
25.024	6.12	48.10	24.945	6.22	52.47	25.380	6.77	78.90	24.750	7.39	106.60
25.511	13.03	5.10	25.503	13.08	5.09	25.498	13.32	6.31	25.474	14.83	102.40
25.783	6.53	56.08	25.702	6.67	60.55	25.124	6.66	86.99	26.196	6.98	93.51
26.313	14.65	5.24	26.312	14.73	5.25	26.302	14.96	6.79	26.279	15.43	5.46
26.541	6.96	63.98	26.458	7.18	68.69	26.868	6.76	92.10	26.918	6.12	81.43
27.118	16.50	5.39	27.115	16.57	5.44	27.101	16.47	7.35	27.079	17.36	5.73
27.299	7.42	71.64	27.212	7.89	76.27	27.610	6.87	93.82	27.638	5.94	68.14
27.919	18.67	5.52	27.917	18.76	5.73	27.906	19.07	7.85	27.878	19.61	6.14
28.055	7.87	78.32	27.965	9.66	82.44	28.351	6.99	91.82	28.357	6.95	55.22

*) Here $^*K_{AR}, K_{AL} \to K_{A(R,L)}(m, \rho) \times 10^3$

Table 2. Influence of dichroism on the absorption coefficient

ρ_1	$\kappa_L=\kappa$; $\kappa_R=\kappa(1+\Delta\kappa/\kappa)$ $\Delta\kappa/\kappa \cdot 100\%$										$\kappa_R=\kappa(1+\Delta\kappa/2\kappa)$, $\kappa_R=\kappa(1+\Delta\kappa/2\kappa)$, $\Delta\kappa/\kappa\cdot 100\%$					
	5%		10%		25%		50%		100%		25%		50%		100%	
	Δ_{AR}	Δ_{AL}	Δ_{AR}	Δ_{AL}	Δ_{AR}	Δ_{AL}	Δ_{AR}	Δ_{AL}	Δ_{AR}	Δ_{AL}	$-\Delta_{AR}$	Δ_{AL}	$-\Delta_{AR}$	Δ_{AL}	$-\Delta_{AR}$	Δ_{AL}
23.692	1.07	3.68	2.26	7.47	5.69	18.7	11.4	37.4	23.0	75.3	6.52	6.52	13.0	13.0	25.7	26.6
24.051	1.01	4.16	1.86	8.18	4.59	20.4	8.9	40.6	17.6	80.7	7.89	8.03	15.8	15.9	31.8	31.6
24.497	1.19	3.58	2.37	7.37	5.96	19.2	11.9	36.4	23.9	73.0	6.18	6.07	12.3	12.2	24.5	25.0
24.862	0.92	3.96	1.85	7.91	4.74	19.8	9.5	39.6	18.8	79.1	7.65	7.52	15.2	15.0	30.2	30.1
25.302	1.29	3.57	2.48	7.13	6.24	17.7	12.5	35.5	25.0	71.1	5.74	6.74	11.5	11.5	22.7	23.3
25.671	0.97	3.86	2.05	7.80	5.07	19.3	10.1	38.7	20.6	78.1	7.24	7.12	14.4	14.2	28.4	29.2
26.105	1.26	3.42	2.61	6.94	6.58	17.3	12.4	34.4	28.8	72.7	5.41	5.32	10.7	10.7	19.4	24.5
26.478	1.10	3.75	2.10	7.55	5.40	18.8	10.8	37.7	22.0	76.2	6.84	6.73	13.6	13.5	26.7	27.6
26.906	1.39	3.35	2.69	6.70	6.78	16.7	13.4	39.4	30.4	71.7	4.98	4.98	10.0	9.96	17.5	23.9
27.285	1.11	3.62	2.21	7.34	5.72	18.5	11.3	36.9	22.6	73.7	6.43	6.33	12.8	12.8	25.5	25.8
27.707	1.40	3.25	2.80	6.49	6.93	16.2	13.8	32.2	30.1	69.1	4.57	4.65	9.22	9.22	15.8	22.3
28.090	1.27	3.65	2.46	7.29	6.01	18.0	11.9	35.9	24.4	72.6	6.01	6.01	12.0	12.0	23.5	25.0
28.506	1.66	3 40	3.06	6.53	7.33	15.8	14.2	31.2	27.4	61.3	4.06	4.44	8.33	8.72	16.8	17.2

In conclusion let us briefly enumerate the main results of the above analysis.

1. Against a background of transformation of the interference component of the dependence $K_E(m, \rho)$ with growth of $\Delta\rho/\rho$ one can clearly make out a tendency toward degeneration of a number of elements of the peak (resonance) structure with simultaneous shift into the region of smaller values of the diffraction parameter.

2. With growth of the degree of birefringence there does not occur a mutual compensation of the diffracting partial waves of the two senses of circular polarization, as a result of which the resonance effect is increased.

3. The characteristic feature of the transformation of the resonance structure of the light scattering coefficients with growth of the 'optical activity of the material of spherical particles is a degeneration of resonances which are caused by the electric components of the partial waves and an increase of the intensity of he resonances in comparison with optically inactive particles for the partial waves of the magnetic mode.

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