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This report deals with the scattering properties of a moving nonspherical object modellized as cluster of spheres. Two particular aspects of the problem are studied:

1. The effect of the changes of shape of the scatterers as a consequence of mutual collisions; and
2. The effect of a very high and very low refractive index on the scattering properties of a particle with possible onset of resonance scattering.

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SCATTERING PROPERTIES OF A MOVING DIELECTRIC OBJECT

Final Technical Report

by

R. Saija

Principal Investigator

December 1984

United States Army

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Scattering Properties of a Moving Dielectric Object

FOREWORD

This report deals with the scattering properties of moving dielectric non-spherical objects, a subject that has been treated in strict collaboration with profs. F. Borghese, P. Denti and G. Toscano, University of Messina, Istituto di Struttura della Materia, and with dr O.I.Sindoni, CRDC, Aberdeen P.G., Md. USA.

The research reported in this document started in November 1983 and is, at present, under further development. In fact, the subject is rather promising and far from exhausted so that I plan to continue along the lines sketched below even after expiration of the present contract.

In view of the vastness of the subject, I will presently deal with two particular aspects of the scattering from moving objects. First of all I will consider the effect of the change of shape of the scatterers as a consequence of mutual collisions. This model is that commonly used to describe chemical reactions and is thus of wide interest. The second aspect I will deal with is that of the scattering properties of particles of very high and very low refractive index, with possible onset of resonance scattering. As will be seen below this phenomenon has been widely studied in the past more as a mathematical curiosity than as a true physical effect. However, as a consequence of general covariance of the Maxwell equations under holonomics space-time transformations, it may happen that, a fast-moving, scattering particle may appear to an observer in the laboratory as an object of very high or very low refractive index. Although the investigation of this particular subject is far from complete, I will present here some preliminary results which may be interesting for other researchers in the field.

PART A

EFFECT OF THE CHANGE OF SHAPE OR OF RECOMBINATION OF SCATTERERS ON THE OPTICAL PROPERTIES OF A POLYDISPERSE MODEL AEROSOL

Let us consider a homogeneous medium within which a number of scatterers are uniformly dispersed. The scatterers need not be all identical to each other but may be of several kinds, as is the case in polydisperse aerosol. I also assume that the scatterers are nonspherical but that their individual scattering properties can be well approximated by modelling them as clusters of spheres. The collective optical properties of the dispersion can be calculated by studying the features of the coherent propagation of the electromagnetic field. Strictly speaking, to be completely correct one should include in the equations for the propagation the effects of multiple scatterings among different scatterers and the effect of motion of the single scatterers. However the former effects can be neglected if one restricts itself to dispersions of low density as is the case of actual aerosols; as for the second effect it is well known to produce broadening of the peaks in the spectra of the single scatterers and no further detectable effects in view of the low speeds attained by the scatterers in actual aerosols. Anyway the effect on the collective spectrum of the dispersion is further smooted on account of the random and uncorrelated motion of the scatterers.
As a consequence of the above consideration our effort has been directed to see if some effect is detectable when, as a consequence of their motion, two or more scatterers collide and recombine to give scatterers of different structure. As will be seen in the following paper, which is the final version of a paper enclosed as a part of the Second Periodic Report, and that has been accepted for publication in Il Nuovo Cimento B, the effort has been successful for the change of shape is quite detectable in spite of the simplifications we assumed to reduce the computations.

Before closing this section I want to stress that the content of the above mentioned paper has been presented at the 1st Int. Conf. on Aerosol Res. and

Tech. held in Minneapolis, Mn. USA from 17 to 21 September 1984.

Furthermore I attended to the 4th Annual Meeting of the sector "Spectroscopy" of the Gruppo Nazionale di Struttura della Materia, held in Alghero, Sardinia, from 11 to 13 October 1984. In this meeting it emerged a new interest in both the experimental and theoretical aspects of physics and chemistry of clusters by the comunity of italian researchers. This circumstance and the results obtained in the past incourages me to continue the research outlined above even after the expiration of the present contract.

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Effect of the « Chemical Reactions » on the Absorption Coefficient of a Polydisperse Model Aerosol (*).

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(ricevuto il 5 Giugno 1984; manoscritto revisionato ricevuto il 18 Settembre 1984)

Summary. — In a preceding paper the absorption coefficient for the propagation of the electromagnetic field through a low-density dispersion of nonspherical scatterers of random orientation was calculated by modelling them as clusters of spheres. In this paper the spheres are allowed to rearrange among the clusters to simulate chemical reactions and the changes induced on the absorption coefficient of the dispersion are discussed in a few significant cases.

PACS. 42.20. - Propagation and transmission in inhomogeneous media.

1. - Introduction.

It is well established through experimental measurements that the refractive index and the absorption coefficient of a dispersion of identical scatterers

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sensibly depend on their shape (1-4). In order to compare the experimental results for the propagation through actual media with the theoretical predictions, two approaches are mainly used: the effective dielectric-constant theories (5-9) (*e.g.* the Maxwell-Garnett theory and the effective medium theory) and the coherent field theories (10-12) (*i.e.* the calculation of the coherent propagation of the electromagnetic field through an assembly of scatterers). Even the effects of the size distribution (5-8), of aggregation of particles (6,13) and of the orientation of the scatterers (11,12) were studied through these approaches.

For what concerns the theoretical investigations several authors dealt in different ways with the scattering by individual nonspherical objects (14-16). In particular, the present authors (17) as well as GERARDY and AUSLOOS (18,19) proposed independently to model some kind of nonspherical scatterers as clusters of spheres. The individual optical response of these model scatterers can be effectively calculated as a function of their orientation with respect to the incident field. Furthermore, in the framework of the coherent field theory we recently devised (12) an original formalism intended to study the effects of the anysotropy of the scatterers on the propagation of the electromagnetic field through a low-density random distribution of clusters of spheres. In our formalism, the choice to use clusters of spheres as model scatterers allowed us, by the use of their specific features, to study the propagation through an assembly of scatterers having a high degree of complexity. On the other hand, the randomness of the distribution and the choice of the low-density limit,

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letting us disregard both the statistical averaging procedures and the effect of multiple scattering among different clusters, nonetheless allowed us to evidence the anysotropy effects in which we are interested. In spite of the prevalent interest in metallic scatterers (20-23), we resolved to include into the clusters only dielectric, *i.e.* nonconducting, spheres with frequency-independent refractive index; in this way the effects of the nonspherical shape of the individual clusters are not masked at all and prove to be quite important.

Since our formalism of ref. (12) also applies to dispersions containing more than one kind of identical scatterers (*e.g.* polydisperse aerosols), the cluster model is very suitable to describe the effects induced on the scattered field by modifications of the individual scatterers. Such modifications can, in turn, be described in the framework of the model by rearrangements of the spheres either within the same cluster or among different clusters. This mechanism could be identified with that of the chemical reactions as long as a cluster of spheres can represent a molecule from the point of view of classical optics.

In this paper we intend to show the effects of the above-mentioned « chemical reactions » on the absorption coefficient of the dispersion of scatterers. Therefore, in the following section, we concisely report our formalism for the absorption coefficient of a low-density random dispersion of clusters. In the last section we apply the formalism to the dispersions of the clusters modelling the « reagents » and the « products » of a few sample « reactions »; then we collect the results of the calculations so that the effects of the « reactions » are profitably displayed.

We conclude these introductory remarks by observing that, even in this paper, we consider only clusters of dielectric spheres, *i.e.* nonconducting, with frequency-independent refractive indices because we want to single out the influence of the structural modifications of the scatterers: in any case our formalism can be applied to clusters of conducting spheres with refractive indices obeying some dispersion relation.

2. – The constants of coherent propagation through a dispersion of clusters.

Referring elsewhere for further details (12), here we concisely recall how the refractive index and the absorption coefficient of a dispersion of identical clusters within a matrix can be calculated. In the formalism reported below the dispersion of clusters is assumed to be random and its number density low

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enough to allow us to disregard the multiple-scattering effects among different clusters. We also assume that the spheres in the clusters as well as the matrix including them are made of isotropic, homogeneous dielectric materials.

Let us consider whatever low-density dispersion of scatterers and send through it the circularly polarized plane-wave field:

$$E_\eta = \sqrt{2}\, E_0\, \boldsymbol{u}_\eta \exp[i\boldsymbol{k}\cdot\boldsymbol{r}]\,, \qquad iB_\eta = \eta E_\eta\,,$$

where

$$\boldsymbol{u}_\eta = (\boldsymbol{\varepsilon}_1 + i\eta\boldsymbol{\varepsilon}_2)/\sqrt{2}$$

with

$$\boldsymbol{\varepsilon}_m\cdot\boldsymbol{\varepsilon}_n = \delta_{mn}$$

and $\eta = \pm 1$ according to the helicity (²⁴). The (complex) refractive index matrix of the dispersion is consequently given by (²⁵)

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + \frac{2\pi}{Vk^2}\sum_\nu f_{\nu,\eta'\eta}\,,$$

where the index ν numbers the scatterers in the volume V and $f_{\nu,\eta'\eta}$ is related to the normalized forward-scattering amplitude of the ν-th scatterer through

$$f_{\nu,\eta'\eta} = \boldsymbol{u}^*_{\eta'}\cdot\boldsymbol{f}_{\nu,\eta}\,.$$

The absorption coefficient and the customary refractive index of the dispersion are given by (²⁵)

$$\gamma_\eta = 2k\,\mathrm{Im}\,(\mathcal{N}_{\eta\eta})\,, \qquad n_\eta = \mathrm{Re}\,(\mathcal{N}_{\eta\eta})\,,$$

respectively.

Now we recall that $\boldsymbol{f}_{\nu,\eta}$ is independent of the location of the ν-th scatterer, but does depend on its orientation with respect to the incident field. Therefore, if the low density of the dispersion is not too low, we can substitute $f_{\eta'\eta}(\theta)$ for $f_{\nu,\eta'\eta}$ and write

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + \frac{2\pi}{k^2}\int N(\theta)\, f_{\eta'\eta}(\theta)\,\mathrm{d}\theta\,,$$

where the argument θ is a collective symbol for the Euler angles that individuate the orientation of the scatterers and $N(\theta)$ is the number density of the scatterers with orientation θ. At this stage we specify the scatterers in the dispersion

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as clusters of spheres and assume that their orientation is random. Then we express $f_{\eta'\eta}(\theta)$ in terms of the amplitudes of the multipole fields scattered by any cluster with orientation θ and, making full use of the features of the cluster model, we succeeded in factorizing this quantity into a part depending only on the structure and a part depending only on the orientation. This allows us to perform the integration over the orientation analytically with the result

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + \frac{N}{4k^3 i}\sum_{LM}\frac{1}{2L+1} W^*_{\eta' LM} W_{\eta LM}\sum_{M'}(\bar{U}^{(A)}_{\eta',LM'LM} + \eta'\bar{U}^{(B)}_{\eta',LM'LM}) . \tag{1}$$

For later purposes we rewrite

$$\mathcal{N}_{\eta'\eta} = \delta_{\eta'\eta} + N\sum_{L}\xi_{\eta'\eta L}S_{\eta'\eta L} , \tag{2}$$

where we define

$$\xi_{\eta'\eta L} = \frac{1}{4\pi i}\,\frac{1}{2L+1}\sum_{M} W^*_{\eta' LM} W_{\eta LM} ,$$

$$S_{\eta'\eta L} = \frac{1}{k^3}\sum_{M'}(\bar{U}^{(A)}_{\eta',LM'LM} + \eta'\bar{U}^{(B)}_{\eta',LM'LM} .$$

In the above equations N is the number density of the clusters; the quantities

$$W_{\eta LM} = 4\pi i^L\sqrt{2}\,u_{\eta}\cdot X^*_{LM}(k) ,$$

where the X_{LM}'s are vector spherical harmonics (24), are proportional to the multipolar amplitudes of the incident plane-wave field; the matrices $\bar{U}^{(A)}_{\eta'}$ and $\bar{U}^{(B)}_{\eta'}$, whose meaning will be discussed below, have a rather complicated expression for which we explicitly rely upon ref. (12).

By their very definition the W's and consequently the ξ's depend only on the direction of the wave vector of the incoming field. The $\bar{U}$'s as well as the S's depend on the magnitude of the wave vector of the plane wave and on the refractive indices of the spheres constituting the (identical) clusters in the dispersion. The $\bar{U}$'s account also for the structure of the clusters and need to be calculated once for all referring to a cluster whatever with arbitrary orientation. As a matter of fact, the $\bar{U}$'s are responsible for almost all of the amount of computations required by eq. (1) because their calculation implies the inversion of a matrix of order $2N_s L_M(L_M + 2)$, where N_s is the number of spheres in a cluster and L_M is the least value to which the sum over L must be extended to obtain well-converged results. The convergence of the multipolar expansion on which eq. (1) is grounded has been discussed elsewhere (17,18); of course we shall return to this subject when considering the specific computation reported in the next section. Finally it is usefull to recall that, if the clusters possess symmetry elements, such as reflection planes and symmetry axes, group theory can substantially help to compute the $\overline{U}$'s for it implies the inversion of matrices of smaller order than otherwise ([16,17]).

3. – Changes in the absorption coefficient caused by « chemical reactions ».

In the case of a low-density dispersion containing more than one kind of clusters, eq. (2) becomes ([15])

$$\mathcal{A}_{\eta\eta'} = \delta_{\eta\eta'} + N \sum_{\alpha,l} c_\alpha \xi_{\eta'\eta l} S_{\alpha\eta\eta' l} ,$$

where the index α refers to the α-th kind of clusters with number density $N_\alpha = Nc_\alpha$.

Now, it is well understood that, if a « chemical reaction » rearranges the spheres among the clusters, it determines, starting with the relative concentrations c_α of the « reagents », the relative concentrations d_β of the « reaction products » as well as those of the « residues of the reaction », c'_α (at least one of these latter is zero when the « reaction » is over). Therefore, we can calculate the absorption coefficients before and after the « reaction » and denote them by

$$\gamma_\eta^{(i)} = N \sum_\alpha c_\alpha Z_{\alpha\eta}$$

and

$$\gamma_\eta^{(f)} = N \left(\sum_\alpha c'_\alpha Z_{\alpha\eta} + \sum_\beta d_\beta Z_{\beta\eta} \right) ,$$

respectively, where

$$Z_{\alpha\eta} = 2k \, \mathrm{Im} \left(\sum_l \xi_{\eta\eta l} S_{\alpha\eta\eta l} \right) .$$

Clearly, a very useful quantity in detecting the structural effects on the absorption is

$$\Gamma_\eta = \frac{p\gamma_\eta^{(f)} + (1-p)\gamma_\eta^{(i)}}{\gamma_\eta^{(i)}} = p\Delta_\eta + 1 ,$$

where p is the completion index of the « reaction » ($0<p<1$), and

$$\Delta_\eta = \frac{\gamma_\eta^{(f)}}{\gamma_\eta^{(i)}} - 1 .$$

In the following part of this section we present the results for a few « chemical reactions ». Before any explanation and comment, however, we want to stress a point to avoid any substantial misunderstanding of the purposes of the present work. Our results refer to sample calculations, indeed, and do not pretend to give a realistic description of actual optical phenomena: too many details should be added to the very schematic cluster model. Consequently one should not bother neither about the choice of the parameters defining the structure of the clusters nor about the possibility of the « reactions » in the assumed low-density limit of the dispersion. We confirm that our question is: if the structure of the cluster changes, how much is the absorption coefficient affected? The results we are going to discuss make us answer that, in general, these changes of structure modify to a significant extent the coherent propagation of the field through the dispersion.

The « reactions » for which Γ_η was actually computed are listed in table 1.

TABLE 1. – *List of the « reactions » considered in this paper.* The refractive indices and radii of the elements A, B and C are $n_A = 1.30$, $n_B = 1.50$, $n_C = 1.40$ and $r_A = 1.0u_l$, $r_B = 0.50u_l$, $r_C = 0.75u_l$, where u_l is an arbitrary unit.

Reaction	Example
1) $3B_2 \rightarrow 2B_3$	$3O_2 \rightarrow 2O_3$
2) $2AB + B_2 \rightarrow 2AB_2$	$2CO + O_2 \rightarrow 2CO_2$
3) $2AB_2 + B_2 \rightarrow 2AB_3$	$2SO_2 + O_2 \rightarrow 2SO_3$
4) $A_2 + 3B_2 \rightarrow 2AB_3$	$N_2 + 3H_2 \rightarrow 2NH_3$
5) $AC + 3B_2 \rightarrow AB_4 + B_2C$	$CO + 3H_2 \rightarrow CH_4 + H_2O$
6) $2AB_3 + C_2 \rightarrow 2ACB_3$	$2PH_3 + O_2 \rightarrow 2POH_3$
7) $C_2 + AB_4 \rightarrow AB_3C + BC$	$Cl_2 + CH_4 \rightarrow CH_3Cl + HCl$
8) $2C_2 + AB_4 \rightarrow AB_2C_2 + 2BC$	$2Cl_2 + CH_4 \rightarrow CH_2Cl_2 + 2HCl$
9) $3C_2 + AB_4 \rightarrow ABC_3 + 3BC$	$3Cl_2 + CH_4 \rightarrow CHCl_3 + 3HCl$
10) $4C_2 + AB_4 \rightarrow AC_4 + 4BC$	$4Cl_2 + CH_2 \rightarrow CCl_4 + 4HCl$

A, B and C label the spheres with the refractive indices $n_A = 1.30$, $n_B = 1.50$ and $n_C = 1.40$, and the radii $r_A = 1.0u_l$, $r_B = 0.50u_l$ and $r_C = 0.75u_l$, respectively, where u_l is an arbitrary unit. The clusters are individuated by their « chemical formulae » in terms of the « elements » A, B and C, and their structures are built so as to match those of the chemical compounds listed under the heading « EXAMPLE ». Since the radii and the refractive indices of the spheres are assumed not to change due to the « reactions », changes of « valence » or « bonds » are not accounted for. In table I the « reactions » are partitioned into three groups: the first group, « reactions » 1÷4, where only the spheres A and B are implied; the second group, « reactions » 5 and 6, where also the spheres C are implied; the third group, that includes the family of « reactions » 7÷10.

In table II we list the clusters as well as the « reactions » in which they are involved according to table I. We also report the co-ordinates of the centres of the spheres to complete the definition of the structures; in this respect one can observe that the neighbouring spheres touch each other.

Figure 1 shows, for $p = 1$, Γ_η with $\eta = 1$ *vs.* k for the « reactions » included in the first of the above-mentioned groups. For completeness we report in fig. 2 the quantity

$$g_{\alpha\eta} = Z_{\alpha\eta}/Z_{0}^{(0)}$$

Fig. 1. – Plot of Γ *vs.* k for $p = 1$ for the «reactions» 1-4 of table I.

Fig. 2. – Plot of g_{α} *vs.* k for the clusters implied in the «reactions» 1-4 of table I. The dashed curves refer to the «reagents» and the solid curves to the «reaction products».

with the same value of η, *vs.* k. In the above equation $N_{\alpha} Z_{\alpha\eta}$ is the absorption coefficient of a dispersion containing only clusters of the α-th kind and α numbers the clusters that occur in the «reactions» of the group under consideration (in this case the first). Furthermore,

$$Z_{\alpha}^{(0)} = \sum_{i \in \alpha} \sigma_{i}^{(\mathrm{tot})},$$

TABLE II. – *Co-ordinates of the centres of the spheres constituting the clusters implied in the «reactions» of table* I. The co-ordinates refer to each sphere in the same order in which it appears in the name of the cluster. The «reactions» in which each cluster is implied according to table I are also listed.

Cluster	Co-ordinates	Reactions
B_2	(−0.5, 0, 0) (0.5, 0, 0)	1, 2, 3, 4, 5
A_2	(−1.0, 0, 0) (1.0, 0, 0)	4
C_2	(−0.75, 0, 0) (0.75, 0, 0)	6, 7, 8, 9, 10
AB	(−1.0, 0, 0) (0.5, 0, 0)	2
AC	(−1.0, 0, 0) (0.75, 0, 0)	5
BC	(−0.5, 0, 0) (0.75, 0, 0)	7, 8, 9, 10
B_3	(0, 0.577 35, 0) (0.5, −0.288 67, 0) (−0.5, −0.288 67, 0)	1
AB_2	(0, 0, 0) (−1.5, 0, 0) (1.5, 0, 0)	2
AB_2	(0, 0, 0) (1.228 73, −0.860 36, 0) (−1.228 73, −0.860 36, 0)	3
B_2C	(1.023 94, −0.716 97, 0) (−1.023 94, −0.716 97, 0) (0, 0, 0)	5
AB_3	(0, 0, 0) (0, 1.5, 0) (1.299 04, −0.75, 0) (−1.299 04, −0.75, 0)	3
AB_3	(0, 0, 1.384 44) (0.5, −0.888 67, 0) (−0.5, −0.288 67, 0) (0, 0.577 35, 0)	4, 6
AB_4	(0, 0, 0) (−0.866 02, −0.866 02, 0.866 02) (−0.866 02, 0.866 082, −0.866 02) (0.866 02, −0.866 02, −0.866 02) (0.866 02, 0.866 02, 0.866 02)	5, 7, [illegible] 9, 10
ACB_3	(0, 0, 1.384 44) (0, 0, 3.134 44) (0, 0.577 35, 0) (0.5, −0.288 67, 0) (−0.5, 0.288 67, 0)	6
AB_3C	(0, 0, 1.384 44) (0, 0.577 35, 0) (0.5, −0.288 67, 0) (−0.5, −0.288 67, 0) (0, 0, 3.134 44)	7
AB_2C_2	(0, −0.375, 1.452 37) (0, 0, 0) (0, −0.375, 2.952 37) (0.75, −1.0, 0) (−0.75, −1.0, 0)	8
ABC_3	(0, 1.520 69, 0) (0, 3.020 69, 0) (0, 0.866 02, 0) (0.75, −0.433 01, 0) (−0.75, −0.433 01, 0)	9
AC_4	(0, 0, 0) (−1.010 36, −1.010 36, 1.010 36) (−1.010 36, 1.010 36, −1.010 36) (1.010 36, −1.010 36, −1.010 36) (1.010 36, 1.010 36, 1.010 36)	10

where σ_i^{tot} is the total cross-section of the i-th sphere belonging to a cluster of the α-th kind, calculated according to the Mie theory (24). Therefore, $N_\alpha Z_\alpha^0$ is the absorption coefficient of the dispersion of the clusters of the α-th kind when the multiple-scattering processes among the spheres within each cluster are disregarded. Γ_η and $g_{\alpha\eta}$ for the reactions of the second and of the third group are shown in the pairs of fig. 3-6, respectively. We notice that, since all the figures report the result for $\eta = 1$, this index has been everywhere dropped.

Fig. 3. - Plot of Γ *vs.* k for $p = 1$ for the «reactions» 5-6 of table I.

Fig. 4. - Plot of g_α *vs.* k for the clusters implied in the «reactions» 5-6 of table I. The dashed curves refer to the «reagents» and the solid curves to the «reaction products».

Fig. 5. - Plot of Γ *vs.* k for $p = 1$ for the «reactions» 7-10 of table I.

Fig. 6. - Plot of g_α *vs.* k for the clusters implied in the «reactions» 7-10 of table I. The dashed curves refer to the «reagents» and the solid curves to the «reaction products».

Figures 2, 4 and 6 are not immediately comparable with fig. 1, 3 and 5, respectively, on account of the different normalization; on the other hand, the chosen normalizations render Γ_η independent of N and $g_{\alpha\eta}$ independent of N_α, although it should be borne in mind that our results are valid only for low-density dispersions.

All our calculations were actually performed for k ranging from $0.002u_l^{-1}$ to $2.0u_l^{-1}$, so that the wave-length of the incident field was allowed to decrease to about the overall dimension of the clusters. However, in all figures, k ranges only from $0.2u_l^{-1}$ to $2.0u_l^{-1}$, the interval in which the results show a more striking k-dependence. We remark, however, that within the whole k-range the individual spheres have no resonance scattering as a consequence of the assumed radii and refractive indices (25).

Before discussing the results, we premise a few words about their convergence. In this respect several authors deserved a particular attention to the effect of the inclusion of higher-order multipolar contributions (26,27) so that it may be useful to point out that our approach (15,28,29) as well as that of ref. (26) is rather different. Actually, our formalism, and Gerardy and Ausloos' as well, require that the amplitudes of the electric and magnetic multipole fields scat-

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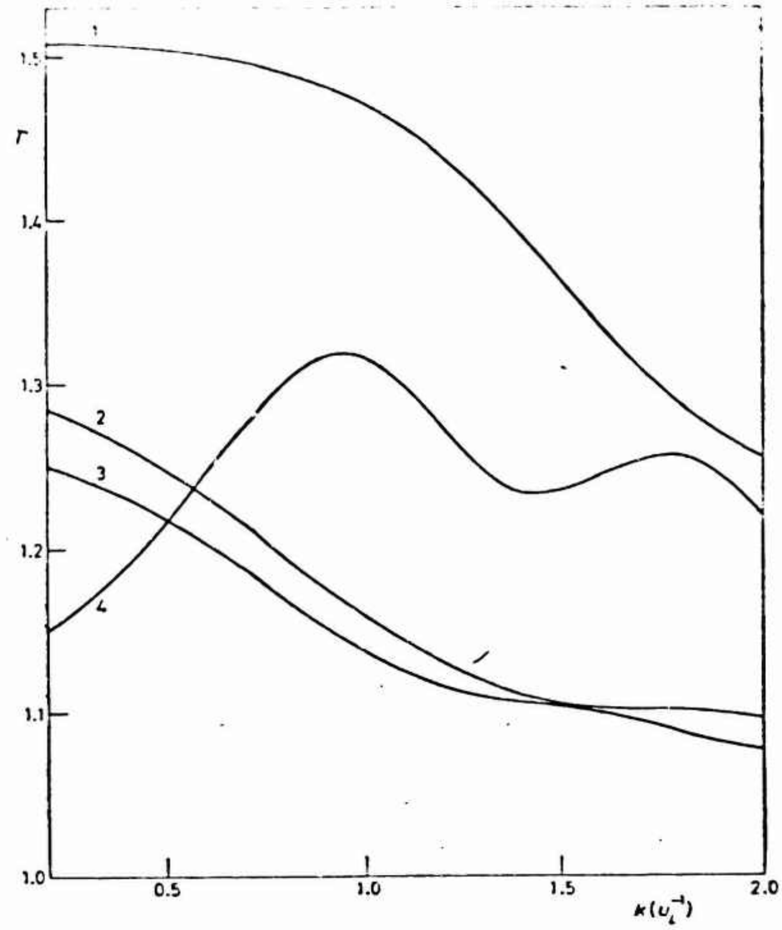
tered by a cluster be calculated simultaneously as the solution of a system of linear equations whose order increases with the truncation value of L, say L_T. As a consequence any calculation with $L_T < L_M$ not only neglects the higher-order multipoles, but also gives inaccurate values for the remaining multipolar amplitudes. Indeed the multiple-scattering processes among the spheres of each cluster enhance the strength of multipole fields of higher order (11,17,19,25,26), so that the L-expansions cannot be truncated to too low a value, whereas too high values imply time-consuming computations and enormous memory requirements. As a matter of fact, we found that a quite reasonable $L_M = 4$ ensures converged results for all the clusters considered here with k ranging from $0.002u_l^{-1}$ to $2.0u_l^{-1}$.

From a general point of view, fig. 2, 4 and 6 show clearly that the multiple-scattering processes among the spheres of a cluster cannot be disregarded without affecting dramatically the correctness of the results. Therefore, it is a very reasonable prediction that structural modifications of the clusters cause appreciable variations of the absorption of the dispersion. As we anticipated, this actually occurs: the Γ-ratios are significantly different from unity for k in the range from $0.002u_l^{-1}$ to $0.2u_l^{-1}$. In fig. 1, 3 and 5, however, we see that, when k increases, the effect of the « reaction » becomes smaller and smaller, although for almost all the « reactions » Γ is significantly different from unity at least up to $k \simeq 1.0u_l^{-1}$. In this respect we remark that, in general, the more the structure of the clusters is modified the more Γ remains different from unity. Even the curves of g_3 in fig. 2, 4 and 6 the less differ from each other the more similar are the structures. As an example, in fig. 1 the curves for CO_2 and SO_2 are indistinguishable from each other. The component spheres are identical for the two clusters AB_2, indeed, and the structural differences are too small to give appreciably different spectra also on account of the randomness of the orientations. We also notice the identity of the spectra of NH_3 in fig. 2 and PH_3 in fig. 4. According to table II, in fact, both « compounds » are AB_3 and have the same piramidal structure. These spectra differ from that of SO_3 in fig. 2, however, this latter « compound » is also an AB_3 but, according to table II, has a planar structure.

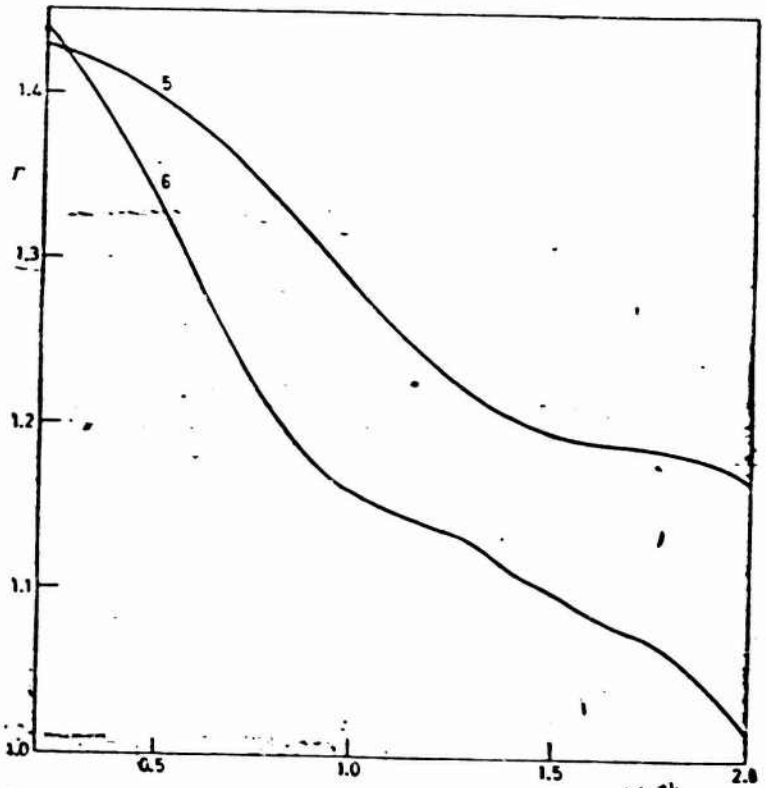
In conclusion, the cluster model accounts for the nonspherical shape of the scatterers so effectively that it renders the structural modifications quite detectable. In this sense, and on account of the possible improvements, the cluster model should be suitable for approaching the experimentally measured properties of real aerosols.

RIASSUNTO

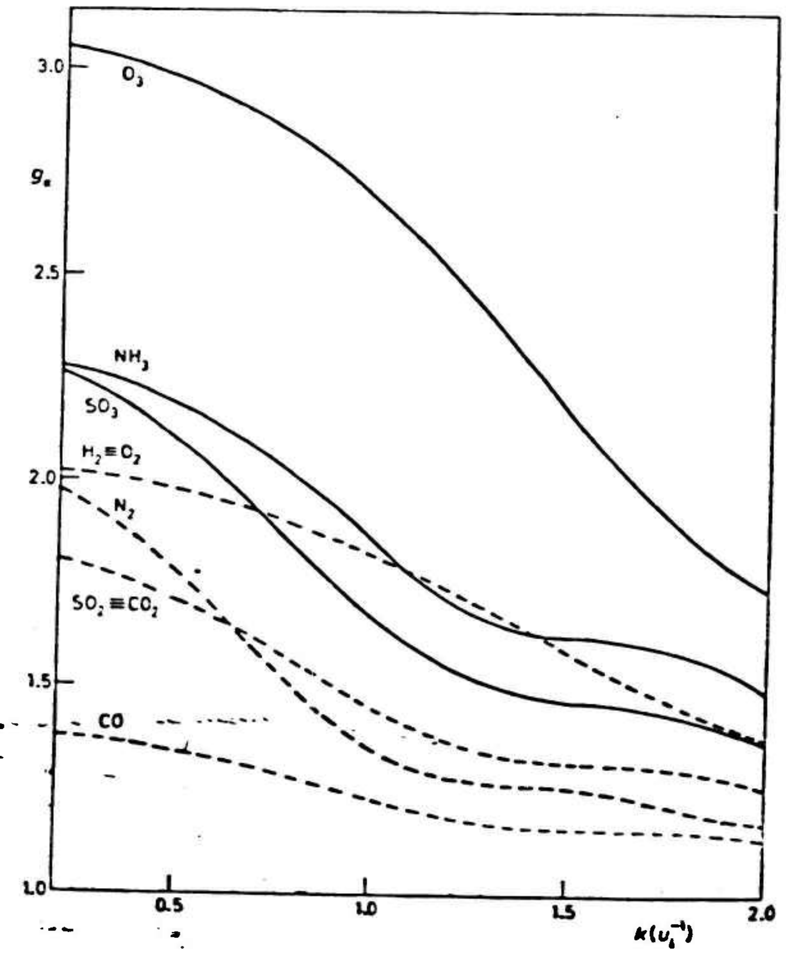
In un lavoro precedente è stato calcolato il coefficiente di assorbimento per la propagazione del campo elettromagnetico attraverso una dispersione a bassa densità di diffusori non sferici modellizzandoli come «cluster» di sfere. In questo articolo si ammette che i cluster possano cambiare struttura in maniera da simulare delle reazioni chimiche e si calcolano le conseguenti variazioni del coefficiente di assorbimento della dispersione in alcuni casi significativi.

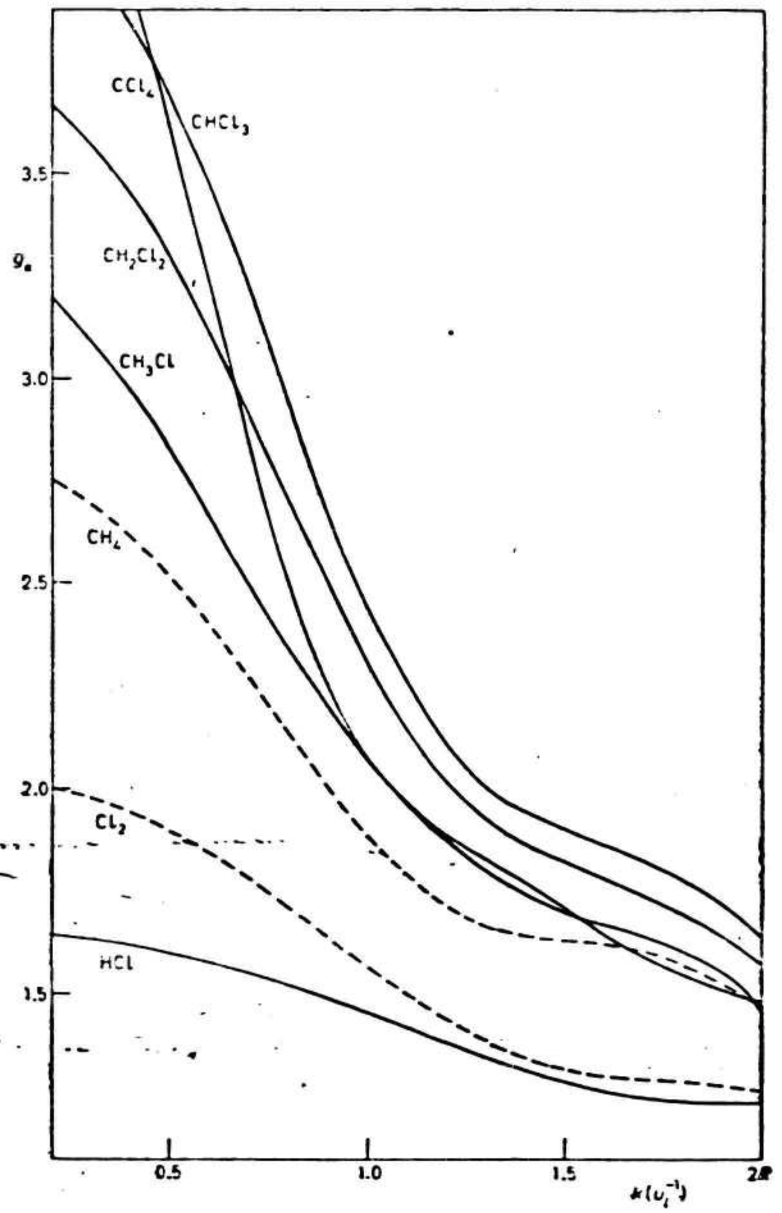


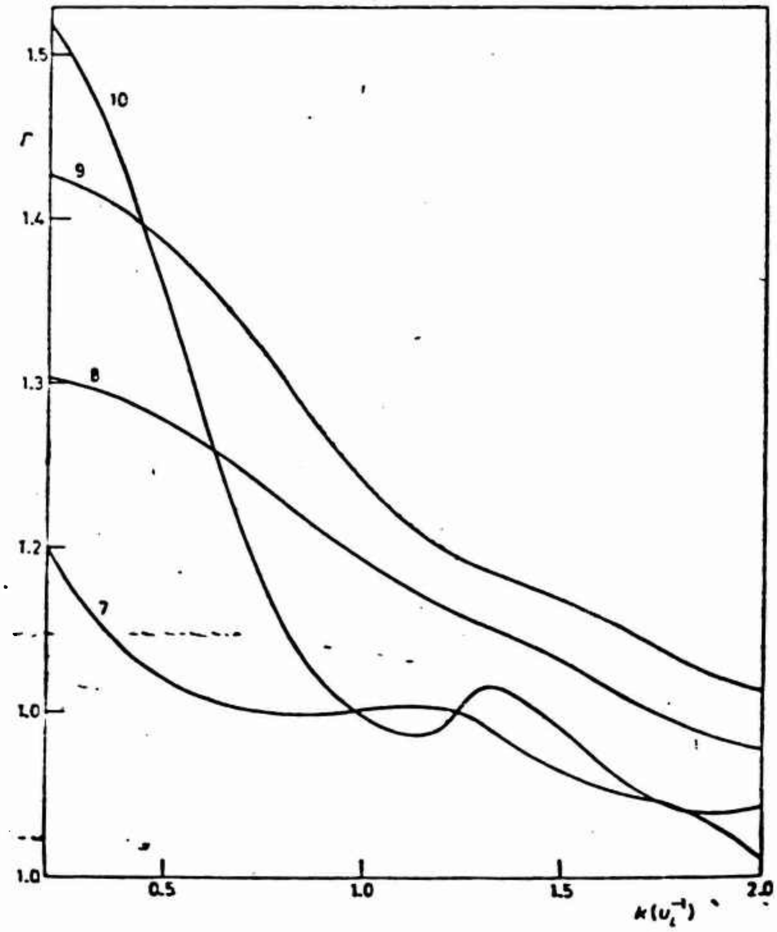
1.5
1.4
1.3
1.2
1.1
1.0
Γ
1
2
3
4
0.5
1.0
1.5
2.0
$k(u_L^{-1})$

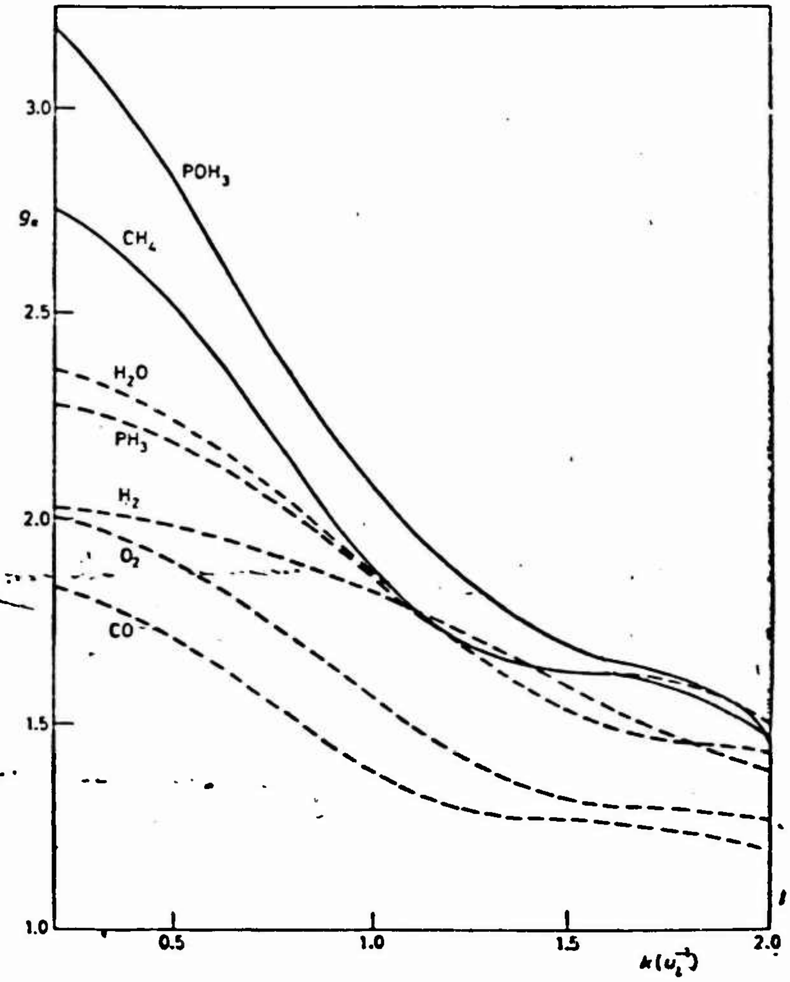


1.4
1.3
1.2
1.1
1.0
Γ
5
6
0.5
1.0
1.5
2.0
$k(u_L^{-1})$


3.0
g_e
2.5
2.0
1.5
1.0
0.5
1.0
1.5
2.0
$k(a_0^{-1})$
O_3
NH_3
SO_3
$H_2 \equiv O_2$
N_2
$SO_2 \equiv CO_2$
CO


CCl$_4$
CHCl$_3$
CH$_2$Cl$_2$
CH$_3$Cl
CH$_4$
Cl$_2$
HCl
g_e
3.5
3.0
2.5
2.0
1.5
0.5
1.0
1.5
2.0
$k(a_0^{-1})$


Γ
1.5
1.4
1.3
1.2
1.0
1.0
0.5
1.0
1.5
2.0
$k(U_L^{-1})$
10
9
8
7


3.0
g_e
2.5
2.0
1.5
1.0
0.5
1.0
1.5
2.0
$k(a_0^{-1})$
POH_3
CH_4
H_2O
PH_3
H_2
O_2
CO

PART B

RESONANCE SCATTERING FROM OBJECTS OF HIGH REFRACTIVE INDEX

1. INTRODUCTION

The study of resonance scattering from dielectric spheres is well known in the literature, and its matematics is well understood(1,2). However this effect actually occur only in spheres of very high refractive index, a feature which in practice is not possessed by any particle of real aerosols. Nevertheless the resonance scattering is able to explain a number of phenomena so that the assumption of high refractive index has been accepted by several workers even without a clear physical justification. We recall in this respect that the highest refractive index turns out to be 9 for water droplets in the far infrared.

Recently, Gérardy and Ausloos(3) pointed out that resonance scattering is an actual occurrence for small metallic particles embedded in a dielectric matrix. In fact, metals have a frequency dependent dielectric constant with a singularity at the plasmon frequency. At this frequency therefore the scattering cross section of small metallic particles presents a sharp peak due to the high value of the refractive index. This phenomenon is well justfied within the framework of solid state theory.

The situation is quite different for dielectric particles where the resonance of the free electron modes do not occur. Therefore in order to justify the existenceof high refractive indexes we took a different approach based on the general-relativistic treatment of the dielectric constant and magnetic permeability of a particle in general motion. According to a paper of Anderson and Ryon(4), indeed, on account of the covariance of Maxwell equations under holonomic space-time transformations, it is possible to write the dielectric tensor of a material medium in a form that takes account of the state of motion of both the medium and the observer. Without going into the calculational details here we simply state that the case in which we are interested is clearly that of an observer at rest while the medium, i.e. the scattering particle, is in general accelarated motion. However, this is not the whole story, for to be able to impose the correct boundary conditions across the surface of the particle we must also consider the case in which both the medium and the observer are in the same state of motion. Combining these two cases through the appropriate space-time transformation, allows us to conclude that for certain states of motion of the particles, an observer at rest sees a scatterer of very high refractive index.

At this stage the formalism of general relativity is no further necessary for the assumption of very high refractive index can be inserted within the customary theory of electromagnetic scattering. However, on account of the above considerations the occurrence of resonance scattering from dielectric particles gains a physical justification.

2. RESONANCE SCATTERING FROM A SPHERE

The theory of resonance scattering has been introduced within the framework of the formalism we developed to study the scattering properties of cluster of spheres. As will be seen later this work has proved to be more hard than expected, so we will give here only an outline and present some preliminary results. However it is first useful to summarize the features of the resonance scattering from a single sphere. To this end we recall that the scattering properties of a sphere of radius r and refractive index n_1 embedded in a homogeneous medium of refractive index n_2, are determined by the amplitudes of the scattered multipoles, a_ℓ and b_ℓ. As shown in standard textbook(2) these can be cast into the form:

$$a_\ell = \frac{\psi_\ell(\alpha)\,\psi'_\ell(\beta) - n\,\psi_\ell(\beta)\,\psi'_\ell(\alpha)}{\zeta_\ell(\alpha)\,\psi'_\ell(\beta) - n\,\psi_\ell(\beta)\,\zeta'_\ell(\alpha)} \qquad (1a)$$

$$b_\ell = \frac{m\,\psi_\ell(\alpha)\,\psi_\ell'(\beta) - \psi_\ell(\beta)\,\psi_\ell'(\alpha)}{m\,\zeta_\ell(\alpha)\,\psi_\ell'(\beta) - \psi_\ell(\beta)\,\zeta_\ell'(\alpha)} \qquad (2a)$$

where

$$\alpha = 2\pi r m_2/\lambda_0 = kr \quad ; \quad \beta = m\alpha$$

λ_0 being the wavelength in vacuo and $n=n_1/n_2$. The Riccati-Bessel functions, ψ_ℓ and ζ_ℓ are defined according to Kerker$^{(5)}$.

Now it is evident that if the denominators of eqs(1) vanish for any value of λ_0 and for a given ℓ, the corresponding a_ℓ and b_ℓ become infinite while $a_{\ell'}$ and $b_{\ell'}$ for $\ell \neq \ell'$ become very small. This phenomenon is known as resonance scattering of order ℓ. Note that, as shown by Van de Hulst$^{(2)}$, $a_\ell \approx b_{\ell-1}$ at resonance. Owing to the presence of the complex functions ζ_ℓ, however, the denominators of eqs.(1) cannot vanish for any real value of α and β. Nevertheless, provided $kr \ll 1$ and $n \gg 1$ there exist complex zeros with a small imaginary part. When one puts the real part of these zeros into eqs(1) one gets values of a_ℓ and b_ℓ that, although very large are not infinite. As a matter of fact one has to put $kr \approx 0.1$ or less and chose n of order 50.

3. RESONANCE SCATTERING FROM A BINARY CLUSTER

The above considerations suggest to study the effect of resonance scattering from a cluster of spheres in order to get information on the optical behaviour of certain kind of nonspherical impurities embedded in a homogeneous matrix. We started our study with a binary cluster in order to reduce the computational effort.

The first stage of the research was to find, for a given kr, the refractive index which produces resonance in the single spheres of cluster. Actually the search was made for $\ell=1,3,5$ and for $kr=0.1$. The results are summarized in fig.1 where the most striking feature is the indistingushability of the curves for different l even for the value of the main peak.The value of the scattering cross section σ_s at resonance is indicated on the figure. The n of resonance, actually ≈ 45 was then introduced into the theory for the scattering from a cluster for several values of the direction of incidence. In practice fig.2 and fig.3 report only the results for broadside (ϑ=90°) and head-on (ϑ=0°) incidence. The most interesting feature of fig.2 is the apparence of staellite peaks for values of n that are not of resonance, while in fig.3 some of these peacks disappear. This disappearence suggest the onset of some kind of shadow effect for head-on incidence, but the very origin of the satellite peaks is rather puzzling, as they were quite unspected on the basis of the general theory. We make the hypotesis that the satellite peaks originate from the concurrent effect of two circumstances. First of all the fact that, as mentioned above, $a_\ell \approx b_{\ell-1}$, at resonance; second, the formalism we use to calculate the scattered field from a cluster, includes the coupling of all multipoles, up to order l, of the constituent spheres. Since even the coupling of electric with magnetic multipoles is taken into account, it may well happen that the sum of the above two circumstances produces peaks in $b_{\ell-1}$ which though not as high as those of a_ℓ are shifted with respect to these latter. In any case this is at present only a reasonable hypotesis whose reliability is to be further investigated.

4. CONCLUSIONS AND LINES OF FUTURE PROGRESS.

The results illustrated in the figures are rather interesting but lack a reliable explanation. A careful analysis of the mathematical circumstances is, in fact, necessary in order to understand correctly the origin of the satellite peaks. This can be done performing the computations for highest values of ℓ

and for clusters of more complicated structures than the simple binary cluster consided thus far. Furthermore, the possible drop in numerical accuracy in the vicinity of the peaks should also be taken into account as a consequence due to the core storage limitations it is necessary to modify the present version of our programs so as to allow treating big clusters and/or large values of ℓ. This has already been done during our stay at the CRDC, last summer. The modified version of the program is at present under test and we plan to use it in the near future.

The details of the structure and the use of the modified program will be described by prof. Borghese in its next report on Contract DAJA37-81-C-0895.

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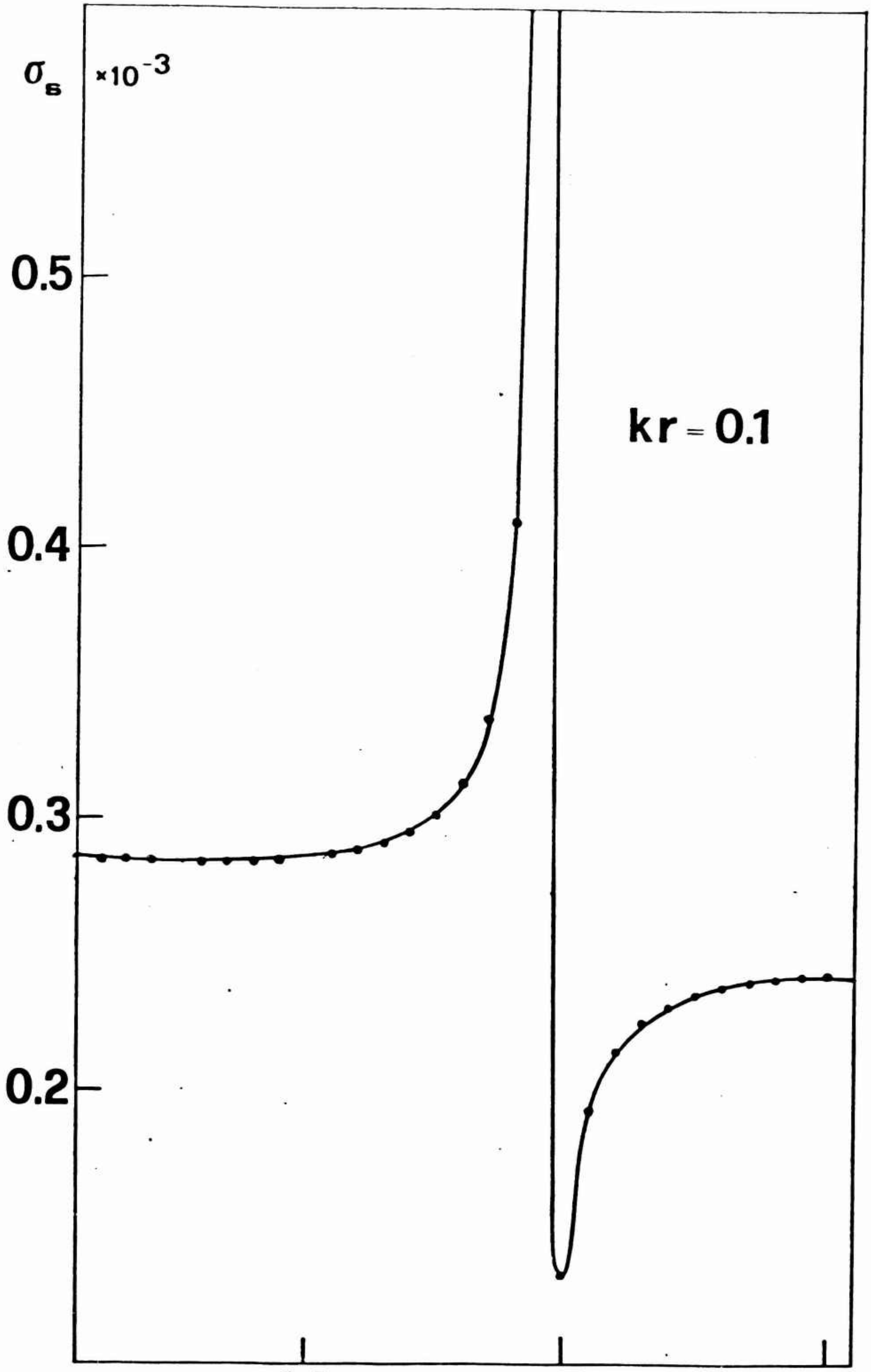
3. J.M.Gerardy and M.Ausloos: Phys. Rev. B25, 4204 (1982).
4. J.L.Anderson and J.W.Ryon: Phys. Rev. 181, 1765 (1969)
5. M.Kerker: "The scattering of light", (Academy Press, New York, 1969).

FIGURE CAPTIONS

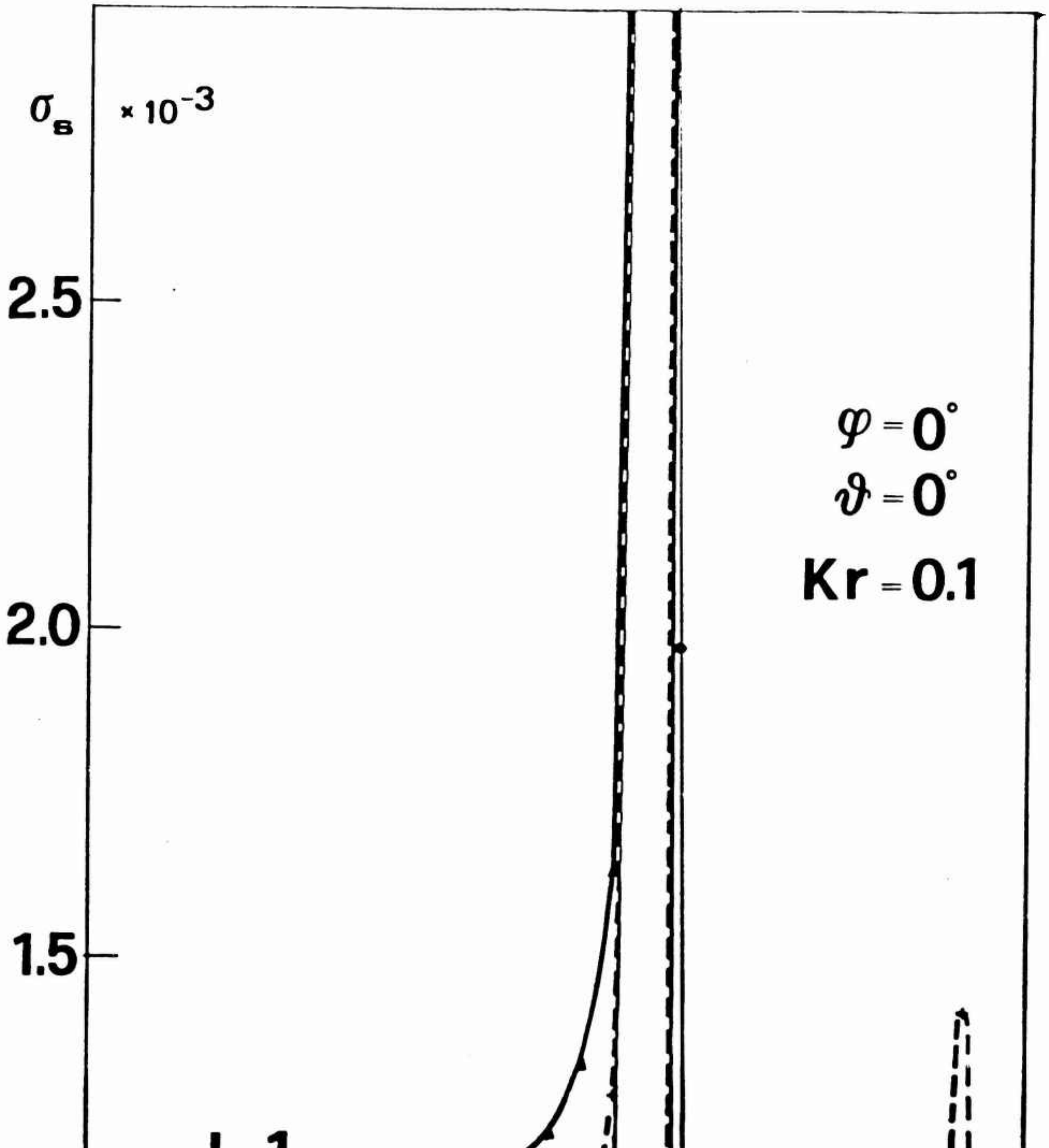
Figure 1: Scattering cross section of a sphere for kr=0.1 as a function of the refractive index n. The curves coincide for $\ell=1,3,5$ and the hight of the main peak is 3.26×10^{-3}.

Figure 2: Plot of σ_s for a binary cluster as a function of n near the resonance for broadside incidence.

Figure 3: Plot of σ for a binary cluster as a function of n near the resonance for head-on incidence.



σ_s ×10⁻³
0.5
0.4
0.3
0.2
kr = 0.1
44
45
n


σ_s ×10^{-3}
2.5
2.0
1.5
1.0
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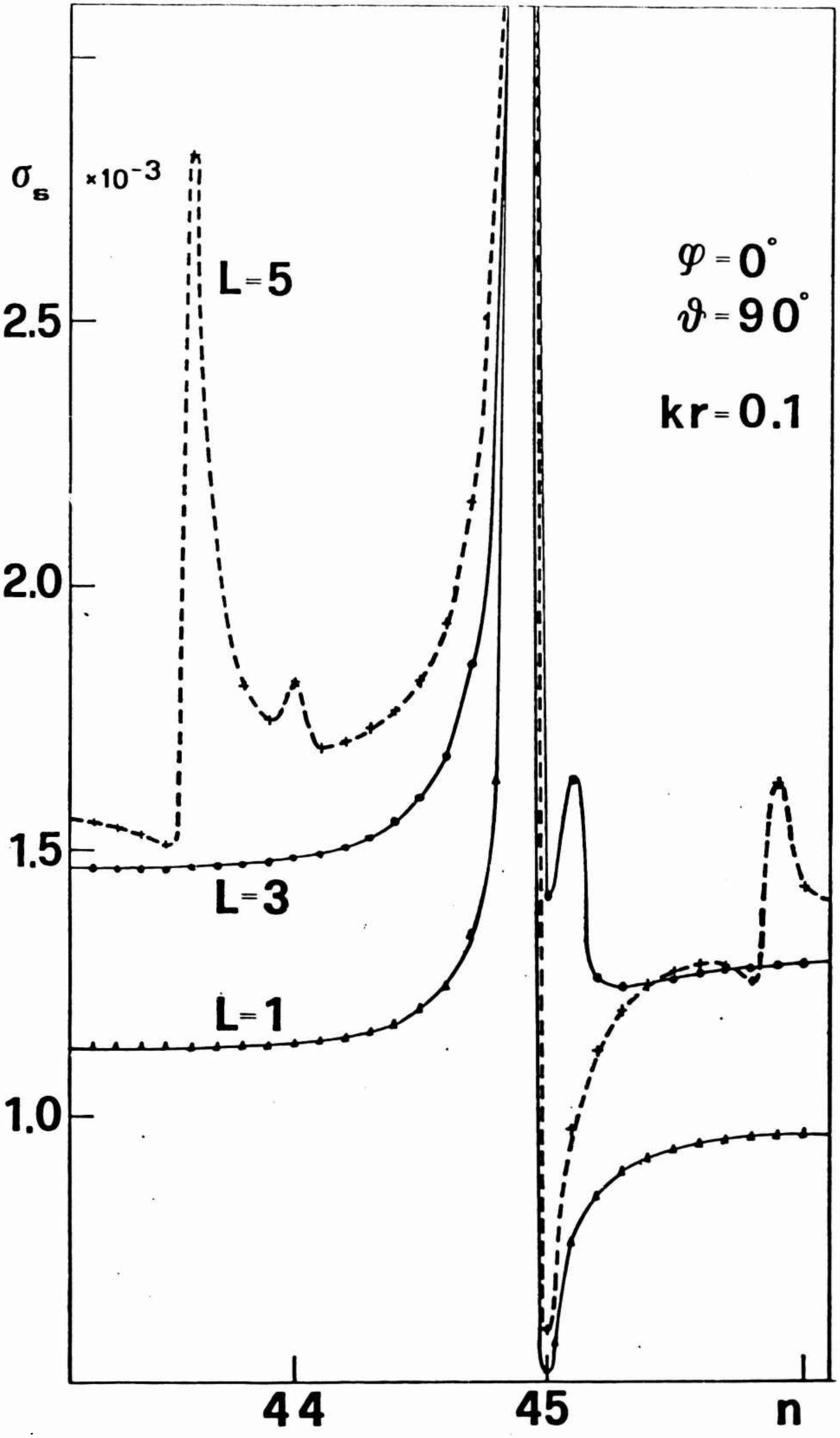
$\vartheta = 0°$
Kr = 0.1
L=1
L=5
L=3
44
45
n


σ_s
$\times 10^{-3}$
2.5
2.0
1.5
1.0
L=5
L=3
L=1
$\varphi = 0°$
$\vartheta = 90°$
kr=0.1
44
45
n