T-matrix approach to calculating circular polarization of aggregates made of optically active (chiral) materials and its applications to cometary dust observations

D. W. Mackowski*[,1], L. Kolokolova[2], and W. Sparks[3]

[1] *Department of Mechanical Engineering, Auburn University, Auburn, AL, USA.*

[2] *Department of Astronomy, University of Maryland, College Park, MD, USA.*

[3] *Space Telescope Science Institute, Baltimore, MD, USA.*

Optical activity is a typical property of the biological materials where left-handed aminoacids and right-handed carbohydrates dominate (so called homochirality). Observationally, optically active materials reveal themselves through the circular polarization in the light they scatter. Thus, circular polarization produced by the optically active particles can serve as a biomarker. This and other applications stimulated a development of the T-matrix code presented in this paper. It allows us to calculate the scattering matrix, and, specifically circular polarization, of the light scattered by aggregated optically active particles. The code can be used for modeling the light scattering by biological objects (e.g., colonies of bacteria, blood cells) and for interpretation of the circular polarization produced by the cosmic dust that contains (pre)biological organic molecules, e.g., comet dust or planetary aerosols.

INTRODUCTION

Many complex organic molecules exist in two forms that are identical except that they pose chirality, i.e., are mirror images of each other. A unique characteristic of life is the homochirality of biological molecules, i.e., predominance of one of the mirror forms of the organic molecules. This characteristic may be manifested on a macroscopic scale through the optical activity of the chiral molecules and, hence, the presence of circular polarization (CP) in the light they scatter. Recently a unique set of data on circular polarization in comets has been accumulated [1]. Characteristics of the cometary CP, specifically a domination of the left-handed polarization in all observed comets, favor the idea that this is evidence of homochiral organics in comet dust similar to that found in meteorites. We have also explored remote-sensing capabilities of circular polarization in the laboratory, studying light scattering from astrobiologically relevant micro-organisms and setting these in the context of abiotic minerals [2]. We have found a dependence of the CP on the dichroism of the materials that results in greater circular polarization in absorption bands.

Theoretical and computational tools are needed to confirm whether the presence of chiral organics can produce the observed characteristics of comet circular polarization and explain the results of our laboratory measurements. A well-known solution exists for an isolated, optically active sphere [3], yet a single sphere model for complex comet dust particles or colonies of bacteria is both unrealistic and incapable of explaining recent results.

Comet dust is known to possess an aggregated structure, and such a structure is also plausible for a variety of biological particles. To better account for the non–spherical na-

*Corresponding author: Daniel Mackowski (mackodw@auburn.edu)

ture of such particles, we have developed a T-matrix code to predict light scattering and absorption by aggregates of optically active spheres. We anticipate that this code will be applicable to a variety of astrobiological problems, including the search for materials containing molecules of prebiological and biological origin in comets, planets, extrasolar planets, and protoplanetary nebulae as well as for studying biological particles in the Earth atmosphere and in the laboratory.

FORMULATION OF THE T-MATRIX CODE FOR OPTICALLY ACTIVE MULTIPLE SPHERES

The existing T-matrix code to calculate light scattering by aggregates assumes that the monomer particles are optically isotropic spheres [4]. However, it is relatively simple to extend the formulation to include aggregates of optically active spheres.

As is the case for isotropic spheres, the electric fields incident on, and scattered by, an optically active sphere can be represented by expansions of regular and outgoing vector wave harmonics (VWH), respectively, which appear as

$$\mathbf{E}_{inc}(\mathbf{r}) = \sum_{n=1} \sum_{m=-n}^{n} \sum_{p=1}^{2} f_{mnp}\, \mathbf{N}^{(1)}_{mnp}(\mathrm{k}\,\mathbf{r}), \tag{1}$$

$$\mathbf{E}_{sca}(\mathbf{r}) = \sum_{n=1} \sum_{m=-n}^{n} \sum_{p=1}^{2} a_{mnp}\, \mathbf{N}^{(3)}_{mnp}(\mathrm{k}\,\mathbf{r}), \tag{2}$$

in which f and a denote the incident-field and scattered-field expansion coefficients, and (m, n, p) denote the degree, order, and mode (TM or TE) of the harmonic. Unlike the isotropic case, however, the optically active sphere results in a coupling of TE and TM modes between the incident and scattered field coefficients for a given harmonic order [3]. That is, the Mie relation for the active sphere appears as

$$a_{mnp} = \sum_{q=1}^{2} \overline{a}_{n;\,pq}\, f_{mnq}, \tag{3}$$

in which the coefficients $\overline{a}$ will be functions of the sphere size parameter ka and the left and right refractive indices.

The modified Mie relation for the active sphere can be incorporated directly into the formulation for a cluster of N_p spheres, and results in the following interaction equations for the sphere scattering coefficients,

$$a^{i}_{mnp} - \sum_{p'=1}^{2} \overline{a}^{i}_{n;pp'} \sum_{\substack{j=1\\ j\neq i}}^{N_p} \sum_{l=1}^{L_j} \sum_{k=-l}^{l} \sum_{q=1}^{2} H^{i-j}_{mnp'\,klq}\, a^{j}_{klq} = \sum_{p'=1}^{2} \overline{a}^{i}_{n;pp'}\, f^{i}_{mnp'}. \tag{4}$$

Above, H^{i-j} is an outgoing harmonic translation matrix, and depends solely on the distance and direction between the origins i and j. Following the procedures developed in [4], a T matrix for the cluster of active spheres can be obtained from the solution of the interaction equations, and the orientation–averaged scattering-matrix elements can be analytically determined from operations on the T matrix.

TESTS AND APPLICATIONS OF THE T-MATRIX CODE FOR OPTICALLY ACTIVE AGGREGATES

A number of consistency checks have been applied to test the veracity of the solution for clusters of optically active spheres. The solution identically satisfy energy conservation (extinction = absorption + scattering) and conforms to equivalent rotations of the cluster and the incident field. In addition, the solution provides a detailed prediction of the electric field in the near-field zones – both within and exterior to the spheres – and this can be used to check the continuity conditions at the surfaces of the spheres. Also, for a single sphere, the solution reduces to the solution from [3] and for isotropic material it provides the standard multi-sphere case from [4].

The results of illustrative test calculations are presented in Fig. 1. Shown are values of the circular polarization ratio S_{14}/S_{11}, as a function of scattering angle, for random–oriented aggregates. For these calculations we set the radius of the monomers to 0.1 µm, which is typical of cometary particles [5], and the incident wavelength is $\lambda = 0.6$ µm which corresponds to the comet red filter. Values of refractive index correspond to $\mathsf{m}_{\mathrm{R}} = 1.55 + 0.0006004\mathsf{i}$ and $\mathsf{m}_{\mathrm{L}} = 1.5500338 + 0.0006000\mathsf{i}$ (R and L refering to left-handed and right-handed, respectively), which are typical for the amino-acids discovered in the Murchison meteorite [6]. According to [7], the specific rotation angle for them is about $100°$, which corresponds to a difference in refractive index for left-handed and right-handed circular polarization of 3×10^{-6}. In our calculations, 10 % of the material is assumed to be optically active.

Two models of aggregates are used in the calculations of Fig. 1, being a fractal–like aggregate with $D_f = 1.8$ and $k_0 = 2.2$ that is characteristic of aggregates formed from cluster–cluster diffusion processes, and clusters of spheres packed randomly and uniformly into a spherical boundary with a volume fraction of 0.5. This latter form would correspond to aggregates formed from evaporation of the liquid phase from droplets of liquid–solid suspensions. Aggregates were generated using a Monte-Carlo method, and the curves represent averages of 20 different realizations of each aggregate for a fixed sphere number N_p. To identically cancel out effects due to random L and R rotation in the aggregate structures – and thereby focus entirely on circular polarization due to optical activity – half of these realizations were mirror images of the other half. Also shown in both plots are the polarization ratios for the single monomer sphere.

The calculation results shown in Fig. 1 are not intended to fit any observational or laboratory data. However, we do see in them some encouraging outcomes. Specifically, the angular change of CP for fractal aggregates (Fig. 1, left) is similar to that observed in comets [1], namely within the range of phase angles $20-120°$ (scattering angles $160-60°$) CP increases almost linearly. Though the values of CP are approximately one order of magnitude smaller than the observed ones (there CP reached 0.4 % at the scattering angle $60°$), at all angles, except the forward scattering area, the results show increase in the CP with the increase of the number of monomers. We expect that for larger aggregates we may reach higher values of CP. The current models of the cometary dust require domination of particles of thousands of monomers [8]. For such large aggregates, the CP may be significantly larger.

The results for the packed sphere clusters (Fig. 1, right) also show a general increase of CP with phase angle and with number of monomers. However, the picture is complicated by the resonant structure of the plots. The reason the packed sphere results show the resonance

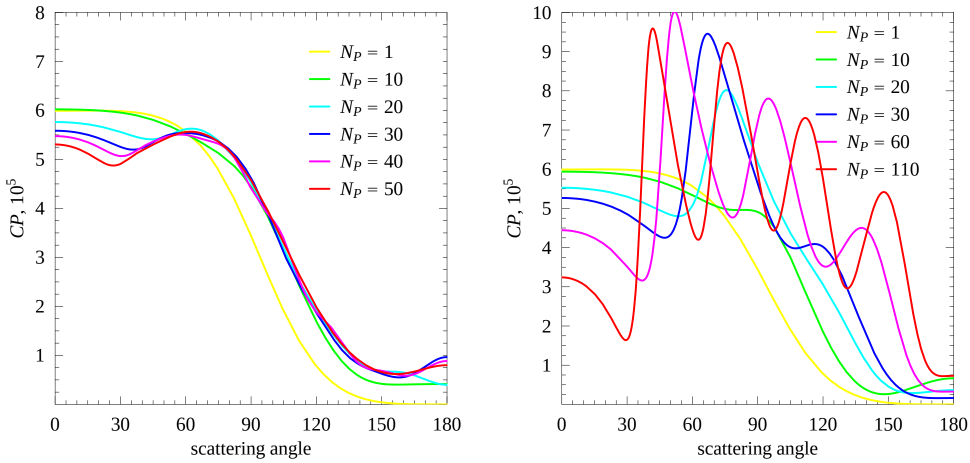

Figure 1. Circular polarization ratios for aggregates of active spheres. Fractal like aggregate (left), and packed–sphere clusters (right).

behavior is because the monomers are forced to fit inside a perfect sphere. This effect would wash out if the enclosing boundary was made random, or when averages were made over different N_p.

One more interesting outcome of our computations is a strong dependence of the CP on the structure of aggregates. It gives us a hope that the new T-matrix code can become a tool to study the structure of cosmic dust particles and planetary aerosols as well as complex biological objects such as cells or microorganisms.

REFERENCES

[1] V. Rosenbush, L. Kolokolova, A. Lazarian, N. Shakhovskoy, and N. Kiselev. Circular polarization in comets: Observations of Comet C/1999 S4 (LINEAR) and tentative interpretation. Icarus **186** (2007).

[2] W.B. Sparks, J.H. Hough, L. Kolokolova, T.A. Germer, F. Chen, S. DasSarma, P. DasSarma, F.T. Robb, N. Manset, I.N. Reid, F.D. Macchetto, and W. Martin. Circular polarization in scattered light as a possible biomarker. JQSRT **110** (2009).

[3] C.F. Bohren. Scattering of electromagnetic waves by an optically active spherical shell. J. Chem. Phys. **62** (1975).

[4] D.W. Mackowski and M.I. Mishchenko, Calculation of the T matrix and the scattering matrix for ensembles of spheres. J. Opt. Soc. Amer. A **13** (1996).

[5] H. Kimura, L. Kolokolova, and I. Mann. Optical properties of cometary dust. Constraints from numerical studies on light scattering by aggregate particles. Astron. Astrophys. **407** (2003).

[6] J.R. Cronin and S. Pizzarello. Amino acids in meteorites. Adv. in Space Research **3** (1983).

[7] S.F. Mason. *Molecular Optical Activity and the Chiral Discriminations*. Cambridge University Press (1982).

[8] L. Kolokolova, H. Kimura, N. Kiselev, and V. Rosenbush. Two different evolutionary types of comets proved by polarimetric and infrared properties of their dust. Astron. Astrophys. **463** (2007).