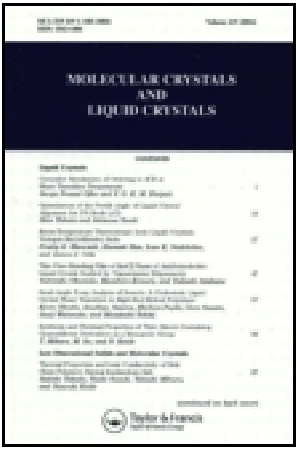

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Polymer-Dispersed Liquid Crystal Droplets: Calculations of Light Scattering

Valery A. Loiko [a] & Vladimir I. Molochko [a]
[a] Stepanov Institute of Physicsn National Academy of Sciences of Belarus 68, F. Skaryna Ave., Minsk, 220072, Belarus

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Polymer-Dispersed Liquid Crystal Droplets: Calculations of Light Scattering

VALERY A. LOIKO and VLADIMIR I. MOLOCHKO

Stepanov Institute of Physics, National Academy of Sciences of Belarus 68, F. Skaryna Ave., Minsk, 220072, Belarus

The extinction efficiency factor and the angular distribution of radiation of polymer-dispersed liquid crystal (PDLC) droplets have been calculated by the discrete-dipole approximation (DDA) and compared with the data obtained by the commonly used Rayleigh-Gans approximation (RGA) and by the anomalous-diffraction approximation (ADA). The errors of calculations by the above-mentioned methods for droplets with homogeneous and droplets with radial director configuration are considered.

Keywords: nematic droplet; light scattering

Introduction

The LC droplet can be considered as an optically «soft» object in the transparency mode and in the scattering mode. Therefore, to describe the optical properties of drop-lets, such well-known methods as the Rayleigh-Gans approximation (RGA) and the anomalous diffraction approximation (ADA)[1] are used traditionally. For small optically «soft» scatterers with internal material anisotropy the RGA is considered in[2-4]. The work[5] considers the ADA for large optically «soft» anisotropic droplets. Based on these approximations for spherical droplets and a number of simple configurations of the director in the droplet analytical formulas have been obtained.

It is impossible to describe analytically configurations that are more complicated. Here, numerical calculations are needed. Recently a method has been developed for calculating characteristics of complex particles based on the discrete-dipole approximation (DDA)[6,7]. It consists in presenting particles in the form of a system of N dipoles, which permits, at an appropriate breakdown, obtaining a result fairly close to the exact result for scatterers of practically any configuration and form. At the same time an essential disadvantage of this method is the large amount of computer resources needed for the calculation of both complex and simple particles as well as a rapid growth of such resources with increasing size of scatterers. Therefore, this method can only be used for not very large scatterers.

Approximate methods of description have such an undoubted advantage as relative simplicity and the calculation rate. At the same time, they give some systematic error which is not always small and which can only be controlled beyond the method. The size of LC droplets R is comparable with the radiation wavelength λ. It includes droplets where it is able to use RGA (submicrometer-size droplets) and ADA (supramicrometer-size droplets). The size parameter $x \equiv 2\pi R/\lambda$ for light in vacuum lies in a fairly wide range, from 0.5 to 50 as a rule. Therefore the question arises of the possibility and exactness of using both the RGA and the ADA for the intermediate range of droplet sizes as well as of the value of the systematic error of these methods and of the influence of the scatterer material anisotropy on the exactness of approximate description. In most works this question is resolved on the basis of interpretation of known conditions (for the RGA $2kR|n-1| << 1, n \approx 1$, for the ADA $kR >> 1, n \approx 1$[1], k is the wave number in vacuum, R is the droplet radius, n is the refractive index of the droplet). Note that for the above range of sizes of scatterers, this interpretation is wordiness and more detailed refinements of both the question of applicability of these approximations and the question of exactness are needed.

Basic equations

Let us consider an LC scatterer with radial configuration placed in a polymer with refractive index n_m, in the light wave field $\vec{E}^{inc} = \vec{e}_0 E_0 \exp(i\vec{k}_0\vec{r})$, $\vec{e}_0$ is the unit vector of polarization. Let us denote the wave vector in the direction of wave scattering by $\vec{k}$, $\vec{k}_s \equiv \vec{k} - \vec{k}_0$. We write the scattered field as usual:

$$\begin{pmatrix} E_\parallel \\ E_\perp \end{pmatrix} = \frac{\exp(ikr)}{-ikr}\begin{pmatrix} S_2 & S_3 \\ S_4 & S_1 \end{pmatrix}\begin{pmatrix} E^{inc}{}_\parallel \\ E^{inc}{}_\perp \end{pmatrix}, \tag{1}$$

where $E_\perp, E_\parallel$ is the vector $\vec{E}$ component perpendicular and parallel to the scattering plane, respectively. We shall use the relative refractive index thus assuming $n_m = 1$. Accordingly, further wave vector $k \equiv 2\pi/\lambda$, size parameter $x \equiv 2\pi R\ \lambda$, where λ is the wavelength in the polymer matrix.

Only for a small and sufficiently «soft» scatterer ($2kR|n-1| << 1, n \approx 1$, here n is the effective refractive index, R is the droplet radius) in the RGA we have[2]:

$$\hat{S} = \left(-ik^3/4\pi\right)\int\left(\hat{\varepsilon} - \hat{1}\right)\exp\left(-i\vec{k}_s\vec{r}\right)d^3\vec{r}\,. \tag{2}$$

For the scattering efficiency factor of axially symmetrical scatterer we have:

$$Q_{sca} = (\pi/k)^2\int(|S_1|^2 + |S_2|^2)\sin\delta d\delta\,, \tag{3}$$

δ is the polar angle of scattering.

In the anomalous diffraction approximation is assumed $kR >> 1, n \approx 1$[1]. Then, in accordance with[5], the scattering matrix $\hat{S}$ will be written in the form:

$$\hat{S} = (2\pi i / \lambda^2) \int \left(\hat{p}(\vec{r}) - \hat{1}\right) \exp\left(-i\vec{k}\vec{r}\right) d^2\vec{r}\,, \tag{4}$$

where the tensor $\hat{p}(\vec{r})$ describes the phase shift and the rotation of the polarization plane of the beam passing through the point $\vec{r}$ of the scatterer projection on the plane perpendicular to the direction of wave propagation. The expressions for $\hat{p}(\vec{r})$ and the results of integration in (4) for a number of director configurations were obtained in[5].

The discrete-dipole approximation is based on the scatterer representation in the form of an array of discrete dipoles with corresponding properties. Each dipole is characterized by its position and polarizability whose choice is a nontrivial task.
In accordance with[7], the problem of finding the polarization of elementary dipoles is reduced to the solution of the following system of linear equations:

$$\vec{P}_j = \hat{\alpha}_j \left[\vec{E}_j^{\,inc} + \sum_{k \neq j} \hat{A}_{jk} \vec{P}_k \right]. \tag{5}$$

Here $\hat{\alpha}_j$ is the j-th dipole polarizability (see[6,7] for expressions for $\hat{\alpha}_j$), the term $\hat{A}_{jk}\vec{P}_k$ represents the field induced by the k-th dipole with polarization $\vec{P}_k$ on the j-th dipole.

As a rule, the solution of the system of equations (5) is reduced to iterations whose physical meaning consists in taking into account greater and greater multiplicity of scattering from the system of N elementary dipoles. Proceeding from the known values of polarization on each elementary dipole one can write expression for the matrix $\hat{S}$ [7].

For the j-th dipole polarizability we have

$$\hat{\alpha}_j = \hat{M}_j^{-1} \hat{\alpha} \hat{M}_j\,, \tag{6}$$

where $\hat{\alpha}$ - is the diagonal tensor of polarizability used in[6,7].

$$\hat{M}_j = \hat{R}_x(\alpha_j) \hat{R}_y(\beta_j) \hat{R}_z(\gamma_j), \tag{7}$$

$\alpha_j, \beta_j, \gamma_j$ - are angles of the j-th dipole rotation about the axes x,y,z, respectively; $\hat{R}_x(\alpha), \hat{R}_y(\beta), \hat{R}_z(\gamma)$ are matrices of rotation about the axes.
For the radial director configuration for these angles we have used

$$\alpha_j = \arctan(z_j, y_j), \beta_j = 0, \gamma_j = \arccos\left(x_j / \sqrt{x_j^2 + y_j^2 + z_j^2}\right). \tag{8}$$

where $\arctan(s,c)=t$ if $s=\sin(t), c=\cos(t)$. The tensor $\hat{\alpha}$ was found in accordance with[6,7] for permittivity $\hat{\varepsilon}=diag(\varepsilon_e,\varepsilon_0,\varepsilon_0)$.

For the scattering efficiency factor equal to the extinction efficiency factor in the case of nonabsorbing scatterers we have[7]

$$Q_{sca}=(4/(xE_0))^2\sum_{j=1}^{N}\mathrm{Im}\left(k^3\vec{P}_j\vec{E}_j^{inc}\right). \tag{9}$$

As the quantity characterizing exactness when comparing the two methods of calculation in next Sections, we have chosen the relative error: for the factor of scattering efficiency Q^{ref} -the quantity

$$\Delta_Q\equiv\left(Q^{ref}-Q\right)/Q. \tag{10}$$

As the function characterizing the angular structure of radiation for axially symmetrical scatterer, we have chosen the dimensionless value I :

$$I\equiv(|S_1|^2+|S_2|^2)/2. \tag{11}$$

This dimensionless value is related to the differential scattering cross-section for natural light by the relation $I=k^2d\sigma/d\Omega$, k is the wave number in vacuum. Then the calculated value I^{ref} is characterized by the error

$$\Delta_I\equiv\left(I^{ref}-I\right)/I. \tag{12}$$

By Q and I are meant the values obtained as more exact ones (in Section "Homogeneous scatterer " -- from the calculation according to the Mie theory, in Section "Liquid-crystal droplet " -- in the DDA).

Homogeneous scatterer

Fig. 1 gives some results of comparing the DDA and the approximate methods RGA, ADA with exact calculations (according to the Mie theory) for spherical homogeneous scatterers. We chose the refractive index $n=1.11$ of the homogeneous droplet as an illustration for relatively "hard" LC droplets.

As can be seen, for the scattering efficiency factor Q_{sca} depending on the droplet size the accuracy of calculations by the DDA method is fairly high within the scope of applicability of the chosen breakdown[7] (at a break-down of $29\times29\times29$ approximately up to $x=24$ with an accuracy better than 5% and up to $x=11$ with an accuracy better than 1%); finer break-down permits describing larger scatterers (at a break-down of $37\times37\times37$ approximately up to $x=34$ with an accuracy better than 5% and up to $x=22$ with an accuracy better than 1%). We have not noticed any possible deviations from this regularity associated with the accumulation of errors with increasing number of both elementary dipoles and iterations for finding the distribution of dipole polarization due to multiple scattering in the calculations (($n \le 1.2$, break-down of up to $40 \times 40 \times 40$ dipoles) for Q_{sca}.

Our comparisons (not presented here) of the calculations for the angular structure of scattering of «soft» homogeneous droplets show that RGA and DDA are not exact enough to describe the angular structure of scattering of such medium-sized droplets. For $x = 5$, $n = 1.05$ the RGA permits describing with an accuracy of 5% the angular distribution up to angles of scattering less than 30°. The ADA gives the result with an error of 10% up to angles of about 20°. For the break-down $29 \times 29 \times 29$ the ADA describes, with an accuracy better than 5%, the structure of radiation up to angles of about 90°, and up to 160° with an accuracy of 10%.

Our comparisons for homogeneous scatterers show that the DDA has a considerably better accuracy than the above-mentioned approximations in the range of parameters characteristic of LC droplets. The exception is the region of scattering by large angles where the description accuracy is limited. From the point of view of the DDA, there is no essential difference between the errors of the system of isotropic and anisotropic elementary scatterers forming the droplet. Therefore, the DDA can be used to estimate the RGA and ADA errors for anisotropic LC droplets. On this basis next section considers the question of exact description of a spherical scatterer with a radial director configuration by the approximations RGA and ADA with respect to the DDA.

Liquid-crystal droplet

Figure 2-5 give the results of the error calculations of the scattering efficiency factor and angular structure of radiation for liquid-crystal spherical droplets with a radial structure of the director. The extraordinary refractive index of the liquid crystal in the droplet was chosen in the range from a «soft» $n_e = 1.08$ to a «hard» droplet $n_e = 1.2$. The ordinary refractive index took on the value $n_0 = 0.97$.

Figure 2 shows plots of the error of calculating the scattering efficiency factor for four values of the extraordinary refractive index of the droplet $n_e = 1.08, 1.11, 1.15, 1.2$. As can be seen from these plots, the situation with the ADA error, as a whole, is similar to that which can be expected proceeding from the analysis of the ADA error for homogeneous scatterers. As with the homogeneous scatterer, in the range of changes in the refractive index from the scatterer size about $x = 10$, the anomalous diffraction approximation can be used to describe the LC droplet with a moderate error (which in our case does not exceed 15%). Note that within the limits of the parameters considered by us the position of this range weakly depends on the refractive index. The ADA can be used, with a model accuracy of 25%, even from the size of the droplet $x = 5$ for both Q and the angular structure of scattering (see Figure 4). Comparison of the error plots for Q with those for homogeneous scatterers shows that the behavior of the radial-structure droplet for the ADA is in general the same as that of the considerably «harder» scatterer than the homogeneous one with the refractive index $n_{eff} = (2n_0 + n_e)/3$. So the error plot for the homogeneous scatterer in Figure 1 (refractive index $n = 1.11$) as analogous to the plot with $n_e = 1.2$ for the droplet with radial configuration. As in the case of homogeneous scatterers, the maximum angle to which the approximation ADA can describe the angular structure of radiation does not exceed 30°, and this angle slightly depends on the scatterer parameters.

Such a picture of changes in the ADA error for LC droplets can be explained by the existence of changes in the refractive index on the path of the beam inside the LC droplet.. This leads to the worsening of the conditions for the ADA applicability as compared to the homogeneous scatterer. At the same time for the radial configuration these changes are not as great as for some other configurations of the director realized in the LC droplet. Therefore, more significant error of ADA descriptions is possible.

The transfer from isotropic to anisotropic droplets (see, for example, Figures 2,3) for the RGA, unlike the ADA, significantly affects the accuracy of this approximation. Although the position of the error minimum for them is shifted toward large scatterers, as with «softer» droplets, its value increases significantly compared to homogeneous scatterers, as with «harder» droplets. So the error value for droplets with radial structure exceeds already 10% even for the «softest» ($n_e = 1.08$) particles, although with the homogeneous scatterer with the refractive index $n = 1.11$ the error is no more than 2%. This points to the growth of the influence of scattering multiplicities higher than the first one on the properties of LC droplets as compared

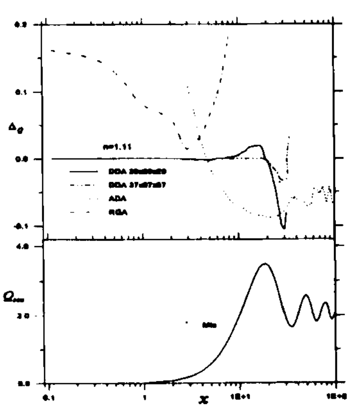

FIGURE 1. Q_{sca} (Mie theory) as a function of size parameter x for a homogeneous sphere with $n = 1.11$ and the error of its calculation Δ_Q for the DDA, RGA, and ADA.

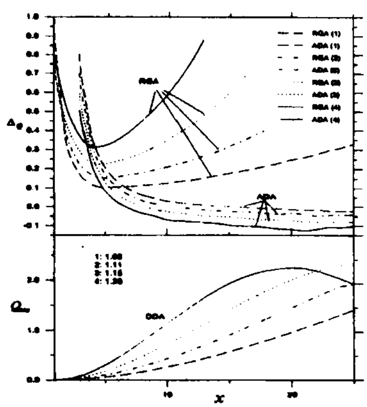

FIGURE 2. Q_{sca} (DDA) as a function of x for a spherical LC droplet with radial director configuration n_0 □ , $n_e = 1.08(1), 1.11(2), 1.15(3), 1.20(4)$ and Δ_Q for the RGA and ADA.

to homogeneous scatterers in the range of sizes characteristic of the RGA. Note the noncharacteristic of homogeneous scatterers high maximum on the plot of the quantity I as a function of angle (Figures 3,4) at $\delta \approx 50^\circ$. For $n_e = 1.08$ and $n_e = 1.11$ its value exceeds the value of I at $\delta \approx 0^\circ$. Such a dependence of the angular structure for homogeneous droplets is absent and the appearance of a high maximum at $\delta \approx 50^\circ$ is due to the peculiarities of scattering from the considered type of inhomogeneous droplets. Such a picture of scattering by the droplet with radial configuration was considered in[5] and attributed to the analogy of the scattering from the radial-configuration droplet with the scattering from an annular screen.

Conclusion

The results presented in this paper permit, in general, answering the question about peculiarities of applicability of the referred approximations for LC droplets.

As can be seen from these results, the DDA can describe the scattering efficiency factor fairly exactly, including that portion of the range of size parameters where neither the RGA nor the ADA give a sufficiently accurate result. As would be expected, the DDA has an apparent advantage from the viewpoint of exactness of the description. At the same time, for the DDA itself for large angles of scattering and large sizes of droplets certain complications of computing are observed.

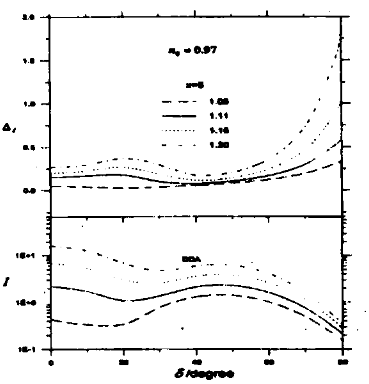

FIGURE 3. I (DDA) as a function of δ for a spherical LC droplet with radial director configuration ($x = 5$, n_0, n_e are as in Fig.2) and the error of its calculation Δ_I, for the RGA.

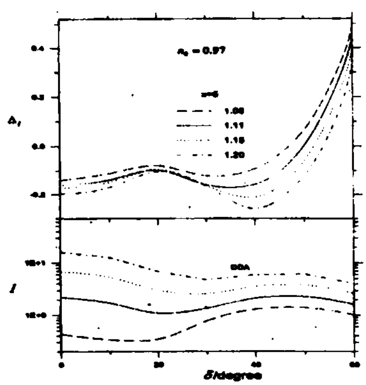

FIGURE 4. I (DDA) as a function of δ for a spherical LC droplet with radial director configuration ($x = 5$, n_0, n_e are as in Fig.2) and the error of its calculation Δ_I, for the ADA.

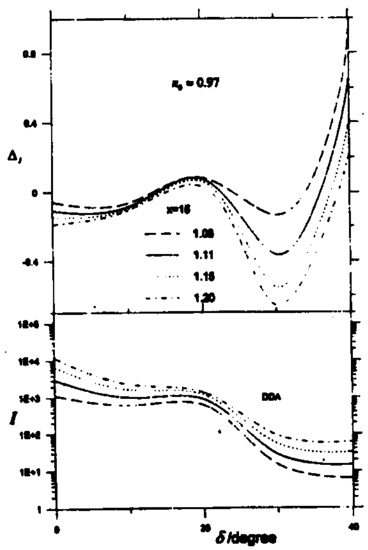

FIGURE 5. I (DDA) as a function of δ for a spherical LC droplet with radial director configuration ($x = 5$, n_0, n_e are as in Fig.2) and the error of its calculation Δ_I, for the ADA.

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