WENTZEL-KRAMERS-BRILLOUIN (WKB) METHOD AS PRINCIPAL APPROXIMATION FOR DESCRIPTION OF LIGHT SCATTERING BY "SOFT" PARTICLES

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It is proved that the Rayleigh, Rayleigh–Hans–Debye, anomalous diffraction, and Fraunhofer diffraction approximations are the corollaries of the integral wave equation in the WKB approximation. The homogeneous sphere is considered as an example.

Approximation methods for solving the light scattering problems are widely applied in optics of colloids, hydrosol and biological suspended particles simpler. They are based on using the well-known physical mechanisms and are simpler for analysis than the exact solution. However, their disadvantage is the limited domain of their correct applicability.

In this connection, it is necessary to develop and apply the methods that combine the simplicity of solution and the possibility of using them in wider optical ranges.

This paper is aimed at the study of the wave equation in the WKB approximation in the areas of legality of the most well-known approximations for "soft" particles (Rayleigh, Rayleigh–Hans–Debye (RHD), anomalous diffraction (AD) and Fraunhofer diffraction (FD) approximations).

Let us consider the integral representation of the scattering amplitude as one of the tools of obtaining the approximate solutions.[1–6]

Using the Hertz vector properties,[2] one can obtain the expression for the field scattered by a particle in the far zone:

$$\mathbf{E}^s(\mathbf{r}) = \mathbf{f}(\mathbf{o},\, \mathbf{i})(e^{ikR}/R); \tag{1}$$

$$\mathbf{f}(\mathbf{o},\mathbf{i}) = \frac{k^2}{4\pi}\int\limits_V -\{\mathbf{o}\times[\mathbf{o}\times\mathbf{E}(\mathbf{r}')]\}[m^2(\mathbf{r}')-1]\exp(-ik\mathbf{r}'\mathbf{o})\mathrm{d}V', \tag{2}$$

where $kR \gg 1$, $k = 2\pi/\lambda$ is the wave number of the disperse medium; R is the distance from the point of observation to the particle along the scattering direction; $\mathbf{i}$ and $\mathbf{o}$ are the unit vectors of the direction of propagation of incident and scattered radiation, respectively; m is the relative refractive index; $\mathbf{E}(\mathbf{r}')$ is the time-independent component of the electric field inside the particle.

The relationship (2) is the exact integral expression for the scattering amplitude in terms of the field $\mathbf{E}(\mathbf{r}')$ inside the particle. In the general case $\mathbf{E}(\mathbf{r}')$ is not known and does not give the closed description for $\mathbf{f}(\mathbf{o},\, \mathbf{i})$. However, based on physical ideas, one often can approximately replace $\mathbf{E}(\mathbf{r}')$ by a known function and thus obtain a useful approximate solution.

Let us consider the WKB approximation, for which $\mathbf{E}(\mathbf{r}')$ inside the particle is approximated by the propagating wave with the wave vector corresponding to the particulate matter. It is also supposed that the direction and amplitude of the wave do not change when passing the scatterer.

Taking into account the aforementioned,

$$\mathbf{E}(\mathbf{r}') = \mathbf{e}_i\exp\Big\{ik\mathbf{r}_1\cdot\mathbf{i} + ik\int\limits_{Z_1}^{Z'} m(z')\mathrm{d}z'\Big\}, \tag{3}$$

where $\mathbf{e}_i$ is the vector of polarization, $Z_1 = (\mathbf{r}_1\cdot\mathbf{i})$ is the input coordinate of the particle surface for the wave passing through the point $\mathbf{r}_1$, $Z' = (\mathbf{r}'\cdot\mathbf{i})$.

Substitution of Eq. (3) into Eq. (1) with small redesignations and regrouping gives

$$\mathbf{f}(\mathbf{o},\, \mathbf{i}) = (k^2/4\pi)\{-\,\mathbf{o}\times[\mathbf{o}\times\mathbf{e}_i]\}VF(\mathbf{o},\, \mathbf{i}), \tag{4}$$

$$F(\mathbf{o},\mathbf{i}) = \frac{1}{V}\int\limits_V [m^2-1]\exp(i\mathbf{k}_s\mathbf{r}')\exp\Big\{ik\int\limits_{Z_1}^{Z'}(m-1)\mathrm{d}z'\Big\}\mathrm{d}V',$$

where $\mathbf{k}_s = k\cdot\mathbf{i}_s = k(\mathbf{i} - \mathbf{o})$ and is directed along the bisector of the complementary scattering angle; $|\mathbf{i}_s| = 2\sin(\theta/2)$; θ is the scattering angle, or the angle between $\mathbf{i}$ and $\mathbf{o}$.

When

$$|m-1| \ll 1, \quad |m-1|\rho_{\max} \ll 1, \tag{5}$$

where $\rho_{\max}$ is the maximum diffraction parameter of the scatterer, is satisfied,

$$\exp\Big\{ik\int\limits_{Z_1}^{Z'}(m-1)\mathrm{d}z'\Big\} \mid 1 \tag{6}$$

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and Eq. (4) coincides with that for RHD:

$$F(\mathbf{o},\ \mathbf{i}) = \frac{1}{V}\int\limits_V (m^2 - 1)\exp(i\mathbf{k}_s\mathbf{r}')\mathrm{d}V'. \qquad (7)$$

Thus, the WKB approximation is the generalization of the RHD approximation taking into account the phase shift, i.e. the “prehistory” of the beam coming to the point $\mathbf{r}'$.

Let us note that the physical scheme of constructing the light scattering mechanism in RHD is based on the usual Rayleigh radiation, i.e. each element of the particle is considered as independent Rayleigh scatterer, the dipole. Since the radiation of different elements is coherent, then the waves scattered by them interfere with each other and partially quench each other due to different positions of elements in space. Formula (4) is constructed based on quite different principles (3), giving however the same result as the physical scheme under the condition (5).

Let us also note that the principle of the Fourier transform of internal field, a fragment of which is used in Eq. (4), is also applied for constructing the exact solution.[7]

For the “soft” particles, whose size is much less than the wavelength,

$$\exp(i\mathbf{k}_s\mathbf{r}') \mid 1. \qquad (8)$$

Taking into account Eq. (6), we obtain $\mathbf{E}(\mathbf{r}') = \mathbf{e}_i$. Substitution of Eqs. (6) and (8) into Eq. (4) leads to the results coinciding with the results of Rayleigh approximation[8] obtained by electrostatic methods:

$$|\mathbf{f}| = (k^2/2\pi)|m - 1|V\sin\chi, \qquad (9)$$

where χ is the angle between $\mathbf{e}_i$ and $\mathbf{o}$.

The error of using Eq. (9) in comparison with the electrostatic formulas in the limiting cases of asphericity ($\varepsilon = 0,\infty$) does not exceed 15.5 and 12.7%, respectively, for $m \le 1.14$ and decreases with decreasing m and asphericity of particles.[9]

Using the properties of particulate softness and continuity of $m(\mathbf{r}')$ and making small regrouping, one can write Eq. (14) in another form:

$$F(\mathbf{o},\ \mathbf{i}) = \frac{2}{ikV}\int\limits_S\int\limits_Z \exp[\bar{F}(z)]\exp[i\mathbf{k}_s\mathbf{r}']\mathrm{d}\bar{F}(z)\mathrm{d}S',$$

$$\bar{F}(z) = ik\int\limits_{Z_1}^{Z'} (m - 1)\mathrm{d}z'. \qquad (10)$$

According to the accepted designations and the scheme of calculation

$$\mathbf{k}_s = \mathbf{k}_{s1}\mathbf{x} + \mathbf{k}_{s2}\mathbf{y} + \mathbf{k}_{s3}\mathbf{z}, \qquad (11)$$

where

$$\mathbf{k}_{s1} = -\ k\ \sin\theta\ \cos\phi;$$
$$\mathbf{k}_{s2} = -\ k\ \sin\theta\ \sin\phi;$$
$$\mathbf{k}_{s3} = k\ (1 - \cos\theta) = 2k\ \sin^2(\theta/2). \qquad (12)$$

Let us note that $(\theta,\ \phi)$ forms the scattering plane identical with $(\mathbf{i},\ \mathbf{o})$ about the plane of incidence $(\mathbf{i},\ \mathbf{e}_i)$.

As is seen from Eqs. (11) and (12), for

$$2kz\ \sin^2(\theta/2) \ll 1, \quad \theta \ll 1 \qquad (13)$$
$$\mathbf{k}_s\mathbf{r}' \mid k_{s1}x' + k_{s2}y', \qquad (14)$$

i.e. does not depend on z. Then we obtain from Eq. (10)

$$F(\mathbf{o},\ \mathbf{i}) = \frac{2i}{kV}\int\limits_S \left\{1 - \exp[ik\int\limits_{Z_1}^{Z_2} (m(\mathbf{r}') - 1)\mathrm{d}z']\right\}\exp(i\mathbf{k}_s\mathbf{r}')\mathrm{d}S'. \qquad (15)$$

Here Z_2 is the exit coordinate of the particulate surface for the wave passing through the point with the radius-vector $\mathbf{r}'$.

Formula (15) holds correct accurate to a small error if one replaces Eq. (13) by the less strong condition:

$$2kZ_3\sin^2(\theta/2) < 0.5, \qquad (16)$$

where $Z_3 = \max\{|Z_1|,\ |Z_2|\}$. The scattering angles, within which practically all the scattered energy is contained for large particles,[8] satisfy the condition (16).

Thus, we obtain

$$k_{\mathrm{ext}} = (4\pi/k)\mathrm{Im}\mathbf{f}(\mathbf{i},\ \mathbf{i}')\mathbf{e}_i =$$

$$= 2\mathrm{Re}\int\limits_S \left\{1 - \exp\left[ik\int\limits_{Z_1}^{Z_2} [m(\mathbf{r}') - 1]\mathrm{d}z'\right]\right\}\mathrm{d}S', \qquad (17)$$

$$|\mathbf{f}(\mathbf{o},\mathbf{i})| = \frac{k}{2\pi}\int\limits_S \left\{1 - \exp\left[ik\int\limits_{Z_1}^{Z_2} [m(\mathbf{r}') - 1]\mathrm{d}z'\right]\right\}\exp(i\mathbf{k}_s\mathbf{r}')\mathrm{d}S',$$
$$\theta \ll 1. \qquad (18)$$

Expressions (17) and (18) are identical to that given by the approximation of anomalous diffraction for the scattering cross section and the small-angle scattering amplitude,[8] respectively, that are based on absolutely different scattering mechanisms. Expressions (17) and (18) in AD approximation are derived on the base of using the Huygens principle in the Fresnel interpretation for the particle projection perpendicular to the sounding radiation, taking into account the phase shifts of corresponding beams coming to it.

For large particles, satisfying the condition

$$|m - 1| \ll 1,$$

$$k\int\limits_{z_1}^{z_2} [m(\mathbf{r}') - 1]\mathrm{d}z' = \Psi(x, y) \neq \mathrm{const} \gg 1,$$

$$k\Delta x, \quad k\Delta y \gg 1, \tag{19}$$

for the majority of scatterers of real shapes, the integrals of the second term in Eqs. (17) and (18) are much less than that of the first term, because the second term is the oscillating sign-changing function, whose absolute value does not exceed unity, while the first term is equal to unity.

Thus, taking into account Eq. (19), we obtain for the small-angle approximation

$$|\mathbf{f}(\mathbf{o}, \mathbf{i})| = \frac{k}{2\pi}\int\limits_S \exp(i\mathbf{k}_s\mathbf{r}')\mathrm{d}S' = \frac{k}{2\pi}SG;$$

$$G = \frac{1}{S}\int\limits_S \exp(i\mathbf{k}_s\mathbf{r}')\mathrm{d}S' . \tag{20}$$

Taking into account Eq. (14), the expression (20) is the absolute value of the Fresnel amplitude function and corresponds to the Fraunhofer diffraction.[8]

Let us demonstrate the legality of the noted general conclusions using the light scattering by a homogeneous sphere as a particular example.

The following expression was obtained in Ref. 10 for a sphere in the WKB approximation:

$$|\mathbf{f}(\mathbf{o}, \mathbf{i})| = \sin\chi(k^2/2\pi)(m - 1)|F(\theta)|; \tag{21}$$

$$F(\theta) = \frac{4\pi a^2}{k_3}\int\limits_0^1 J_0(\rho\sin\theta\sqrt{1 - t^2}\sin[\rho(m - \cos\theta)t]\times$$

$$\times \exp(it\Delta/2)t\mathrm{d}t,$$

where $k_3 = k(m - \cos\theta)$; $J_0(x)$ is the Bessel function of the zero order.

The real part of the integral (21) can be reduced to the Sonin integral, that yields

$$\mathrm{Re}[F(\theta)] = (2\pi a^2/k_3)\{(B_1/U_1^3)[\sin(U_1) - U_1\cos(U_1)] -$$

$$- (B_2/U_2^3)[\sin(U_2) - U_2\cos(U_2)]\}, \tag{22}$$

where

$$B_1 = \Delta/2 + \rho(m - \cos\theta); \quad B_2 = \rho(\cos\theta - 1);$$

$$U_1 = \sqrt{\rho^2 \sin^2\theta + B_1^2};$$

$$U_2 = \sqrt{\rho^2 \sin^2\theta + B_2^2}.$$

Obviously, the real part of the light scattering amplitude (21) makes the principal contribution at the small phase shift, i.e. in the RHD domain. Then it follows from Eq. (22)

$$B_1 \approx - B_2, \quad U_1 \approx U_2 = 2\rho\sin(\theta/2), \tag{23}$$

i.e. the light scattering amplitude is equal to

$$|\mathbf{f}(\mathbf{o}, \mathbf{i})| = \sin\chi 2(m-1)\rho^2 a\big[(1/U_2^3)(\sin U_2 - U_2\cos U_2)\big], \tag{24}$$

that corresponds to the RHD light scattering amplitude.

In the case of AD approximation, in the small-angle region ($\theta \ll 1$),

$$B_2 \approx - \rho\, \theta^2/2 \ll 1, \quad B_1 \approx \Delta \tag{25}$$

and the light scattering amplitude has the form:

$$f(\mathbf{o}, \mathbf{i}) = \rho a\Big\{\int\limits_0^1 J_0(\rho \sin\theta\sqrt{1 - t^2}) \sin(\Delta t)t\mathrm{d}t +$$

$$+ i\int\limits_0^1 J_0(\rho \sin\theta\sqrt{1 - t^2})[1 - \cos(\Delta t)]t\mathrm{d}t\Big\}, \tag{26}$$

that corresponds to the AD formula given in Ref. 8, if one takes $t = \sin\tau$.

Let us analyze in more detail the imaginary part of the scattering amplitude for $\Delta \gg 1$. Obviously (see Eqs. (21), (22), and (26)), the real part in this case does not significantly affect the shape of the scattering phase function. Then one can write the integral (21) in the form

$$|F(\theta)| = (2\pi a^2/k_3) \times$$

$$\times \int\limits_0^1 \{\cos(B_2 t) - \cos(B_1 t)\}J_0(\rho\sin\theta\sqrt{1 - t^2})t\mathrm{d}t. \tag{27}$$

If the function $\cos(Bt)$ is expanded into the Bt power series (the terms of the series are derived by means of the first Sonin integral), then we obtain

$$|F(\theta)| = \frac{2\pi a^2}{k_3}\sum_{n=1}^{\infty}(-1)^n\frac{J_{n+1}(z)}{z^{n+1}}\frac{1}{(2n - 1)!!}\{B_2^{2n} - B_1^{2n}\}, \tag{28}$$

where $z = \rho \sin\theta$.

In the small-angle approximation $\theta \ll 1$, the last series reduces to the series suggested in Ref. 8 for AD:

$$|F(\theta)| =$$

$$= \frac{2\pi a^2}{k_3}\left\{\Delta^2\frac{1}{z^2}J_2(z) - \frac{\Delta^4}{1{\cdot}3}\frac{1}{z^3}J_3(z) + \frac{\Delta^6}{1{\cdot}3{\cdot}5}\frac{1}{z^4}J_4(z)\ \ldots\right\}. \tag{29}$$

One can represent the expression (27) in the form of another series, if using the expansion of the Bessel function $J_0(\rho \sin\theta\sqrt{1-t^2})$. Then we obtain

$$|F(\theta)| = \frac{2\pi a^2}{k_3} \frac{\sqrt{\pi}}{2} \sum_{n=0}^{\infty} \frac{(-z^2/4)^n}{n!} \times$$

$$\times\{(B_2/2)^{-n-1/2} H_{n+3/2}(B_2) - (B_1/2)^{-n-1/2} H_{n+3/2}(B_1)\}, \quad (30)$$

where H_ν is the Struve function.[11]

Under the condition $\Delta \gg 1$ and $\rho(1-\cos\theta) \ll 1$ one can reduce the last series to a simple expression

$$|F(\theta)| = \frac{2\pi a^2}{k(m-1)} \left\{ \frac{J_1(z)}{z} + \frac{1}{4} (K(B_1) - 2) \right\}, \quad (31)$$

where

$$K(B_1) \approx K(\Delta) = 2 - \frac{4\sin\Delta}{\Delta} + \frac{4}{\Delta^2} (1 - \cos\Delta).$$

Let us note that $K(\Delta)$ is the factor of extinction efficiency for nonabsorbing particles. In particular, it is seen that the small-angle scattering phase function depends on Δ similarly to the factor of light scattering efficiency.

$K_{ext} = 2$ for $\Delta \to \infty$ and, hence

$$f(\mathbf{o}, \mathbf{i}) = \frac{i\rho a}{2} \left(2 \frac{J_1(\rho \sin\theta)}{\rho \sin\theta} \right), \quad (32)$$

that corresponds to the Fraunhofer diffraction.

In Ref. 8, for the small-angle region the expression was obtained that immediately follows from Eq. (26)

$$\mathrm{Im}[F(\theta)] = \frac{2\pi a^2}{k_3} \left\{ \frac{1}{z} J_1(z) + \frac{\Delta}{U^2} \sqrt{\frac{\pi U}{2}} N_{3/2}(U) + \right.$$

$$\left. + \frac{1}{\Delta^2} J_0(z) + \frac{1\cdot 3}{\Delta^4} z J_1(z) + \ldots \right\}, \quad (33)$$

where $U = \sqrt{\Delta^2 + z^2}$; $N_{3/2}(U)$ is the Neumann function.

Restricting ourselves to the two first terms ($\Delta \gg 1$), we obtain

$$|F(\theta)| = \frac{2\pi a^2}{k_3} \left\{ \frac{1}{z} J_1(z) - \frac{\Delta}{U^2} \left(\sin(U) + \frac{\cos(U)}{U} \right) \right\}, \quad (34)$$

that practically coincides with Eq. (31).

Then it is seen therefrom that the positions of extrema of the scattering phase function (in the $z = \rho \sin\theta$ coordinates) execute damped oscillations about the positions of extrema corresponding to FD, as Δ ($\Delta \gg 1$) increases.

The initial start positions of the extrema are defined by the RHD approximation (in particular, minima are at the points $z = 4.49$; 7.73;...), and the final positions are at the points corresponding to FD (in particular, minima are at $z = 3.83$; 7.01;...). In this case the distance between neighboring minima remains practically constant and equal to π in the coordinates of z.

Thus, it is proved, based on the study performed, that the Rayleigh, RHD, AD, and FD approximations are the corollaries of the wave equation in the WKB approximation. The general scheme for structure formation of the light scattering phase function is constructed in this case on the basis of RHD scheme that undergoes the corresponding linear shift toward the FD scheme with subsequent damped oscillations about it as Δ increases.

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